

Fatigue Crack Growth and Localisation of Strain in Zircaloy-4

Daniel Joseph Long

Royal School of Mines
Department of Materials
Imperial College London

Submitted in part fulfilment of the requirements for the degree of
Doctor of Philosophy
2024

Under the supervision of Prof. Fionn P.E. Dunne and Dr Stella Pedrazzini

Page would be left blank if not for this note.

in memory of william o'malley

Page would be left blank if not for this note.

Abstract

Short fatigue crack growth in engineering alloys is among the most prominent challenges in micromechanics. The multifarious nature of crack growth at microstructure length scales necessitates advanced modelling techniques to predict fatigue life accurately. A key contributor to this is crack path, which is also challenging to predict. To address these issues in the context of Zircaloy-4, this thesis presents (i) a critical appraisal of the stored energy density criterion within a crystal plasticity modelling framework for crack growth rate prediction, (ii) the development of new energy-based criteria for crack path, and (iii) a novel mechanistic analytical model for crack growth rate using a stored energy density fracture criterion. Subsequent chapters address environmental degradation issues and their impact on structural integrity, viz., new models for synergistic coupling of irradiation damage and thermomechanical loads, and measurement of fatigue crack growth and cyclic strain ratcheting effects in microstructures containing hydride precipitates. Stored energy density is shown to accurately capture major microstructure-driven differences in crack growth rate; direct comparisons of simulations with experiments enable estimation of the critical stored energy density for crack growth. This is later used to predict crack propagation rates via the new analytical model, which is shown to accurately capture experimental measurements while offering significant computer processing time reductions compared with state-of-the-art numerical methods. The synergistic coupling of irradiation damage and thermomechanical loads, compared with conventional post-irradiation testing, is shown to lead to localisation of quantities linked with crack nucleation including geometrically necessary dislocations and stress. Lastly, experimental measurements of fatigue cracking through hydrided microstructures reveal new crack propagation mechanisms, which on average, accelerate crack propagation rates. Measurements of static hydride precipitation strains demonstrate a discernible strain field directionality, while cyclic thermomechanical loads are shown to promote hydride strain ratcheting, leading to the development of geometrically necessary dislocation networks.

Statement of Originality

I hereby declare that this thesis contains original research which was completed in the Department of Materials, Imperial College London between October 2020 and December 2023. All work presented in this thesis is my own unless referenced or stated otherwise at the beginning of a chapter.

Copyright Declaration

The copyright of this thesis rests with the author. Unless otherwise indicated, its contents are licensed under a Creative Commons Attribution Non-Commercial 4.0 International Licence (CC BY-NC). Under this licence, you may copy and redistribute the material in any medium or format. You may also create and distribute modified versions of the work. This is on the condition that: you credit the author and do not use it, or any derivative works, for a commercial purpose. When reusing or sharing this work, ensure you make the licence terms clear to others by naming the licence and linking to the licence text. Where a work has been adapted, you should indicate that the work has been changed and describe those changes. Please seek permission from the copyright holder for uses of this work that are not included in this licence or permitted under UK Copyright Law.

Acknowledgements

Firstly, I would like to thank Prof. Fionn Dunne for his supervision, expert mentoring, and for cultivating independent thinking during my time at Imperial College. It is a privilege to have been one of his last PhD students. I would also like to thank my co-supervisor, Dr Stella Pedrazzini, for her assistance with experimental aspects of the project and dedication to sustaining an interactive research environment. For sponsorship of my project, I extend my appreciation to Rolls-Royce plc. and their technical specialists (particularly Salah Muflahi, Robert Marshall, and Michael Rogers), who have helped in many ways to refine my research objectives. Others who have contributed to the evolution of my project are Prof. David Dye, who asked thought-provoking questions during my early-stage assessment, and Dr Ben Britton, whose rigorous explanations of EBSD sample preparation methods were very helpful. My sincere thanks also go to friends and workmates, Yang Liu, Vasileios ‘Bill’ Karamitros, Yilun Xu, Angelo Battistini, Cameron MacDonald, and Ryan Stroud for their willingness to thrash out and exchange ideas at any time. In the lab, David Shepherd, Ben Poole, Di Wang, Cheng-Jung Lin, and Neil MacFadyen have been incredibly helpful.

Beyond focused research, there are many people in my personal life who have supported me along the way. I am particularly grateful for my partner, Amy, who moved to London with me and has since been a source of unwavering encouragement. My parents, Denis and Kathleen and sister, Shannon, have also been very supportive throughout; their phone calls, visits, and warm welcomes home have been a great source of rest. I am also grateful to Éamon, Esther, and old friends from Galway for their words of encouragement.

Publications

The following peer-reviewed journal papers have been published as part of this thesis:

- a. Daniel J. Long, Yang Liu, Chris Hardie, Fionn P.E. Dunne, Synergistic coupling of thermomechanical loading and irradiation damage in Zircaloy-4, Modelling and Simulation in Materials Science and Engineering, Volume 32, 2024, <https://doi.org/10.1088/1361-651X/ad312b>.
- b. Daniel J. Long, Fionn P.E. Dunne, On the mechanistic driving force for short fatigue crack path, Journal of the Mechanics and Physics of Solids, Volume 179, 2023, <https://doi.org/10.1016/j.jmps.2023.105368>.
- c. Daniel J. Long, Yang Liu, Weifeng Wan, Fionn P.E. Dunne, A microstructure-sensitive analytical solution for short fatigue crack growth rate in metallic materials, International Journal of Mechanical Sciences, Volume 253, 2023, <https://doi.org/10.1016/j.ijmecsci.2023.108365>.
- d. Daniel J. Long, Weifeng Wan, Fionn P.E. Dunne, The influence of microstructure on short fatigue crack growth rates in Zircaloy-4: Crystal plasticity modelling and experiment, International Journal of Fatigue, Volume 167, Part A, 2023, <https://doi.org/10.1016/j.ijfatigue.2022.107385>.
- e. Chris Hardie, Daniel J. Long, Eralp Demir, Edmund Tarleton, Fionn P.E. Dunne, A robust and efficient hybrid solver for crystal plasticity, International Journal of Plasticity, Volume 170, 2023, <https://doi.org/10.1016/j.ijplas.2023.103773>.

The following paper, which was also written as part of this thesis, is currently under review:

- a. Daniel J. Long, Thibaut Dessolier, Ben T. Britton, Stella Pedrazzini, Fionn P.E. Dunne, Delta Hydrides: Influence on Localisation and Fatigue Crack Growth under Thermomechanical Load, submitted to International Journal of Plasticity (under review).

Contents

Abstract	5
Statement of Originality	6
Copyright Declaration	6
Acknowledgements	7
Publications	8
Contents.....	9
Nomenclature	15
Glossary	17
1 Introduction	19
1.1 A brief timeline of nuclear power and policy in the United Kingdom	19
1.2 Basic principles of nuclear power.....	21
1.2.1 Overview of nuclear fission.....	21
1.2.2 Harnessing nuclear power	23
1.3 Zirconium alloys in the nuclear industry	24
1.3.1 Introduction	24
1.3.2 Cladding design.....	24
1.3.3 Zirconium alloy development.....	25
1.4 Thesis.....	27
2 Literature Review.....	29
2.1 Driving forces for fatigue crack growth.....	29
2.1.1 Introduction	29
2.1.2 Classical methods.....	29
2.1.3 Fatigue indicator parameters.....	33
2.1.4 Discrete dislocation methods	35
2.1.5 Stored energy density	38

2.2	Irradiation damage.....	41
2.2.1	Irradiation-induced lattice defects	41
2.2.2	Channel clearing phenomenon and influence on mechanical properties.....	42
2.2.3	Irradiation growth.....	44
2.2.4	Models for irradiation damage.....	44
2.3	Hydrides.....	46
2.3.1	Introduction	46
2.3.2	Morphology and microstructure interaction.....	48
2.3.3	Delayed hydride cracking and embrittlement	50
3	Methodology	53
3.1	Introduction	53
3.2	Computational	53
3.2.1	Crystal plasticity-based constitutive model	53
3.2.2	The eXtended Finite Element Method (XFEM).....	56
3.2.3	Finite element implementation of strain gradient plasticity for GND density..	57
3.2.4	The stored energy density fracture criterion.....	59
3.2.5	Criterion for short fatigue crack path: to be continued.....	60
3.2.6	User-material subroutine implementation of the constitutive model	61
3.2.7	Zircaloy-4 material properties.....	62
3.3	Experimental.....	63
3.3.1	Micro-thermo-mechanical testing.....	63
3.3.2	In-situ digital image correlation	71
3.3.3	Microstructure characterisation.....	76
3.3.4	Sample preparation	79
4	The Influence of Microstructure on Short Fatigue Crack Growth Rates and Path	83
4.1	Introduction	83

4.2	Methodology	88
4.2.1	Single crystal simulations	88
4.2.2	Polycrystal simulations	88
4.2.3	Implementation of existing and development of new criteria for crack path	90
4.3	Results	93
4.3.1	A crack growth rate study using the maximum slip criterion for crack path ...	93
4.3.2	A study to evaluate and compare the performance of various crack path drivers 105	
4.4	Discussion	114
4.4.1	Crack growth rates.....	114
4.4.2	Crack paths.....	117
4.5	Conclusions	119
5	An analytical solution for short fatigue crack growth	121
5.1	Introduction	121
5.2	Model development and implementation	124
5.2.1	Development of an analytical model for cyclic stored energy density at a crack tip	124
5.2.2	Analytical solution for crack growth rate	128
5.2.3	Incorporation of dislocation shielding	130
5.2.4	A simplified method for crack path selection and full-field implementation...	133
5.3	Results and discussion.....	135
5.3.1	Case study: crack path prediction and comparison with experiments	135
5.3.2	Case study: simulated crack growth rates and comparison with experiments	137
5.3.3	Influence of crystal orientation and dislocation shielding with application to stage II cracks	140
5.4	Conclusions	143
6	Synergistic Coupling of Thermomechanical Loads and Irradiation Damage.....	145

6.1	Introduction	145
6.2	Methodology	146
6.2.1	Adaptation of existing crystal plasticity formulation for irradiation hardening and softening.....	146
6.2.2	Development of a simplified model for loop density evolution during irradiation	148
6.2.3	Development of a model for dislocation loop annihilation at elevated temperatures	150
6.2.4	Case study: a global cladding model for representative loading conditions	153
6.3	Results and discussion.....	156
6.3.1	Influence of loading on key damage variables.....	156
6.3.2	Dislocation loop density evolution.....	159
6.3.3	Sub-study of transient behaviour at various irradiation dose rates at 293 K .	161
6.4	Conclusions	163
7	Delta Hydrides: Influence on Fatigue Crack Growth and Localisation under Thermomechanical Loads	165
7.1	Introduction	165
7.2	Development of a heat treatment device for hydride redistribution	168
7.2.1	Introduction	168
7.2.2	Design and manufacture of a device for thermal gradient heat treatments	169
7.2.3	Analytical modelling for spatial and temporal hydrogen redistribution	170
7.2.4	Optical assessment of spatial redistribution of hydrides	173
7.3	Results and discussion.....	174
7.3.1	Room temperature short fatigue crack growth	174
7.3.2	Influence of annealing twin features on hydride precipitation and fatigue	181
7.3.3	Hydride ratcheting effects during thermomechanical fatigue	186
7.4	Conclusions	197

8	Conclusions	199
9	Future Work: Irradiation and Fatigue Crack Growth	201
9.1	Introduction	201
9.2	Modelling methodology for irradiation damage and short crack growth	202
10	References.....	205

Page would be left blank if not for this note.

Nomenclature

A	Paris' law constant
$\bar{A}_{\text{loop}}$	Dislocation loop area
a	Crack length
Δa	Crack length extension
a_e	Effective crack length
α	Dislocation loop thermal annihilation constant
B	Modified Paris' law constant
b	Burgers vector magnitude
b'	Basquin exponent
$\mathbf{C}$	Stiffness matrix
C	Hydrogen concentration
c	Coffin-Manson exponent
c_0	Thermal equilibrium concentration of interstitials in bulk
c_1	Interstitial concentration
c_p	Specific heat capacity
c_v	Vacancy concentration
$\mathbf{D}$	Global stiffness matrix
D	Hydrogen diffusion coefficient
E	Elastic modulus
E'	Elastic modulus for plane strain conditions
E_p	Plastic strain energy density
E_s	Stored energy density (volumetric)
e	Error
ε_1	Maximum principal strain
ε_3	Minimum principal strain
ε'_f	Coffin-Manson fracture strain
$\boldsymbol{\varepsilon}^e$	Elastic strain tensor
$\Delta \varepsilon_e$	Elastic strain range
$\Delta \varepsilon_p$	Plastic strain range
$\boldsymbol{\varepsilon}^p$	Plastic strain tensor
$\varepsilon_n^{p,i}$	Normal component of plastic strain for slip plane i
$\boldsymbol{\varepsilon}^{\text{th}}$	Thermal strain
$\boldsymbol{\varepsilon}_{\text{vm}}$	Volumetric misfit strain due to hydride precipitation
η	Irradiation defect efficiency factor
$\mathbf{F}$	Total deformation gradient
$\mathbf{F}^e$	Elastic deformation gradient
$\mathbf{F}^p$	Plastic deformation gradient
ΔF	Activation energy
γ^e	Elastic shear strain
γ^p	Plastic shear strain
$\dot{\gamma}^i$	Rate of plastic shear on i^{th} slip system
$\Delta \gamma_{\text{max}}^p$	Fatemie-Socie maximum plastic shear range
γ_{net}^p	Modified Fatemie-Socie net cumulative plastic shear
γ^{st}	Dislocation density evolution constant
G	Stored energy density

G_0	Crack tip stored energy density after some crack extension
G_c	Critical stored energy density
G_{NL}	Non-local stored energy density
G_{MER}^i	Maximum energy release rate for slip system i
G_{NSED}^i	Normal stored energy density for slip system i
$\mathcal{G}_c$	Griffith critical energy release rate
J	J-integral
$\mathbf{J}$	Jacobian matrix
J_{Fick}	Fick hydrogen flux
J_{Soret}	Soret hydrogen flux
ΔK	Stress intensity factor range
K_I	Mode I stress intensity factor
K_{II}	Mode II stress intensity factor
ΔK^∞	Remote applied stress intensity factor range
ΔK_{Jpc}	J-plasticity-corrected stress intensity factor range
K_d	Derivative gain
K_i	Integral gain
K_p	Proportional gain
K_θ	Orientation dependent stress intensity factor
$K_\theta^\perp$	Shielding stress intensity factor due to a dislocation
k	Fatemie-Socie scaling parameter
k_B	Boltzmann constant
k_{cond}	Thermal conductivity
k_{GB}	Modified Fatemie-Socie scaling parameter
$\mathbf{L}^p$	Plastic velocity gradient
l_d	Irradiation defect spacing
l_s	Slip line length
$\mathbf{\Lambda}$	Nye's dislocation tensor
M	Total number of slip systems
μ	Shear modulus
N	Number of cycles (to crack length or failure)
N_{CI}	Tanaka-Mura number of cycles to crack initiation
$\mathbf{n}^i$	Unit normal vector for plane of slip system i
n	Irradiation defect efficiency
ν	Poisson's ratio
P	Concentrated load
p	Paris' law exponent
Q_D	Energy for hydrogen transport
Q^*	Heat of transport
q	Modified Paris' law exponent
q''_{conv}	Convective heat flux loss
q''_{rad}	Radiation heat flux loss
r_1	Outer measurement radius for maximum energy release rate criterion
r_2	Mesh-sensitive measurement radius for maximum energy release rate criterion
r_{loop}	Dislocation loop radius
r_p	Plastic zone radius
ρ_{GND}	Density of geometrically necessary dislocations

ρ_{loop}	Density of dislocation loops
$\rho_{\text{loop},0}$	Initial density of dislocation loops
ρ_{m}	Density of mobile dislocations
ρ_{SSD}	Density of statistically stored dislocations
ψ	Residual stress (error)
ψ^k	Dislocation loop annihilation probability
φ	Irradiation dose
$\mathbf{s}^i$	Unit slip vector for slip system i
σ	Stress tensor
σ_{a}	Stress amplitude
$\Delta\sigma_{\text{ff}}$	Far field stress range
σ_{f}	Griffith fracture stress
σ'_{f}	Basquin fracture stress
σ_{n}^i	Normal stress for slip plane i
$\sigma_{\text{n,max}}$	Fatemie-Socie maximum normal stress
σ_{y}	Yield stress
T	Temperature
$T_{0.5}$	Temperature constant for dislocation loop annihilation
T_{d}	Derivative time constant
T_{i}	Integral time constant
T^{∞}	Ambient air temperature
t	Time
τ	Shear stress
τ^i	Resolved (applied) shear stress on i^{th} slip system
τ_{c}^i	Critical resolved shear stress for i^{th} slip system
$\tau_{\text{c},0}^i$	Initial critical resolved shear stress for i^{th} slip system
τ_{loop}	Dislocation loop line tension
$\tau_{r\theta}$	Shear stress in polar coordinates
ΔV	Activation volume
V_{at}	Characteristic atomic volume
w	Elastic energy released per vacancy emission
χ	Fraction of critical stored energy density remaining after crack extension
ω	Dislocation escape attempt frequency
ξ	Fraction of recoverable plastic strain energy

Glossary

AGR	Advanced gas-cooled reactors
BCT	Body centred tetragonal
BIB	Broad ion beam
CEGB	Central Electricity Generating Board
CD	Cluster dynamics
CPFE	Crystal plasticity finite element
CRA	Creation relaxation algorithm
CRSS	Critical resolved shear stress
ΔCTSD	Crack tip sliding displacement range

DDP	Discrete dislocation plasticity
DFT	Density functional theory
DFZ	Dislocation free zone
DHC	Delayed hydride cracking
DIC	Digital image correlation
DPA	Displacement per atom
DSC	Differential scanning calorimetry
EBSD	Electron back-scatter diffraction
EBSP	Electron backscatter diffraction patterns
EDM	Electrical discharge machine
FCC	Face centred cubic
FCG	Fatigue crack growth
FIP	Fatigue indicator parameters
GB	Grain boundary
GND	Geometrically necessary dislocations
HCP	Hexagonal close packed
HRDIC	High resolution digital image correlation
IPF	Inverse pole figure (orientation dependent)
IR	Infrared
ISO-LH	Isotropic linear hardening
MD	Molecular dynamics
MSCDFM	Micromechanical Short Crack Driving Force Metrics
ND	Normal direction
PFM	Phase field modelling
PID	Proportional-integral-derivative
PWR	Pressurised water reactor
RD	Rolling direction
ROI	Region of interest
SED	Stored energy density
SEM	Scanning electron microscope
SIF	Stress intensity factor
SRSCD	Spatially resolved stochastic cluster dynamic
SSD	Statistically stored dislocations
TD	Transverse direction
TEM	Transmission electron microscope
TSSD	Terminal solid solubility for dissolution
TSSP	Terminal solid solubility for precipitation
UDMGINI	User-defined damage initiation
XFEM	eXtended Finite Element Method
XRD	X-ray diffraction

1 Introduction

1.1 A brief timeline of nuclear power and policy in the United Kingdom

The atomic bomb gave rise to the nuclear power industry, having brought together talented scientists who ended up working for the organisations which themselves were born of defence-dominated ambitions. Initially, nuclear reactors were constructed to manufacture plutonium for military weapons programs. The emphasis later transitioned to electricity generation. While the first electricity generating nuclear power plant was the Experimental Breeder Reactor I in Idaho, USA, Britons were the first to benefit from commercial nuclear power produced at Calder Hall power station in 1956. Calder Hall was the first of a fleet of gas-cooled reactors across Britain. At this stage, nuclear power promised to provide inexpensive electricity for millions of homes, which would garner support for government during the post-war era. However, it transpired that initial estimates were highly optimistic and nuclear power would not revolutionise the cost of electricity at this early stage in its history. A second wave of reactors, advanced gas-cooled reactors (AGRs), were introduced, the first of which was constructed at Dungeness. In terms of cost and time to build, the Dungeness prototype, and later, the entire AGR programme proved unsuccessful. Following a spate of attempts by various UK governments and the Atomic Energy Authority to deliver a successful reactor programme, by the mid-1980s, the Thatcher administration introduced dramatic changes to economic policy which, alongside growing discontent for nuclear power globally, would have serious consequences for the industry.

Over the past 40 years, the UK may be viewed as a test case for measuring how the level of government involvement in financing nuclear new builds impacts their viability. During this period, the UK has witnessed dramatic changes to the energy market. In the early years of civil nuclear power in the UK, it was the responsibility of the Central Electricity Generating Board (CEGB) to generate, transmit and sell electricity across England and Wales. This was until market liberalisation was introduced as part of free-market reforms during the Thatcher administration, which brought with it widespread disruption, but also changes which might otherwise be viewed today as beneficial. The final nuclear power plant build financed by the CEGB was Sizewell B, a pressurised-water reactor (PWR) which is currently still in operation. Being financed by the CEGB meant that public money was invested, and therefore the risks associated with the project were shared amongst taxpayers. Market liberalisation meant that

private investors would have to bear these risks to finance nuclear new builds on their own. The cost of nuclear energy differs substantially from other common (mainly fossil-fuels-based) sources in that the greatest expenditure is in construction. Contrariwise, the cost to build a combined cycle gas turbine, for example, accounts for just a small fraction of its levelized cost of energy. To the private investor, this means the following: (i) upfront costs and associated risks are much lower and (ii) a return on investment is likely within a much shorter timeframe than for financing a new nuclear build. Hence, with market liberalisation, the UK witnessed the decentralisation of the energy market, large-scale replacement of coal-powered plants with gas-powered technology, the importation of fossil fuels, and the gradual regression of nuclear energy.

Market liberalisation has inadvertently had a great positive effect on the level of emissions in the UK over the years due to the relative cleanliness and efficiency of gas-fired power plants compared with coal. While climate change was not the most pertinent matter in energy policy during the 1980's, it has become so in recent years. With this, the demand for clean energy is much greater than it ever has been. To many, this means that renewable energy is the way forward. Where policy is concerned, energy security is equally important, and for this, nuclear energy is also highly favourable. In consequence, the attitudes of government began to shift once more towards enabling nuclear new builds. In 2016, the government decided to help fund the development of Sizewell C (alongside energy company, EDF), the UK's newest nuclear power plant. This was enabled by the implementation of Contracts for Difference, which are intended to incentivise low carbon energy generation in the UK by contributing to capital costs for the benefit of the energy provider. This somewhat hybrid approach to nuclear financing appears successful, as Sizewell C is set to start generating power by 2026. The UK government also recently announced changes to how future nuclear energy projects may receive funding with the Regulated Asset Base model. This approach will enable the investor to share some of the nuclear new build construction costs with the consumer, which should lower financing costs significantly. It seems as though over the past 40 years, governmental attitudes to state subsidy of the energy market have found an appropriate medium between total control and none whatsoever.

Globally, nuclear power began to gain traction by the mid-1970s. France positioned itself as a world leader by 1980, with nuclear now contributing to 40% of its primary energy mix [1] (see

Figure 1) and up to 70% of its electricity demand [2]. By contrast, several countries have phased out their nuclear programmes including Italy and more recently, Germany, due to risks associated with the technology. Despite the purported risks, other countries such as Japan continue to expand their nuclear energy capacity, aiming to achieve an energy share contribution of 20% by 2030 [3].

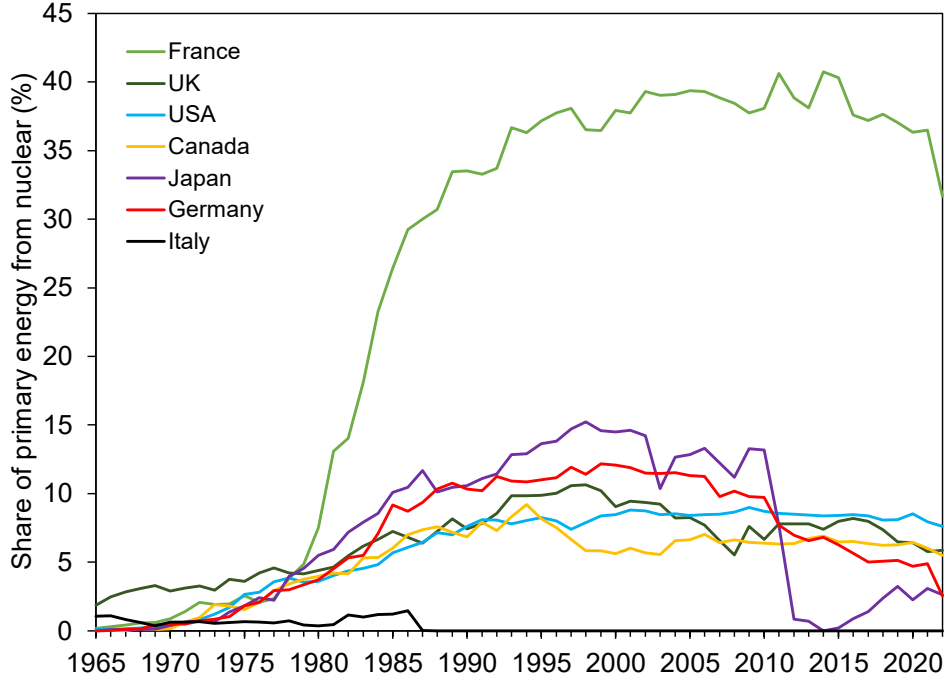


Figure 1: Share of primary energy from nuclear for various countries from 1965 to 2022. Data from Energy Institute Statistical Review of World Energy (2023) [1].

Fissile nuclear power will play a vital role in decarbonising the global energy mix for years to come. To increase its economic viability and maintain a competitive advantage over e.g., intermittent renewable sources, it is vital that technological and scientific innovation persists, particularly for next generation civil nuclear reactors, including small modular reactors (SMRs) etc. The work presented in this thesis aims to contribute to the development of safety cases for the nuclear industry through improved scientific understanding of failure mechanisms relevant to structural fuel cladding materials. With reduced uncertainty in reactor design and operation, plant lifetime extension is made feasible.

1.2 Basic principles of nuclear power

1.2.1 Overview of nuclear fission

Nuclear energy refers to the release of energy from atomic nuclei during a fission events (it can also refer to the release of energy during a fusion reaction). When a high-energy neutron is

captured by a large, “unstable” nucleus, the surface energy of the nucleus is increased, which in turn reduces its binding energy. This further reduces the stability of the nucleus and excites it beyond the fission energy barrier. Upon splitting a nucleus into smaller, more stable fission products, energy is released due to the mass-energy equivalence principle [4], which is described by,

$$E = mc^2 \quad (1.1)$$

where E is the energy released, m is the change in mass of the system, and c is the speed of light in a vacuum. Hence, the mass of the products of the fission reaction can be marginally lower than the mass of the reactants, giving rise to the release of kinetic energy in the products (see Figure 2).

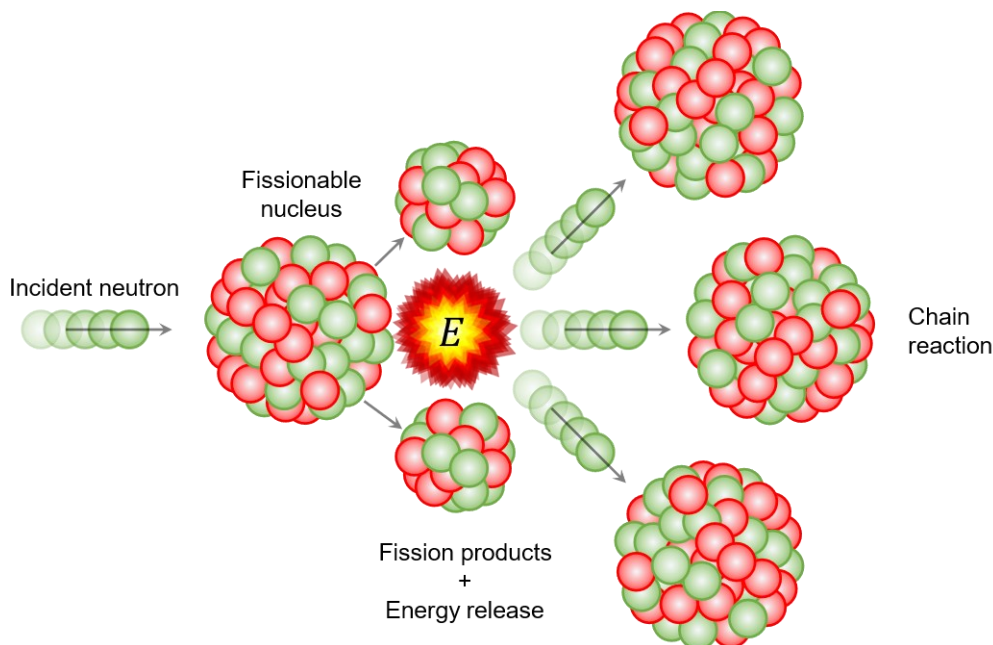
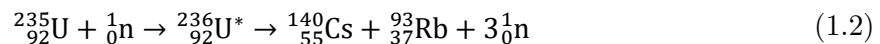


Figure 2: Schematic representation of fission chain reaction.

A common example of a fission reaction, and one which is utilised by the nuclear industry, is that of uranium-235, which is given by Equation 1.2.



Here, the caesium, rubidium, and neutron fission products have a combined mass which is 0.085% lower than that of the reactants, resulting in roughly 187 MeV of kinetic energy release per fission event. Due to the release of surplus high-energy neutrons, fission events can initiate a chain reaction (see Figure 2). The field of study of reactor kinetics deals with the transient behaviour and control of nuclear chain reactions. From an industrial perspective, the release

of high energy neutrons has important implications for the structural integrity of safety-critical materials (including Zircaloy-4), which will be discussed throughout this thesis.

1.2.2 *Harnessing nuclear power*

Over the past 60 years, there have been many reactor designs and concepts. Through collisions and interactions of high energy fission fragments, heat is generated, which forms the basis for virtually all nuclear reactor designs. In Figure 3, a schematic of the PWR design is used for illustration due to its relevance to this thesis.

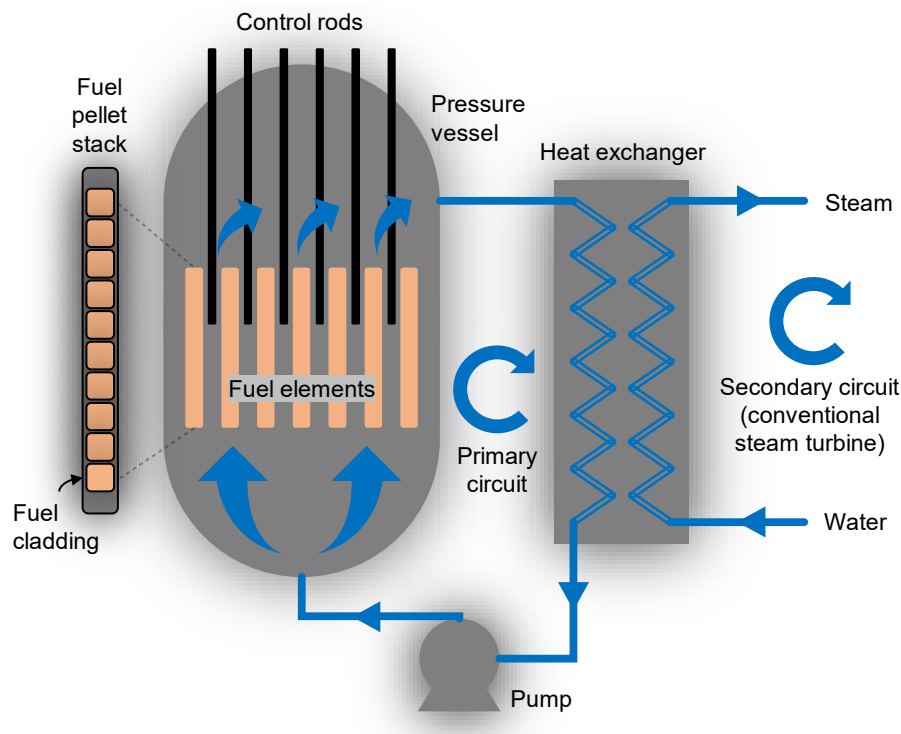


Figure 3: Overview of PWR design. Heat from the primary circuit water is transferred via a heat exchanger to a secondary steam turbine circuit.

Figure 3 shows that the reactor core, which is contained within the pressure vessel, is comprised of a series of fuel rods/elements, which constitute a stack of fuel pellets (typically made of compressed uranium dioxide powder). The fissile chain reaction between fuel elements is controlled by the control rods, which are raised or lowered to accelerate, inhibit, or sustain the rate of reaction. Pressurised water within the primary circuit is pumped through the reactor core, longitudinally by the fuel elements and serves two main purposes: (i) to transfer heat from the reactor core to the secondary circuit via the heat exchanger and (ii) moderation, which is to reduce the kinetic energy, i.e., velocity of neutrons and thereby increase the probability of neutron capture by uranium nuclei. Most heat is generated at the centre of the fuel rod ($>1000^{\circ}\text{C}$), which is transferred radially to the fuel cladding, and then by convection

to the flowing water ($>300^{\circ}\text{C}$). As most fuel rod diameters are of the order of 10 mm, significant thermal spatial gradients are generated radially through fuel and structural fuel cladding (which is typically just 0.6 mm thick). Crucially, this leads to cladding stresses, which can vary cyclically depending on reactor operating conditions. Moreover, the corrosive high temperature environment of the reactor core can lead to additional phenomena (oxidation, hydrogen ingress, hydride embrittlement) which affect cladding structural integrity and will be discussed in subsequent sections.

1.3 Zirconium alloys in the nuclear industry

1.3.1 *Introduction*

A primary focus in the design of nuclear reactors is to ensure a high neutron economy, and hence to select a fuel cladding material which has a low neutron absorption cross-section. This highlights one reason why zirconium alloys are selected for this application over more conventional materials, including steels. Based on this criterion alone, other appropriate material choices may include magnesium or beryllium. However, when considering formability, mechanical strength, service temperature, creep resistance, neutron radiation resistance, and corrosion resistance, zirconium alloys are often the preferred solution.

1.3.2 *Cladding design*

In PWR fuel assemblies, cladding and other structural components are usually made of Zircaloy-2 or Zircaloy-4. Depending on reactor design, geometries can vary, but claddings are typically 4 to 5 m in length, 9 to 12 mm diameter, and have wall thicknesses in the range 0.6 to 0.8 mm [5]. The typical PWR fuel assembly, which is illustrated in Figure 4 (a) consists of a grid of closely spaced clad fuel rods. Other Zr-based components such as grids and guide tubes are used to form the skeleton of the fuel assembly. Figure 4 (b) shows the end of a fuel rod. Fuel pellets, which are typically 11 mm long, are stacked longitudinally through several metres of cladding. Plugs and temperature-resistant springs are inserted at either end to seal the fuel rod while also accommodating changes in length due to fuel degradation.

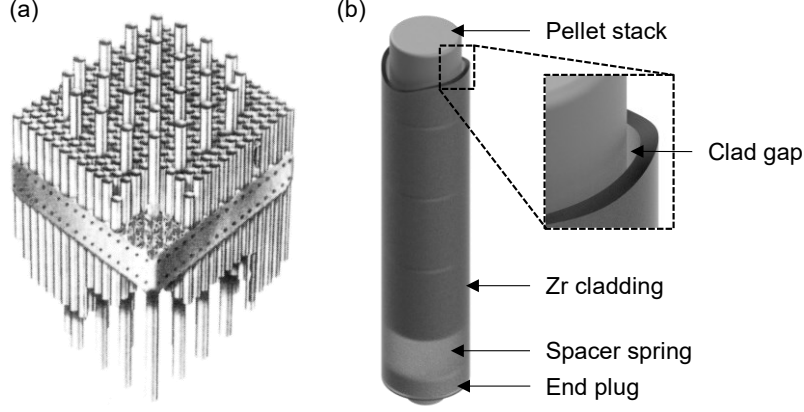


Figure 4: (a) Typical PWR fuel assembly (FRAGEMA) [5], and (b) details of fuel rod construction.

Figure 4 (b) also shows the fuel clad gap, which separates the cladding and fuel pellets. Fission gases released from fuel pellets are transported via this gap to the end of the pellet stack. This helps to maintain an appropriate fuel temperature. The gap is carefully designed to be large enough to allow fission gas transport, yet small enough to permit efficient heat transfer through the cladding. Over time, zirconium cladding can undergo severe creep deformation. Specifically, the creep-down phenomenon [6] causes the fuel-cladding gap to close, which is detrimental. This effect can be accelerated significantly by increased fuel temperatures. The release of fission gases can also drive an increase in cladding internal pressure, which is an important consideration for structural integrity assessment, particularly at high levels of burn-up.

1.3.3 Zirconium alloy development

There have been several major successes and failures in the development of zirconium alloys for fuel cladding. The primary alloys and their compositions are listed in Table 1. Those listed can be separated into two main categories based on phase; the single phase (α) Zircalloys and the dual phase (α/β) zirconium alloys which contain niobium.

Table 1: Compositions of most common zirconium alloys in the nuclear industry [7].

ASTM reference	R 60802	R 60804	R 60901	R 60904
Common name	Zircaloy-2	Zircaloy-4	Zr-Nb	Zr-Nb
<i>Alloying elements</i> (mass %)				
Sn	1.2 – 1.7	1.2 – 1.7	-	-
Fe	0.07 – 0.2	0.18 – 0.24	-	-
Cr	0.05 – 0.15	0.07 – 0.13	-	-
Ni	0.03 – 0.08	-	-	-
Nb	-	-	2.4 – 2.8	2.5 – 2.8

Naturally, the first Zircaloy, which was developed in the early 1950s was labelled Zircaloy-1 (Zr-2.5%Sn). Tin was added to inhibit the damaging effects of impurities, but the alloy was

deemed insufficient for long-term corrosion resistance. Later in the 1950s, Zircaloy-2 was developed; tin content was reduced slightly, but nickel, iron, and chromium were added to improve corrosion resistance. Zircaloy-3 was then developed with a further tin content reduction (due to inefficiency in terms of corrosion resistance) and removal of nickel due to increased hydrogen pick-up. However, it had very poor mechanical performance and was not brought into production. Lastly, Zircaloy-4 was developed to counteract the removal of nickel with an increased iron content. Hence, it has improved corrosion resistance, reduced affinity for hydrogen pick-up, and improved mechanical properties over previous Zircaloys. Zr-Nb alloys were primarily developed for CANDU reactor (Canadian deuterium uranium) pressure tubes; niobium is added as it is a β phase stabiliser, and is found to significantly improve mechanical strength, which is a key consideration for pressure tube design (while also resulting in a reduction in corrosion resistance). As discussed previously, hydrogen ingress and hydride formation are important factors affecting the structural integrity of zirconium alloy cladding materials. While not an element by design, waterside high-temperature cladding oxidation leads to the production of hydrogen (see Equation 1.3), a proportion of which is absorbed into the bulk of the material.



Hydrogen atoms diffuse to interstitial sites within the hexagonal close packed (HCP) lattice of α zirconium. The phase diagram in Figure 5 shows that at sufficient concentrations, the precipitation of zirconium hydrides can occur.

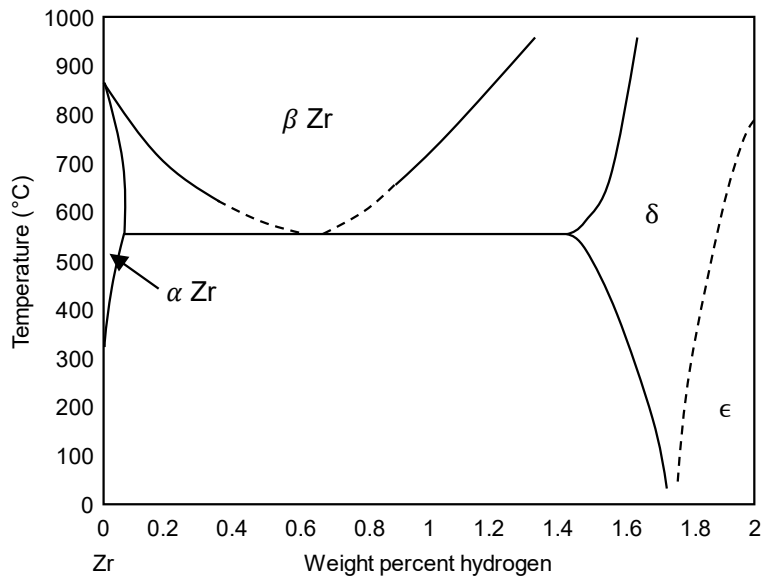


Figure 5: Zirconium-hydrogen phase diagram. Adapted from Zuzek et al. [8].

Under normal reactor operating conditions, hydrogen precipitates as the δ hydride phase, which is characterised by a face centred cubic (FCC) crystal structure, and stoichiometry around $\text{ZrH}_{1.66}$. At higher cooling rates, metastable body centred tetragonal (BCT) phases precipitate also. Hydride precipitation leads to the embrittlement and performance degradation of zirconium alloys; specifically, at certain temperatures and loading conditions, delayed hydride cracking (DHC) can occur, which is a gradual electro-chemo-mechanical crack growth mechanism. More generally, hydrides can lead to and accelerate crack initiation and subsequent fatigue crack propagation under cyclic loads, which is a core theme of this thesis.

1.4 Thesis

The overarching aim of this work is to develop mechanistic understanding of the primary factors affecting structural integrity in fission fuel cladding materials for enhancement of failure prediction standards. Chapters 2 and 3 of this thesis present a review of existing literature in a fuel cladding structural integrity context, and an overview of the methods used in subsequent chapters, respectively. Beginning with the fundamental issue of short fatigue crack propagation in Chapters 4 and 5, the role of microstructure in governing propagation rates and crack direction is investigated, leading to the development of improved models for crack path prediction and an analytical solution for crack rate. In Chapter 6, new models describing temperature, stress, and flux-dependant irradiation damage evolution are developed to construct a framework for simulating conditions which are more representative of real reactors. Lastly, Chapter 7, which is mostly experimental, addresses issues of fatigue crack propagation and localisation of strain in hydrided Zircaloy-4. The results from this chapter are intended to provide key insights into damage localisation due to hydride precipitation and subsequent crack propagation mechanisms for subsequent physically based modelling.

Page would be left blank if not for this note.

2 Literature Review

2.1 Driving forces for fatigue crack growth

2.1.1 Introduction

With an ever-increasing supply of grid electricity from renewables, nuclear power can represent an appropriate energy baseload provider in a net-zero economy. However, the intermittency of renewables will require nuclear power plants to operate in a more variable and dynamic way than ever before [9]. As a consequence, fatigue becomes a more important consideration for structural reactor core components including fuel cladding. Specifically, for precise life assessment and development of rigorous codes and standards, fatigue crack initiation and microstructurally short fatigue crack propagation are most relevant, particularly when considering sub-millimetre Zircaloy cladding dimensions. Fortunately, fatigue and fracture mechanics have been recognised as important considerations in engineering design for almost 200 years [10]; today, there are a plethora of analytical, numerical, and experimental methods available, upon which the current work is built. In this section, a review of existing predictive modelling methods for fatigue life are presented and discussed in the context of this thesis.

2.1.2 Classical methods

Where cyclic stresses are introduced from fluctuating temperature, conditions for low-cycle fatigue are established; low-cycle fatigue is regarded as being dominated by cyclic strains. The Coffin-Manson relationship [11], which is given by Equation 2.1, was developed based on the phenomenological relationship between the total (plastic) strain range and the number of cycles to *failure*.

$$\frac{\Delta\varepsilon_p}{2} = \varepsilon'_f(2N)^c \quad (2.1)$$

From Equation 2.1, $\Delta\varepsilon_p$ represents the plastic strain range, N the number of cycles to failure, and ε'_f and c are fitted power law constants; ε'_f is approximately equal to the uniaxial tensile fracture strain. Typically, the exponent c will vary between -0.5 and -0.7 for metals [10], and hence captures the dramatic fatigue life reduction associated with increased levels of plasticity. The Coffin-Manson equation has been shown to predict *failure* due to low-cycle fatigue for a range of metallic materials (under a specific set of conditions) [12], [13], [14]. High-cycle fatigue is regarded as being dominated by stress, since strain magnitudes are low, and therefore

predominantly elastic. Hence, the earlier Basquin equation [15], which bears resemblance to the Coffin-Manson law, accounts for the elastic strain range, and is given by Equation 2.2,

$$\sigma_a = \frac{\Delta \varepsilon_e}{2} E = \sigma'_f (2N)^b \quad (2.2)$$

where σ_a is the alternating stress amplitude, $\Delta \varepsilon_e$ the elastic strain range, E the elastic modulus, and σ'_f and b are fitted constants; σ'_f is approximately equal to the monotonic fracture stress. While useful in simple engineering design, there are three key weaknesses of the Coffin-Manson and Basquin equations:

- i) Applicability only to specific regimes of deformation, i.e., fully elastic, or predominantly plastic. It is clear from several studies [16], [17], [18] that the Coffin-Manson equation breaks down when applied outside of its limits; it should be noted however that Morrow [19] later formulated a model based on the total strain amplitude which could capture behaviour across both regimes.
- ii) Reliance on vast experimental datasets to determine fitted constants for every material.
- iii) The inherent definition of *failure*; both models consider failure to correspond to test specimen fracture, without regard for the early stages of crack initiation and growth etc., which are arguably more important for safety-critical components.

Around the same time as the establishment of the Coffin-Manson equation, an empirical formula was published which would facilitate *failure* prediction on the basis of a specific crack length, rather than total fracture. The equation is known as Paris' law [20] and is given by Equation 2.3,

$$\frac{da}{dN} = A(\Delta K)^p \quad (2.3)$$

where a is crack length, ΔK is crack tip factor (SIF) range, and A and p are fitted constants. This approach is more attractive for fatigue life assessment as it does not require a *failure* definition as a precondition. However, it still suffers from drawbacks i) and ii) listed above. Specifically, it is only applicable to stage II fatigue crack growth (FCG), which is otherwise known as the Paris regime, and hence does not account for crack nucleation or initiation, which dominate fatigue life for most materials. As will be outlined in greater detail in subsequent chapters, stage II FCG describes when cracks are at such a length that crack tip stress is dominant; when crack lengths are at the microstructural scale, dislocation slip and plasticity

are known to have a substantial effect on the FCG rate, $\frac{da}{dN}$ [21], [22], [23] and hence, Paris' law is inapplicable. The crack tip SIF range is primarily a function of applied stress or load, crack length, and cracking mode; the three basic modes of cracking (I. opening, II. in-plane shear (sliding), and III. out-of-plane shear (tearing)) are illustrated in Figure 6.

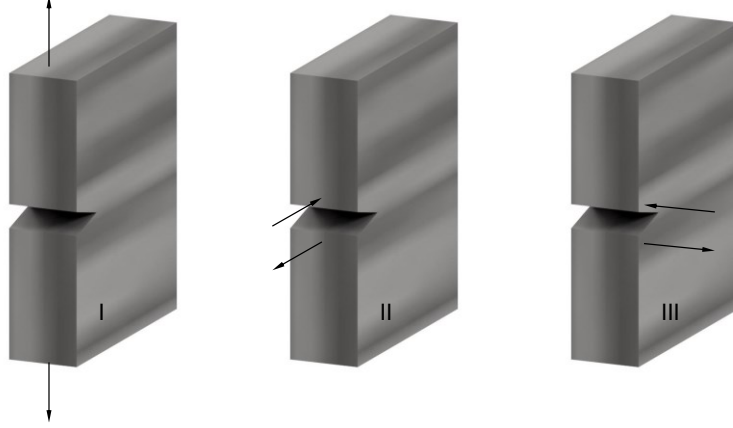


Figure 6: The basic modes of cracking: I. opening, II. sliding, and III. tearing.

Several attempts [24], [25], [26] have been made to modify the SIF parameter to accommodate effects due to plasticity. For example, Dai and Li [24] developed a plasticity-corrected SIF range, ΔK_{pc} , which adapts Eshelby's inhomogeneity theory [27] to transform the inhomogeneous elastic-plastic crack front to a homogeneous region. This approach was shown to eliminate variability due to different loading ratios but was also limited to the Paris regime due to the effects of microstructural inhomogeneity.

In fracture mechanics, Griffith theory [28] was the first to successfully capture experimental observations. According to Griffith, the fracture stress is given by,

$$\sigma_f \approx \left(\frac{EW_p}{a} \right)^{1/2} \quad (2.4)$$

where W_p is the *plastic* work required to extend a crack of length $2a$. This was later modified by Irwin [29], [30] to eliminate the abstruse *plastic* work term and replace it with a measurable quantity: the crack-extension force *or* critical energy release rate, $\mathcal{G}_c$, which has units J/m². Irwin's modified expression is given by Equation 2.5.

$$\sigma_f = \left(\frac{E\mathcal{G}_c}{\pi a} \right)^{1/2} \quad (2.5)$$

The energy release rate is then given by Equation 2.6.

$$\mathcal{G} = \frac{\pi a \sigma^2}{E} \quad (2.6)$$

When arranged in this way, it becomes clear that the Griffith-Irwin approach is only applicable to brittle “elastic” materials, and hence, will not be discussed further in the context of FCG. Rice [31] later introduced the J-integral for ductile materials. The J-integral may be interpreted as the rate of change of potential energy with respect to crack length, which is $J = \frac{\partial U}{\partial a}$; this is equivalent to Irwin’s energy release rate, $\mathcal{G}$, for “elastic” materials. Rice defined the J-integral as the path-independent line integral of the difference between strain energy and rate of work by the stress field around a crack, and is expressed as,

$$J = \oint_{\Gamma} \left(W dy - \mathbf{T} \frac{\partial \mathbf{u}}{\partial x} ds \right) \quad (2.7)$$

where $W = \int \boldsymbol{\sigma} : d\boldsymbol{\varepsilon}$ is the strain energy per unit volume, Γ is the closed path of the integral around the crack (see Figure 7), $\mathbf{T}$ the traction vector acting on the path, $\mathbf{u}$ the displacement vector, ds the increment around the path, and x and y are Cartesian directions. Due to its relative complexity (compared with e.g., Griffith [28]), practical implementation of the J-integral requires numerical solvers, e.g., finite element.

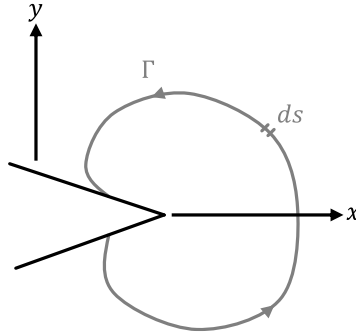


Figure 7: Graphical representation of J-integral measurement at a crack tip.

As discussed above, the J-integral is equivalent to the energy release rate for fully elastic deformation and can therefore be considered a plasticity-corrected energy release rate. Under the same conditions used to derive Equation 2.6, SIF is given by $K = \sigma\sqrt{\pi a}$, and hence, Equation 2.6 may be rewritten more generally as $\mathcal{G} = \frac{K^2}{E'}$, where $E' = E$ for plane stress, and $E' = \frac{E}{1-\nu^2}$ for plane strain. By extension of $J - \mathcal{G}$ equivalence, the J-integral range under cyclic loading may be written in terms of a J plasticity-corrected SIF range as follows,

$$\Delta J = \frac{(\Delta K_{\text{Jpc}})^2}{E'} \quad (2.8)$$

where ΔK_{Jpc} is the modified SIF range. This enables an elegant coupling of the cyclic J-integral with Paris' law, where ΔK is replaced by its J plasticity-corrected counterpart, as follows,

$$\frac{da}{dN} = A(\sqrt{\Delta J E'})^p = B(\Delta J)^q \quad (2.9)$$

where B and q are fitted constants. This pseudo-J-integral Paris' law method has been successfully applied to several metallic systems [32], [33], but typically within the stage II FCG, small scale yielding regime. Moreover, the Paris-like approach suffers from reliance on extensive experimental data to obtain the fitting constants for each material, which ultimately have no physical meaning. However, Wilson et al. [34] used crystal plasticity finite element (CPFE) modelling to study the microstructural sensitivity (for stage I FCG) of the J-integral as a fracture criterion in HCP single crystals. While it was shown to capture some microstructure-linked variability, the J-integral was ultimately deemed inadequate compared with more physically based methods (which will be discussed later).

2.1.3 Fatigue indicator parameters

With recognition that there are a multitude of factors responsible for the initiation and propagation of fatigue cracks, fatigue indicator parameters (FIPs) were developed as correlative proxies for driving forces and are primarily based on correlated data. FIPs could often predict the number of cycles to *failure* based on a wide range of loading conditions and material properties. Several formulations were recently reviewed by Stopka et al. [35]. Among the first to establish a FIP were Brown and Miller [36], who theorised that a combination of shear strain and tensile strain normal to the plane of maximum shear could characterise fatigue failures. The FIP was expressed mathematically as,

$$\text{FIP}_{\text{BM}} = \frac{\varepsilon_1 - \varepsilon_3}{2} = f\left(\frac{\varepsilon_1 + \varepsilon_3}{2}\right) \quad (2.10)$$

Where $\varepsilon_1 > \varepsilon_2 > \varepsilon_3$ are the principal strains, $\frac{\varepsilon_1 - \varepsilon_3}{2}$ is the maximum shear strain, $\frac{\varepsilon_1 + \varepsilon_3}{2}$ is the tensile strain normal to the plane of maximum shear, and f is some function (determined by fitting to experimental data). Graphical representations of this correlation were used to establish the bounds of fatigue life from experiments under multiaxial strain conditions (which was a great leap from the aforementioned Coffin-Manson [11] and Basquin equations [15]).

However, the curves used to represent fatigue life limits were unintuitive and without physical basis and would therefore require extensive experimental datasets for each material. Building on this work, Fatemi and Socie [37] developed the widely known Fatemi-Socie FIP, which accounts for the maximum tensile stress normal to the plane of maximum cyclic plastic shear strain. The Fatemi-Socie FIP is given by,

$$\text{FIP}_{\text{FS}} = \frac{\Delta\gamma_{\text{max}}^{\text{p}}}{2} \left[1 + k \frac{\sigma_{\text{n,max}}}{\sigma_{\text{y}}} \right] \quad (2.11)$$

where $\Delta\gamma_{\text{max}}^{\text{p}}$ is the maximum plastic shear strain range (cyclic), $\sigma_{\text{n,max}}$ the maximum tensile stress normal to the plane maximum cyclic plastic shear strain, σ_{y} the yield stress, and k is a fitted constant which scales the influence of the normal stress. The mathematical formulation for this FIP is compatible with experimental observations of fatigue crack nucleation in metallic materials, which demonstrate that intragranular fatigue cracks tend to nucleate on crystallographic slip planes (which experience damage due to high levels of cyclic plastic shear, $\Delta\gamma_{\text{max}}^{\text{p}}$). The normal stress term is argued to drive crack opening on highly damaged planes. The greatest body of experimental work on this topic is within materials for aerospace applications [38], [39], [40], [41]. The Fatemi-Socie FIP is well established and has been applied to both millimetre-scale fatigue [42], [43] and micron-scale fatigue crack nucleation [41], [44], [45], [46], [47] under multiaxial loading conditions. However, while appealing due to simplicity and ease of implementation, it has rarely captured e.g., quantitative experimental measurements of crack nucleation or FCG rate. Moreover, it has been suggested that the fitting parameter k is dependent upon both material and loading conditions [48], i.e., at higher stress magnitudes, k is known to increase. An adaptation of the Fatemi-Socie FIP for intergranular fatigue crack nucleation [35] is given by,

$$\text{FIP}_{\text{GB}} = \frac{\gamma_{\text{net}}^{\text{p}}}{2} \left[1 + k_{\text{GB}} \frac{\sigma_{\text{n}}}{\sigma_{\text{y}}} \right] \quad (2.12)$$

where $\gamma_{\text{net}}^{\text{p}}$ is the cumulative plastic shear strain at a grain boundary, σ_{n} is the grain boundary normal stress, and k_{GB} is a fitted parameter. This FIP is limited to failure governed by grain boundary decohesion but is applicable to both micro-scale crack nucleation and propagation. It is implementable using a cohesive zones approach [49]. As with the original Fatemi-Socie term, the intergranular fracture approach has only really been used to indicate microstructural fracture “hot-spots”, without extensive experimental validation.

Using crystal plasticity modelling in combination with high-resolution short FCG rate and path data, Rovinelli et al. [50] investigated the solidity of several FIPs via probabilistic post-processing. The spectrum of FIPs included simple terms such as slip system with maximum slip and slip plane with maximum slip, and more involved FIPs such as dissipated energy and energy-based Fatemi-Socie terms. Results showed that on the whole, each of the eight FIPs performed terribly, with mean precision (based on correlation with experimental observations) not exceeding 31%, with total plasticity terms performing the worst. Liu et al. [51] recently published a comparative study of various conventional and new FIPs within a CPFE modelling framework. The authors proposed a new FIP based on the relative accumulation of dislocations in each grain by comparison with its neighbours, and is given by,

$$\text{FIP}_{\text{Dis}} = \max(\Delta\rho_{\text{tot}}^i) = \max\left(\Delta(\rho_{\text{for}}^i + \rho_{\text{deb}}^i + \rho_{\text{GND}}^i)\right) \quad (2.13)$$

where i refers to the grain number, $\Delta\rho_{\text{tot}}^i$ is the total dislocation density discrepancy between grain i and neighbouring grains, ρ_{for}^i the forest (immobile) dislocation density, ρ_{deb}^i the debris dislocation density, and ρ_{GND}^i the density of geometrically necessary dislocations (GNDs). This dislocation-density-based FIP was found to correlate well with experimental observations of crack nucleation sites in near- α titanium alloys [52], due to its ability to capture the detrimental effect of hard-soft grain orientation combinations (with respect to the primary loading direction). However, the FIP was not shown to provide quantitative predictions for e.g., number of cycles to crack initiation or rate of crack growth. In summary, FIPs have been demonstrated as useful indicators of crack nucleation location across length-scales but often lack physical understanding.

2.1.4 Discrete dislocation methods

The importance of dislocations in the nucleation and growth of fatigue cracks has been widely recognised in the literature for more than half a century [53], [54]. Weertman [54] proposed a critical stress criterion for fatigue crack nucleation and propagation which accounts for work hardening due to dislocation pileups. The critical stress was linked to Griffith's [28] surface energy but accounted for the effect of the crack tip plastic zone, within which a distribution of dislocations was assumed. This gave an analytical solution for FCG rate, which is,

$$\frac{da}{dN} = \left(\frac{\pi^3 \alpha}{8W_p \mu}\right) \left(\frac{r_p \sigma_A^2}{\sigma_c}\right)^2 \quad (2.14)$$

where $\alpha = 1$ for screw dislocation cracks (mode III) and $\alpha = 1 - \nu$ for edge dislocation cracks (modes I and II), ν is Poisson's ratio, μ the shear modulus, r_p the plastic zone size, σ_A the externally applied stress, and σ_c is the critical stress for fracture. This approach links the cyclic accumulation of dislocations with a local increase in stress until such a point that the critical stress is reached. Analytical methods such as this are highly attractive from an industrial perspective as they demand little computational expense, while solving a complex problem. However, Weertman's approach was not validated against experimental test data, and is an unlikely candidate for microstructurally sensitive short FCG, since important local quantities such as dislocation slip, and crystallography are unaccounted for. The first coherent theory of micromechanical fatigue crack nucleation was presented by Tanaka and Mura [55]. This approach is based on the irreversible accumulation of dislocations on slip bands within grains and led to the establishment of a mechanistic energy criterion for crack initiation. Figure 8 shows the dislocation motion in a favourably oriented grain of size $2a$ with a dislocation source at its centre. Where slip bands meet a free surface, interstitial and vacancy dipoles lead to the progressive development of surface intrusions and extrusions, respectively. In this case however, dislocations are assumed to pileup at grain boundaries, leading to recoverable (elastic) strain energy storage. The derived model for number of cycles to crack initiation is given by,

$$N_{CI} = \frac{8AW_s}{ba(\Delta\tau - 2\tau_{BS})^2} \quad (2.15)$$

where $A = \frac{\mu b}{2\pi(1-\nu)}$, W_s is a Griffith-like surface energy term for fracture, b the Burgers vector magnitude, a is half of the grain size (see Figure 8), $\Delta\tau$ the applied shear stress range, and τ_{BS} the shear back stress due to dislocations.

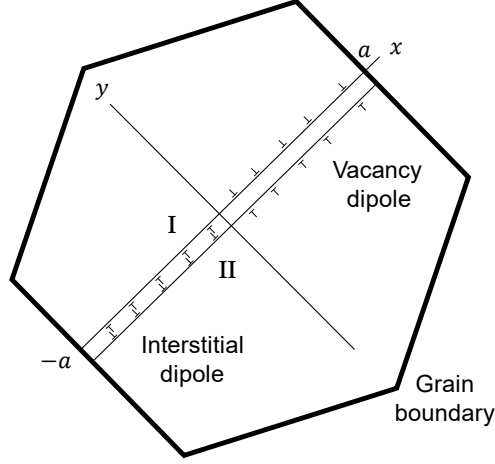


Figure 8: Dislocation motion in a most favourably oriented grain, which forms the basis for the Tanaka-Mura Fatigue crack nucleation model [55].

This simple, yet effective approach has since been widely adopted and applied to modern fatigue crack nucleation problems [56], [57], [58]. However, the very particular formulation and various associated assumptions render the method inapplicable to short FCG, which is a core theme in this thesis (N.B. crack nucleation in cladding materials is assumed to be dominated by environmental effects, which will be discussed later).

Purely analytical dislocation analyses were extended by Van der Giessen and Needleman [59] in the development of plane strain discrete dislocation plasticity (DDP). In DDP, the collective motion of discrete dislocations in an elastic medium is modelled to solve small-strain plasticity problems, accounting for nucleation, annihilation, and pileup of dislocations. Deshpande et al. [60], [61] used DDP modelling and a cohesive zones traction separation law to simulate mode I cyclic crack propagation in single crystals under plane strain conditions. The model could capture the transition between two distinct regimes of FCG: the threshold regime (which is commonly associated with stage I cracking) and the Paris regime (stage II) as crack length increased. Gurratxaga-Lerma et al. [62] used DDP modelling to study the effect of crack tip dislocations on stress states and shielding ahead of static cracks. Example dislocation distributions ahead of crack tips from Deshpande et al. [61] are shown in Figure 9.

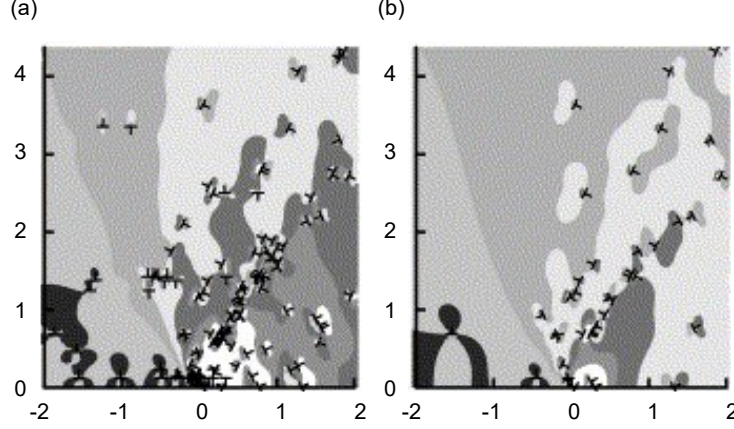


Figure 9: Symmetric dislocation distributions ahead of (a) 10 μm and (b) 3 μm edge cracks under cyclic loading [61]¹. In each case, the crack is horizontal, and the crack tip is located at coordinates (0,0).

More recently, Déprés et al. [63] used 3D discrete dislocation dynamics simulations to capture stage II FCG behaviour in FCC single crystals. Based on this, a semi-analytical model for stage II crack growth was developed, which could capture experimental trends. While possible to model 3D polycrystals in the stage I regime using DDP, it is prohibitively computationally expensive [64], and increasingly so as plasticity develops (due to increasing accommodation of motion of individual dislocations); even 2D DDP is highly computationally expensive, despite being limited to highly simplified and non-physical plane-strain infinite medium boundary conditions. DDP is therefore currently deemed an unsuitable methodology for accurate stage I FCG analyses in polycrystalline materials. However, the semi-analytical surrogate modelling approach developed by Déprés et al. [63] for stage II cracks raises the question: can this be done for microstructurally short FCG? This question will be revisited in Chapter 5.

2.1.5 *Stored energy density*

The nucleation and propagation of short fatigue cracks in metallic polycrystals is highly microstructurally sensitive due to strong interactions between cracks and localised plasticity [65], [66]. This is illustrated diagrammatically in Figure 10 (a), which shows that for stage I FCG, the crack length, plastic zone size, and microstructural feature length (i.e., grain size) are of the same order of magnitude, leading to strong interactions between the crack and its surrounding microstructure [22]. Figure 10 (b) highlights that as crack length is increased beyond the stage I regime, the plastic zone can span several grains, resulting in spatially homogeneous, non-microstructurally sensitive interactions.

¹ Reprinted from [61] (Figure 9), with permission from Elsevier (license number 5820771442221).

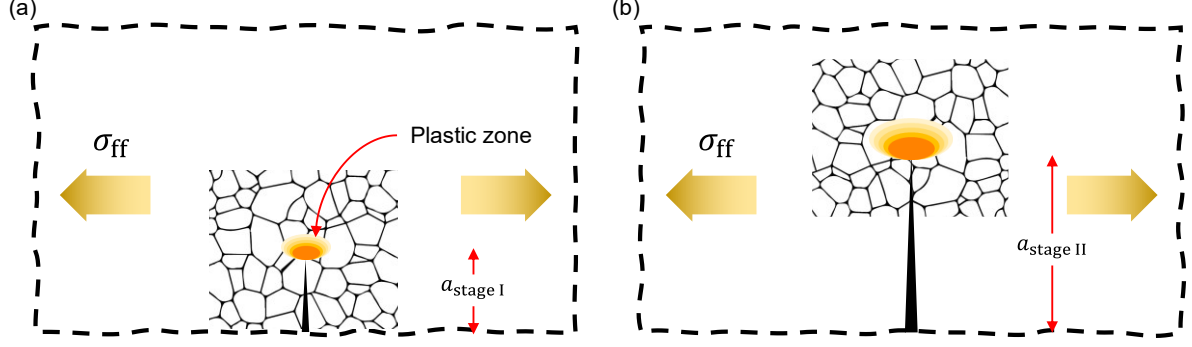


Figure 10: Interactions between crack tips and the local microstructure for (a) stage I and (b) stage II cracks. σ_{ff} refers to the far-field stress (independent of crack length).

To fully understand and predict how microstructure can impact crack nucleation and short FCG, it is therefore important to implement a fracture criterion which is fully microstructurally sensitive. Wan et al. [67] presented the stored energy density (SED) criterion for fatigue crack nucleation, which is reported to satisfy this requirement. SED is associated with plastic strain energy density, which is imparted on the material during plastic deformation, and is given by,

$$E_p = \int \boldsymbol{\sigma} : d\boldsymbol{\epsilon}^p \quad (2.16)$$

where $\boldsymbol{\sigma}$ and $\boldsymbol{\epsilon}^p$ are the stress and plastic strain tensors, respectively. E_p has units Jm^{-3} . Much of the plastic strain energy dissipates immediately as heat or sound, leaving a fraction which is stored. While the precise fraction is unknown, it is approximated as a constant, $\xi = 0.05$ [68], which gives Equation 2.17 for volumetric SED.

$$E_s = \xi \int \boldsymbol{\sigma} : d\boldsymbol{\epsilon}^p \quad (2.17)$$

The energy stored per unit volume is argued to be stored elastically (i.e., is recoverable) within dislocation structure, which accumulates cyclically in e.g., persistent slip bands, or as GNDs at grain boundaries or crack tips to accommodate highly localised lattice curvature. Hence, the energy required to extend a fatigue crack by some area, A , is related to energy stored per unit volume via the mean free distance (between dislocations), λ , as, $G = \lambda E_s = \xi \int \lambda \boldsymbol{\sigma} : d\boldsymbol{\epsilon}^p$, where $\lambda = \rho^{-1/2}$ and ρ is the total dislocation density (λ is included within the integral since dislocation density will evolve during plastic deformation). The volume associated with energy storage, $V = \lambda A$, is therefore highly length scale dependent. This is illustrated in greater detail in Figure 11. Hence, the SED fracture criterion term is given by Equation 2.18.

$$G = \xi \int \frac{\boldsymbol{\sigma} : d\boldsymbol{\varepsilon}^p}{\rho^{1/2}} \quad (2.18)$$

SED is most often implemented at the microscale using numerical CPFE modelling. Wan et al. [67] first demonstrated its microstructural sensitivity and ability to capture experimental crack nucleation data in notched ferritic steel samples, and again later in polycrystalline nickel superalloys [69], [70], [71]. Zheng et al. [72] presented the dislocation configurational energy formulation in DDP modelling, which is akin to SED, but is discrete in that it accounts for the energy contribution from each dislocation. Quantitative comparisons of SED and dislocation configurational energy distributions in polycrystals showed strong overall agreement, despite the DDP formulation leading to more intense localisation due to its discreteness.

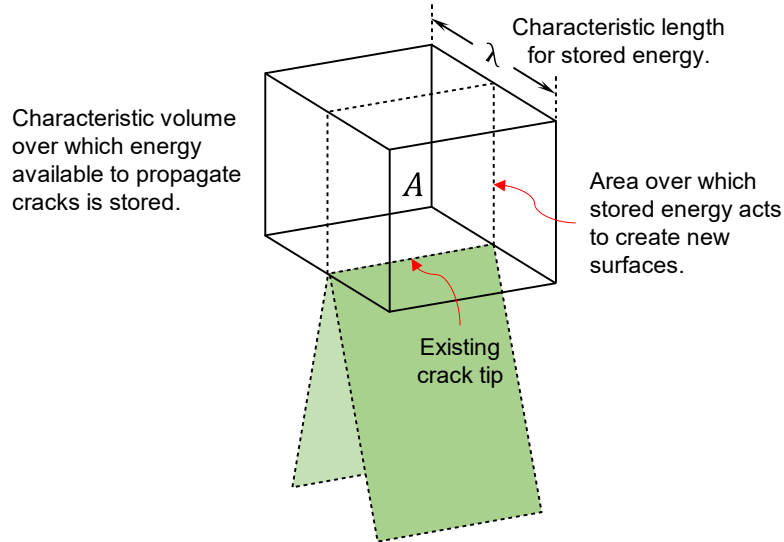


Figure 11: Physical basis for SED as a fracture criterion during FCG.

Hence, SED is arguably the primary driver for fatigue crack nucleation in metallic materials. However, this work focuses primarily on microstructurally short FCG. Wilson et al. published a series of studies [34], [73], [74] to evaluate the performance of SED for predicting rates of FCG in various metallic materials. Results showed that experimental crack paths [75] were reasonably well captured using a maximum slip criterion and that the SED fracture criterion would predict microstructure sensitive FCG rates. However, SED has yet to be shown to capture experimental FCG rate data. Experimental digital image correlation (DIC) data from Wan [76], in combination with short FCG rate data for Zircaloy-4 indicate a strong link between SED and microstructure sensitive FCG; this motivates Chapter 4 of this thesis. Specifically, FCG rates in two strongly textured large grain samples demonstrated significant

anisotropy in terms of crystal orientation. In a sample where the crack propagation direction was approximately parallel to the c-axes of individual grains, FCG rates were approximately twice as high as those in the counterpart where crack propagation direction was predominantly orthogonal to c-axes. In each case, crack paths were crystallographic and mostly along prismatic planes. Hence, the aims for Chapter 4 are as follows: to (i) determine if SED is the dominant driving force for short FCG in Zircaloy-4 by attempting to capture texture sensitive experimental data, (ii) understand the role of microstructure (aside from crystallography) in affecting FCG rates; specifically, how features such as grain boundaries and triple junctions influence crack propagation both prior to and during interactions, and (iii) quantify the critical SED for FCG in Zircaloy-4.

2.2 Irradiation damage

2.2.1 *Irradiation-induced lattice defects*

Operation of a nuclear reactor relies on the steady-state production of high-energy neutrons in the reactor core. While the theoretically ideal reactor fuel cladding material would be *transparent* to high-energy neutrons, such materials do not exist. Hence, collisions take place which result in momentum transfer from high-energy neutrons to zirconium atoms (these are referred to primary knock-on collisions), resulting in atomic displacements which can have knock-on effects for neighbouring atoms [5]. Brinkman [77] was among the first to theorise the impact of high energy incident particles on the atomic structure of metals; the principles of vacancy-interstitial pair (Frenkel defect) formation described by Brinkman and others [77], [78], [79], [80], [81] formed the basis for current understanding of irradiation damage. Figure 12 illustrates how a high energy particle, i.e., neutron can lead to the formation of single and double displacements, and by the same processes, defect cascades within metals [77]. Defect cascades refer to the localised nature of energy deposition due to first (primary knock-on atom) and second order displacements.

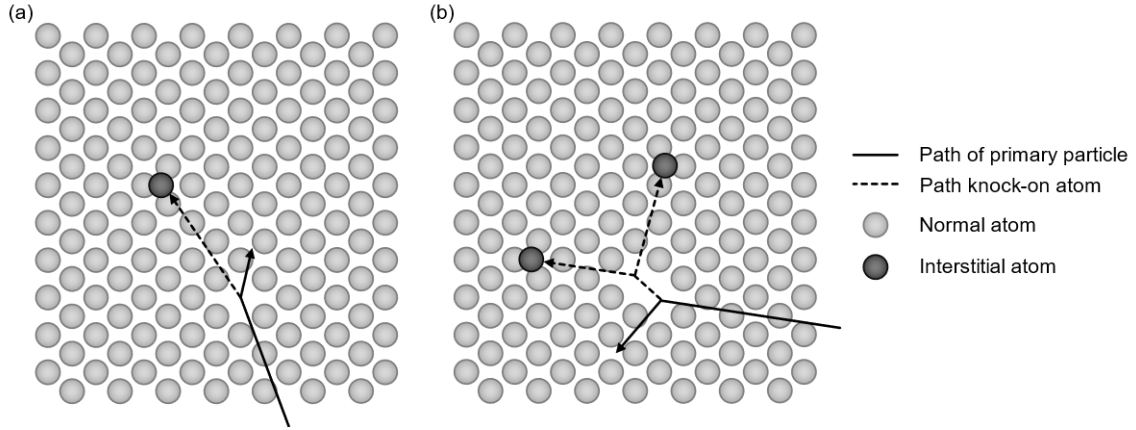


Figure 12: Two-dimensional schematic representation of simple irradiation damage processes, adapted from [77]. (a) Production of a single displacement. (b) Production of a double displacement.

It is now known that the primary mechanism by which irradiation affects material behaviour is the coalescence of vacancy and interstitial defects to form vacancy and interstitial dislocation loops [82]. In zirconium alloys, dislocation loops are characterised as either $\langle a \rangle$ or $\langle c \rangle$ type which lie on prismatic or basal planes, respectively [83]. Typically, $\langle a \rangle$ loops are of interstitial type [84], while $\langle c \rangle$ loops only constitute vacancies [85]. The most important difference between $\langle a \rangle$ and $\langle c \rangle$ loops is scale; $\langle c \rangle$ loops are typically > 10 nm in diameter, while $\langle a \rangle$ loop diameters are in the range 1-3 nm [84], [86]. Dislocation loops developing under irradiation are mostly $\langle a \rangle$ type, particularly at low irradiation doses [87].

2.2.2 Channel clearing phenomenon and influence on mechanical properties

Dislocation loops pin mobile dislocations and inhibit slip, leading to irradiation hardening [88]. At sufficient stress levels, dislocation slip is achieved which leads to the rapid annihilation of dislocation loops and hence, strain softening [89]. Early work by Martin and Weir [78] showed how such phenomena translate to component-scale tensile properties in stainless steel, with irradiation leading to both a substantial yield stress increase and fracture strain reduction; at elevated test temperatures, only the fracture strain reduction was apparent. Farrell et al. [90] investigated the tensile properties of various neutron-irradiated nuclear structural alloys including Zircaloy-4. It was shown for very low irradiation doses, e.g., 0.001 displacements per atom (dpa), that the tensile behaviour of Zircaloy-4 is impacted significantly. Transmission electron microscopy (TEM) of the same samples after straining showed traces of approximately uniformly spaced dislocation channels (see Figure 13), which accommodate plastic deformation and promote strain softening [90].

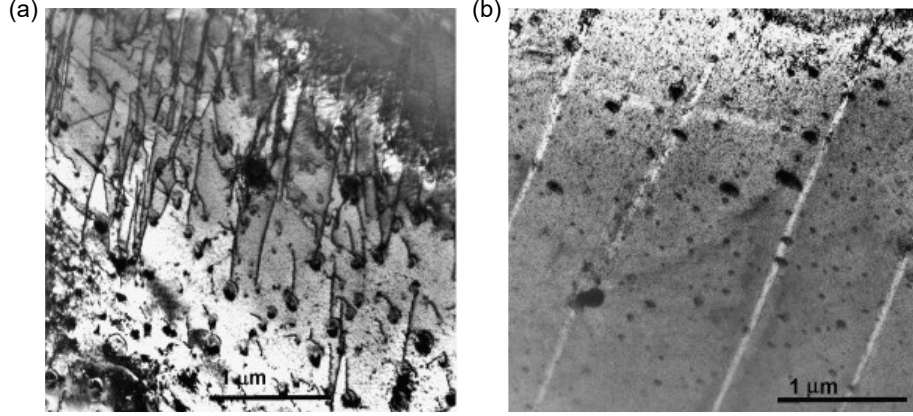


Figure 13: Deformation microstructures in unirradiated and irradiated Zircaloy-4 from [90]². (a) Dislocation structure in unirradiated Zircaloy-4 after 3% macro tensile strain. (b) Fully developed channels at 6.3% macro tensile strain in Zircaloy-4 irradiated up to 0.8 dpa.

More recent work by Thomas et al. [91] investigated the impact of proton irradiation on the local microstructural response in Zircaloy-4 using in-situ high resolution digital image correlation (HRDIC). Figure 14 shows a comparison of the free surface strain distributions of unirradiated and proton irradiated (0.1 dpa) regions of microstructure which highlights marked contrast due to increased localisation along discrete shear bands spanning entire grains after irradiation; the shear bands, which lie parallel with slip trace directions correspond to regions in which dislocation channel clearing has occurred. A statistical analysis of slip trace directions demonstrated that both unirradiated and irradiated plastic deformation was predominantly accommodated by prismatic slip.

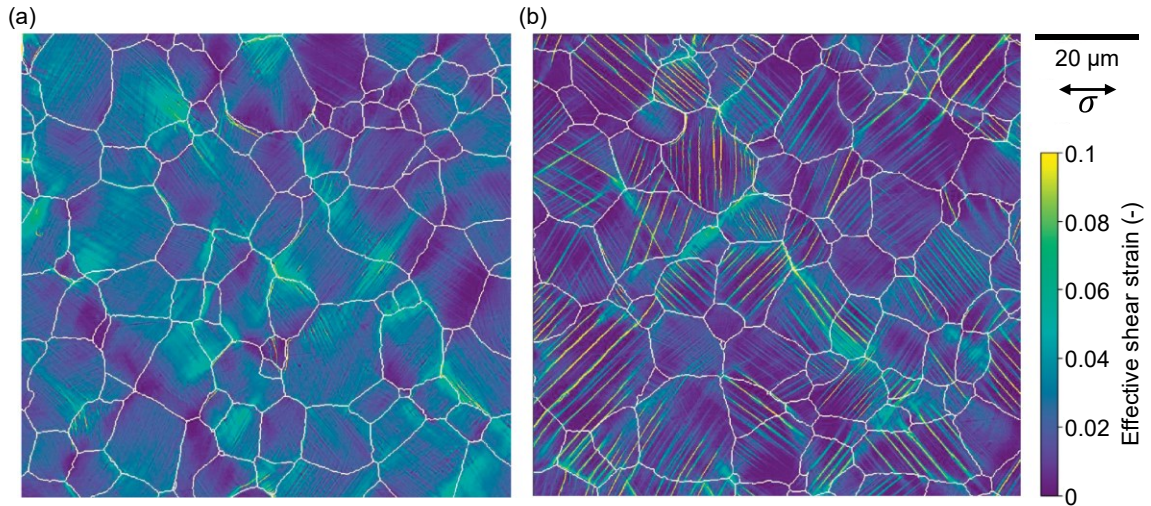


Figure 14: Strain distribution maps from HRDIC by Thomas et al. [91]³ in (a) non-irradiated and (b) irradiated Zircaloy-4 up to 2.59% and 2.04% strain in the loading direction, respectively.

² Reprinted from [90] (Figure 8 (b)), with permission from Elsevier (license number 5820771255614).

³ Reprinted from [91] under the Creative Commons Attribution 4.0 International Public License.

2.2.3 *Irradiation growth*

Irradiation growth refers to the gradual development of strain in unstressed materials due to irradiation damage (this differs from irradiation creep which is a material response to combined stress and irradiation conditions) [92]. Within zirconium single crystals, irradiation growth manifests itself as expansion along the $\langle a \rangle$ directions and contraction in the $\langle c \rangle$ direction [5]; this is broadly true for strongly textured zirconium polycrystals also. Cladding tube manufacture is typically such that c -axes are aligned with the radial and circumferential directions [93], meaning that irradiation growth results in longitudinal expansion and radial and circumferential shrinkage. Fidelis [92] showed that irradiation growth strain is strongly dependent on material (demonstrating significant differences even between zirconium alloys), microstructure, i.e., grain size and dislocation density, irradiation temperature, and irradiation dose rate. Post irradiation annealing for one hour at 783 K was shown to lead to up to 95-100% recovery of irradiation growth strain in recrystallized microstructures, and just 25% for cold-worked alloys. While not a core theme of this thesis, irradiation growth highlights the high microstructure sensitivity of irradiation damage evolution in zirconium alloys, which is relevant to Chapter 6.

2.2.4 *Models for irradiation damage*

Given the length and timescales over which irradiation damage processes occur, it is not surprising that most irradiation damage models are at the atomistic level. Most fundamentally, density functional theory (DFT) has been used to study the nature of nanoscale irradiation defects, clustering, and migration [94]. DFT is the dominant method for quantum mechanical simulations of metallic systems [95]; the approach solves for the ground state energy of a system of atoms, accounting for kinetic energy, external potential interactions, and electron-electron interactions for each individual atom. This method therefore becomes exponentially computationally expensive as the number of atoms in the system is increased, and hence most simulations are limited to a system consisting of around 500 atoms in the absence of exascale computing capabilities [96]. DFT models can provide useful and reliable information about the time-dependant evolution of collision cascades but are limited to relatively low impact energies due to system size constraints [97]. Consequently, DFT has been used to fit models for interatomic potentials for application to molecular dynamics (MD) simulations [98], which can model much larger systems consisting of hundreds of thousands of atoms [99]. Hence, MD simulations have been used to study defect, cluster, and dislocation loop density evolution at

different temperatures and irradiation dose rates. However, the well documented microstructural effects on defect evolution [100], [101], [102] are currently impossible to quantify using MD models, making it difficult to establish links between atomistic and engineering length scales. Moreover, the nanosecond timeframes over which MD simulations are carried out have little relevance to the hour to year-long cyclic operating conditions in a reactor core [103]. A time-independent atomistic scale modelling methodology known as the creation-relaxation algorithm (CRA) was recently developed by Derlet and Dudarev [104] in an attempt to circumvent computationally expensive MD modelling. Succinctly, CRA involves the randomised removal and re-insertion of individual atoms (at new random locations) according to a probability distribution, and hence is similar to Frenkel pair formation. An energy minimisation method is then applied after each atom displacement to solve for a potential energy minimum over the entire system. By discounting computationally expensive and time-dependant cascade events, this simplified approach has enabled direct linkage between damage in terms of dpa for even larger systems (millions of atoms), and the evolution of dislocation loop density in pure zirconium up to industrially relevant irradiation doses [105]. However, since CRA is based only upon potential energy minimisation, the kinetic energy (i.e., thermodynamics) of each atom is unaccounted for, and hence, CRA corresponds to a zero Kelvin system which is non-physical. The effects of this simplification are evident for irradiation doses up to 5 dpa, where CRA-predicted dislocation loop densities [105] were more than an order of magnitude greater than those measured experimentally [106]. Currently, the only available physically based methodology for transient irradiation damage which is applicable to industrially relevant length and time scales is cluster dynamics (CD). CD models were first proposed by Gillespie [107] for simulating the transient behaviour of a homogeneous mixture of multi-chemical species, i.e., developed within a chemical kinetics framework. The models were subsequently enhanced by Marian and Bulatov [108] to tackle irradiation defect problems in metals. To integrate microstructural effects, Dunn et al. [109] developed the so-called spatially resolved stochastic cluster dynamics (SRSCD) approach, which incorporates nano- and microscale dependencies, accounting for the proximity of defects to one another and grain boundaries, etc. The basic rate equations used in SRSCD are given by,

$$\begin{aligned}\frac{dC_{v,n}}{dt} &= K_{v,n}(t) + J_v(n-1, t) - J_v(n, t) \\ \frac{dC_{i,n}}{dt} &= K_{i,n}(t) + J_i(n-1, t) - J_i(n, t)\end{aligned}\tag{2.19}$$

where C is the concentration of vacancy (v) and interstitial (i) clusters of size n , K is the generation rate for either vacancy or interstitial clusters of a given size, J -terms are the rates of conversion of clusters from $n - 1$ to n , and from n to $n + 1$, respectively. The rate equations for each cluster type and size are rooted in classical statistical mechanics, which enables computationally efficient simulation of large volumes, despite nano- and microscale discretisation being required to quantify spatially resolved damage. The SRSCD model has been validated against more fundamental methods [110] with great success.

A major literature gap which was recently addressed by Hardie et al. [111] was the disconnect between irradiation damage models and micromechanical behaviour, despite the well-established experimental linkage [112], [113], [114]. Hardie et al. [111] developed a Taylor hardening-based model within a CPFE framework accounting for irradiation hardening and subsequent softening based on probabilistic dislocation loop annihilation. The model could capture the experimental macroscale response as well as micromechanical observations of channel clearing in post-irradiated (various irradiation doses) Zircaloy-4 samples under uniaxial tensile loads. However, significant developmental challenges and scientific questions for coupled behaviour are yet to be addressed, which include: (i) quantifying the impact of irradiation damage on short FCG, (ii) incorporating an irradiation damage evolution model within a micromechanical modelling framework (as opposed to a priori and independent specification of an initial dislocation loop density [111]), (iii) understanding the role of temperature in irradiation damage evolution and associated mechanical behaviour, and (iv) assessing the impact of synergistic irradiation and thermomechanical loads over conventional post-irradiation testing. Challenges (i) – (iv) are revisited in Chapter 6 and briefly in Chapter 9.

2.3 Hydrides

2.3.1 Introduction

As discussed in Chapter 1, waterside cladding oxidation during reactor operation leads to the production of hydrogen (H^+) radicals. Most of them recombine to form hydrogen gas molecules (which is an issue in its own right [115]), while some manage to ingress in the zirconia (oxide) and diffuse through the metallic zirconium matrix. At very low concentrations and temperatures, hydrogen atoms occupy interstitial HCP lattice sites. At sufficient concentrations however, hydrogen radicals react with zirconium to form hydrides, the phases of which can vary depending upon environmental conditions [116]. The hydrogen content in

the bulk required to precipitate hydrides is strongly temperature dependant, as illustrated in Figure 15, which shows the terminal solid solubility for dissolution (TSSD) and precipitation (TSSP) of hydrogen over a broad temperature range, based on differential scanning calorimetry (DSC) measurements [117].

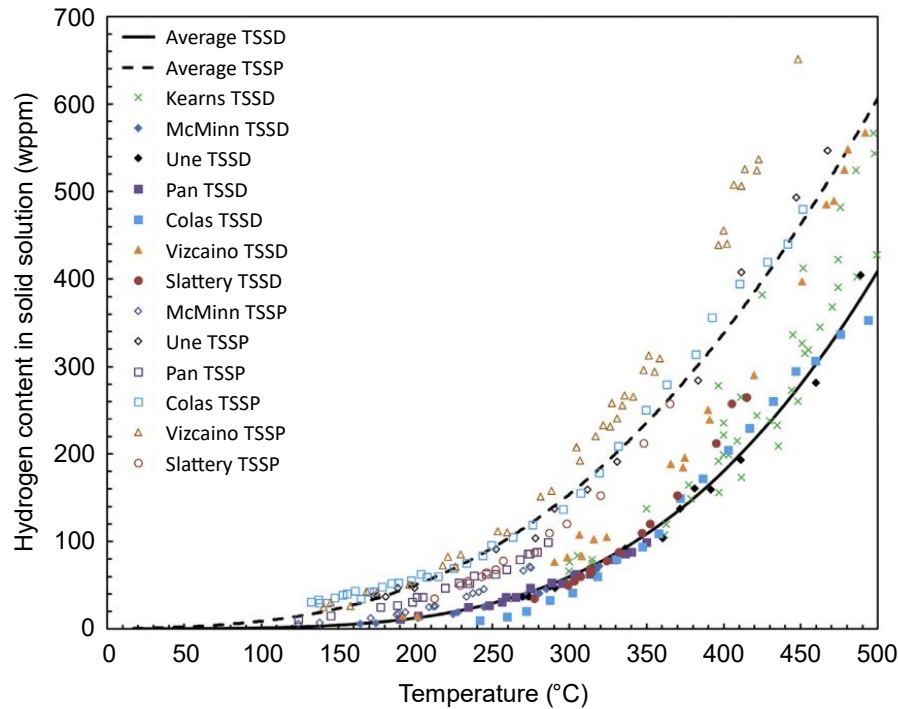


Figure 15: Data from various sources [118], [119], [120], [121], [122], [123], [124] showing the temperature dependence of the terminal solid solubility for dissolution and terminal solid solubility for precipitation of hydrogen in zirconium, collated by Motta et al. [117]⁴.

It is clear from Figure 15 that for a given concentration of hydrogen, the temperature at which hydrides precipitate can be substantially lower than for hydride dissolution. The undercooling required for precipitation is understood to be due to the volumetric or dilatational misfit between e.g., FCC hydrides and the HCP zirconium matrix [125], i.e., there is an associated strain energy for precipitation. In solution, hydrogen transport through cladding materials is driven primarily by the following three factors [126]:

- i) gradients of hydrogen concentration, i.e., Fick's law [127],
- ii) temperature gradients, i.e., the thermophoretic effect,
- iii) and stress gradients.

Note that factors i) – iii) listed above can be described conveniently using a chemical potential gradient formulation [128]. This section describes how through these hydrogen transport

⁴ Reprinted from [117] (Figure 4), with permission from Elsevier (license number 5820771014244).

mechanisms in combination with strong microstructure interactions, hydride precipitation can have detrimental effects on reactor fuel cladding structural integrity, both in in-service and in post-service dry storage conditions. The literature presented here will be focused on the more prevalent delta FCC hydride phase [129].

2.3.2 Morphology and microstructure interaction

The FCC delta hydride phase is the most widely observed and has the most stable atomic structure [129]. Within the zirconium lattice, delta hydrides exhibit specific morphological and crystallographic characteristics. As shown in Figure 16, in their most elementary form, hydrides are thin platelet structures which have an orientation relationship with the underlying matrix. In the early years of fuel cladding development, various researchers investigated the habit plane of hydrides in zirconium, producing a range of contradicting findings which made it a somewhat controversial topic [130]. Langeron and Lehr [131] were the first to publish results in this field, observing hydride plate formation on $\{10\bar{1}0\}$ prismatic planes. In Zircaloy-2 and Zircaloy-4, Westlake [132] first identified the $\{10\bar{1}7\}$ habit plane family, which is inclined to the basal plane at an angle of around 14.7° . Morphologically, it is now understood that such hydrides are actually comprised of multiple layered nano-hydride discs (in what is often referred to as a *deck of cards* configuration), whose orientation is misaligned with the overall macro-hydride *stringer* [133]. Nano-hydride platelets actually lie on HCP basal planes, i.e., $(0002)_\alpha \parallel (111)_\delta$. Perovic et al. [134] presented strong evidence for the *deck of cards* configuration in pure zirconium (see Figure 16 (b)).

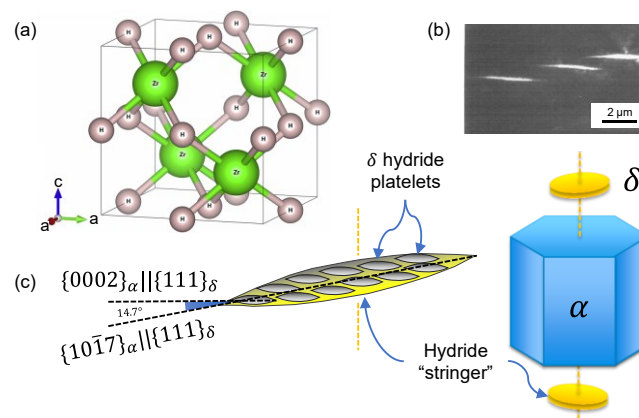


Figure 16: Morphology and orientation relationships of hydrides. (a) Atomic structure of FCC delta hydrides from [135]⁵. (b) Micrograph showing stacking arrangement of hydride platelets [134]⁶. (c) Diagrammatic illustration of orientation relationship between delta hydride platelets, stacked platelets (stringers) and the zirconium matrix.

⁵ Reprinted from [135] (Figure 1), with permission from Elsevier (license number 5820770648499).

⁶ Reprinted from [134] (Figure 13), with permission from Elsevier (license number 5820770854526).

Hydride stringers can however undergo morphological reorientation in the presence of stress (particularly when in combination with thermal cycling); this is known as stress reorientation. Generally, under a tensile stress above some threshold level (typically 70 – 100 MPa [136]), the stacking configuration is altered such that hydride stringers precipitate orthogonally to the stress direction [137]. This is linked with the propensity for hydrogen to diffuse towards regions of elevated hydrostatic stress [138], [139]; tensile hydrostatic stress leads to lattice expansion and hence promotes the accommodation of hydrogen atoms in interstitial (tetrahedral) sites. Cui et al. [140] showed that in notched zirconium-niobium samples, thermomechanical cycling at industrially relevant levels can lead to substantial redistribution and reorientation of hydrides. Figure 17 shows optical micrographs of a notched region of a sample before and after 50 thermomechanical cycles had been applied, which led to substantial redistribution and morphological reorientation of hydrides. This is understood to be due to a cyclic ratcheting effect, whereby the residual hydrostatic stress field (due to local plastic deformation) around a hydride promotes diffusion of additional hydrogen within each successive cycle, and hence hydride growth. It is important to note that the crystallographic orientation relationship (which is controlled by hydride platelet orientation) remains satisfied even after such substantial macroscale developments [141].

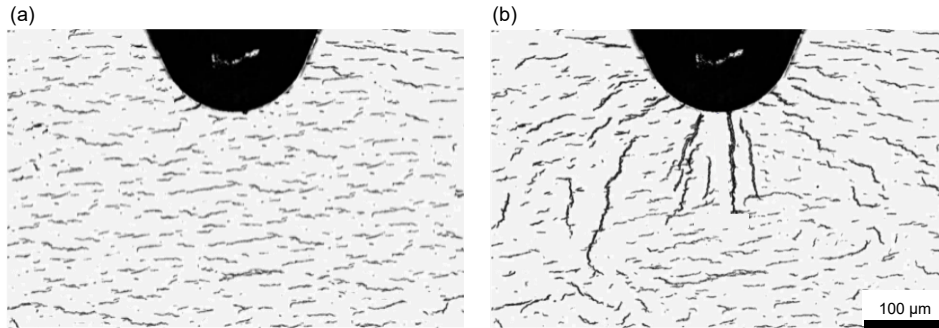


Figure 17: Distribution of hydrides about a stress concentration feature (a) before and (b) after exposure to thermomechanical cycles from [140].

As will be discussed in the next subsection, the hydride stringer configuration (orthogonal to the remote loading direction) shown in Figure 17 (b) is detrimental to cladding structural integrity due to the brittle nature of hydrides [142]. It is largely for this reason that the pilgering process used to manufacture cladding tubes is designed to promote strong textures with HCP c-axes aligned with the radial direction, i.e., such that hydrides which form preferentially on basal planes are parallel with and not orthogonal to the circumferential stress direction [143]. However, macroscale observations such as this provide little insight into the role of microstructure in structural integrity, which is essential for predictive modelling. For

example, grain size can strongly influence how hydrides are distributed throughout the microstructure; where there are large grains ($> 50 \mu\text{m}$), hydrides tend to form predominantly at intragranular locations due to a reduced grain boundary surface area to volume ratio, as compared with fine grain microstructures which are dominated by intergranular hydride precipitation [144]; Qin et al. [145] used thermodynamic modelling to show that it is more energetically favourable for hydrides to precipitate at grain boundaries. Wang et al. [146] demonstrated using electron back-scatter diffraction (EBSD) that grain boundary hydrides often have complex substructures. As shown in Figure 18, grain boundary hydride features can be polycrystalline, consisting of four hydride orientations, i.e., two variants for each HCP grain orientation, both of which satisfy the orientation relationship. Due to symmetry the of orientation variants, some hydride-hydride boundaries are effectively twin boundaries (orientations 3 & 4 and 5 & 6, respectively).

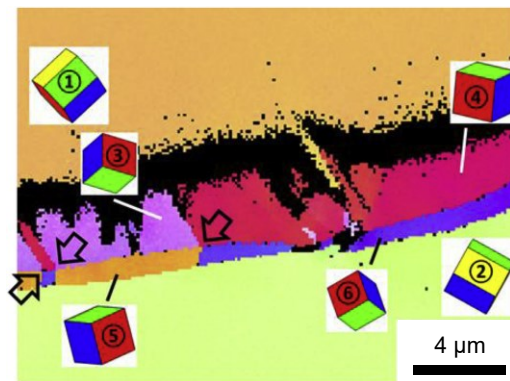


Figure 18: Crystal orientation map of hydride packet feature at a Zircaloy-4 grain boundary [146]⁷.

Hence, fatigue and other failure mechanisms linked with intergranular hydrides and intragranular hydrides (which tend to have a single orientation consistent with the orientation relationship) are likely to differ from one another, though little attention has been given to this distinction in the published literature.

2.3.3 Delayed hydride cracking and embrittlement

In the fuel cladding structural integrity research community, the issue of DHC has been most prevalent, particularly in recent years [147], [148], [149], [150], [151]. DHC is a complex electro-chemo-mechanical crack growth process. Under an applied load, morphological defects, e.g., notches created by non-uniform corrosion or hydride blistering at the cladding outer wall, give rise to stress concentrations. Due to the propensity for hydrogen to diffuse towards regions of

⁷ Reprinted from [146] (Figure 7), with permission from Elsevier (license number 5820770129122).

high tensile stress, these features promote local hydride precipitation. With the appropriate environmental conditions, this steady-state process can drive hydride growth ahead of a stress concentration feature until such a point that brittle fracture will occur spontaneously. Hence, an even greater stress concentration is created, and the process is repeated. DHC is most pervasive during dry (in air) storage of spent nuclear fuel due to an imbalance of internal and external pressures, giving rise to increased circumferential stress [152]. Moreover, the diffusion of hydrogen along negative temperature gradients (from high to low temperature regions) results in a radially non-uniform hydrogen & hydride distribution [5], i.e., hydrogen concentrates at the cladding outer wall, which exacerbates the problem. Recently, Colldeweih et al. [148] used in-situ high-resolution neutron radiography to capture spatially resolved hydrogen concentration measurements during DHC of Zircaloy-2 at various temperatures (ranging between 200°C and 360°C). Sections of cladding were fatigue pre-cracked and charged with hydrogen to 200 wppm (in the bulk). Under an applied load, neutron radiography measurements showed that the local crack tip hydrogen concentration would range between 1000 to 3000 wppm, with higher values corresponding to higher temperatures. A previous study by Colldeweih et al. [149] used a similar methodology, but with an increasing applied load to measure the critical SIF for DHC to occur. The critical value was shown to be effectively independent of bulk content, and mildly dependant on temperature, in the range 6 – 10 MPa√m. While DHC is an interesting problem, it has been reasonably well characterised in terms of process parameters, etc. Moreover, it is largely a problem most fitted to continuum level analyses and hence, there is arguably little room for further scientific innovation.

For plant life extension, cladding performance during in-service conditions is most relevant. The issues of fatigue crack nucleation and short FCG in hydrided zirconium alloys have been recognised recently as key strategic research topics [153], [154]. Li et al. [154] addressed the effect of hydride stress reorientation on fatigue crack initiation and growth behaviour. Specifically, the behaviour of clad tubing with circumferential hydrides was compared with tubing with a 4:1 circumferential-radial hydride distribution. The reduction in fatigue life of hydride-reoriented material was quantified, and advanced characterisation work was used to understand the mechanisms responsible for the reduction, including more generally, crack growth mechanisms in intergranular and intragranular hydrides. However, the influence of hydrides on fatigue performance at the microstructural level has not yet been quantified by comparison with non-hydrided materials; this will be addressed in Chapter 7 of this thesis.

Moreover, on the issue of hydride ratcheting, published experimental studies tend to focus on qualitative observations at the macroscale, without mechanistic understanding or quantitative measurements to inform microstructural computational models [155], [156]. Hence, this understanding gap will also be addressed in Chapter 7 of this thesis.

3 Methodology

3.1 Introduction

This chapter provides an overview of computational and experimental methods which are applied within subsequent thesis chapters. The methods presented here are broadly existing micromechanical methods and have been presented previously. New methods and adaptations to existing methods which have been developed for this thesis will be presented as such in subsequent chapters. The crystal plasticity formulation (and its implementation as a user-material subroutine in *Abaqus*) presented here is mostly based on work by Dunne et al. [157] in which a user-element subroutine code was developed for titanium alloys. The current code has been developed over the past decade and a half by various researchers at Oxford University and Imperial College London. Similarly, much of the experimental methodology and equipment used here is the result of years of developmental work by previous researchers.

3.2 Computational

3.2.1 *Crystal plasticity-based constitutive model*

Large scale deformation of solids is described here by the deformation gradient, which is given in Equation 3.1; this accounts for deformation of a material both in terms of elongation (along a set of orthogonal axes) and rigid body rotation.

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}} = \begin{pmatrix} \frac{\partial x}{\partial X} & \frac{\partial x}{\partial Y} & \frac{\partial x}{\partial Z} \\ \frac{\partial y}{\partial X} & \frac{\partial y}{\partial Y} & \frac{\partial y}{\partial Z} \\ \frac{\partial z}{\partial X} & \frac{\partial z}{\partial Y} & \frac{\partial z}{\partial Z} \end{pmatrix} \quad (3.1)$$

where $\mathbf{x}$ describes the deformed state (orientation and scale) at a point and $\mathbf{X}$ the undeformed state. The constitutive framework of the crystal plasticity model implemented in this thesis makes use of Lee's multiplicative decomposition [158] of the deformation gradient from continuum mechanics, as shown in Equation 3.2.

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p \quad (3.2)$$

where $\mathbf{F}^e$ and $\mathbf{F}^p$ are the elastic and plastic deformation gradients, respectively. The elastic deformation gradient is used to determine the stress state via Hooke's Law (see Equation 3.3) and is given by inverting the plastic deformation gradient, $\mathbf{F}^e = \mathbf{F} \mathbf{F}^p{}^{-1}$. The elastic (Green)

strain tensor is then simply $\boldsymbol{\varepsilon}^e = \frac{1}{2}(\mathbf{F}^{eT}\mathbf{F}^e - \mathbf{I})$. In Equation 3.3, the three-dimensional stress ($\boldsymbol{\sigma}$) and strain ($\boldsymbol{\varepsilon}^e$) tensors are represented using Voigt notation (enabled by symmetry of the shear terms); in doing so, the stiffness matrix, $\mathbf{C}$, is reduced from having 81 components to just 36. Of the 36 terms, just 21 unique terms are required to produce a full stiffness matrix (also due to symmetry), e.g., for anisotropic HCP Zircaloy-4. In Equation 3.3, the shear terms in $\boldsymbol{\varepsilon}^e$ represents the mathematical shear strains (as opposed to engineering shear strains, which are used in *Abaqus*).

$$\boldsymbol{\sigma} = \mathbf{C}\boldsymbol{\varepsilon}^e = \begin{bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{31} \\ \sigma_{12} \end{bmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & C_{14} & C_{15} & C_{16} \\ C_{21} & C_{22} & C_{23} & C_{24} & C_{25} & C_{26} \\ C_{31} & C_{32} & C_{33} & C_{34} & C_{35} & C_{36} \\ C_{41} & C_{42} & C_{43} & C_{44} & C_{45} & C_{46} \\ C_{51} & C_{52} & C_{53} & C_{54} & C_{55} & C_{56} \\ C_{61} & C_{62} & C_{63} & C_{64} & C_{65} & C_{66} \end{bmatrix} \begin{bmatrix} \varepsilon_{11}^e \\ \varepsilon_{22}^e \\ \varepsilon_{33}^e \\ \varepsilon_{23}^e \\ \varepsilon_{31}^e \\ \varepsilon_{12}^e \end{bmatrix} \quad (3.3)$$

In the current model, it is assumed that all plastic deformation is accommodated for by dislocation glide or slip. This is a process in which the application of a local shear stress (of sufficient magnitude) results in the activation of a dislocation slip system and hence, dislocation motion. The following mechanistic slip rule [159] is introduced to capture the rate of plastic shear on a given slip plane, i .

$$\dot{\gamma}^i = \rho_m \omega b^2 \exp\left(\frac{-\Delta F}{k_B T}\right) \sinh\left(\frac{(\tau^i - \tau_c^i)\Delta V}{k_B T}\right) \quad (3.4)$$

where $\dot{\gamma}^i$ is the rate of plastic shear strain, ρ_m is mobile dislocation density, ω is frequency of attempts for dislocations to surmount thermal energy barriers, b is the systeurgers vector magnitude, ΔF is activation energy. τ_c^i is CRSS (and τ^i is applied shear stress for the corresponding slip system), ΔV is activation volume and k_B and T have their usual meanings. This slip rule originates from Gibbs' [160] statistical mechanics work on thermally activated escape of pinned dislocations. To glide, pinned dislocations must overcome thermal energy barriers presented by obstacles, the probability of which is controlled by the activation energy (or Helmholtz energy), ΔF ; together with the frequency of 'jump' attempts, ν , and work done by the stress field, $(\tau^i - \tau_c^i)\Delta V$, the rate of plastic shear can be determine. Activation energy and activation volume are also therefore the most important terms controlling strain-rate sensitivity of the material [161]. Physically, the activation volume represents the volume of material involved in the thermally activated glide of a dislocation [162], and is therefore linked

with the pinning distance, l [157], [163]. It is important to note that Equation 3.4 only applies where the applied shear stress applied over a given slip system is greater than the corresponding critical value; otherwise, $\dot{\gamma}^i = 0$. The contributions of individual slip systems to overall plastic strain are then accounted for within the plastic velocity gradient, $\mathbf{L}^p$, which is calculated using Schmid's Law, as shown in Equation 3.5.

$$\mathbf{L}^p = \sum_{i=1}^M (\dot{\gamma}^i \mathbf{n}^i \otimes \mathbf{s}^i) \quad (3.5)$$

where M is the total number of slip systems and $\mathbf{n}^i$ and $\mathbf{s}^i$ represent the individual slip system normal and slip direction vectors, respectively. The $\otimes$ function represents the Kronecker product of vectors $\mathbf{n}^i$ and $\mathbf{s}^i$, and produces a tensor. The plastic velocity gradient is then related back to the plastic deformation gradient by Equation 3.6.

$$\mathbf{L}^p = \dot{\mathbf{F}}^p \mathbf{F}^{p-1} \quad (3.6)$$

The various HCP slip systems are illustrated in Figure 19 in relation to the atomic crystallography of HCP materials.

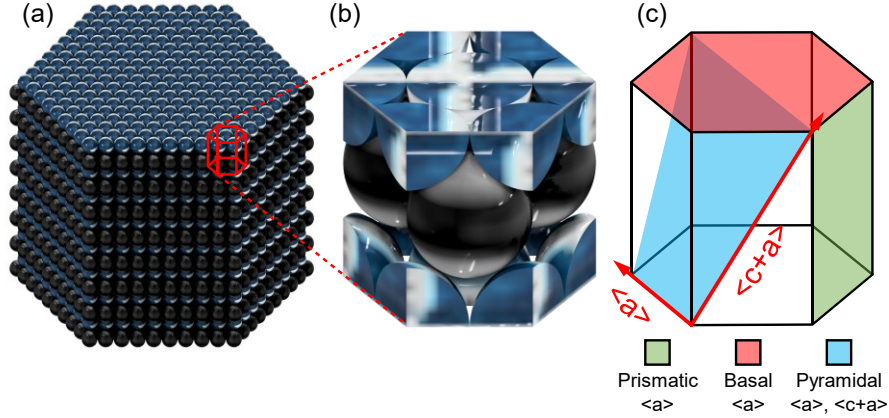


Figure 19: HCP crystallography. (a) Schematic representation of HCP atomic structure. (b) Repeating unit cell of HCP crystal. (c) Major HCP slip systems represented within unit cell (only first order pyramidal slip is shown here).

There are a total of 30 HCP slip systems, and 5 slip system types (prismatic, basal, first order pyramidal ($\langle a \rangle$ and $\langle c+a \rangle$), and second order pyramidal ($\langle c+a \rangle$)). Evidently, the slip strengths, τ_c^i , are highly important material properties in the current model, and have therefore been obtained from direct experimental measurements in the literature [164]. The classic Taylor hardening model [165] is used within the crystal plasticity framework to account for plastic strain hardening due to dislocations, as shown in Equation 3.7,

$$\tau_c^i = \tau_{c,0}^i + \mu b \sqrt{\rho_{\text{SSD}} + \rho_{\text{GND}}} \quad (3.7)$$

where $\tau_{c,0}^i$ is the intrinsic ‘obstacle-free’ CRSS from experiments, μ is shear modulus and ρ_{SSD} and ρ_{GND} are total area densities of statistically stored dislocations and GNDs, respectively. Here it is assumed that all dislocations contribute to hardening across all slip systems [166]. The evolution of SSD density is controlled by a simple linear hardening rule, $\rho_{\text{SSD}} = \rho_{\text{SSD},0} + \gamma^{\text{st}} p$ [73], where $\rho_{\text{SSD},0}$ is the initial SSD density (pre-deformation), γ^{st} is a material constant and p is total cumulative plastic strain. Calculating GND densities which arise from lattice curvature is much more involved and is described in the next section. It is assumed that both SSDs and GNDs contribute equally to hardening on all slip systems.

3.2.2 The eXtended Finite Element Method (XFEM)

XFEM is an extension of the traditional numerical finite element method which was developed to deal with discontinuities imposed by cracks, for example. With the conventional finite element method, cracks can be represented by the surrounding mesh, i.e., nodes positioned along the crack plane and at crack tips. To capture the asymptotic stress and strain fields associated with cracks, significant mesh refinement is required in the vicinity of crack tips. In addition, to model crack propagation, continual remeshing is required to maintain local refinement around the new crack path. Projection of results from old to new meshes is therefore required, which is highly computationally expensive. Hence, XFEM was developed by Belytschko and Black [167] to overcome this problem; XFEM uses the partition of unity concept, which is that a set of n shape functions N_i satisfy the following relationship,

$$u(x) = \sum_{i=1}^n N_i(X) u_i \quad (3.8)$$

where u_i are the finite element degrees of freedom. Using the well understood characteristics of crack tip displacement fields, for example, XFEM enriches the finite element space local to the crack tip as follows,

$$u(x) = \sum_{i=1}^n N_i(X) u_i + \sum_{i=1}^n N_i(X) \sum_{j=1}^k p_j a_{ij} \quad (3.9)$$

where p_j are the enrichment functions and a_{ij} are the added degrees of freedom. The *Abaqus* implementation of XFEM applies enrichment functions to elements which are fully bisected by cracks. Hence, so-called jump enrichment functions are added to create a discontinuity at

either side of a bisected element. The main advantages of this method are that (i) the solution is relatively mesh independent and (ii) continual remeshing is not required.

3.2.3 Finite element implementation of strain gradient plasticity for GND density

3.2.3.1 GND density from plastic strain gradients

The elastic and plastic anisotropy of HCP metals results in non-homogeneous grain-scale deformation under load. As a result, spatial gradients of deformation are imposed by microstructures, and are concentrated at grain boundaries and other microstructural features. This is also true for geometric inhomogeneities such as cracks, which generate severe strain localisation and hence strain gradients. GNDs arise to accommodate plastic deformation gradients [168] to enable compatibility and minimise long-range stresses between e.g., neighbouring grains. Ashby [169] showed how GND density is linked with spatial gradients of plastic strain for a simple case of single slip in a material under pure bending. The 1D analytical solution is given in Equation 3.10,

$$\rho_{\text{GND}} = \frac{1}{b} \frac{\partial \gamma}{\partial s} \quad (3.10)$$

where γ is the shear strain and s is the direction of single slip in this idealised scenario. For 3D analyses, Nye's dislocation tensor [170], [171], $\mathbf{\Lambda}$, is introduced, and is given by the curl of the plastic deformation gradient as follows,

$$\mathbf{\Lambda} = \nabla \times \mathbf{F}^p = \sum_{i=1}^M \rho_{\text{GND},s}^i \mathbf{b}^i \otimes \mathbf{s}^i + \rho_{\text{GND},et}^i \mathbf{b}^i \otimes \mathbf{t}^i + \rho_{\text{GND},en}^i \mathbf{b}^i \otimes \mathbf{n}^i \quad (3.11)$$

where $\rho_{\text{GND},s}^i$, $\rho_{\text{GND},et}^i$, and $\rho_{\text{GND},en}^i$ are (decomposed) groups of screw and two edge components (tangential and normal) of GND density, respectively; $\mathbf{t}^i$ is the edge dislocation tangent vector group given by $\mathbf{t}^i = \mathbf{s}^i \times \mathbf{n}^i$. To solve for the 90 unique components of $\boldsymbol{\rho}_{\text{GND}}$, we can write $\hat{\mathbf{\Lambda}} = \mathbf{A} \boldsymbol{\rho}_{\text{GND}}$, where $\hat{\mathbf{\Lambda}}$ is the vectorised Nye tensor (9×1) and $\mathbf{A}$ is a 9×90 matrix containing the coefficients $\mathbf{b}^i \otimes \mathbf{s}^i$, $\mathbf{b}^i \otimes \mathbf{t}^i$, and $\mathbf{b}^i \otimes \mathbf{n}^i$. This produces an underdetermined system of equations, since there are 90 independent $\boldsymbol{\rho}_{\text{GND}}$ components and just 9 equations. Hence, the L_2 norm minimisation strategy from Cheng and Ghosh [171] is adopted here, subject to the $\mathbf{A} \boldsymbol{\rho}_{\text{GND}} = \bar{\mathbf{\Lambda}}$ constraint, giving Equation 3.12.

$$\rho_{\text{GND}} = \sqrt{\sum_{i=1}^M (\rho_{\text{GND},s}^i)^2 + (\rho_{\text{GND},et}^i)^2 + (\rho_{\text{GND},en}^i)^2} \quad (3.12)$$

Only this scalar representation of GND density, ρ_{GND} , will be used hereafter.

3.2.3.2 The sub-element method for calculating GND density with linear shape functions

As will be demonstrated in this thesis, GND density is a highly important physical quantity for analysis of short FCG. However, coupling the methods described in the previous section with XFEM (used here to model crack growth) is non-trivial, particularly since XFEM is only currently available with finite elements with linear shape functions, i.e., C3D8 eight-node linear brick elements with full integration ($2 \times 2 \times 2$) are used in this thesis. Hence, this presents a problem, as it is not possible to calculate second order spatial derivatives using linear shape functions alone. Dunne et al. first presented the sub-element method [157] for eight-node reduced integration 2D elements. As illustrated in Figure 20, (for 3D elements), the method involves defining a sub-element internally within each finite element with eight new nodes at each Gauss point, where the plastic deformation gradient, $\mathbf{F}^p$ is known.

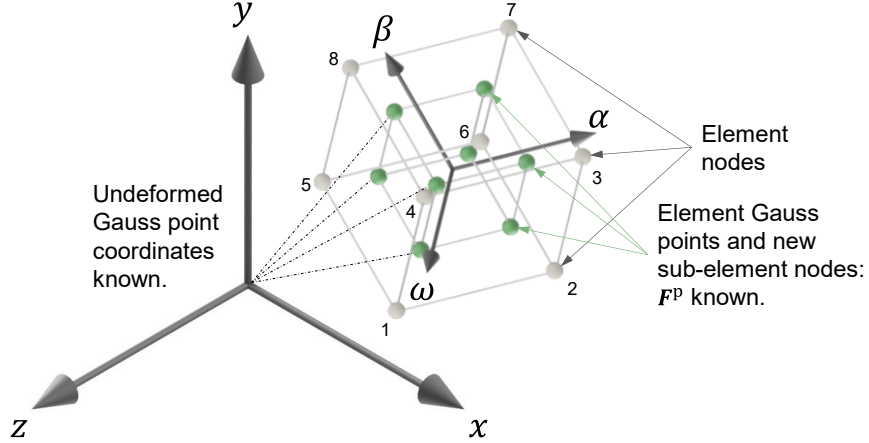


Figure 20: Schematic showing sub-element method used to calculate spatial gradients of plastic deformation in 3D eight-node linear brick elements (C3D8). The figure shows that the nodes of the internal (sub) element are defined at the Gauss points of the original elements.

There is then a spatial distribution of plastic deformation between nodes of the sub element, which can be described simply using linear shape functions, $\mathbf{r}$, within the local coordinate system, $\boldsymbol{\alpha}$. The derivatives of plastic deformation within the global coordinate system, $\mathbf{x}$, are then related to the local coordinate system as follows [157],

$$\frac{\partial \mathbf{F}^p}{\partial \mathbf{x}} = \frac{\partial}{\partial \mathbf{x}} (\mathbf{r} \mathbf{F}_j^p) = \frac{\partial \mathbf{r}}{\partial \mathbf{x}} \mathbf{F}_j^p = \frac{\partial \mathbf{r}}{\partial \boldsymbol{\alpha}} \mathbf{J}^{-1} \mathbf{F}_j^p \quad (3.13)$$

where $\mathbf{F}_j^p$ is the plastic deformation gradient at Gauss point, j . $\mathbf{J}$ is the Jacobian matrix, which maps from the external to the internal elements and is given by $\mathbf{J} = \mathbf{G}^x \frac{\partial \mathbf{r}}{\partial \boldsymbol{\alpha}}$, where $\mathbf{G}^x$ is a 3×8 matrix containing the global Gauss point coordinates for each element.

3.2.4 The stored energy density fracture criterion

3.2.4.1 Stored energy density

As discussed in Chapter 2, SED has been identified as and demonstrated as the most reliable and microstructure-sensitive driver currently available for predicting both fatigue crack nucleation [67], [172] and short fatigue crack propagation [34], [73]. Hence, it is utilised here as the fracture criterion in studies of short crack growth. SED is a measure of the energy required to nucleate or extend a crack per unit area of crack extension and is measured in J / m². The mechanistic basis for the SED fracture criterion is loosely based on Griffith's theory [28], which states that there is some critical level of energy required to create new surfaces during crack growth; in ductile metals, the required energy is stored elastically (and hence recoverably) in dislocation structure and accumulates cyclically during plastic deformation. Since CPFE is a numerical method, quantities such as total slip for a given slip system are computed locally and incrementally, using numerical integration, giving spatial and temporal resolution. Similarly, SED is calculated within the CPFE code using numerical integration of SED increments as follows,

$$G = \xi \int \frac{\boldsymbol{\sigma} : d\boldsymbol{\varepsilon}^p}{\sqrt{\rho_{SSD} + \rho_{GND}}} \quad (3.14)$$

where $\boldsymbol{\sigma}$ is the stress tensor and $d\boldsymbol{\varepsilon}^p$ is plastic strain increment (corresponding to step increment in finite element modelling). For very short cracks ($< 10 \mu\text{m}$), GND density is thought to be the dominant source of crack tip dislocation structure due to highly localised lattice curvature [173]. ξ is the recoverable fraction (stored) of plastic strain energy, which is stored elastically; its value is not known precisely, but is assumed here to equal 5%, and to be independent of loading conditions etc. [68]. Crack propagation is simulated using XFEM and is enabled when the value for SED ahead of the crack tip exceeds a critical value, G_c , which is a material constant. For this, the user-defined damage initiation (UDMGINI) subroutine is employed; when the value for SED at the centroid of the finite element ahead of the crack tip

exceeds the predefined critical value, the element will fracture. In Chapter 4, significant changes are made to the code to incorporate new crack path prediction methods.

3.2.4.2 *The non-local averaging method*

The local SED is calculated using Equation 3.14. To mitigate erroneous SED calculations at integration points, due to either spurious GND density calculations (due to use of linear elements) and / or finite element mesh anomalies, Wilson and Dunne [34] introduced a non-local averaging scheme based on the work of Nguyen et al. [174]. This is done using a summation (volume integration) of local SED values, which is carried out about each integration point over a given distance, l . A weighting function is implemented which takes a maximum value of 1 at distance, $r = 0$ and minimum at $r = l$, and is given by $g(r) = \exp\left(\frac{-\pi r^2}{l^2}\right)$. The non-local SED is given by Equation 3.15.

$$G_{\text{NL}} = \sum_{i=1}^n G_i \cdot g(r) \quad (3.15)$$

where G_i is local SED at integration point i and n is the total number of integration points within the non-local volume. G_{NL} is used as the fracture criterion here in crack growth studies. Clearly, the nonlocal radius l is an important parameter, and is therefore selected based on the approximate size of the GND density field ahead of a microstructurally short crack (since most energy available to a crack tip is stored in the GND structure which surrounds it). It has been shown [73] that even across various anisotropic HCP single crystal orientations, GNDs are mostly confined to a local radius l about crack tips. The use of a SED fracture criterion is highly advantageous as it is relatively mesh independent due to the non-local averaging scheme and since crack tip SED is non-asymptotic [175].

3.2.5 *Criterion for short fatigue crack path: to be continued*

As suggested by the title of this subsection, the criterion for crack path presented here will be challenged in a subsequent chapter. To begin with, the maximum slip criterion for short crack path is used and has been used with reasonable success in various previous studies [74], [176], [177]. However, as is demonstrated in the next chapter, the maximum slip criterion is not always sufficient and hence, alternative methods are developed which will be described in detail. From Equation 3.4, the total accumulated slip is calculated for each HCP slip system i , as follows,

$$\gamma^i = \int \dot{\gamma}^i dt \quad (3.16)$$

where dt is the time increment. Hence, according to the maximum slip criterion, the crack will extend along the slip plane of the system with maximum slip magnitude ahead of the crack tip, the orientation of which is given by its unit normal vector $\mathbf{n}$ (which depends upon grain orientation). The mechanistic basis for this is that planes with greatest damage, i.e., slip are most susceptible to cracking.

3.2.6 User-material subroutine implementation of the constitutive model

To solve for the highly non-linear force-displacement behaviour within Zircaloy-4 microstructures under plastic deformation, an implicit (iterative) integration scheme [178] is employed here, and hence, all quantities are computed at the end of each time increment, $t + \Delta t$. For each iteration, a trial stress, σ^{tr} , is calculated, and is adjusted by the plastic corrector term as follows,

$$\sigma = D\epsilon^e = D(\epsilon_t^e + \Delta\epsilon_{t+\Delta t}^e) = D(\epsilon_t^e + \Delta\epsilon_{t+\Delta t}^e) - D\Delta\epsilon_{t+\Delta t}^p = \sigma^{\text{tr}} - D\Delta\epsilon_{t+\Delta t}^p \quad (3.17)$$

where D is the global stiffness matrix, given by rotating the local stiffness matrix, C , and $D\Delta\epsilon^p$ is the plastic corrector term. Since the plastic strain increment is unknown, the trial stress is approximated by linear elastic extrapolation using the total strain; note that for simulations in which there are temperature changes, the inelastic thermal strains are deducted from the total strain when calculating the trial stress as follows, $\sigma^{\text{tr}} = D(\epsilon_t^e + \Delta\epsilon_{t+\Delta t}^e - \Delta\epsilon_{t+\Delta t}^{\text{th}})$, where $\Delta\epsilon_{t+\Delta t}^{\text{th}}$ is the thermal strain increment. Hence, an error function based on residual stress is given by Equation 3.18.

$$\psi = |\sigma - \sigma^{\text{tr}} - D\Delta\epsilon_{t+\Delta t}^p| = 0 \quad (3.18)$$

A Newton-Raphson iterative scheme is employed which solves for stress by minimising the error function; hence, the stress is updated within each iterative loop by the derivative of the error function with respect to stress as, $\frac{\partial\psi}{\partial\sigma} = \left[I + \frac{\partial\epsilon_{t+\Delta t}^p}{\partial\sigma} \right]$, which gives,

$$\psi + \frac{\partial\psi}{\partial\sigma} \delta\sigma = |\sigma - \sigma^{\text{tr}} - D\Delta\epsilon_{t+\Delta t}^p| + \left[I + \frac{\partial\epsilon_{t+\Delta t}^p}{\partial\sigma} \right] \delta\sigma = 0 \quad (3.19)$$

where $\delta\sigma$ is the stress corrector term and is given by $\delta\sigma = \delta\sigma^{\text{tr}} - \mathbf{D}\delta\Delta\epsilon_{t+\Delta t}^{\text{p}}$, which yields Equation 3.20 [157].

$$\delta\sigma = - \left[\mathbf{I} + \frac{\partial \epsilon_{t+\Delta t}^{\text{p}}}{\partial \sigma} \right]^{-1} \mathbf{D}(\Delta\epsilon_{t+\Delta t}^{\text{e}} + \Delta\epsilon_{t+\Delta t}^{\text{p}} - \Delta\epsilon_{t+\Delta t}) \quad (3.20)$$

Within each Newton loop, the updated stress gives the resolved shear stress for each slip system and hence, the rate of plastic shear for each slip system also. This then gives the plastic velocity gradient, which in turn updates the plastic deformation gradient. As discussed earlier, the elastic deformation gradient can be computed simply using Lee’s multiplicative decomposition [158]. The stress is then updated, and the iterative process continues until $\psi \leq \text{tol}$, where **tol** is some very low tolerance value (10^{-12} MPa).

3.2.7 Zircaloy-4 material properties

To accurately capture grain-scale deformation of materials (particularly HCP materials), it is vital that crystallographic elastic anisotropy is fully accounted for. While most modelling work presented in this thesis is at room temperature, both room temperature and temperature dependent elastic constants are given in Table 2. The values presented here were previously reported and validated against macroscopic experimental stress-strain data [161]. Temperature-dependent relationships were extracted from Fisher and Renken’s work [179] and were approximated as linear functions over the temperature range 293 K to 623 K.

Table 2: Room temperature and temperature dependent anisotropic elastic properties of single crystal Zr alloys.

Property	Value at room temperature	Temperature T (K) dependent value [179]	Units
E_1	98.308	$-0.0755T + 120.4$	GPa
E_3	132.208	$-0.0372T + 132.9$	GPa
G_{12}	32.010	$-0.0233T + 38.8$	GPa
ν_{12}	0.4006	$3.4273 \times 10^{-4}T + 0.3002$	-
ν_{13}	0.2375	$-9.1182 \times 10^{-5}T + 0.2642$	-

Just as with elastic deformation, grain-scale plasticity in Zr alloys is highly anisotropic. Room temperature CRSS values for various slip systems which were measured experimentally by Gong et al. [164] using micro cantilever testing are used here (see Table 3). Subsequent high temperature modelling work by Liu et al. [161] enabled extraction of a temperature dependent prismatic CRSS; assuming the ratio of prismatic CRSS to that of other slip systems remains constant at elevated temperatures, a simple linear approximation is needed to obtain high

temperature basal and pyramidal slip strengths, as shown in Table 3. The values reported here were previously shown [180] to yield excellent agreement with published experimental data [181].

Table 3: Room temperature and temperature dependent CRSS values for Zr alloys.

Property	Value at room temperature	Temperature dependent value	Units
$\tau_{c,0}^{\text{prism}}$	153	$26.3 \times \exp(514.7/T)$	MPa
$\tau_{c,0}^{\text{basal}}$	203	$1.33 \tau_{c,0}^{\text{prism}}(T)$	MPa
$\tau_{c,0}^{\text{pyramidal}<c+a>}$	532	$3.48 \tau_{c,0}^{\text{prism}}(T)$	MPa

For the mechanistic slip rule (Equation 3.4), the various physical constants are presented in Table 4. Due to there being negligible change to the strain-rate sensitive parameters, ΔV , ΔF , at elevated temperatures [182], it is assumed that these values are constant in this work (in the temperature range 293 – 613 K). Similarly, Burgers vector magnitudes and mobile dislocation density are assumed unaffected by temperature. Mobile dislocation density ρ_m is assumed constant since the rates of dislocation nucleation, annihilation, and pinning are unknown at CPFE length scales.

Table 4: Constants used in mechanistic slip rule, assumed temperature independent.

Property	Symbol	Value	Units
Mobile dislocation density	ρ_m	0.01	μm^{-2}
Escape attempt frequency	ω	1.0×10^{11}	Hz
Type <a> Burgers vector	$b_{<a>}$	3.2×10^{-4}	μm
Type <c+a> Burgers vector	$b_{<c+a>}$	$1.593 b_{<a>}$	μm
Activation energy	ΔF	0.32	eV
Activation volume	ΔV	20.93	b^3
Boltzmann constant	k_B	1.381×10^{-23}	JK^{-1}

Lastly, a very important material property for predicting rates of crack growth in Zircaloy-4 is the critical SED for fracture, G_c . The first estimate of its value, and the methodology by which it is extracted are later presented in this thesis.

3.3 Experimental

3.3.1 Micro-thermo-mechanical testing

3.3.1.1 Notched three-point bending

For studies of short fatigue crack propagation and thermo-mechanical ratcheting effects in hydrided Zircaloy-4, force-controlled three-point bending is used. As illustrated in Figure 21,

samples are notched at the centre to generate stress concentrations sufficient to initiate and propagate cracks within hundreds of cycles, and to ensure stress-driven hydrogen redistribution during thermo-mechanical testing.

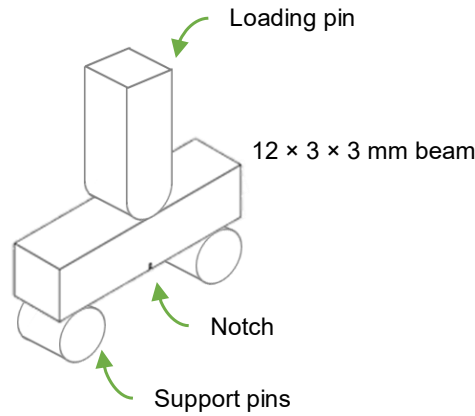


Figure 21: Illustration of three-point bending configuration. The notch, which is visible at the front surface, extends through the sample thickness (nominally 3 mm). Support pins are 3 mm in diameter and the loading pin tip radius is 1.5 mm. Spacing between support pins is 10 mm. Notch height and radius are approximately 300 μm and 100 μm , respectively.

The $12 \times 3 \times 3$ mm beam design enables samples to be easily mounted on SEM stubs for imaging and EBSD characterisation both prior to and after testing. Notch geometry and applied load are used to tailor the notch tip stress (or equivalent SIF for FCG) for each test. As discussed in subsequent sections, in-situ DIC is performed across a region of interest local to the notch for each test. EBSD characterisation of the same region of interest enables linkage between micromechanical test data and microstructure and crystallography for mechanistic insights at the appropriate length-scale into factors which influence localisation of strain and cracking in hydrided Zircaloy-4.

3.3.1.2 High temperature testing

The three-point bending rig was designed to allow beam heating for coupled force and temperature control [183]. As shown in Figure 22, air-cooled 250 W infrared (IR) lamps (from *Precision Control Systems Ltd.* [184]) are positioned at either end (3×3 mm ends) of the beam; this configuration was previously shown to provide reasonably uniform heating across the beam length [183]. For temperature control, K-type thermocouples are spot-welded to the rear face of each beam, which feed into a full proportional-integral-derivative (PID) control loop [185]. A *Eurotherm 2416* controller is used to control both the power provided to the lamps, and pneumatic solenoid valves which control air flow to the lamps as well as a separate cold air

supply for quenching (quenching is not used here). An air compressor with appropriate filtration is used to provide a steady flow of air at approximately 30 L/min per lamp.

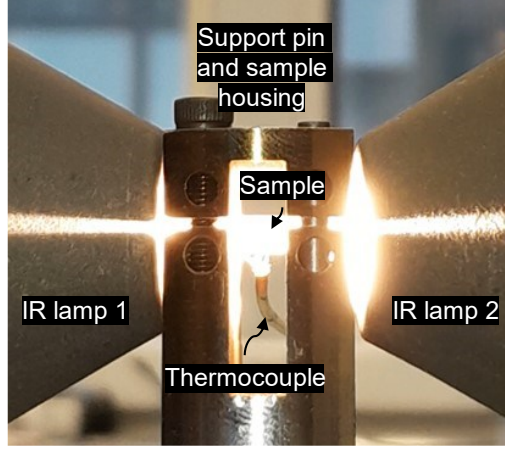


Figure 22: IR lamp configuration in heated and unloaded state (load pin is not in contact with sample).

PID control is a highly robust and responsive feedback-based control mechanism and is widely used for a range of industrial applications. Each element of PID refers to the adjustments made to the control variable (which is lamp current, here) in response to the process variable (temperature) error measurements. The following equation [185] describes how the process variable, I , varies according to each of the three elements of PID control,

$$I(t) = K_p e(t) + K_i \int e(t) dt + K_d \frac{de(t)}{dt} \quad (3.21)$$

where K_p is the proportional gain, K_i the integral gain, K_d the derivative gain, and e is the error (which is the difference between the set-point temperature and measured temperature for this application). The proportional component adjusts the control variable directly against the error magnitude, and hence K_p must be carefully selected to avoid lag (if K_p is too low) or substantial overshoot, leading to instability (if K_p is too high). Time dependence is incorporated by the integral and derivative functions, which consider the error history, and error-time gradient, respectively. Generally, Equation 3.21 is written in terms of the proportional gain as follows,

$$I(t) = K_p \left(e(t) + \frac{1}{T_i} \int e(t) dt + T_d \frac{de(t)}{dt} \right) \quad (3.22)$$

where T_i and T_d are the integration time constant ($T_i = \frac{K_p}{K_i}$), and derivative time constant ($T_d = \frac{K_d}{K_p}$), respectively, and have a physical meaning. For an initial estimate of the PID control

constants required to control beam temperature, a simple thermal model is developed based on Poole's work [183]. A lumped capacitance approach [186] is first used which considers heat transfer to the beam based on lamp heat flux, q''_{lamp} , and convective and radiation heat transfer losses away from the beam (see Figure 24 (a)). Convective heat transfer losses are estimated using Newton's law of cooling [187], which is given by,

$$q''_{\text{conv}} = h(T - T^\infty) \quad (3.23)$$

where h is the convective heat transfer coefficient, T is temperature, and T^∞ is the coolant (ambient air) temperature. The convective heat transfer coefficient is taken as $30 \text{ Wm}^{-1}\text{K}^{-1}$, which is typical for convective air heat transfer. Radiation heat transfer is modelled using the Stefan-Boltzmann law of radiation [188], which is given by,

$$q''_{\text{rad}} = \epsilon\sigma_{\text{SB}}(T - T^\infty)^4 \quad (3.24)$$

where ϵ is emissivity, and σ_{SB} is the Stefan-Boltzmann constant ($\sigma_{\text{SB}} = 5.6 \times 10^{-8} \text{ Wm}^{-2}\text{K}^{-4}$). Writing Equations 3.23 and 3.24 to into a balanced time dependant heat flow equation gives,

$$q = q_{\text{lamp}} - q_{\text{conv}} - q_{\text{rad}} = q''_{\text{lamp}}A_{\text{heated}} - (q''_{\text{conv}} + q''_{\text{rad}})A_{\text{beam}} = m_{\text{beam}}c_p \frac{dT}{dt} \quad (3.25)$$

where q represents the heat flow rate, A_{ends} and A_{beam} represent the heated and total surface areas of the beam, respectively, m_{beam} the beam mass, and c_p the specific heat capacity. Using a simple time-step integration scheme in *Matlab*, and the properties given in Table 5, the transient behaviour of the system is modelled at 4% of the maximum lamp heat flux ($q''_{\text{lamp,max}} = 1.7 \times 10^6 \text{ Wm}^{-2}$). The ambient and initial air temperature are set at 293 K.

Table 5: Zircaloy-4 beam properties used in lumped capacitance model and thermal finite element analyses.

Property	Symbol	Value	Units
Emissivity [189]	ϵ	0.45	-
Beam mass	m_{beam}	7.08×10^{-4}	kg
Specific heat capacity [190]	c_p	362	$\text{Jkg}^{-1}\text{K}^{-1}$
Thermal conductivity [191]	k_{cond}	25	$\text{Wm}^{-1}\text{K}^{-1}$

A simple thermal finite element model is created in the same way as the lumped capacitance model, but which accounts for the non-uniform temperature distribution throughout the beam

and internal heat conduction, as well as the notch geometry. The resultant temperature evolution over time is plotted in Figure 23 for both models (data is taken from the centre of the beam in the finite element model).

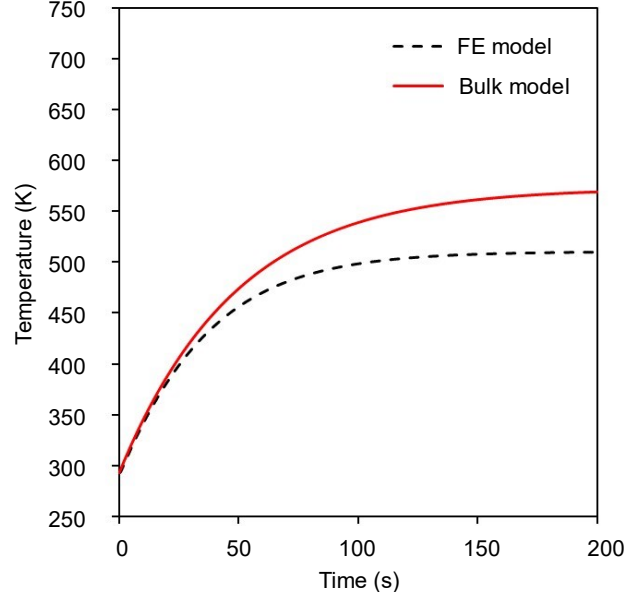


Figure 23: Comparison of temperature evolution within a Zircaloy-4 beam at 4% lamp power predicted using the lumped capacitance model and a thermal finite element model.

Figure 23 shows that the lumped capacitance model over-predicts the saturation temperature by almost 100 K due to not considering spatial temperature variation or notch geometry etc. Hence, only the thermal finite element model is used from here forward. Figure 24 (a) gives an overview of the model boundary conditions. For high-temperature testing work presented in this thesis, peak test temperatures are in the range 570 – 600 K; Figure 24 (b) shows the predicted temperature distribution throughout the beam upon achieving saturation in this temperature range, which suggests that the total temperature variation over 12 mm is around 10 K, which is acceptable, particularly since the DIC region of interest is typically around 0.3 mm wide (corresponding to <1 K temperature variation). The 10 K temperature variation is close to an experimentally measured (for the same sample geometry and heating) 15 K range in *Nitronic 60* steel [183]. The predicted time-temperature relationships at the beam midpoint are presented in Figure 24 (c) at different power levels and indicate that relatively low levels of lamp power are required to reach target temperatures.

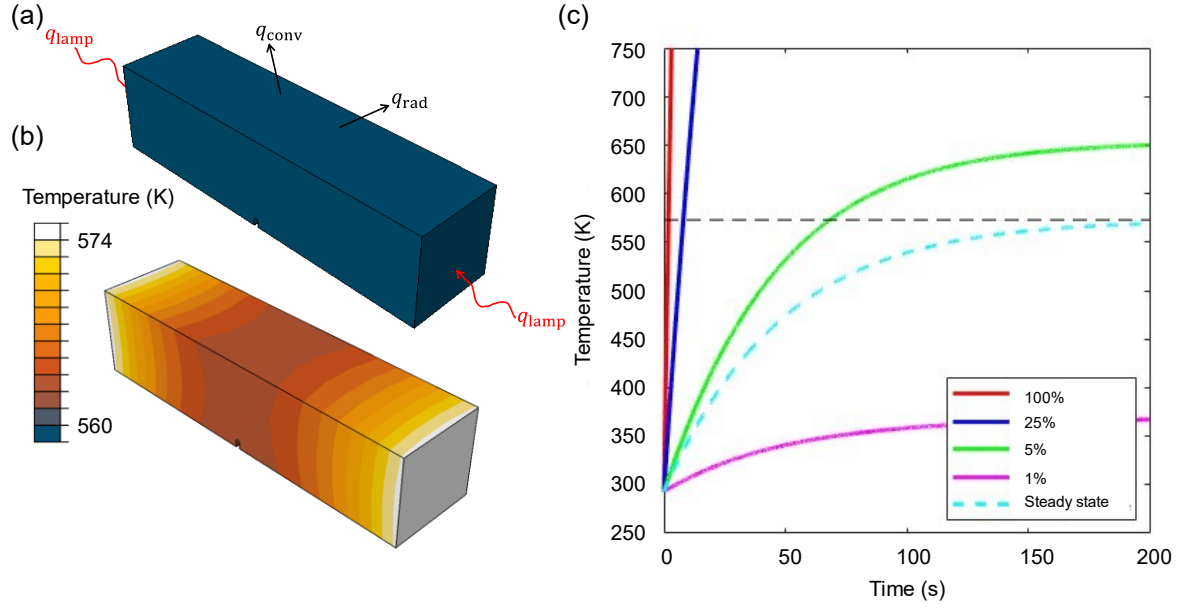


Figure 24: (a) Schematic representation of thermal finite element model boundary conditions. (b) Predicted temperature distribution throughout beam upon achieving saturation at approximately 570 K. (c) Temperature saturation curves at varying levels of input power; the steady state curve corresponds to part (b), where the horizontal dashed line represents the target temperature.

Hence, with a rudimentary understanding of the appropriate lamp power range, simulations are tailored to extract initial estimates for suitable PID control constants. The approach taken here makes use of the Ziegler-Nichols process reaction curve method [192]. This involves running the model in an open-loop state, i.e., the control variable takes a fixed value, until the process variable reaches a saturated state. At this point, a step change is made to the control variable (ΔCV), which results in a gradual change from the original process variable (temperature) saturation point to a new saturation temperature (which gives ΔPV). This is illustrated diagrammatically in Figure 25, which shows the predicted beam temperature response for a heat flux step change from 4% to 5% of the maximum value.

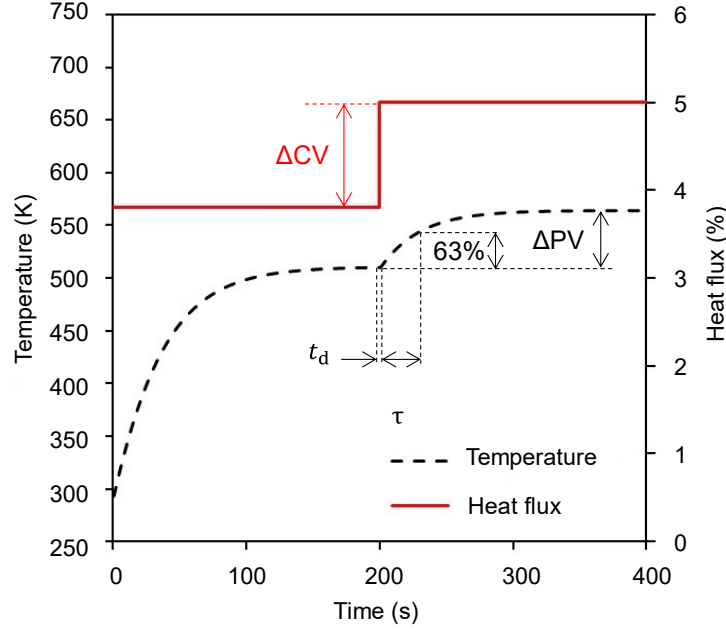


Figure 25: Simulated control variable and process variable evolution for preliminary estimate of appropriate PID controller constants.

As shown in Figure 25, three measurements are taken, which include the saturation temperature change, ΔPV , the time constant (a widely adopted characteristic of exponential behaviour), τ , and the dead time, t_d , which characterises the delay between the control variable input and significant change to the process variable. The process gain is defined as $g_p = \frac{\Delta PV}{\Delta CV}$. Based on these measurements, the rules outlined by Ziegler and Nichols [192] enable calculation of appropriate PID control constants as follows.

$$K_p = \frac{1.2\tau}{g_p t_d} \quad (3.26)$$

$$T_i = 2t_d \quad (3.27)$$

$$T_d = 0.5t_d \quad (3.28)$$

Using initial estimates for the control constants, the heating system is subjected to an initial trial, in which a thermal history is specified for a ‘dummy’ Zircaloy-4 beam, with the loading pin in contact with the sample and held at a constant load of 100 N. The thermal history consists of a linear temperature increase from room temperature to 573 K at an arbitrary rate of 30 K/min, followed by a one-minute dwell period at 573 K, which is then followed by cooling at 2 K/min (typical cooling rate for delta hydride precipitation [193]) to 473 K. As will be shown subsequently, this initial trial demonstrated a period of significant instability, resulting in poor overall temperature control. Shortcomings of the thermal finite element model

including absence of conduction heat transfer losses, neglecting lamp air coolant effects, etc., are likely to have resulted in inaccurate predictions of transient behaviour, and therefore, poorly tuned PID control settings. Hence, the Ziegler-Nichols method is applied directly to experimental measurements of temperature over time with a step change in lamp current. The temperature measurements, which are recorded using a *Pico Technology TC-08* data logger with a 1 Hz sampling frequency are presented in Figure 26.

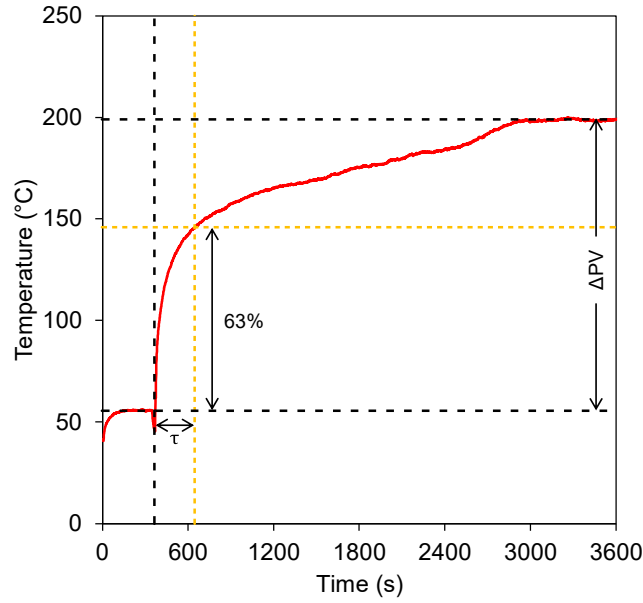


Figure 26: Experimental temperature measurements over time shown in red with step change in lamp current (proportional to electrical power consumption, which is a proxy for heat flux) at the vertical black dashed line.

As before, using Equations 3.26, 3.27, and 3.28, the PID control constants are calculated, now using direct experimental measurements, which are also subjected to a trial using the same thermal history. Significant improvements in control are observed, as shown in Figure 27.

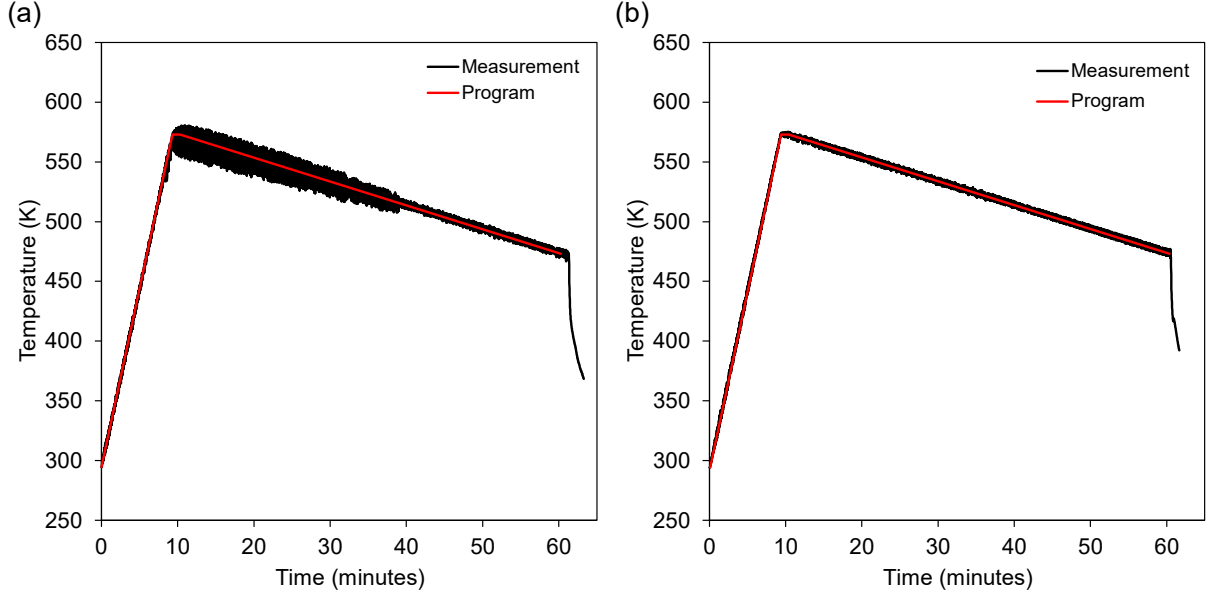


Figure 27: (a) Programmed and measured thermal history for initial temperature control system trial using PID control settings selected based on finite element modelling. (b) Programmed and improved measured thermal history for control system trial using revised PID control settings based on direct experimental measurements.

Figure 27 (a) shows significant instability in temperature control during the transition from heating to dwell and during cooling and restabilises at lower temperatures. Using experimentally determined PID control constants however, the trial demonstrates strong correlation with the programmed thermal history, as shown in Figure 27 (b). Hence, for actual thermomechanical testing, the experimental PID control constants, which are given in Table 6, are used.

Table 6: PID control constants determined using experiment-based Ziegler-Nichols calculations.

Property	Symbol	Value	Units
Proportional gain	K_p	42.73	-
Integral time	T_i	6.0	s
Derivative time	T_d	1.5	s

3.3.2 In-situ digital image correlation

3.3.2.1 Strain measurement using digital image correlation

DIC is an optical full-field displacement measurement technique (primarily optical) which tracks the evolving coordinates of features on a 2D, or 3D surface. Evolution of the coordinate fields enables calculation of displacements and hence strains and strain gradients by first and second order spatial derivation, respectively. Fundamentally, DIC requires a minimum of two images of the region of interest (i) prior to (reference image) and (ii) after deformation. Pattern

recognition software is used to identify a set of regularly spaced subsets (containing multiple pixels and unique features) within the region of interest of the reference image, which are later identified within the post-deformation image; in most cases, a randomised speckle pattern is applied to the sample surface to ensure subsets are easily distinguishable from one another. Hence, the coordinates of the subsets are updated, allowing for calculation of the 2D or 3D local displacement vectors, $\mathbf{x}$. The deformation gradient, $\mathbf{F}$, which is given by Equation 3.1 can then be determined at each DIC measurement point. The basic algorithm used here to determine components of the deformation gradient are shown in Figure 28.

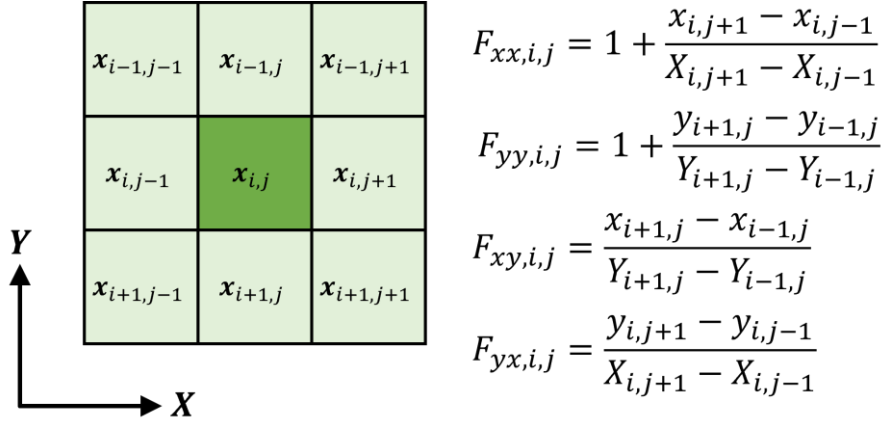


Figure 28: Point-wise algorithm used to determine components of deformation gradient (in 2D) based on displacement vectors $\mathbf{x}$. Deformation gradient at point i, j is based on nearest neighbouring displacements, and not on displacements measured at point i, j .

A single camera system is used here for 2D-DIC analysis of Zircaloy-4 beam samples under mechanical and thermomechanical loads, since most deformation is confined to the plane of bending. A digital camera (8 megapixel 12-bit *QIClick*) coupled with a long-range optical microscope (*Questar QM-100*) is used to image the front surface of each sample during testing, as shown in Figure 29. A fibreoptic illuminator is used to provide coaxial illumination of the sample. The image resolution, and hence DIC resolution is approximately $3 \mu\text{m}$.

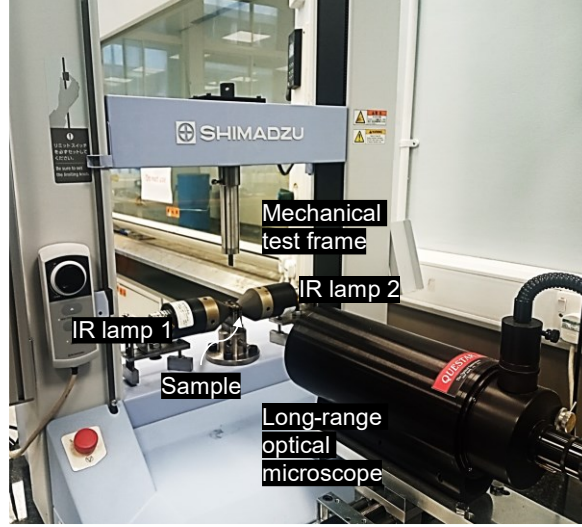


Figure 29: Micro-thermo-mechanical test setup. Beam samples are placed on two horizontal support pins at the base of the 10 kN Shimadzu mechanical test frame; a screw-driven crosshead applies a downward load to the centre of the beam for three-point bending. Infrared lamps are positioned at either end of the beam for heating. The long-range optical microscope is positioned directly in front of the centre of the sample.

For fatigue testing at high frequency, a high sampling rate is required to ensure sufficient resolution when attempting to capture data at the peaks and troughs of each cycle (reasons for needing this data are made clear later); for example, for a test frequency of 0.1 Hz, a sampling rate of 1 Hz is used. After testing, the images are processed using *Ncorr*, which is an open-source *Matlab*-based 2D-DIC program. Upon obtaining the displacement fields, the deformation gradient is calculated using a separate code based on the methodology outlined in Figure 28. The strain tensor (2D) is then simply $\boldsymbol{\varepsilon} = \frac{1}{2}(\mathbf{F}^T \mathbf{F} - \mathbf{I})$.

3.3.2.2 Surface speckle pattern application

As mentioned previously, the resolution of the in-situ DIC equipment used in this work is approximately 3 μm . Hence, the speckle pattern applied to the surface of the sample should contain features of the same length-scale. Here, a polishing solution from *Struers*, containing 1-micron single crystal diamond particles is used, and is applied to the surface of each sample as an aerosol. The diamond particles are suspended in an ethanol-based solution, most of which is removed by rinsing (in acetone) and drying after spraying. An optical microscope is used to carefully assess the pattern quality after each spray. According to iDICs' good practise guide for DIC [194], pattern density should be around 50%, i.e., 50% of the sample surface area should be covered with particles. An example speckle pattern with this pattern density, using the method described here is shown in Figure 30.

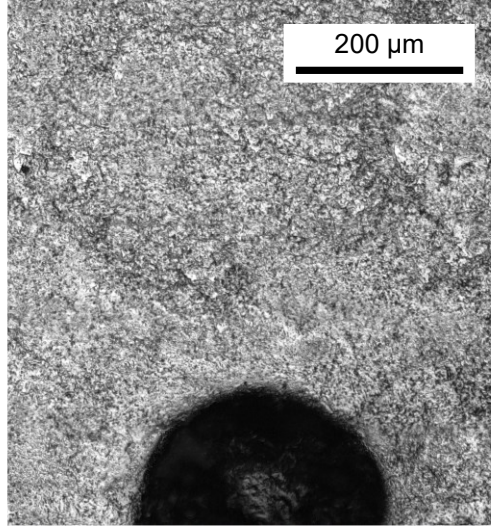


Figure 30: Example DIC speckle pattern around notched region of a hydrided Zircaloy-4 sample.

A basic test used to estimate the error of DIC displacement measurements and resultant strains is to image the region of interest before and after applying a “rigid body” displacement. In theory, the DIC software should capture the uniform displacement, and hence, the displacement gradients and strains should equal zero. Calculating strain in this way gives a reasonable estimate of the experimental error before testing and serves as a proxy for speckle pattern quality. Using the speckle pattern method described here, the maximum experimental error is approximately $\pm 0.05\%$ strain at room temperature, which is significant compared to the strain at yield, and has strong implications for stress calculations. However, such large errors are confined to regions where speckle density is either too high or low; the mean error range is $< \pm 0.01\%$, which is acceptable.

3.3.2.3 *Estimating stress*

An advantage of in-situ DIC over ex-situ DIC is that strain measurements can be made while the material is under an applied load. This feature is exploited here to approximate the full field 2D stress state. Central to this approximation is the assumption that in the unloaded state, strains measured with respect to the reference (undeformed) state are assumed to comprise plastic strains only (assuming residual microstructural elastic strains are negligible). For a given fatigue cycle, the elastic strain can be approximated by subtracting the strain in the unloaded state from the strain in the loaded state, as illustrated in Figure 31.

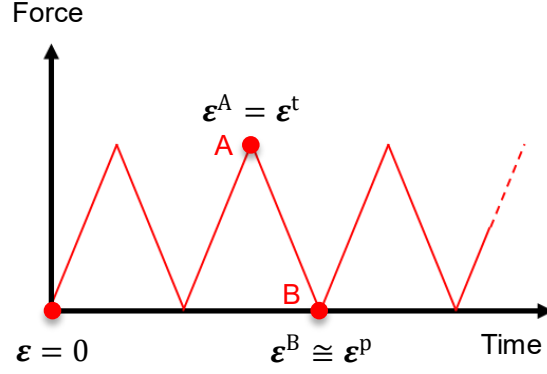


Figure 31: Illustration of typical unidirectional ($R = 0$) loading history, with points A and B, at which the total and plastic strains are calculated, respectively.

Hence, the 2D elastic strain tensor is given by,

$$\boldsymbol{\varepsilon}^e = \boldsymbol{\varepsilon}^t - \boldsymbol{\varepsilon}^p \cong \boldsymbol{\varepsilon}^A - \boldsymbol{\varepsilon}^B \quad (3.29)$$

where $\boldsymbol{\varepsilon}^t$ is the total strain (elastic + plastic), $\boldsymbol{\varepsilon}^p$ is the plastic strain, and $\boldsymbol{\varepsilon}^A$ and $\boldsymbol{\varepsilon}^B$ represent the strains measured at the loaded and unloaded states of a given fatigue cycle, respectively. Upon determining the elastic strain fields, the components stress at the free surface (assuming plane stress, $\sigma_{13} = \sigma_{23} = \sigma_{33} = 0$) are calculated using Hooke's Law, which is given by Equation 3.30 for 2D,

$$\boldsymbol{\sigma} = \mathbf{D}^{2D} \boldsymbol{\varepsilon}^e = \begin{bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{12} \end{bmatrix} = \begin{bmatrix} D_{11} & D_{12} & D_{14} \\ D_{12} & D_{22} & D_{24} \\ D_{14} & D_{24} & D_{44} \end{bmatrix} \begin{bmatrix} \varepsilon_{22}^e \\ \varepsilon_{22}^e \\ \varepsilon_{12}^e \end{bmatrix} \quad (3.30)$$

where $\mathbf{D}^{2D}$ is a matrix containing components of the full 3D stiffness matrix in the sample's frame of reference. Naturally, this requires orientation data for each grain, which is obtained using EBSD. A *Matlab* program is used to align the elastic strain fields with the local EBSD data, and hence, calculate the stress fields.

3.3.2.4 Estimating GND density

The assumption that residual strain measured in the unloaded state (no external forces) only comprises plastic strain is extended to enable full-field measurements of GND density. The deformation gradient in the unloaded state, e.g., state B, is calculated using the method described in Section 3.2.3.1, which gives four independent deformation gradient components in 2D. Since the individual slip system contributions to plastic deformation are unknown in experiments, Ashby's analytical solution [169] (see Equation 3.10) is generalised in 2D and is given by Equation 3.31.

$$\rho_{\text{GND},kl} = \boldsymbol{\rho}_{\text{GND}} = \frac{1}{b} \nabla \times \mathbf{F}^p = \frac{1}{b} \cdot \begin{bmatrix} \frac{\partial F_{xx}^p}{\partial x} & \frac{\partial F_{xx}^p}{\partial y} \\ \frac{\partial F_{yy}^p}{\partial x} & \frac{\partial F_{yy}^p}{\partial y} \\ \frac{\partial F_{xy}^p}{\partial x} & \frac{\partial F_{xy}^p}{\partial y} \\ \frac{\partial F_{yx}^p}{\partial x} & \frac{\partial F_{yx}^p}{\partial y} \end{bmatrix} \quad (3.31)$$

To determine the spatial gradients of deformation gradient, the same algorithm used to determine the deformation gradient using the displacement fields is adopted, e.g., $\frac{\partial F_{xx}^p}{\partial x}_{i,j} = \frac{F_{xx,i,j+1}^p - F_{xx,i,j-1}^p}{x_{i,j+1} - x_{i,j-1}}$, where subscripts i and j represent the row and column of a point in the 2D dataset. The scalar GND density is then given by taking the 2nd order norm of its components, i.e., minimising dislocation energy, as follows.

$$\rho_{\text{GND}} = \sqrt{\sum_{l=1}^2 \sum_{k=1}^4 \rho_{\text{GND},kl}^2} \quad (3.32)$$

This formulation is an adaptation of that which was reported by Xu et al. [175], in which GND density was approximated as the curl of the plastic strain tensor, i.e., $\boldsymbol{\rho}_{\text{GND}} = \frac{1}{b} \nabla \times \boldsymbol{\varepsilon}^p$. Using the current revised formulation is more precise however, since the asymmetric rotation components are accounted for in full (unlike the plastic strain tensor, which is symmetric).

3.3.3 Microstructure characterisation

3.3.3.1 Optical microscopy

For rapid characterisation of hydrogen-charged and hydrided Zircaloy-4 microstructures, an *Olympus BX51* optical microscope is used. This method is particularly useful for the current work since a heat treatment, which is described later, is used to promote significant grain growth (hundreds of microns) prior to the introduction of hydrides. Using a polarising light filter, grain orientation contrast and phase contrast make microstructural features readily identifiable. This is particularly useful when, for example, tracking the evolution of hydride precipitates with respect to the underlying Zircaloy-4 microstructure between successive thermomechanical cycles. Sample surface preparation steps taken for polarised light microscopy are listed in Table 7.

Table 7: Sample surface preparation steps taken for polarised light microscopy. Samples are mounted in 30 mm diameter thermoset polymer cylinders.

Step	Abrasive / solution	Applied load (N)	Time (minutes)
Grinding	800 grit SiC abrasive pad	10	As required
	1200 grit SiC abrasive pad		4
	2000 grit SiC abrasive pad		8
	4000 grit SiC abrasive pad		16
Polishing	Colloidal silica suspension (0.04 μm particles)		30

3.3.3.2 Scanning electron microscopy and electron backscatter diffraction

For increased imaging resolution and phase contrast at lower length scales, a *Zeiss Sigma 300* SEM is used. This method is particularly useful for post deformation analysis of hydrided samples which, e.g., have undergone fatigue cracking to assist the identification of potential crack growth mechanisms. In Figure 32, SEM imaging is used to locate and identify delta hydride platelets and stringers (within a blocky-alpha Zircaloy-4 microstructure) by their orientation relationships relative to the crystal orientation of the Zr matrix.

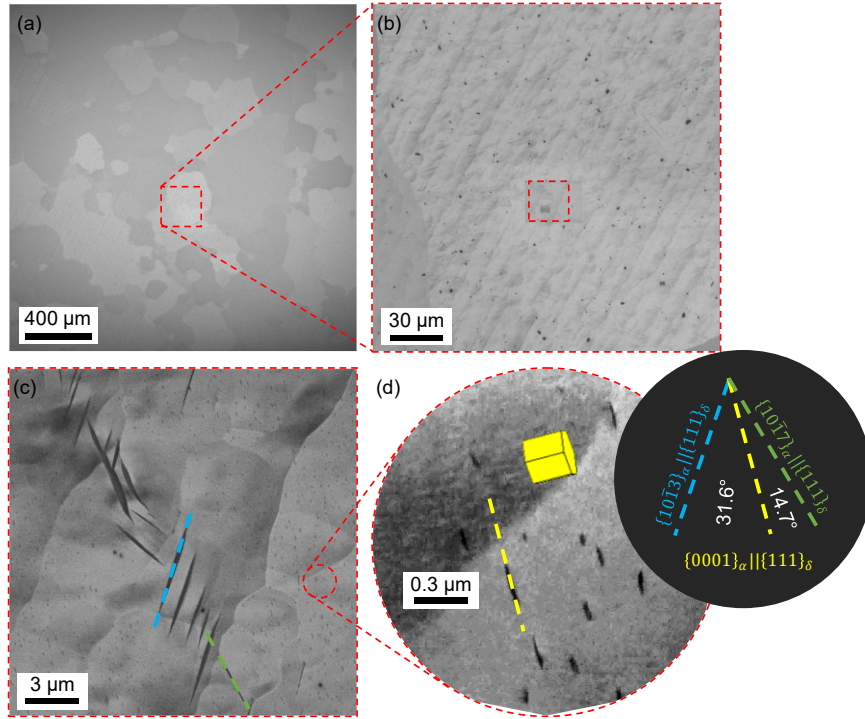


Figure 32: (a) – (c) Example SEM images increasing in magnification of a region of Zr-4 microstructure containing intragranular hydrides; (c) and (d) identification of delta hydrides and corresponding orientation relationships [195] with basal plane of matrix orientation.

Figure 32 (d) shows the crystal orientation of the local Zr-4 matrix, which was determined using EBSD analysis. EBSD is an advanced SEM-based characterisation technique which enables investigation of phase, crystal orientation, and other microstructural characteristics

including grain morphology, distribution, size, texture, etc. Here, it is mostly used to obtain information about matrix and hydride crystal orientations, since all experimental work presented in this thesis is focused on local microstructural effects. The technique relies on the collection of electron backscatter diffraction patterns (EBSP), using a detector which is oriented at usually 20° relative to the sample surface (which itself, is tilted at 70° relative to the horizontal plane). Some of the electrons which are scattered from the sample surface are incident on lattice planes, and are diffracted towards the detector, producing high-intensity bands (Kikuchi bands). These electrons satisfy Bragg's Law [196], which is given by,

$$n\lambda = 2d_{hkl} \sin \theta \quad (3.33)$$

where n is an arbitrary integer, λ the wavelength of incident electrons, d_{hkl} the interplanar spacing of lattice plane hkl , and θ is the Bragg angle. Constructive interference, and hence bands of high intensity are also produced when this law is satisfied. *Bruker* software is used to detect the Kikuchi bands (typically 8 to 12 bands), and their points of intersection; a Hough transformation is then performed to calculate the interplanar angles. A look-up table consisting of possible interplanar angles for the phase(s) of interest is used to cross-reference the measured values and determine the crystal orientation. Orientation data is saved pointwise for each EBSD measurement, which is used to produce an orientation map. Data is saved as a set of Euler angles using the Bunge (ZXZ) convention. The rotation matrix, which is used extensively in the current crystal plasticity code (for determining slip system normal orientations etc.), using the Bunge convention is given by,

$$\mathbf{R} = \begin{bmatrix} \cos \varphi_{z2} & -\sin \varphi_{z2} & 0 \\ \sin \varphi_{z2} & \cos \varphi_{z2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos \varphi_{x1} & -\sin \varphi_{x1} \\ 0 & \sin \varphi_{x1} & \cos \varphi_{x1} \end{bmatrix} \begin{bmatrix} \cos \varphi_{z1} & -\sin \varphi_{z1} & 0 \\ \sin \varphi_{z1} & \cos \varphi_{z1} & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (3.34)$$

where φ_{z1} and φ_{z2} are the first and second Z-axis rotations, and φ_{x1} is the rotation about the X-axis. Careful consideration is given to ensure that the EBSD reference orientation is known prior to applying the rotation.

Since the acquisition of EBSD data relies on nanoscale crystallographic information, a very flat surface, *free of deformation* (strictly, not free of deformation, but free of severe deformation at the surface due to e.g., grinding and polishing) is required. While the final polishing stage listed in Table 7 is sufficient for some materials, e.g., Ni superalloys [197], Zr alloys, particularly hydrided Zr alloys are far less docile; this is typical of soft materials, which are susceptible to

high levels of surface damage during mechanical polishing. Previous Zircaloy-4 characterisation work by Wan [76] relied on electropolishing in a perchloric acid solution, which yielded excellent results. However, working with perchloric acid, which is a strong oxidiser, is highly dangerous due to risk of fire, explosion, severe burning etc. [198], and hence, an alternative method is preferred. It was found that an additional step of ion beam polishing using the *Gatan PECS II* system at cryogenic temperatures also yielded excellent EBSD pattern detection. Polishing conditions, which were optimised for Zircaloy-4 by Fang et al. [199] are used here and are listed in Table 8.

Table 8: Ion beam polishing parameters used for sample preparation for EBSD.

Incident polishing angle (°)	Ion energy (keV)	Sample temperature (°C)	Duration (minutes)
8	8	<-50	30

Ion beam polishing, specifically broad ion beam (BIB) polishing, uses highly energetic ions to sputter and remove material from the sample surface. It is a highly robust method which introduces significantly less surface damage than mechanical polishing, though it can cause surface radiation damage at high energy levels. There are clear topological effects on the sample surface, which are evident in Figure 32 (c), showing localised pitting; this is not ideal for SEM imaging, but does not affect EBSD data acquisition. To maximise conductivity between the sample and the SEM and inhibit drift during EBSD [200], samples are mounted to 12 mm diameter stubs using silver paint from *RS*.

3.3.4 Sample preparation

3.3.4.1 Sample manufacture

Beam samples are manufactured from Zircaloy-4 rolled plate material in the as-received condition (from *Rolls-Royce plc.*). Samples are cut using a *Fanuc* wire electrical discharge machine (EDM) with a wire diameter of 120 μm . Notches are also machined using wire EDM. Utilising the rolled plate’s strong texture, samples are machined in two distinct configurations to ensure the texture with respect to the notched surface of each sample is characterised by c-axes which are either predominantly in-plane or out-of-plane. Each configuration, A, B, is illustrated in Figure 33.

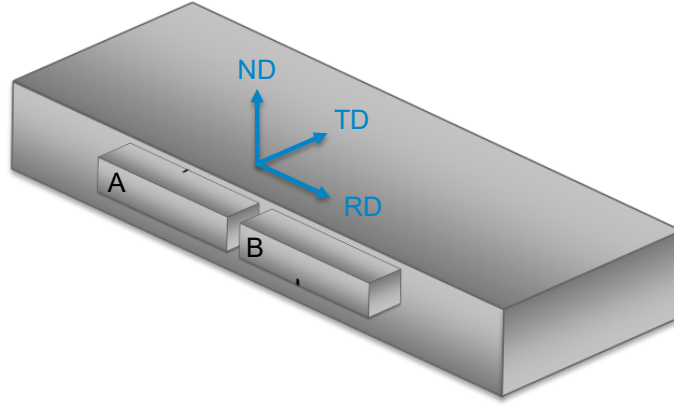


Figure 33: Configuration of samples (A and B) to control texture of notched face with respect to the rolled plate coordinate system.

Figure 33 shows the rolled plate's local coordinate system, RD-TD-ND, the axes of which represent the rolling direction (RD), transverse direction (TD), and normal direction (ND), respectively. Depending on the level of deformation during rolling, a strong texture can be produced [201]; the plate used here was manufactured such that the c-axes of grains are predominantly in alignment with the ND, and hence, A type samples, whose notched faces lie in the TD-RD plane have a mostly basal texture, while c-axes are predominantly in the plane of B type sample faces. An advantage of machining samples in this way is that when tested, the textures can help elucidate important aspects of crystallography / microstructure which impact micromechanical performance.

3.3.4.2 Blocky alpha grain growth heat treatment

Given the resolution of the current in-situ DIC methodology is limited to 3 μm , to maximise its potential for understanding the role of crystallography in deformation, a heat treatment is applied to each sample to generate very large, blocky alpha grains, the features of which can be easily resolved in-situ. The heat treatment used here was developed by Tong [202] and was designed to achieve maximum possible grain growth. The material is heated to 800 $^{\circ}\text{C}$, which is just below the alpha-beta transus temperature (the transus temperature is around 820 $^{\circ}\text{C}$ for Zircaloy-4 [203]) and is held at this temperature for 336 hours (14 days). To prevent oxidation during each heat treatment, samples are encapsulated in quartz tubing and backfilled with argon gas.

3.3.4.3 Electrolytic hydrogen charging

Following the grain growth heat treatment, an electrolytic hydrogen charging procedure is used to introduce hydrogen to the matrix and form hydride precipitates. The Lapage technique

[204] is adopted here, which involves the gradual deposition of hydride on the sample surface, while submerged in an electrolytic bath. An altered version of this technique, which was described by Birch et al. [144] is used in this work. The method is illustrated diagrammatically in Figure 34.

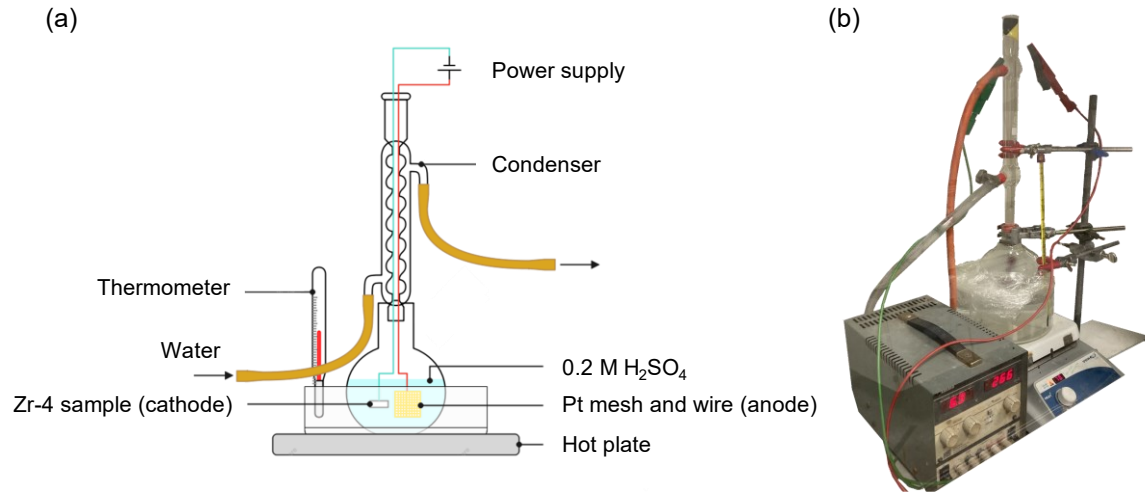


Figure 34: (a) Schematic illustration of electrolytic hydrogen charging rig. (b) Actual hydrogen charging rig used for this work.

As shown in Figure 34, a voltage is applied across the electrolyte (0.2 molar H₂SO₄) between the Zircaloy-4 (sample) cathode and platinum mesh anode; the cathode is connected to the negative wire terminal using a spot welder. The electrolyte is preheated to 65 ± 5 °C to increase the reaction rate, within a round-bottom flask, which itself is submerged in a basin of water to ensure uniform heating. The flask is connected to a water-cooled condenser to maintain a constant molar concentration of H₂SO₄. The voltage is adjusted using a DC power supply unit to generate current density of 2 kA/m² (relative to the charged area of the sample) for 24 hours. Here, only a single surface is charged, such that measurements of the deposited hydride layer thickness can be made after charging; this is later used to assess the success of the procedure. Prior to charging, each sample is ground to 2000 grit level, primarily to remove oxides and ensure consistency between samples, and an insulating varnish is used to coat all non-charged surfaces.

3.3.4.4 Post-charging homogenisation heat treatment

Upon generating a sufficiently thick hydride layer / rim (which is assessed using an empirical model for hydrogen content [204], with a target concentration of 150 – 200 wppm), a homogenisation heat treatment is used to uniformly distribute hydrogen, and hence hydrides throughout the sample. This procedure involves raising the sample temperature to a sufficient

level to bring the hydrogen into solution, and holding the sample at this temperature for some time to allow the hydrogen to diffuse until an equilibrium concentration is reached throughout. According to Lepage et al. [204], samples should be held at 400 °C for 7 hours to ensure a uniform hydride distribution at the mm length-scale. As with the grain growth heat treatment, samples are encapsulated quartz tubing in an inert environment to prevent high temperature oxidation. During subsequent high temperature testing, it is assumed that the bulk hydrogen content remains relatively unchanged due to the inhibition of surface losses of soluble hydrogen owing to oxide layer formation.

4 The Influence of Microstructure on Short Fatigue Crack Growth Rates and Path

4.1 Introduction

Most models for short FCG fail to capture the effects of microstructure on growth rate. The first part of this chapter explores whether SED can fulfil this role. Specifically, the current work is aimed at understanding the key mechanistic drivers promoting microstructural dependence of FCG rate in Zircaloy-4. It is known that texture, particularly in anisotropic HCP materials, is a key factor affecting short crack growth [205], [206], [207]. This is also made clear by experimental data [205], [206], [207] presented here, which highlights the significant influence of microstructure on short FCG rate in two strongly textured blocky alpha Zircaloy-4 samples. In experimental crack paths through grains where the c-axis is in-plane and approximately parallel with the crack propagation direction, the crack path is often straight, propagating along prismatic planes, perpendicular to their Burgers vector directions. In cases where the HCP c-axis is orthogonal to the loading direction (and free surface), cracks are also observed to propagate along prismatic planes (parallel with the Burgers vector direction) and oscillate between them. Zeng et al. [208] studied short FCG behaviour in zirconium with equiaxed grains measuring 15 μm in diameter. Results showed that cracking was predominantly transgranular, and that crack directions within single grains could be characterised by the traces of individual slip planes (mostly prismatic and pyramidal). Results also showed that a key factor affecting crack propagation rate across grain boundaries is the twist angle; twist angles greater than 40° were suggested to inhibit crack propagation entirely. Similarly, Zhai et al. [209] showed that crack plane twist at grain boundaries is mainly responsible for growth rate retardation at grain boundaries, and that large angles can have a lasting effect on growth rate after the crack has crossed the boundary. The authors also presented a mechanism for crack propagation across grain boundaries which enables compatibility between incoming and outgoing crack planes. The mechanism involves the formation of crack steps on the outgoing plane, which have been observed in experiments, and can help explain growth rate retardation, particularly due to high angles of twist. Likewise, Wen et al. [210] performed experiments to quantify the resistance of boundaries to FCG in an Al-Cu alloy, in terms of the twist angle. The study showed a strong linear correlation between twist angle and the change in crack growth rate, $\Delta(da/dN)$ at the grain boundary.

As discussed by Rovinelli et al. [211], [212], since the length of a stage I (short) crack is comparable with the size of the plastic zone at the crack tip, and with that of surrounding microstructural features (e.g., grain size), its rate of growth is strongly influenced by local plasticity. Additionally, since plastic deformation is concentrated on individual slip systems experiencing the highest shear stress, short cracks tend to propagate along these highly damaged systems [213], [214], often inclined at a sharp angle relative to the primary loading direction. Hence, classical stress-based fracture mechanics approaches such as Paris Law [20] cannot capture the associated microstructural-sensitivity of short cracks [215], which is illustrated by the wide degree of growth rate variability observed within the regime. However, various attempts have been made to correct the conventional (global) SIF to account for the plastic zone to mitigate this issue [24], [216], [217]. Pataky et al. [218] used DIC full field displacement data to determine the effective mode I and II SIFs for short cracks in single crystal 316L stainless steel, accounting for local plasticity. Results showed marked contrast between the global SIF range and the effective SIF range in terms of growth rate prediction. As anticipated, high variability is observed between growth rate trends among differing crystal orientations when using the global SIF range, which is much less apparent for the effective SIF range. Despite this improvement, the method lacks microstructural sensitivity as it relies on several fitting parameters and remains ineffective for specific crack propagation directions. Rovinelli et al. [211] carried out detailed modelling analyses to investigate the viability of various Micromechanical Short Crack Driving Force Metrics (MSCDFMs), which are essentially the same as FIPs but which also account for monotonic behaviour. Using crystal plasticity modelling, various random microstructures were simulated under monotonic loading incorporating a range of crack lengths. The results showed significant stress concentrations ahead of the crack tip and at grain boundaries, which the authors referred to as hot spots. These hot spots were alluded to play a key role in promoting fluctuating crack growth rates and directions within the short crack growth regime. Six MSCDFMs were compared with one another; three of which were shear strain-based, the others energy-based. The authors chose to work primarily with an energy-based term (which was based on the Fatemi-Socie FIP [37] in that it accounts for energy dissipated on the critical plane) due to maximised microstructural sensitivity. A key finding from the study was that the change in MSCDFM with respect to an increase in crack length for simulations with different texture was analogous to the variability observed within the short crack growth regime (ΔK vs. da/dN). However, this should be viewed

as problematic, since this is what makes short FCG difficult to characterise using the conventional SIF range.

For zirconium and its alloys, there remains a lack of mechanistic understanding of the key factors affecting FCG rates, i.e., grain boundaries and triple junctions, grain orientation, and more generally, texture. For fatigue crack nucleation and propagation work, SED and similar quantities have shown very promising results in terms of predictive capability [219], [220], [221]. SED is a measure of the energy stored in dislocation structure, including GNDs, which accommodate local lattice curvature at crack tips, and statistically stored dislocations (SSDs). The release of this energy is argued here to provide the driving force for short FCG in Zircaloy-4. Rates of short FCG in Zircaloy-4 have not been simulated until now; this chapter assesses the microstructural sensitivity of SED in terms of capturing experimental microstructure-linked growth rate fluctuations and grain orientation dependence. Rigorous assessment of this methodology in Zircaloy-4 will enable progression towards more complex modelling challenges, such as DHC [222], [223].

Short crack paths in polycrystalline microstructures can strongly influence crack growth rate but cannot be easily predicted. Based on findings from the current crack growth rate study, this chapter also presents a critical appraisal of three microstructure-sensitive criteria for crack path. Experimental evidence suggests that short or stage I cracking is controlled by crystallographic slip [224], [225], [226], [227]. Laird [213] first highlighted observations of short cracks following paths inclined at 45° to the loading direction, compared with 90° for long cracks. Naturally, for an idealised single crystal with orientation such that slip planes are oriented at 45° (relative to the remote loading direction), slip is maximised due to increased shear stress. Neumann [228], [229] later added refinement to this observation by demonstrating the proclivity for very short cracks to oscillate between visibly active slip systems. An in-situ SEM study by Zhang et al. [230] of crack initiation and short propagation in single crystal nickel showed that crack nucleation occurred within shear bands containing multiple octahedral slip traces. The tortuous short cracks which followed were apparently controlled by the same phenomenon. Similar observations were recorded by Carroll et al. [231]. Recent work by Rovinelli et al. [232] used high resolution 3D short crack path data in combination with crystal plasticity modelling to establish correlations between candidate mechanistic crack path drivers and experimental path. Bayesian networks were used to analyse the data and identify

two high-correlation drivers in the order: (i) misalignment between the principal stress axis and the slip direction and (ii) maximum resolved shear stress, which corresponds somewhat with the well documented maximum slip driver [213]. However, (iii) maximum accumulated shear strain was also analysed and showed lower correlation with the experimental crack path. Nevertheless, maximum accumulated shear strain was shown to be strongly linked with crack growth rate. Similar investigations were carried out by Pierson et al. [233] using a large 3D crack path dataset. While no micromechanical field variable was shown to definitively predict short fatigue crack path, both stress and strain gradients were implicated as important factors.

With the use of numerical modelling methods to predict short crack growth, there has been near consensus on the use of the maximum slip criterion [177], [234], [235], [236]. Lin et al. [234] were among the first to use the maximum slip criterion for short crack path within CPFE models, which showed strong microstructure-sensitivity in polycrystalline materials. However, intragranular variations in crack path were not captured since the local stress state was not updated according to the crack tip position. Building on this, Zhang et al. [177] simulated crack growth in single crystal nickel using CPFE modelling combined with XFEM [167]. Maximum slip was used as the criterion for crack path and was shown to capture experimental phenomena such as tortuosity, but the overall segment lengths and oscillation frequencies disaccorded with experiments. There is also a range of experimental evidence which demonstrates crystallographic FCG along planes which show less slip activity than others (see Figure 35). Farukh et al. [237] used a maximum principal strain criterion (non-crystallographic) to predict crack direction in a polycrystalline nickel superalloy for comparison with experiment. Despite the use of crystal plasticity, the simulated crack path was essentially orthogonal to the remote loading direction and was in poor agreement with the experimental path indicating poor microstructure sensitivity. Several energy-based criteria for crack path have also been proposed [238], [239], [240]. However, implementation of such criteria is often at the continuum level and also lacks microstructure sensitivity. Methods such as maximum energy release rate [241] were developed based on Griffith theory [28] from classical continuum fracture mechanics but have not yet been investigated in the context of short fatigue crack paths. There is experimental evidence in the literature, some of which is presented in Figure 35, which suggests that maximum slip may not be the exclusive determinant for short fatigue crack path. Ravi et al. [242] have also shown experimentally that both mode I and II

mechanisms contribute to the opening of short fatigue cracks, demonstrating the importance of crack tip stress. Thus, there is a need to explore new and alternative predictive methods.

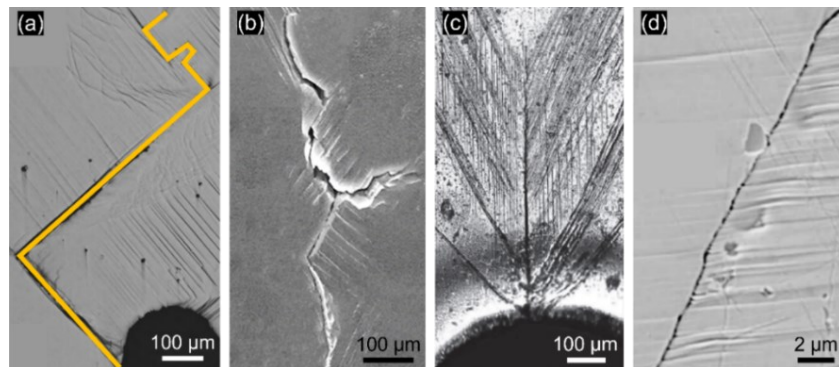


Figure 35: Experimental evidence for short fatigue crack propagation in a range of metallic materials where the crack direction is visibly different to slip trace directions corresponding to high slip activity. (a) Microstructurally short FCG in a Ni-based superalloy [243]⁸. The crack path is controlled by octahedral planes. There is clearly preferential activation of a single octahedral slip system, despite the tortuous crack path following several planes. (b) An experimental crack path in a polycrystalline Al alloy [244]⁹. The crack path is partly controlled by grain boundary decohesion (intergranular) and intragranular crystallographic cracking. There are several examples of slip activity with traces orthogonal to the crack path. (c) A straight basal fatigue crack path in a Mg single crystal [245]¹⁰. There are clear basal slip traces, but also microcracking and high slip parallel to pyramidal planes. (d) A high magnification image of the end of a fatigue crack path in a Zr alloy [76]. The crack path is parallel to a prismatic plane, but higher levels of slip are observed on another plane.

In the second part of this chapter, it is hypothesised that short crack paths are controlled partially, but not entirely by crystallographic slip (it is not disputed, however, that crack nucleation (small cracks) is dominated by slip activity [246]). According to McDowell [247], small cracks differ from short cracks by their ratio of crack length to the characteristic microstructure length scale, i.e., grain size for most metallic materials. Microstructurally small cracks are ones in which the crack length is on the same order as individual microstructure features; microstructurally short cracks are characterised by cracks with length on the order of several repeating microstructure features. The hypothesis is tested by comparing CPFE-simulated microstructurally short paths, in combination with XFEM, controlled by (i) the maximum slip criterion, (ii) maximum energy release rate and (iii) a new maximum normal SED criterion, directly with experiments. Crack paths for (ii) and (iii) are constrained by considering active crystallographic slip planes only. The hypothesis is developed on the basis that dislocation slip contributes to damage and hence weakening of the plane, but that some other factor, e.g., stored energy is responsible for opening the crack on the damaged plane.

⁸ Reprinted from [243] under the Creative Commons Attribution 4.0 International Public License.

⁹ Reprinted from [244] (Figure 2 (a)), with permission from Elsevier (license number 5820761253775).

¹⁰ Reprinted from [245] under the Creative Commons Attribution 4.0 International Public License.

4.2 Methodology

4.2.1 Single crystal simulations

To understand how FCG rates differ across different microstructures, simplified single crystal crack growth studies are first carried out for two crystal orientations, which exhibit strongly different mechanical properties. Additional single crystal orientations are investigated in the subsequent crack path study. Details of the finite element model are given in Figure 36 (a). The single crystal crack growth model is partitioned into CPFE and isotropic plastic regions (described in Section 4.2.2); this is to minimise computational expense, as elements within the isotropic plastic regions do not require a user-material subroutine. All elements within the CPFE region are 10 μm in length. A cyclic far field stress of 200 MPa is applied to the model with loading ratio, $R = 0$. An initial planar edge crack is inserted perpendicular to the free surface, which gives an initial SIF range of 7.8 MPa $\sqrt{\text{m}}$. This is calculated using the analytic solution for SIF from Liu et al. [248], given by Equation 4.1.

$$\Delta K^\infty = \Delta\sigma_{\text{ff}}\sqrt{\pi a} \left[1.12 - 0.23 \left(\frac{a}{b_w} \right) + 10.6 \left(\frac{a}{b_w} \right)^2 - 21.7 \left(\frac{a}{b_w} \right)^3 + 30.4 \left(\frac{a}{b_w} \right)^4 \right] \quad (4.1)$$

where a is projected crack length, b_w is total vertical width of the sample, and $\Delta\sigma_{\text{ff}}$ is the remote applied stress range. Note that this expression for SIF range applies only to the single crystal edge crack specimen. The two crystal orientations studied within the first results section, denoted A and B in Figure 36 (a), are oriented such that their HCP c-axes lie orthogonal to the x-z and the x-y plane, respectively, i.e., are hard and soft grains with respect to the remote loading direction.

4.2.2 Polycrystal simulations

To assess whether SED can accurately capture differences in FCG rate observed in real microstructures, two samples (AA and BB) from previous experimental work [76] are modelled here in detail. The c-axis directions of orientations A and B are approximately the same as those of grains in samples AA and BB respectively (with respect to the crack growth direction). Hence, it is anticipated that the growth rate behaviour of orientations A and B will resemble the experiments to some degree. In 3-point bending experiments, cracks were propagated through strongly textured samples. These polycrystal models are also used to assess the performance of each crack path driver. Details of the finite element model used to represent these experiments are presented in Figure 36 (b) and (c).

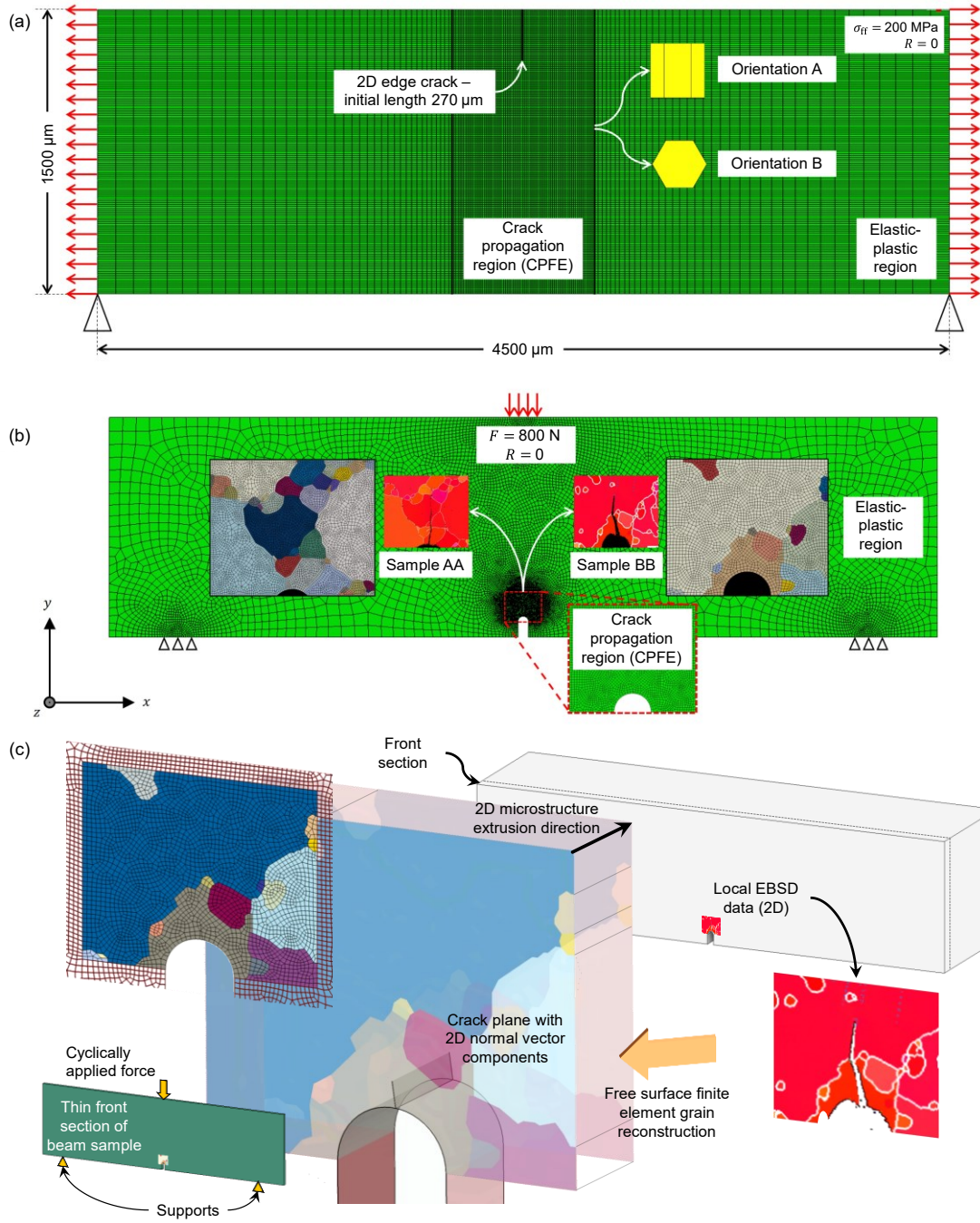


Figure 36: (a) Single crystal edge crack model setup. (b) 3-point bend modelling configuration. (c) Post-mortem free surface EBSD data is used to reconstruct the 2D microstructure. A 3D model is constructed, which is just 150 μm thick to accurately represent the free surface stress state. Free surface microstructure is simply extruded due to absence of 3D data. As shown, crack planes are also represented as extrusions of their traces. Vertical boundary conditions are used to represent support pins in the 3-point bend configuration. A distributed cyclic load is applied to a region at the top surface of the sample, which is representative of the experimental loading pin.

The 3-point bending configuration is represented in the finite element model using y-direction boundary conditions to simulate the support pins, and a distributed load is applied across several nodes along the top surface to simulate the loading pin. From experiments, only 2D grain distribution and crack path data (at the free surface) are obtained. Within this modelling

work, only crack growth at the free surface is considered, and crack plane normal directions are constrained to have zero components along the z-direction. Hence, to minimise computational expense, the beam is represented as a thin plate, with free surface plane stress conditions. The 2D approximation may mean that important substructure and crack plane orientation effects are omitted. However, it is conceivable that since blocky alpha microstructures are modelled here, substructure has little effect on the crack path observed at the free surface. As shown in Figure 36 (b) and (c), there is significant mesh refinement within the ROI ($3 - 5 \mu\text{m}$ elements), where crystal orientation data for each microstructure is assigned. As with the single crystal simulations, the area outside of the ROI is modelled using a simplified isotropic plastic model, incorporating elastic anisotropy, with elastic properties from single crystal A used for sample AA, and elastic properties from single crystal B used for sample BB. In each case, the isotropic (Von Mises) yield stress is set to 435 MPa, incorporating linear strain hardening to 500 MPa at 1.6% strain. Nevertheless, in both single crystal and polycrystalline models, yielding within these elastic-plastic regions is not achieved as they are sufficiently far removed from the crack tips. The remote SIF range may also be calculated for the 3-point bending configuration and is given by Equation 4.2 from Anderson [249],

$$\Delta K^\infty = \frac{P}{B_t \sqrt{W}} f\left(\frac{a_e}{W}\right) \quad (4.2)$$

where P is the applied force, W and B_t are the sample width and thickness, S is the distance between the support pins and a_e is the effective crack length (notch length + projected crack length). The function, $f\left(\frac{a_e}{W}\right)$ is given by,

$$f\left(\frac{a_e}{W}\right) = \frac{3\left(\frac{S}{W}\right)\sqrt{\frac{a_e}{W}}}{2\left(1 + 2\frac{a_e}{W}\right)\left(1 - \frac{a_e}{W}\right)^{\frac{3}{2}}}\left\{1.99 - \frac{a_e}{W}\left(1 - \frac{a_e}{W}\right)\left[2.15 - 3.93\left(\frac{a_e}{W}\right) + 2.7\left(\frac{a_e}{W}\right)^2\right]\right\} \quad (4.3)$$

4.2.3 Implementation of existing and development of new criteria for crack path

4.2.3.1 Maximum slip criterion for crack path

Within the aforementioned CPFE user-material subroutine, the total accumulated slip is calculated for each HCP slip system i , by the following equation,

$$\gamma^i = \int \dot{\gamma}^i dt \quad (4.4)$$

where $\dot{\gamma}^i$ is the rate of plastic shear and dt the time increment. Hence, according to the maximum slip criterion, the crack will extend along the slip plane of the system with maximum slip magnitude ahead of the crack tip, the orientation of which is given by its unit normal vector (which depends upon grain orientation). The mechanistic basis for this is that planes with greatest damage, i.e., slip are most susceptible to cracking.

4.2.3.2 Maximum energy release rate criterion for crack path

In this section, a new method to predict short fatigue crack path based on the maximum energy release rate criterion is presented. The method involves utilisation of the crack tip SED field to select a crack path enabling maximal release of energy, i.e., SED. However, the key factor which differentiates this method from others is that it accounts for the crystallographic nature of short cracks by only considering paths controlled by crystallographic slip planes. An additional constraint, which ties with the current hypothesis, is that only active (damaged) slip systems are considered. The non-local methodology is described in greater detail in Figure 37 (a). As shown in Figure 37 (a), a semi-circular measurement zone is defined ahead of the crack tip, throughout which SED measurements are recorded. It is assumed that for some measurement zone radius, r_1 , the crack path of maximal energy release is given by the path which traverses the greatest total SED. For simulation of 2D cracks, each slip system is compared by considering the release of energy given by the free surface slip plane trace of each active slip system. However, since the current CPFE approach involves SED calculation at discrete integration points, k , the total SED release along a given slip trace is unknown. Hence, circular measurement subzones, with radius r_2 , are uniformly spaced along each active slip system trace to encompass local SED measurements, enabling an approximation of the potential energy release. In single crystal case studies, the value of r_2 was found to be mesh insensitive if selected within reasonable limits; specifically, provided r_2 is large enough to encompass at least one integration point per circle and small enough such that the circles do not overlap, the predicted crack path is reproducible. The basis for selecting r_1 is discussed later. Since the total number of encompassed integration points M may vary, the release of energy for slip system is normalised as, $G_{\text{MER}}^i = \sum_{k=1}^M \frac{G_k^i}{M}$. The mesh-independent unique solution is given by Equation 4.5.

$$G_{\text{MER}}^i = \frac{1}{r_1} \int_0^{r_1} G^i dr \quad (4.5)$$

Hence, the short crack path which gives maximum energy release rate is given by the maximum value for G_{MER}^i from the set of i active slip systems. Criteria for r_1 and r_2 selection are presented in the results section.

4.2.3.3 Maximum normal stored energy density criterion for crack path

Thirdly, a new criterion for crack path is presented in this section, which also follows from the hypothesis that short crack paths are partially controlled by slip activation, but that another factor drives crack formation on active slip planes. As with the maximum energy release rate implementation, only active slip systems are considered using this method, but with maximum SED normal to the slip plane controlling crack path. This criterion bears resemblance to the maximum principal stress criterion, but an important difference (aside from considering only active slip planes) is that cyclic plasticity effects are accounted for, which are expected to be significant for short crack growth.

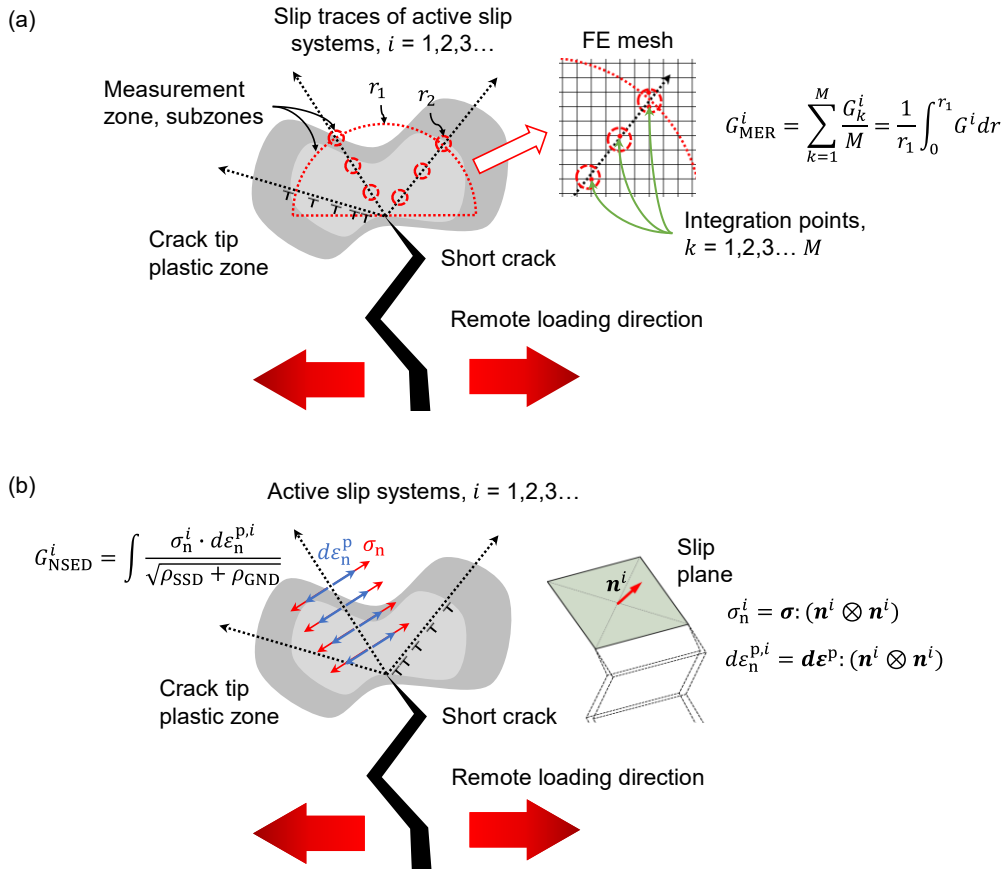


Figure 37: (a) 2D illustration of implementation of maximum energy release rate criterion using a finite element mesh. The measurement zone refers to the semi-circular region centred at the crack tip. The measurement sub-zones are the circular regions spanning the individual slip traces and are used to encompass integration points from the finite element mesh. (b) Illustration describing normal SED criterion for crack path. Red and blue arrows represent component of stress and plastic strain tensors, which act normal to the crack plane, respectively. The normal components are calculated by considering the crack plane orientations in 3D, given by their unit normal vectors, $\mathbf{n}^i$.

As shown in Figure 37 (b), the slip plane normal stress and normal plastic strain increment are first calculated, considering full 3D plane orientation and stress tensor etc. For a given slip plane orientation i , the normal stress is given by $\sigma_n^i = \boldsymbol{\sigma} : (\mathbf{n}^i \otimes \mathbf{n}^i)$, and the normal plastic strain increment $d\epsilon_n^{p,i} = d\boldsymbol{\epsilon}^p : (\mathbf{n}^i \otimes \mathbf{n}^i)$, where $\mathbf{n}^i$ is the slip system unit normal vector, and $\otimes$ is the dyadic product. Hence, numerical integration enables calculation of each slip system normal SED as follows.

$$G_{\text{NSED}}^i = \int \frac{\sigma_n^i \cdot d\epsilon_n^{p,i}}{\sqrt{\rho_{\text{SSD}} + \rho_{\text{GND}}}} \quad (4.6)$$

As before, crack path is controlled by the i^{th} slip plane corresponding to the maximum energy value. However, implementation of this method is local to the crack tip, i.e., energy terms calculated within the element directly ahead of the crack tip control crack path.

4.3 Results

4.3.1 A crack growth rate study using the maximum slip criterion for crack path

4.3.1.1 Single crystal crack paths

Crystallographic HCP crack paths in single crystals and bicrystals were previously studied in detail using the current modelling approach [73]. It was shown for orientation B-type crystals, the slip fields which developed ahead of the crack tip would lead to an alternating pattern of maximum slip between two competing prismatic slip systems. Based on the maximum slip crack path criterion, this effect was shown to produce a tortuous crack path, which is often observed in experiments for various materials [250], [251]. The single crystal crack paths from this work for orientations A and B are shown in Figure 38 based on simulations with an arbitrary critical SED of 7.25 J/m^2 ; the corresponding SED fields around the crack are also shown. The critical SED has no influence on crack path within the range considered here.

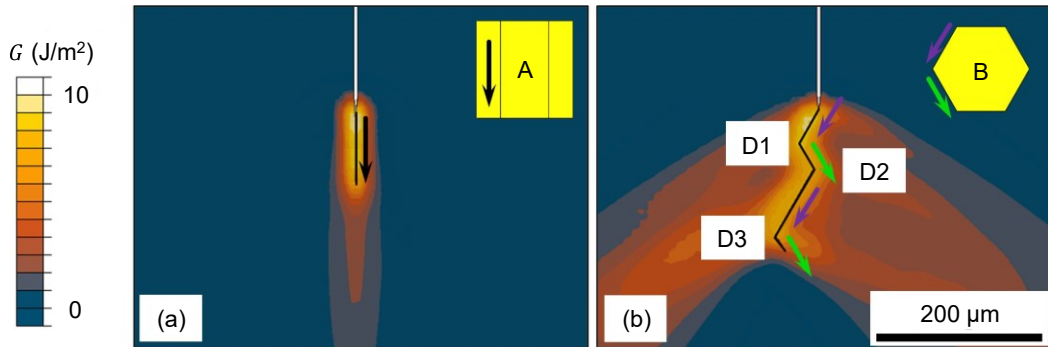


Figure 38: Crack path results and SED fields for (a) crystal orientation A and (b) crystal orientation B.

For orientation A, the crack path is entirely straight and perpendicular to the loading direction and is propagated along prismatic planes perpendicular to their Burgers vectors. Conversely, for orientation B, the crack path is tortuous and oscillates between two competing prismatic slip planes, as indicated by the purple and green arrows. As will be discussed later, crack path tortuosity can aid to diminish crack growth rate. To understand, using the maximum slip criterion, why tortuosity occurs in more depth, it is useful to analyse the evolution of shear stress ahead of the crack tip, since this is the key driver for slip activation. Figure 39 shows the cyclic evolution of the in-plane shear stress, τ_{xy} at points of crack deflection, D1 and D2 for crystal orientation B.

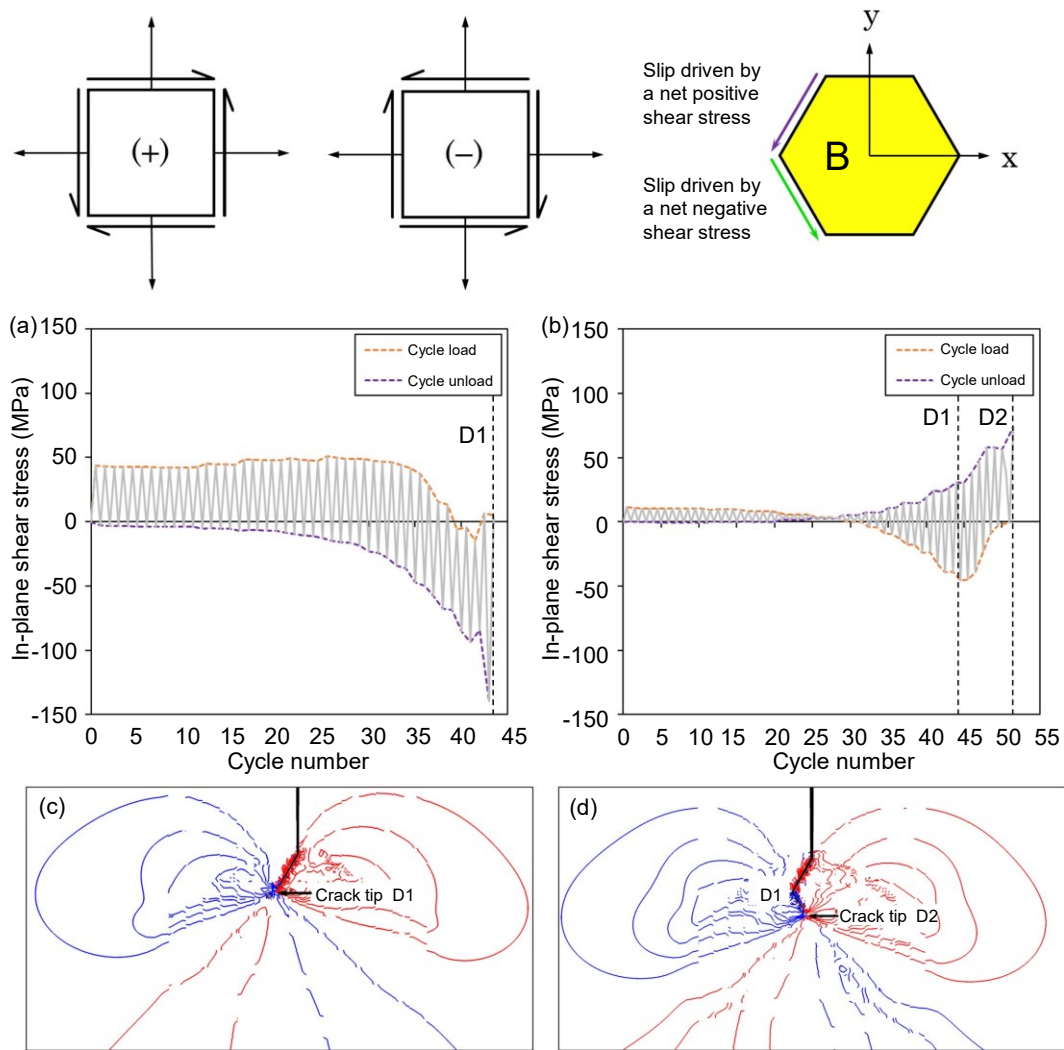


Figure 39: Single crystal orientation B – in-plane shear stress evolution with cycle number ahead of the crack tip at (a) D1 and (b) D2 and corresponding in-plane shear stress contour fields at (c) cycle 44 and (d) cycle 52, indicating regions of positive (red) and negative (blue) shear stress.

Figure 39 (a) shows the cyclic evolution of in-plane shear stress at the fixed point, D1 (up to the cycle before crack deflection), which is where the first crack deflection for orientation B

occurs. As the propagating crack approaches D1 with increasing cycle number, the local in-plane shear stress decreases rapidly and becomes dominated by a net negative shear stress. As illustrated at the top of Figure 39, a net negative shear stress corresponds to a plane of maximum shear which is inclined at a negative angle with respect to the x and y-axes. This drives maximum slip on the prismatic slip system with negative slip plane inclination, which results in the crack deflection D1. Similarly, at deflection D2, the shear stress rapidly increases and becomes positive as the crack approaches, which in turn drives maximum slip along the positively inclined prismatic slip plane, giving deflection D2. In each case of deflection, the maximum shear stress (magnitude) is driven by the unloaded peak of the cycle (far field applied stress, $\sigma_{ff} = 0$), i.e., a residual stress. Hence, the plasticity induced by shear stress in a single direction, results in a residual shear stress which will ultimately drive slip in the opposite direction. Figure 39 (c) and (d) show that at the point of deflection, the residual shear stress causes a zone of either positive (c) or negative (d) shear stress ahead of the crack tip, due to plasticity. Additionally, as the crack length increases, the positive / negative shear stress bias at the crack tip becomes more subtle, which gives rise to shorter crack lengths between deflections and therefore, increased frequency of crack oscillation.

4.3.1.2 *Single crystal crack growth rates*

Paris Law is among the most widely used models to predict FCG but is most practically applied to stage II cracks due to their dependency on stress. It is expressed via a power law relationship with SIF range, as given by Equation 2.3. Despite the strong dependence of short cracks on local plasticity, Paris' Law is used here as a tool to study the differences observed in FCG rates for crystal orientations A and B across a range of critical SED values (between 2 and 10 J/m²). Using methods described in Section 4.2.1, data are obtained for various critical values for SED. Table 9 shows that by increasing the critical SED, the Paris' Law exponent, p , remains relatively unchanged for each crystal orientation. Changes in FCG rate are instead reflected by decreasing Paris' Law constant, A with increasing critical SED. However, differences in exponent between orientations A and B are large, and hence, rates of growth in orientation A are significantly higher (excluding where ΔK is very low). Specifically, p , which governs the rate of change of FCG rate with increasing crack length, is much higher for orientation A, which reflects experimentally observed growth rate gradient differences observed in [252].

Table 9: Fitted Paris' Law constants for simulated crack growth data in single crystal orientations A and B across a range of critical SED values.

Orientation	Critical SED (J/m ²)	A ($\times 10^{-6}$)	p
A	2	250	6.01
	4	40	5.43
	6	13	5.31
	8	6	5.09
	10	3	5.93
B	2	870	3.37
	4	470	3.16
	6	150	3.74
	8	120	3.37
	10	98	3.43

To assess how the current SED-based fracture criterion captures this behaviour, the cyclic evolution of SED ahead of the growing crack tip is studied in Figure 40.

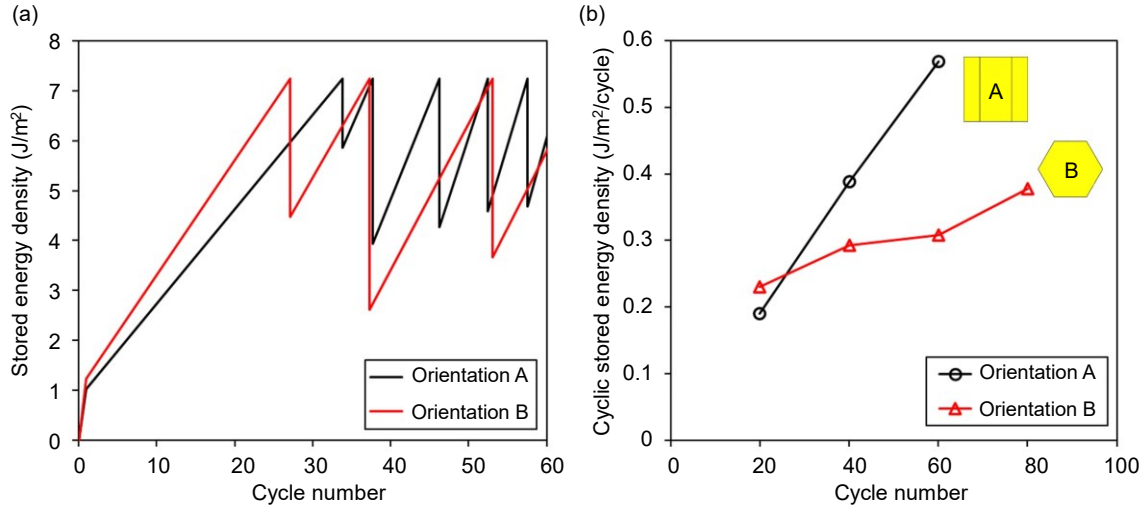


Figure 40: Cyclic evolution of SED ahead of the crack tip for crystal orientations A and B using critical SED of 7.25 J/m². (b) Corresponding cyclic SED rates plotted against cycle number for A and B.

Figure 40 (a) shows the evolution of SED ahead of growing crack tips within crystal orientations A and B. The critical SED used in this case is 7.25 J/m². For each orientation, SED increases approximately linearly with cycle number (excluding the first cycle, where there are large plastic strain increments) until the critical value is reached. At this point, the crack extends, and the crack tip is redefined, and hence the SED ahead of the new crack tip is lower; the SED ahead of the new crack tip is referred to here as remaining SED. A key difference between orientations A and B is that the remaining SED after crack advance is consistently much lower for orientation B, which impacts the crack growth rate as it takes a greater number

of cycles to reach the critical value. The remaining SED term may be expressed in terms of the critical SED to be more general, as $G_0 = \chi \cdot G_c$, where χ represents the percentage of the critical value remaining after the crack has extended by a fixed crack length, Δa . For orientation A, χ is equal to approximately 66%, compared with just 46% for orientation B (based on two single crystal simulations with critical SED values of 7.25 and 10 J/m² for each case with a constant crack extension length, $\Delta a = 10 \text{ } \mu\text{m}$). The other key factor which influences FCG rates is the rate of accumulation of SED ahead of the crack tip, dG/dN , otherwise called cyclic SED. Cyclic SED is plotted against cycle number for both orientations in Figure 40 (b). Although the cyclic SED is initially lower for orientation A, it increases more rapidly with increasing number of cycles (corresponding to an increased crack length), resulting in higher crack growth rates. Hence, since there is an approximate linear relationship between cycle number and SED, the critical SED may be expressed as $G_c = G_0 + \Delta N \cdot \frac{dG}{dN}$, where ΔN is the number of cycles to reach the critical value. For a constant crack extension length, Δa , the number of cycles required to achieve this is linked with crack growth rate as $\Delta N = \Delta a \cdot \left(\frac{da}{dN}\right)^{-1}$. Since $G_0 = \chi \cdot G_c$, this gives $G_c = \chi \cdot G_c + \Delta a \cdot \left(\frac{da}{dN}\right)^{-1} \cdot \frac{dG}{dN}$, which is rearranged to give Equation 4.7.

$$G_c = \frac{1}{1 - \chi} \Delta a \left(\frac{da}{dN}\right)^{-1} \frac{dG}{dN} \quad (4.7)$$

To use Equation 4.7 to estimate the true value for critical SED in Zircaloy-4 would require experimental in-situ measurement of remaining and cyclic SED corresponding to measured crack extension and crack growth rate. However, where crack growth in a real material is modelled explicitly using the current CPFE formulation, Equation 4.7 may be applied to both experiment and simulation. Dividing the expressions and removing the common terms gives the following expression for estimating critical SED,

$$G_{c,\text{exp}} = G_{c,\text{sim}} \cdot \left(\frac{\frac{da}{dN}_{\text{sim}}}{\frac{da}{dN}_{\text{exp}}} \right) \quad (4.8)$$

where subscripts exp and sim represent experiment and simulation, respectively. Hence, this pragmatic approach for estimating the true critical SED only requires the ratio of simulated and experimental crack growth rates. Unlike SIF range, SED is microstructurally sensitive as

it accounts for local variation in material properties and microstructural features such as grain boundaries. It is therefore a much more promising candidate for predicting rates of short FCG. In Figure 41, SED and cyclic SIF range are plotted separately against crack growth rate for single crystal orientations A and B.

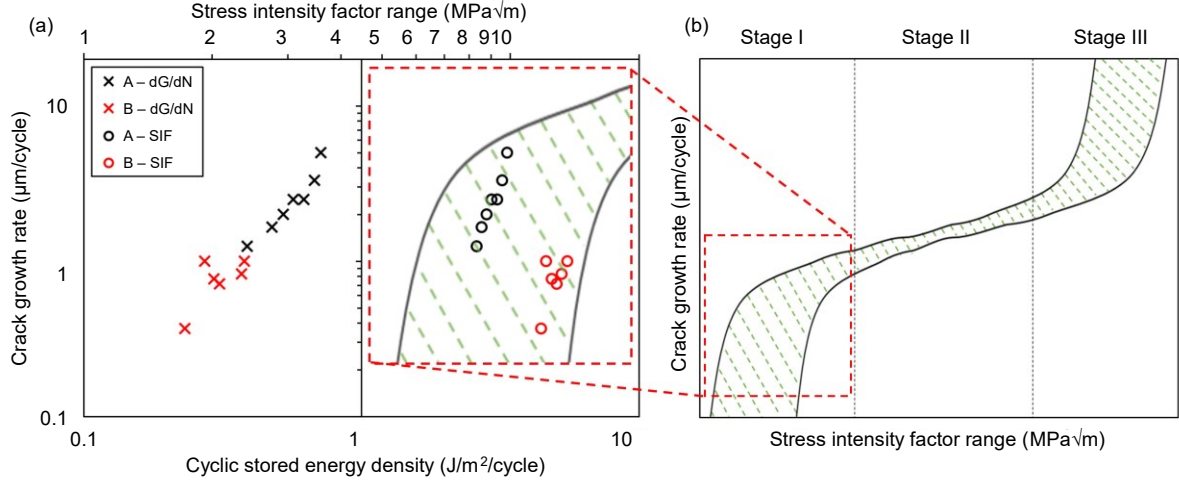


Figure 41: Comparison of cyclic SED with SIF plotted against crack growth rate for crystal orientations A and B (simulated using the same value for critical SED). (b) Illustration outlining large degree of variability associated with SIF vs. crack growth rate within the short crack growth regime.

Figure 41 (a) shows that for orientations A and B, there is a significant difference between the trends observed for cyclic SED and SIF range. Specifically, SIF range plotted against crack growth rate gives two distinct trends for each crystal orientation, whereas there is a more continuous trend between cyclic SED and crack growth rate across both orientations. Hence, the well-known large degree of scatter [253] associated with stage I or short FCG rates (see Figure 41 (b)) may be addressed using SED. Additionally, Figure 41 (a) suggests that a single value for critical SED should be capable of capturing FCG rates within materials, regardless of texture.

4.3.1.3 Polycrystalline crack growth: influence of microstructure

In this section, the influence of microstructure on FCG rates is studied based on experimental crack growth data [175], [252] from two Zircaloy-4 samples (AA and BB) with strongly differing textures. These results are particularly interesting because of a clear difference in growth rate trend between the samples, and because of apparent fluctuations in growth rate as the crack crosses several microstructural features in sample AA. Sample AA is the focus of this study because of (i) the clear link between experimental growth rate and microstructural features and (ii) the maximum slip criterion was unsuccessful in accurately predicting crack path (which

is not the focus of this part of the chapter) in sample BB. The simulated crack path for sample AA is presented in Figure 42.

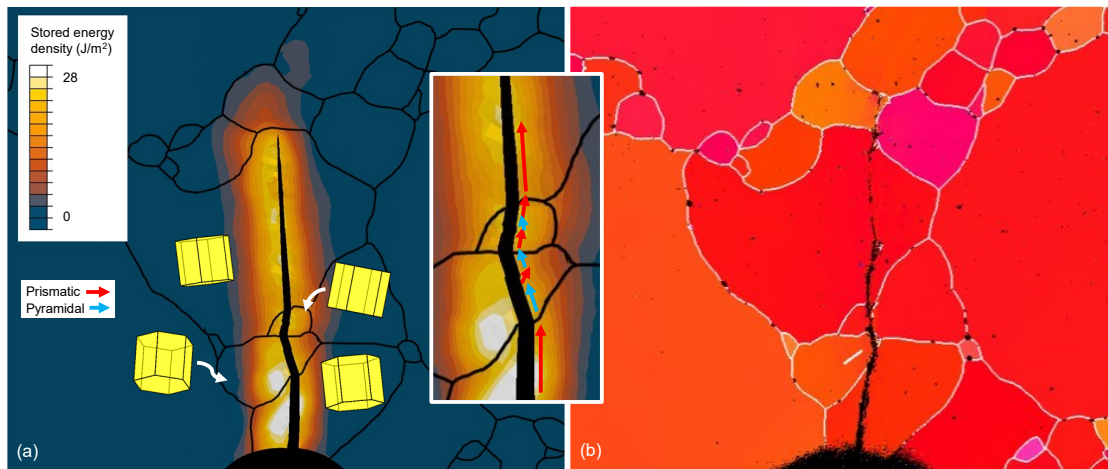


Figure 42: Entire simulated crack path in polycrystalline sample AA with SED field. Red and blue arrows represent crystallographic crack plane type. (b) Corresponding experimental crack path in polycrystalline sample AA from Wan [76], [252].

Figure 42 (a) shows the simulated crack path in sample AA, using a critical SED of 20 J/m² (which is an arbitrary value selected to ensure several fatigue cycles between crack advance through any given element); the corresponding SED distribution is also shown. In Figure 42 (b), the experimental path is presented for comparison. Within the first grain (starting at the notch), the simulation predicts crack growth along a prismatic plane, giving a near-vertical crack path. In the experiment, however, the crack path tends to skew rightward, indicating that the true crack path may predominantly follow pyramidal planes. This affects the point at which the crack crosses the first grain boundary and enters the second grain. In consequence, there is little experimental crack path data to study in this grain. However, the model predicts mostly pyramidal crack growth (with a single oscillation on a prismatic plane), which appears to correspond with the experimental crack direction. Within the third grain, the experimental crack path appears to follow the grain boundary quite closely before crossing the grain's uppermost triple junction. Due to the initial skew in the simulated crack path, however, the crack is simply propagated along prismatic and pyramidal slip systems before crossing at the grain boundary. Within the fourth grain, the model predicts crack growth entirely along prismatic slip planes. This correlates well with large portions of the experimental crack direction, though there are some deviations and oscillations between prismatic and pyramidal planes. In summary, the maximum slip criterion used in the simulation captures the overall experimental crack path reasonably well, with the greatest difference being in the first grain.

Analysis of predicted slip activity in this grain shows that several prismatic and $\langle a \rangle$ pyramidal slip systems are active, but not dominant, which might otherwise have enabled more precise predictions.

Using projected crack length and cycle data from this simulation, the predicted crack growth rate trend is compared directly with experimental results (see Figure 43).

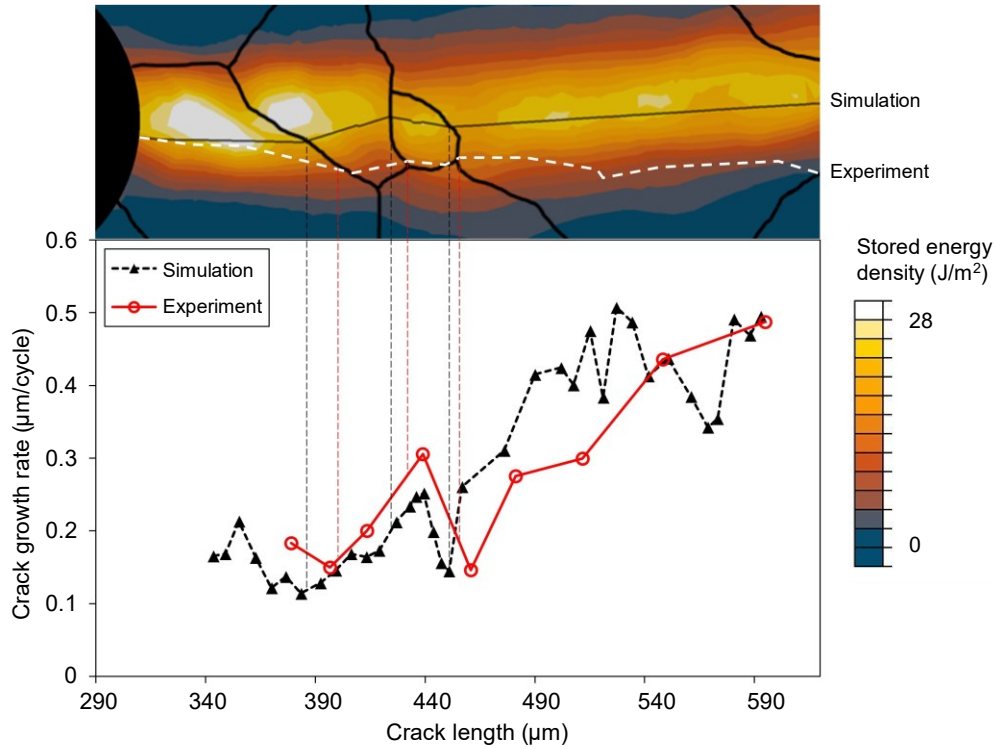


Figure 43: Simulated and experimental crack growth rates plotted against crack length in polycrystalline sample AA. Experimental crack path (white dashed line) is overlaid on simulated crack path (black line) above. Vertical dashed lines correspond with where crack tip interacts with microstructural features.

In Figure 43, the simulated FCG rates are presented using a three-point moving average trendline due to there being a greater number of data points than available from the experiment. While data acquisition frequency can have some slight impact on the overall FCG rate trend, the surplus data points from simulations are maintained due to differences between simulated and experimental crack paths. Additionally, simulated growth rates have been adjusted to account for differences between the experimental (true) critical SED and the value used in the simulation via Equation 4.8. The value used in the simulation was chosen arbitrarily such that results were generated within a reasonable computational time. This will be discussed in more detail in subsequent sections. At the top of Figure 43, exact differences in crack path become clear, and hence, it can be anticipated that growth rate trends will not be predicted

precisely. Beginning in the first grain, the simulation shows a gradual FCG rate retardation prior to the boundary, which is followed by a gradual increase in rate immediately after crossing the boundary. The experimental data show the same phenomenon. However, as the crack tip approaches and crosses the second grain boundary, the rate continues to increase for both experiment and simulation, before reaching a peak within the third grain. This is followed by a dramatic reduction in FCG rate until the crack crosses the third grain boundary. Beyond this point, the model predicts several growth rate fluctuations, with a generally increasing trend, as is observed experimentally. Hence, despite differences in overall crack path, the model has captured the large fluctuations in FCG rate and trend reasonably well.

To understand FCG rate trends in more detail, it is useful to study the evolution of SED at microstructural features (along grain boundaries and at triple junctions), as this is the key factor affecting growth rate. The third grain is particularly interesting in this sense, as significant differences in rate are observed prior to crack propagation across each boundary. As the crack approaches boundary 2, it accelerates, and continues to do so until it reaches (approximately) the centre of the grain, at which point, the crack decelerates significantly as it approaches boundary 3. After the crack crosses boundary 3, it begins to accelerate again. Hence, the evolution of SED along each of the three grain boundaries formed with this grain is analysed. The results are presented in Figure 44. Figure 44 (a) shows the region of interest for this analysis; around the third grain, three paths have been selected corresponding to the three grain boundaries of interest, which lie between each of the grain's three triple junctions (TJs), labelled 1-3. Figure 44 (b) shows the cyclic evolution of SED along the boundary between TJ1 and TJ2. The maximum cycle number (115) in this case is selected prior to crack propagation across the first grain boundary (path 1). At cycles 60, 90, and 100, maximum SED is measured at TJ1, which decreases continuously up to TJ2. At cycles 110 and 115, the influence of the growing crack becomes evident, as SED is maximised nearest the crack tip. Figure 44 (c) shows SED evolution between TJ2 and TJ3. With increasing cycle number, SED appears to increase almost uniformly along the grain boundary, at a magnitude equivalent to approximately half of that at TJ1. Lastly, Figure 44 (d) shows SED evolution between TJ1 and TJ3. Here, SED decreases significantly from TJ1 to TJ3.

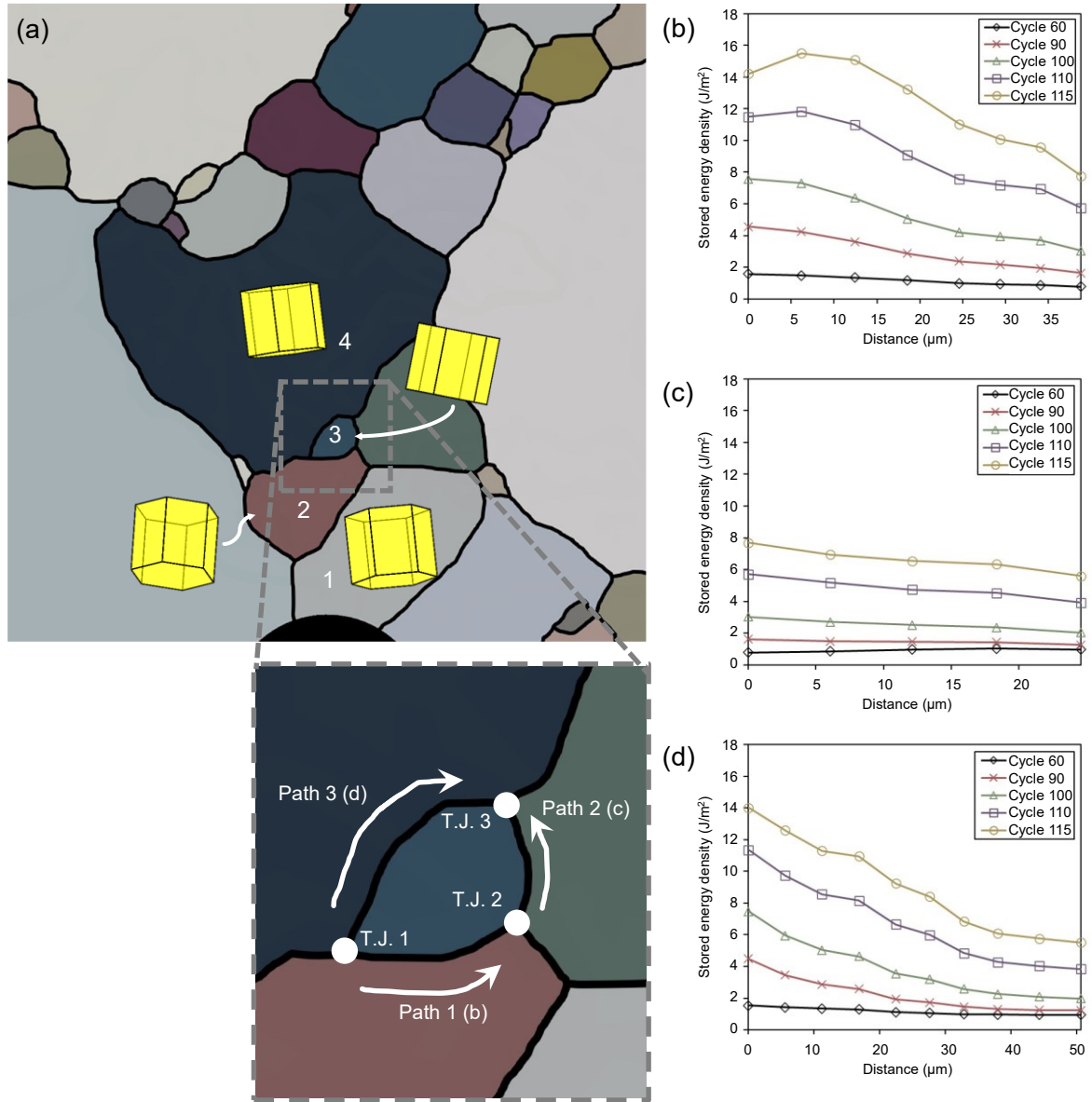


Figure 44: Illustration of entire polycrystal sample with ROI around grain 3, including triple junction sites and paths around the grain. (b), (c), and (d) show SED distribution along paths 1, 2, and 3, respectively for a range of cycle numbers (note that maximum cycle number plotted is prior to the crack entering grain 3). The critical SED used in this analysis is also 20 J/m².

Overall, Figure 44 indicates that throughout the simulation, there is high SED localisation at TJ1, with minimum SED at TJ3. This observation coincides with the FCG rate trend across this grain, as the crack accelerates ahead the first boundary (high SED) and decreases ahead of the second boundary (low SED). However, the role of each boundary in affecting SED is not yet clear.

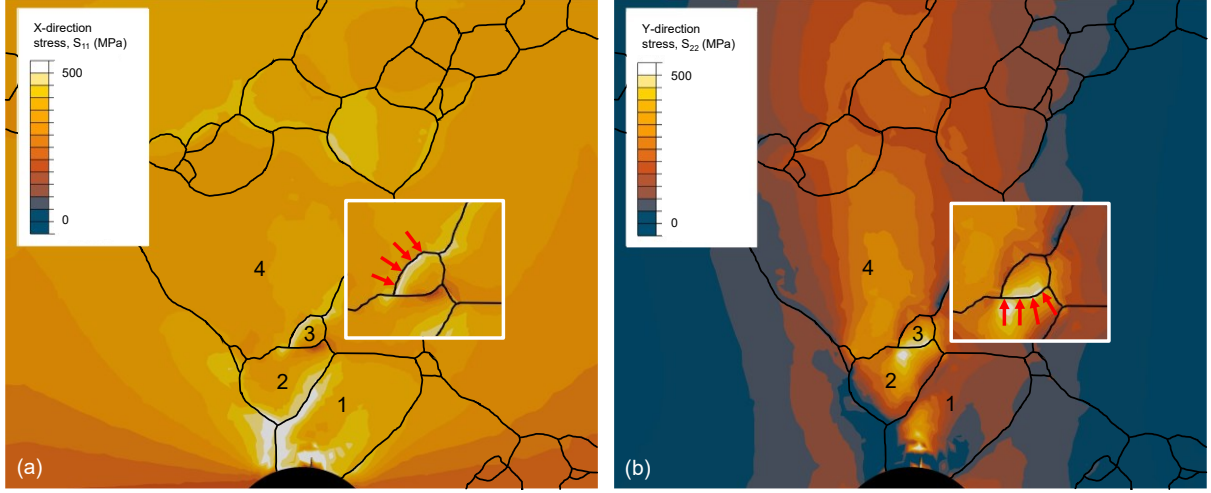


Figure 45: (a) x-direction stress distribution at the peak of the first cycle. (b) y-direction stress distribution at the peak of the first cycle. Grain 3 is highlighted in each case to illustrate stress concentrations along two of the boundaries.

The x-direction stress field (S_{11}) in Figure 45 (a) shows that around the third grain, there are regions of stress localisation and regions of comparatively low stress. Specifically, there is some S_{11} localisation along the lower portion of the TJ1→TJ3 boundary. Similarly, Figure 45 (b) shows significant S_{22} localisation along the TJ1→TJ2 boundary, and hence local stress redistribution resulting in S_{22} minima around the upper region of the grain. Elastic and yielding property differences across each of these boundaries are presented in Table 10. Yield stress is estimated for each grain using Schmid factors and initial (zero dislocation density evolution) CRSS values for each slip system.

Table 10: Elastic modulus and yield stress mismatch between grain pairs across each grain boundary for grain 3 in x- and y-directions.

Boundary	C_{11} mismatch (%)	C_{22} mismatch (%)	σ_{y11} mismatch (%)	σ_{y22} mismatch (%)
TJ1→TJ2	2.2	12.0	3.4	70.8
TJ2→TJ3	2.1	1.5	2.7	1.2
TJ1→TJ3	1.5	0.7	1.6	3.7

Table 10 shows the mismatch in stiffness and yield stress between grains corresponding to each grain boundary for grain 3. This highlights a significant difference in the y-direction modulus across the TJ1→TJ2 boundary, which explains the y-direction (perpendicular to the boundary) stress localisation in this region. Additionally, there is a very large (70.8%) yield stress mismatch across this boundary in the y-direction, which gives plastic strain localisation. These factors lead to local cyclic accumulation of SED along the grain boundary, prior to interaction with the crack. Therefore, the rapid crack acceleration across the boundary and into grain 3 is

driven primarily by elastic and plastic TJ1→TJ2 boundary interactions. Localisation of x-direction stress along the TJ1→TJ3 boundary is likely a secondary stress effect, due to relatively low property mismatch. Additionally, stress localisation leads to local stress redistribution resulting in low stress and plastic strain and consequently low SED along the upper portion of the TJ1→TJ3 boundary, across which the elastic and yield stress properties have low mismatch, which explains the dramatic FCG rate reduction as the crack approaches it.

4.3.1.4 Estimating the critical stored energy density

Despite the maximum slip criterion failing to accurately predict crack path in sample BB, the crack growth rate trend may indicate how well the SED driving force can capture differences in FCG rate between samples AA and BB.

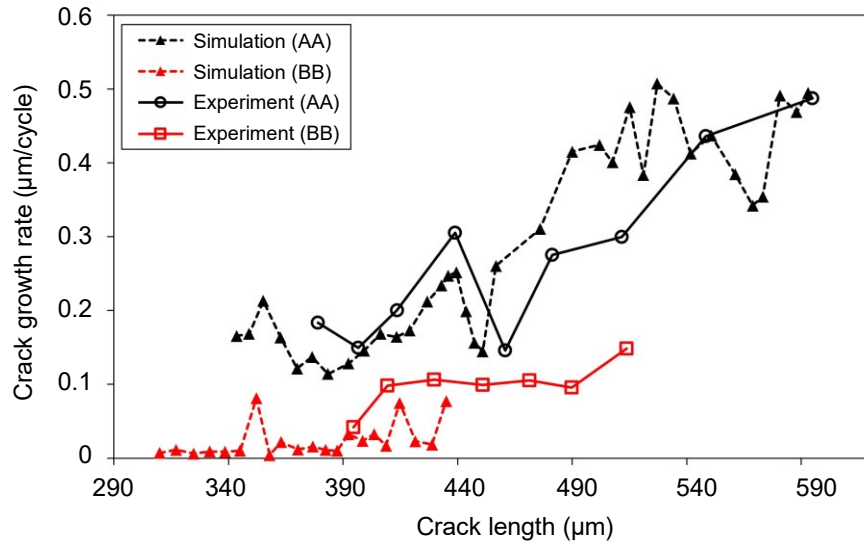


Figure 46: Simulated and experimental crack growth rates plotted against crack length in polycrystalline samples AA and BB.

Simulated and experimental FCG rates for sample AA and BB are compared in Figure 46. Note that less simulation data is available for B due to computational time constraints and differences in crack path. As before, the simulated rates are first adjusted to account for differences in critical SED between simulation and experiment. The same critical SED is used for simulating samples AA and BB. Firstly, comparing simulated rates between AA and BB, the model captures both the very large difference in growth rate magnitude as well as the steeper growth rate gradient in sample AA. However, due to lack of availability of simulated rate data and crack path differences in sample BB, experimental and simulated FCG rates cannot be compared in detail. Despite this, these results indicate that the simulated to

experimental growth rate ratio may be assumed constant for samples AA and BB, hence allowing for prediction of the true experimental critical SED for Zircaloy-4 using Equation 4.8. This gives $G_{\text{c,exp}} = 250 \text{ J/m}^2$. This scaling approach enables estimation of the true critical SED, but direct simulation of crack growth using this value is made impossible due to severe computational expense.

4.3.2 A study to evaluate and compare the performance of various crack path drivers

4.3.2.1 Establishing a non-local measurement radius for the maximum energy release rate criterion

As discussed in previous sections, the mechanistic basis for the SED driving force for crack growth is that elastically recoverable energy, which is stored in crack tip dislocation structure, specifically GNDs, is released when it exceeds a critical value. Hence, for the maximum energy release rate criterion for crack path, the non-local distance over which measurements are taken is linked with the distribution of crack tip GNDs. Several single crystal CPFEM simulations were carried out in which a remote stress of 200 MPa was applied to edge-cracked material with crack lengths ranging from 1 to 250 μm (crack lengths within the physically and microstructurally short crack growth range). Figure 47 (a) shows the spatial distributions of GND density ahead of each crack. It should be noted that due to the limitations of XFEM in *Abaqus*, linear brick elements are used in these analyses. Hence, since GND density is related to the second order spatial derivative of displacement, it is not easily calculated using conventional linear elements. The sub-element method [157] for calculating spatial gradients of plastic deformation and GND density is used here, which is described in detail in Chapter 3. As described by Xu et al., [175] a one-dimensional analytical solution for crack tip GND density can be derived via the radial plastic strain gradient [169], $\rho_{\text{GND}} = \frac{1}{b} \frac{\partial \gamma^{\text{p}}}{\partial r}$, where b is Burgers vector magnitude, γ^{p} , is plastic shear strain, and r is a measurement of distance (in this instance, radial distance from the crack tip). Assuming a blocked slip system, where GNDs are contained between the crack tip dislocation free zone (DFZ) and the plastic-elastic boundary, the analytical model [175] is,

$$\rho_{\text{GND}} = \frac{K_{\theta}}{2b\mu(2\pi)^{\frac{1}{2}}} r^{-\frac{3}{2}} \quad (4.9)$$

where K_{θ} is an orientation-specific shear stress intensity factor term (which is directly proportional to the global crack tip stress intensity factor) and μ is shear modulus. Hence,

Figure 47 (b) shows the radial distributions of GNDs, normalised for each crack length by crack tip stress intensity factor, i.e., $\frac{\rho_{\text{GND}}}{\sqrt{\pi a}}$, since $K_{\theta} \propto \sqrt{\pi a}$.

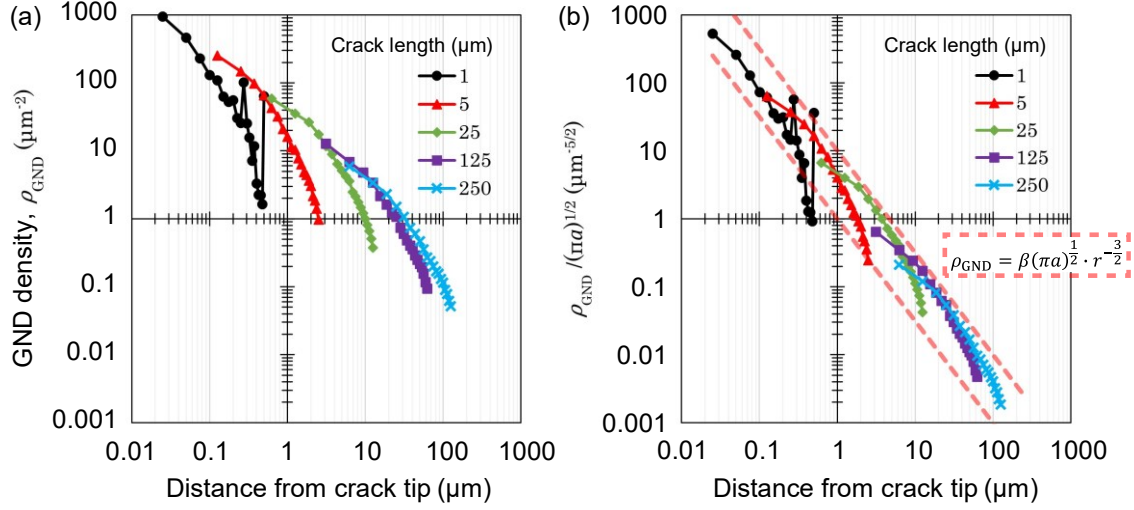


Figure 47: CPFE modelling of GND density at crack tips using single crystal orientation A; GND density is measured vertically downward from the crack tip along the plastic strain field direction. (a) Predicted distribution of GND density ahead of cracks of varying lengths. The remote applied stress is constant for each case. (b) Accounting for variations in crack length to establish a general relationship between crack tip GND density and distance from the crack tip. The dashed lines follow the power law trend with exponent $-3/2$.

Figure 47 (b) shows that by normalising results against stress intensity factor, an approximate convergence for each crack length is achieved, which is demonstrated to conform to the power law relation as $\frac{\rho_{\text{GND}}}{\sqrt{\pi a}} = \beta r^{-3/2}$, where $\beta = \frac{1}{2b\mu(2\pi)^{1/2}}$. Therefore, using this analytical relationship, an estimate is obtained for the distance from the crack tip in which the majority of GNDs are contained. It is estimated that 99% of GNDs are contained within 18.97 μm from the crack tip. Hence, a non-local radius, $r_1 = 20 \mu\text{m}$ is selected for SED measurement using the maximum energy release rate criterion. As radius, r_2 has no physical meaning, it is chosen on the basis of mesh size alone, such that at least three integration points are accounted for with each measurement. Here, the measurement radius is set to equal the mean element size, which for the microstructure simulations presented is 4 μm .

4.3.2.2 Comparison of methods in single crystals

The maximum slip criterion for crack path has been shown in several studies to capture short crack phenomena such as tortuosity [176], [177], which is a key feature of short crack growth in HCP materials. This is most commonly observed in HCP grains which are oriented such that the prismatic plane Burgers vector directions lie in the plane of crack growth and are

inclined relative to the mean crack growth direction. In Figure 48, which presents results from additional single crystal studies, the first orientation (from left to right) meets this description. Note that crystal orientation is represented by yellow HCP unit cell orientations, corresponding to the orientation observed from the sample free surface. Single crystal orientations A and B are in the first and third positions, respectively, while those in second and fourth positions are unique to the crack path driver study.

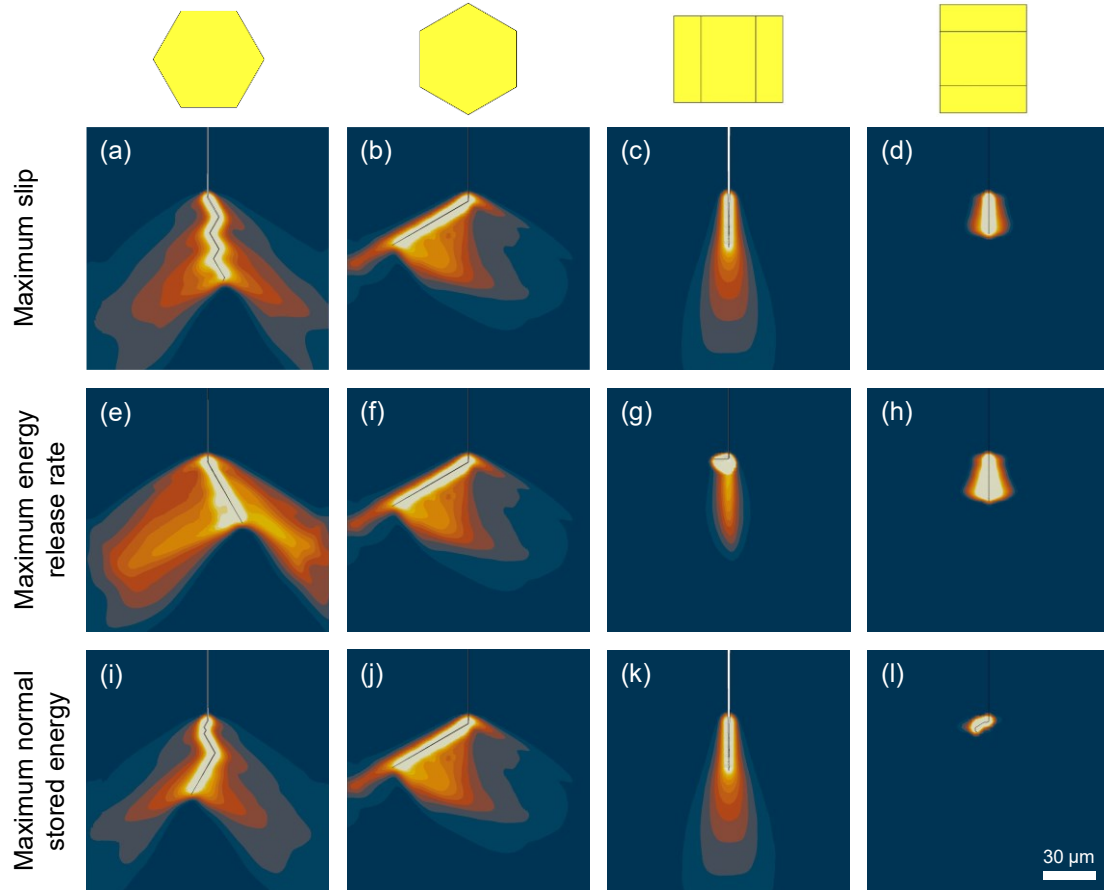


Figure 48: Comparison of single crystal crack path predictions using the maximum slip (a – d), maximum energy release rate (e – h), and maximum normal stored energy (i – l) criteria. The corresponding SED fields (with arbitrary units) are also shown for illustration, which are particularly important for the maximum energy release rate criterion.

The predicted crack paths for the first single crystal orientation are shown in Figure 48 (a), (e), and (i). Similar results using the maximum slip criterion have been presented by Wilson et al. [73] in previous studies. Hence, as anticipated, a tortuous crack path which oscillates between symmetric prismatic planes (parallel with their Burgers vector directions) is predicted in Figure 48 (a). Another key feature of this crack path is that the frequency of oscillations tends to increase with increasing crack length, which has been discussed here in detail. Interestingly, the maximum energy release rate criterion (Figure 48 (e)) predicts crack growth

along a single prismatic plane direction (the selection of which is arbitrary due to symmetry) and does not predict tortuosity. The SED field around the crack shows that the highest levels are not ahead of the crack tip but are to the left of the inclined crack. However, the predicted crack path is based on energy ahead of the crack tip (i.e., energy which can affect crack path) only, which directs the crack path linearly. This suggests that if crack nucleation were included within the current model, crack bifurcation may be predicted, for which there is ample experimental evidence [226], [254], [255], [256]. The maximum normal stored energy criterion does predict a tortuous path for the first orientation, which differs slightly from the maximum slip result in that the segment length between oscillations increases, and therefore the frequency of oscillations decreases with increasing crack length. For the second orientation, each of the three methods produced the same result, which is a straight crack path along a prismatic slip plane. Due to symmetry, each model could have equally predicted growth along the prismatic plane which slopes downward and to the right. Due to near inactivity of the third (vertical) prismatic slip system, crack growth is not predicted along it using any method. However, if the slip activity constraint were not applied when using the maximum normal energy criterion, it is foreseeable that a vertical crack path along this plane may have been initially predicted. For the third crystal orientation considered, a vertical crack path following prismatic planes (perpendicular to their Burgers vector directions) is predicted using both the maximum slip and normal stored energy criteria. However, the maximum energy release rate method predicts basal cracking (Figure 48 (g)), resulting in a crack path which is parallel with the remote loading direction. Lastly, maximum slip and maximum energy release rate agree for the final orientation, predicting basal cracking, while a tortuous path along basal, prismatic, and pyramidal planes is predicted using the maximum normal stored energy criterion.

4.3.2.3 *Comparison of methods applied to real microstructures*

In this section, the three criteria for short crack path are compared directly with the two experimental datasets for samples AA and BB. The experimental crack path from sample AA is compared against the theoretical criteria in Figure 49. Without excessive repetition, Figure 49 (a) shows reasonable agreement between the maximum slip model and experimental results; the key difference here is that the simulated path deviates leftward initially due to crack growth along a prismatic plane, rather than an apparent rightward crack path along a pyramidal plane. General crack path trends are captured within individual grains after this point, particularly in the final grain, where prismatic cracking is dominant.

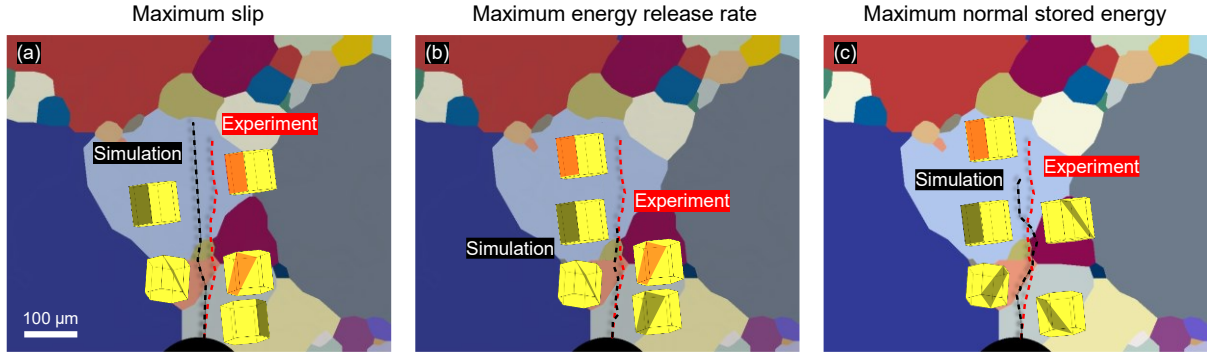


Figure 49: Comparison of three short crack prediction methods with experimental crack path in sample AA. Apparent experimental crack planes are shown with red shading, model-predicted crack planes are shown with black shading. (a) Simulated crack path using maximum slip criterion. (b) Simulated crack path using the crystallographic maximum energy release rate criterion. The simulation terminated prematurely due to local convergence issues. (c) Simulated crack path using the crystallographic maximum normal SED criterion.

Figure 49 (b) shows strong agreement between the model using the maximum energy release rate criterion and experiment (up to the point at which the simulation terminates). Specifically, in the first grain, the predicted crack path first oscillates and is then propagated along a pyramidal plane, which corresponds closely with the apparent experimental crack plane. Moreover, in the second and third grains, a combination of pyramidal and prismatic cracking, respectively, is predicted. This is in accord with the maximum slip model. Little maximum energy release rate data is available for cracking in the last grain; however, the predicted crack segment agrees with the maximum slip model and the (majority of the) experimental path. The maximum normal stored energy criterion is compared with the experiment in Figure 49 (c). While the overall crack path trend is reasonably captured here, most individual crack segments disagree. In the first grain, the crack path is pyramidal, but deviates leftward, rather than rightward. Contrarily, like the experiment, the simulated path in the second grain is also pyramidal, but deviates rightward rather than leftward, as shown in the experiment. In the final grain, crack tortuosity is predicted, which is followed by prismatic cracking, which is in conformity with the experiment and the other theoretical criteria considered.

The experimental crack path from sample BB is compared against the theoretical criteria in Figure 50. The maximum slip criterion is shown in Figure 50 (a) to fail dramatically at capturing the experimental path.

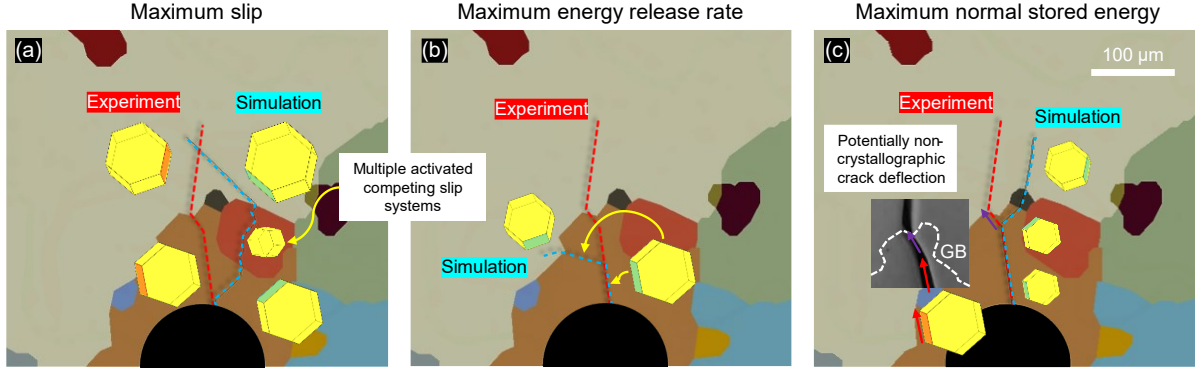


Figure 50: Comparison of three short crack prediction methods with experimental crack path in sample BB. Apparent experimental crack planes are shown with red shading, model-predicted crack planes are shown with cyan shading. (a) Simulated crack path using maximum slip criterion. The initial crack direction is such that it propagates into a grain for which there is no experimental data for comparison. (b) Simulated crack path using the crystallographic maximum energy release rate criterion. (c) Simulated crack path using the crystallographic maximum normal SED criterion. Experimental crack path deflection near the grain boundary is shown in detail with a SEM image.

The experimental crack path in sample BB is dominated by a single prismatic slip plane direction in both grains through which it propagates. As the crack first approaches the grain boundary, it deviates slightly leftward, seemingly along a pyramidal plane. Figure 50 (a) shows that in both grains, the maximum slip criterion also predicts prismatic-dominated paths, but which differ significantly from the experiment. Naturally, the model predicts maximum shear strain (slip) on planes which are inclined closest to 45° relative to the remote loading direction (in 3-point bending, this is the horizontal direction, though in reality, the stress-state is multiaxial due to the notch and microstructure). A simple Schmid factor analysis, based on uniaxial loading in the horizontal direction reveals that the prismatic slip systems controlling the experimental crack path have the lowest Schmid factors among all prismatic systems (0.18 and 0.16 for the first and second grains, respectively). By contrast, the Schmid factors for the maximum slip criterion predicted cracks were highest, at 0.48 and 0.42, respectively. The maximum energy release rate criterion predicts an entirely different result, shown in Figure 50 (b). In the first grain, there is initially concurrence with the experiment (crack growth along low Schmid factor prismatic plane), until there is a leftward deflection along the third (intermediate Schmid factor) prismatic plane, before crossing into the second grain. At this point, a downward deflection is predicted, resulting in artefactual self-termination of crack growth; this is discussed later in greater detail (see Figure 52). Figure 50 (c) shows the maximum normal stored energy criterion predicts a markedly improved crack path compared with the other methods. The initial crack direction matches the experiment closely until it approaches the grain boundary; just as in the experiment, there is a deflection close to this

grain boundary, but in a different direction to the experiment. Note that most of the crack growth in the first grain is controlled by the prismatic system shown in Figure 50 (c), but cracking is also predicted on pyramidal planes, which effectively produce the same crack path at the free surface. In the final grain, maximum normal stored energy predicts the experimental prismatic crack path precisely. Hence, the experimental crack path for sample BB is reasonably captured using this new method, including the microstructure-driven crack deflection prior to the grain boundary.

A fuller presentation of the prismatic slip system Schmid factor analysis is given in Figure 51 for the first (a) – (c) and second (d) – (f) grains in sample BB (grain numbering is according to the experimental crack path).

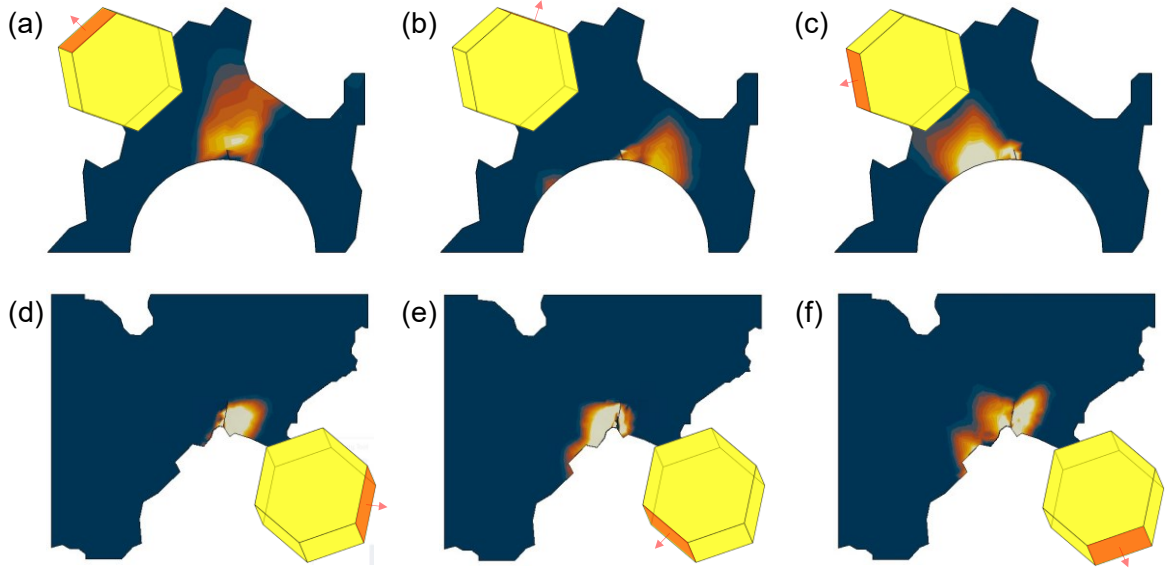


Figure 51: Differences in prismatic slip activity at two stages of crack growth in sample BB (using the maximum normal stored energy criterion). (a) – (c) predicted prismatic slip fields in the first grain after 10 fatigue cycles. (d) – (f) predicted prismatic slip fields in the second grain (with reference to the experimental crack path) after several hundred fatigue cycles.

Based on the maximum slip simulation of sample BB, shown in Figure 50 (a), subfigures (a) and (c) of Figure 51 are of most interest, since the predicted crack propagates along the plane shown in Figure 51 (a), while the experimental path appears to propagate along plane shown in Figure 51 (c). The slip fields indicate clearly that the prismatic system with maximum slip ahead of the specified crack initiation location (which is according to the experiment) is that which is presented in Figure 51 (a). However, it should be noted that due to the notch geometry, the overall maximum slip magnitude within the first grain (at this stage of crack

growth) occurs on the slip system shown in Figure 51 (c), but in a different region to the crack initiation site. Meanwhile, lowest prismatic slip is predicted to occur on the slip system shown in Figure 51 (b). Hence, it could be speculated that notch geometry or inaccurate crack initiation site definition may adversely affect the final crack path using the maximum slip model; to address this, four equally spaced crack initiation sites spanning 100 μm of the notch are considered. In some cases, particularly those where the crack initiation site is towards the left side of the notch, there is up to 30 μm of crack growth in a direction which agrees with the experiment. However, each case ultimately results in cracking along the plane shown in Figure 51 (a), and hence the final crack path is effectively independent of initiation site and notch geometry. Figure 51 (d) – (f) shows prismatic slip activity in the second grain; using the maximum slip criterion, crack growth is predicted to occur on the plane shown in (e), whereas the experimental path, and that predicted using the maximum normal stored energy criterion is controlled by the prismatic plane shown in (d).

Results at different stages of crack growth in sample BB using the maximum energy release rate criterion are presented in Figure 52. Figure 52 (a) shows that after several fatigue cycles, the region of highest SED within the measurement zone is slightly to the left of the specified crack initiation site, which results in the prismatic crack path shown in Figure 52 (b). Up to this point, the model is consistent with the experiment. Due to depletion of stored energy ahead of the crack (i.e., by maximum rate of release), the crack is propagated leftward along an alternative prismatic plane, as shown in Figure 52 (c). This crack path results in the accumulation of energy ahead of the initial crack path, rather than the crack tip; however, since the measurement zone using this method is kept ahead of the crack tip, this is unaccounted for. This is considered in greater detail in the discussion section of this chapter for prediction of short crack bifurcation phenomena. Lastly, as the crack traverses the grain boundary (see Figure 52 (d)), it is propagated in a downward facing direction, resulting in crack arrest.

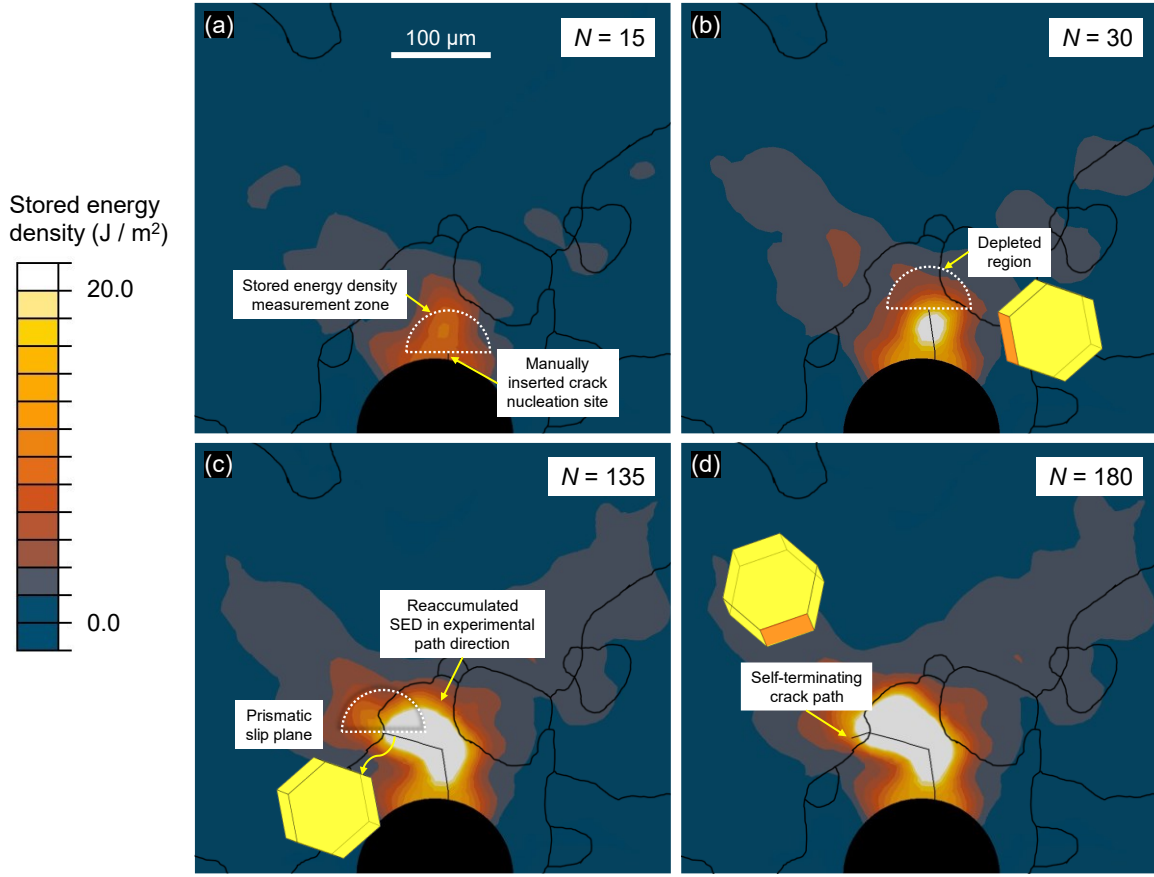


Figure 52: SED fields in microstructure BB at key four key stages of crack growth using maximum energy release rate criterion. (a) SED field at 15th fatigue cycle ($N = 15$), prior to crack propagation. Energy measurement semi-circle is shown ahead of the manually inserted edge crack. Direction corresponding to maximum energy release agrees with the experiment. (b) Energy field at first point of crack deflection. Within the semi-circular measurement zone, there is a region which is depleted of energy in the original crack growth direction. (c) Energy field at $N = 135$, where the crack first interacts with a grain boundary. The crack is clearly growing away from the region of highest SED, as there is reaccumulated energy in the experimental crack growth direction. (d) Final crack path, as crack artefactually propagates in a self-terminating direction due to cross-over of measurement zone between grains.

For sample BB, the maximum normal SED criterion is shown to best capture the experimental crack path; the main difference is that before traversing the grain boundary, the crack is deflected towards the right, rather than the left, as it does in the experiment. To understand the combined influence of microstructure features and the crack tip on the predicted crack path using this method, Figure 53 shows the cyclic evolution of predicted crack plane at two points in the first grain.

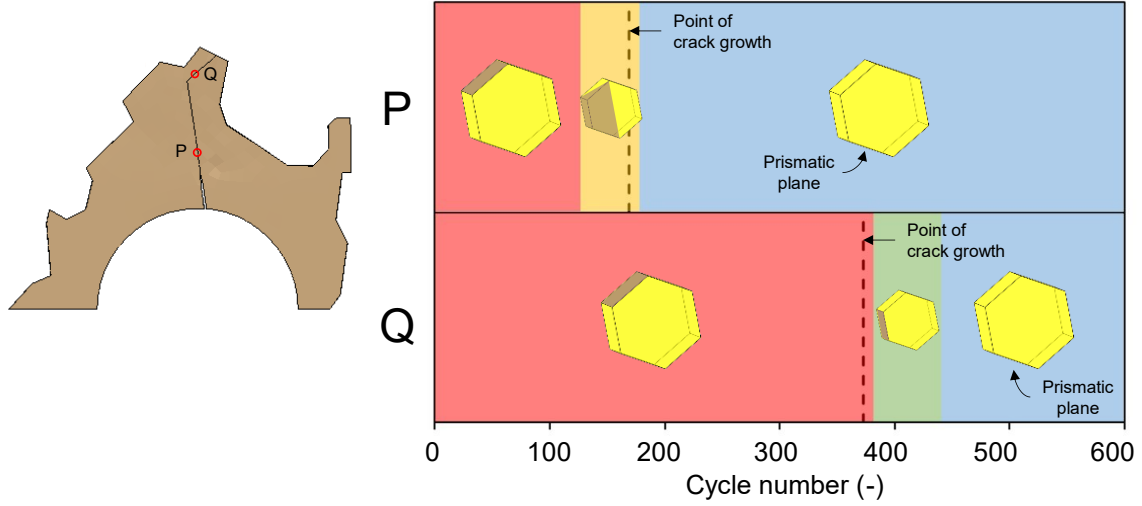


Figure 53: The evolution of predicted crack path planes during crack growth for the first 600 cycles in sample BB using the maximum normal SED criterion. Two points (P and Q) in the first grain of sample BB are selected for analysis. The cycle number at which crack growth occurs for each location is indicated using a vertical dashed line, i.e., the ‘point of crack growth’.

At point P in Figure 53, it is shown that for over 100 cycles before the crack reaches this point, the plane with maximum normal SED is the one on which maximum slip is predicted, i.e., agrees with the maximum slip model. However, as the crack length increases, and the crack tip stress fields begin to take greater effect, the predicted crack plane switches to pyramidal (which is one of the predicted crack planes which agrees with the experiment) for approximately 50 cycles before the crack passes through point P. As expected after cracking, there are large redistributions of stress, and hence, a different prismatic crack plane is predicted at point P thereafter. The evolution of predicted crack plane at point Q is more interesting. As with point P, the initial predicted crack plane at point Q agrees with the maximum slip criterion. However, due to its adjacency to the grain boundary, cyclic accumulation of SED due to grain-grain interactions results in the path at this point being less affected by the crack tip than at point P. Hence, this is a highly microstructure-sensitive method.

4.4 Discussion

4.4.1 Crack growth rates

SED is highly microstructurally sensitive and is shown here to capture FCG rate fluctuations associated with grain boundaries, and differences in FCG rate magnitude and gradient in strongly contrasting textures. This is first indicated by single crystal simulation results, which show that rates of crack growth in grains where the c-axis is parallel with the crack growth direction (orientation A) is significantly higher than in grains where the c-axis is perpendicular

to the crack growth direction (orientation B). This is reflected by Paris' Law fitting results for these single crystals (see Table 9), which also show that the rate of change of crack growth is consistently higher for the former (indicated by higher Paris' Law exponent); both phenomena were observed experimentally [175]. Simplified single crystal simulations have also enabled understanding of the key factors driving these differences, which are described using Figure 40. The first is that cyclic SED increases much more rapidly with increasing crack length (or cycle number) in orientation A; this is linked primarily with there being higher in-plane elastic modulus and "yield stress" due to the orientation of individual slip systems with respect to the loading direction, increasing the stress locally. In addition, orientation A enables activation of additional $\langle a \rangle$ pyramidal slip systems, compared with B. The second key factor is crack propagation direction; specifically, crack direction in orientation A results in the formation of new crack tips in regions of high remaining SED (after crack advance), which is lower for orientation B due to (i) a more diffuse SED distribution, (ii) misalignment between crack propagation direction and direction of maximum SED, and (iii) crack path tortuosity / meandering. This factor is generalised for these orientations in terms of critical SED using χ .

The CPFE simulation of polycrystalline sample AA shows strong agreement with the experiment in terms of FCG rate trend. Dominant experimental variations in rate corresponding to crack interactions with grain boundaries are captured by the model and are shown to be strongly linked with cyclic accumulation or lack thereof of SED. Crucially, growth rate acceleration and retardation within grain 3, followed again by acceleration within grain 4 is captured. Elastic anisotropy and yield stress mismatch at grain boundaries are shown to be dominant drivers for this, leading to stress localisation, and slip localisation within grain 3, near the boundary. This leads to local cyclic accumulation of SED as the crack propagates, ultimately enabling rapid crack propagation within grain 3, which is followed by retardation before crossing the grain 3-4 boundary. This linked with low elastic property mismatch between grains and local stress redistribution, accommodating grain 2-3 boundary localisation, and hence, minimisation of cyclic SED. Due to the nature of the free surface (2D) modelling configuration used here, the simulations have no knowledge of twist angle between incoming and outgoing crack planes, which is shown experimentally to be strongly linked with FCG retardation and acceleration at grain boundaries [209], [210]. The twist angle has been shown to affect short FCG rate most significantly. Hence, FCG rate fluctuations may not always be captured precisely using the current 2D modelling configuration.

However, the key objective for this part of Chapter 4 is to assess whether SED can capture microstructurally sensitive FCG rate, and while crack path does have an effect, it is not the dominant factor. The significant difference in rate between strongly textured samples AA and BB is captured using SED, and the ratio of simulated to experimental rate appears consistent, demonstrating that a single critical value for SED can capture FCG trends in Zircaloy-4 regardless of texture. In addition, SED captures more subtle details linked with microstructural variations reasonably well (e.g., the effect of grain boundary interactions).

The study of crack path oscillations in single crystal orientation B have provided insights for the mechanism responsible for prismatic crack path tortuosity in grains with similar orientation. In summary, it is local accumulated plastic strain ahead of the crack tip which produces a residual shear stress, opposite in sign to the in-plane shear stress responsible for driving the initial crack direction (i.e., the initial maximum slip direction). Hence, more slip is then driven on the prismatic slip system which is favoured by the new maximum in-plane shear stress direction, and the process repeats; note that the mechanism(s) driving crack tortuosity using new criteria differ. Another observation is that as crack length increases, the shear stress range between the loaded and unloaded states decreases. This indicates that fewer loading cycles are required to cause the transition from one maximum shear stress direction to the other, which increases the frequency of crack oscillation. This same phenomenon has been observed in experiments [76]. In terms of crack path prediction in polycrystalline samples AA and BB, the model is reasonably successful for AA, and highly unsuccessful in BB. In sample AA, the main differences between simulation and experiment are in grains 1 and 4. In grain 1, the simulation predicts a near-vertical crack path along a prismatic plane, whereas in the experiment, it is skewed rightward, and appears to propagate along a pyramidal plane, affecting the overall crack path. In grain 2, crack direction is captured by the model, despite it being affected by crack path in grain 1. In grain 3, simulated crack path is predominantly prismatic, but there is a lack of information from the experiment, due to its proximity to the grain boundary. In grain 4, the simulated crack path is prismatic, and aligns with crack direction observed in the experiment, excluding several oscillations. In sample BB, the model predicts a crack path which is dominated by a slip system which is highly misaligned from the true crack growth direction. Despite this, simulated FCG rate comparisons are made with experiments using projected crack lengths (which are independent of crack direction), since the focus of this part of the chapter is FCG rate sensitivity.

4.4.2 Crack paths

Simulated crack paths in single crystals with various orientations show both examples of agreement and contradiction between the maximum slip, maximum energy release rate, and maximum normal SED criteria. The HCP crack path tortuosity phenomenon is captured using both the maximum slip and normal SED criteria, and not maximum energy release rate. However, it is interesting to note that while the maximum energy release rate criterion predicts a straight prismatic crack path (Figure 48 (e)), there are regions with significant localisation of SED which are not ahead of the crack tip. Figure 54 (a) shows this crack path again, with arrows indicating regions with localised SED; the idea of crack path bifurcation is alluded to here, where these so-called ‘hot spots’ could act as potential sites for crack bifurcation, and hence crack growth in another direction. In a grain with similar orientation, Figure 54 (c) shows several prismatic crack bifurcations [76], which could potentially be explained using the maximum energy release rate criterion. Bifurcation could also enable the tortuous crack path phenomenon to be captured. However, since XFEM is used in the current studies, it is not possible to simulate bifurcated cracks explicitly. More advanced methods such as combined XFEM and cohesive zones modelling [257] could potentially enable this in the future. Another interesting example of single crystal cracking using maximum energy release rate is given in Figure 54 (b). Here, unlike the other methods where prismatic cracking is predicted, there is basal cracking parallel to the remote loading direction. This prediction reflects experimental observations shown in Figure 54 (d). As discussed with the prismatic cracking case, if crack bifurcation were enabled using some critical SED for initiation of new cracks, it is likely that an alternative crack path would be predicted using maximum energy release rate, where bifurcation from the basal crack would lead to vertical prismatic crack growth.

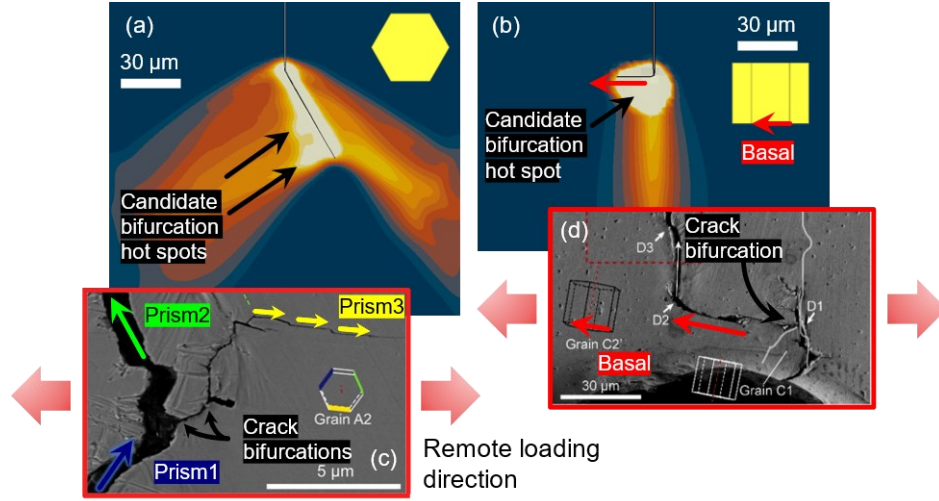


Figure 54: Single crystal observations using the maximum energy release rate criterion in the context of the literature. (a) Single prismatic crystal crack path highlighting two potential locations for crack bifurcation to occur based on stored energy field. (b) Single crystal basal crack path, where the crack growth direction is parallel with the remote applied loading direction. The SED field indicates crack bifurcation may be favourable. (c) Several examples of prismatic crack bifurcation in Zircaloy-4 [76]. (d) Example of crack bifurcation and crack growth along a basal plane which is approximately parallel to the remote loading direction [76].

Advancing to the polycrystal studies, each of the three criteria predicted the experimental crack path in sample AA reasonably well. Despite the simulation ending prematurely, it could be argued that the maximum energy release rate criterion demonstrates the greatest overall agreement with experiment. Interestingly however, the maximum normal stored energy criterion predicts pyramidal cracking (which appears to occur in the experiment also) in both the first and second grains, but in wrong directions. As a consequence, the predicted crack path using the maximum normal SED is most tortuous. The crack growth simulations of sample BB reveal that the maximum normal SED criterion is unequivocally superior to the other methods. It captures the straight prismatic (and pyramidal) cracks in the first and second grains, as well as the microstructure-driven crack deflection (in the wrong direction) prior to crossing the grain boundary. Though the crack deflection is predicted in a direction which differs from the experiment, it is significant that a deflection is predicted using this microstructure-sensitive method; the free surface extrusion approximation could potentially play a role here, since there are substructural effects which may have guided the experimental path. It is also shown that while the crack tip stress and strain states have a strong impact on the subsequent crack path, cyclic interactions between neighbouring grains can also have an important effect. It is interesting to highlight that within the first grain of sample BB, without the crack tip present, the predicted crack plane agrees with the maximum slip method. Hence,

for microstructurally small cracks, i.e., for crack initiation, it is anticipated that maximum slip and maximum normal SED criteria would yield similar results. Therefore, it is conceivable that the maximum normal SED model could act as a reliable crack path predictor across microstructurally and physically small and short fatigue cracks. Nonetheless, the new maximum energy release rate criterion initially gives promising results in sample BB also. As discussed previously, regarding Figure 52, with the introduction of some crack bifurcation capability, this criterion has potential to predict results with greater agreement with experiments.

4.5 Conclusions

Microstructural sensitivity of rates of FCG in single crystal and polycrystalline Zircaloy-4 is captured well using SED as the driving force within the current modelling framework; a single critical SED is shown to capture significant differences in FCG rate observed experimentally in samples AA and BB. From this, the experimental critical SED is estimated to equal 250 J/m². Comparison of SED with SIF range vs. growth rate demonstrates that microstructure linked short FCG rate variability is eliminated using SED. Moreover, microstructurally linked crack growth rate fluctuations are captured by the model and are strongly associated with elastic anisotropy at grain boundaries leading to cyclic accumulation of SED. In HCP single crystals, crack path tortuosity is captured using the maximum slip criterion and is linked with the cyclic accumulation of a residual shear stress due to local plasticity. In polycrystalline modelling, using the maximum slip criterion, crack path is reasonably captured in one sample, but not in another. Hence, two new microstructurally sensitive crack path criteria have been established based on the hypothesis that dislocation slip activity plays a key role but is not the dominant factor. This chapter has compared these new modelling methods and the conventional maximum slip criterion with experimental results. In overall terms, the novel maximum normal SED criterion is demonstrated as the most reliable and robust method (considered in this work) for prediction of crack paths in real microstructures. Further, it is shown to be capable of capturing the crack path tortuosity phenomenon in HCP single crystals. The maximum energy release rate criterion predictions are also shown to compare well against experimental crack paths but would likely require crack bifurcation capabilities to reach full potential. In summary, this chapter demonstrates that there is a requirement for alternative crystallographic crack path prediction methods for microstructurally short fatigue cracks, which are not based on dislocation slip activity alone.

Page would be left blank if not for this note.

5 An analytical solution for short fatigue crack growth

5.1 Introduction

The intricacy of the short FCG problem has meant that growth rates are not easily predicted, leaving a knowledge gap which can only be addressed through mechanistic understanding. This chapter presents a deterministic analytical solution to overcome this obstacle. Since the beginning of the industrial revolution, conventional fracture mechanics methods for FCG have enabled the progressive improvement of engineering design through failure mechanism understanding [258]. For example, Paris' Law [20], which relates crack propagation rate to SIF has been widely used to characterise the stage II FCG behaviour of metallic materials. However, for modern and future industrial applications (e.g., nuclear reactor fuel cladding [259], pressure vessels [260], aerospace turbine components [81], [261], submarine power cables [262]), it is recognised that component lifetimes are dominated by stage I, viz., crack nucleation and short FCG, as illustrated in Figure 55. Hence, since the length-scales for these processes are generally of the same scale as microstructure and the crack tip plastic zone, microstructure variations and plasticity become crucial; this has been discussed at length in Chapter 4.

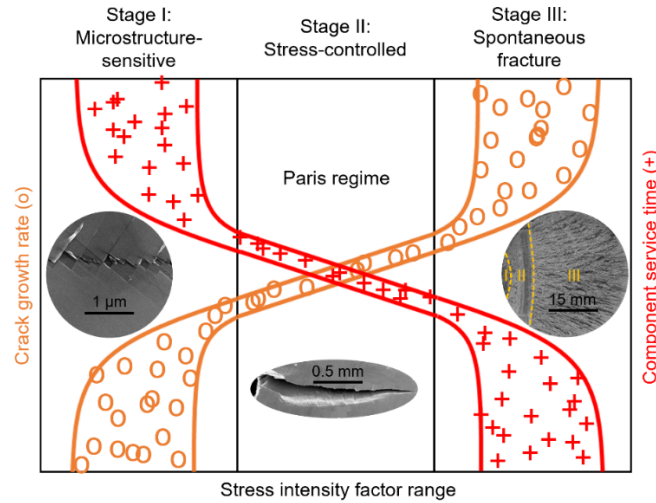


Figure 55: Illustration showing evolution of FCG rate trends with increasing SIF range from stage I (short cracks) to stage III (unstable growth). The corresponding and inversely proportional component service time trend is also shown, demonstrating that component lifetimes are dominated by stage I cracking. Stage II, denoted the ‘Paris regime’, is clearly strongly linked with SIF, and hence, is easily characterised. Conversely, stage I suffers from data scatter (when applying classical fracture mechanics) due to microstructure sensitivity. A stage I micrograph [225]¹¹ also demonstrates microstructure sensitivity of crack path (which is clearly linked with crystallography), while stage II paths [230]¹² are typically orthogonal to the remote applied stress. The stage III figure of a fracture surface is from Milan et al. [263]¹³.

¹¹ Reprinted from [225] (Figure 4 (a)), with permission from Elsevier (license number 5820760272383).

¹² Reprinted from [230] under the Creative Commons Attribution 4.0 International Public License.

¹³ Reprinted from [263] (Figure 3), with permission from Elsevier (license number 5820760547903).

Consequently, classical approaches are made redundant, and the necessity for new microstructure-sensitive fracture mechanics becomes clear. In this chapter, microstructurally short fatigue cracks follow the definition adopted by McDowell [247], in which the length of the crack is of the same order as several microstructure features. In addition, only physically short cracks are considered, i.e., where crack length is less than 1 mm. Microstructural features such as grain boundaries add further complexity to short FCG modelling as they can strongly influence the local behaviour in terms of rate and path due to impedance of dislocation motion, pileup of dislocations, and stress localisation. For example, atomistic modelling by Sangid et al. [264] was used to investigate the behaviour of a range of grain boundary types in terms of the energy barriers for and hence resistance against dislocation transmission. Trends were compared with in-situ TEM data at grain boundaries, showing strong agreement. However, approaches such as this demand significant computer processing capabilities. Chowdhury et al. [265] used a similar approach in modelling the FCG resistance of other microstructural obstacles such as nanoscale twin boundaries. Atomistic modelling was used to generate energy barrier data, which was used as an input to a dislocation mechanics based FCG model; the methodology addresses key aspects of microstructure sensitive crack growth including slip irreversibility and the threshold SIF range but lacks direct validation against experimental data. Furthermore, the approach is inefficient as it relies on atomistic modelling for specific microstructure interactions. Chowdhury et al. [266], [267] also presented detailed mechanistic models for short FCG in nano-twinned materials based on twin-slip interactions, slip irreversibility and the Peierls-Nabarro model. A continuum model for FCG was developed based on the threshold SIF range; results compared favourably with a range of experimental data, but the model could not be applied to explicitly predict rates of growth in real materials.

To capture microscale material behaviour, CPFE modelling is a potential candidate, as demonstrated in Chapter 4. This approach was used in combination with the XFEM by Zhang et al. [177] to simulate short FCG in a nickel-based superalloy. Crack path phenomena such as tortuosity were predicted using a maximum slip criterion, albeit using representative modelling, rather than direct simulation of experiments. Growth rate trends were reasonably captured, though not precisely, using critical cumulative shear strain. An arbitrary scaling factor was required to ‘fit’ simulated rates to experiment, demonstrating a lack of mechanistic understanding. XFEM enables simulation of crack paths in any direction without a priori specification, while minimising computational cost, since remeshing is not required. However,

practically, linking the CPFE-XFEM approach with the component scale remains inaccessible due to the associated increase in processing requirements [268]. Recent modelling work by Shen et al. [269] aimed to bridge microstructural and component length-scales for crack nucleation in α/β Ti-64 microstructures; fatigue life predictions were found to be consistent with experimental data. However, an obvious drawback of the approach is that it requires extensive fitting and calibration for each material. Cohesive zones modelling linked with CPFE has been widely used [270], [271], [272] to predict microscale cracking but comes with the drawback of crack path confinement by mesh geometry.

Modern numerical methods used to predict short FCG are enormously computationally expensive. A key factor responsible for the computational cost of XFEM is that the governing equations which solve for crack tip displacements are in differential form [273]. Bond-based peridynamics modelling of FCG offers a more rapid alternative to this, since it is formulated using spatial integrals. The method was first introduced by Silling [274], and has been successfully implemented to simulate FCG in numerous materials [275], [276], [277]. However, the approach accounts only for linear elasticity, and requires coupling with e.g., CPFE models, to capture the microscale plasticity associated with short FCG. Crystal plasticity code within phase field modelling (PFM) has also been used to capture micromechanical material behaviour [278], [279]. Ai et al. [279] presented a multi-physics PFM approach to predict crack nucleation and short FCG in battery electrode particles. Experimental observations including nucleation sites were captured, but not growth rate, and the scale of the model was not sufficiently large to predict realistic material performance. High fidelity PFM approaches, which are underpinned by mechanistic understanding, are inherently computationally expensive, as they require complex numerical integration schemes [280]. In summary, high fidelity analytical models, such as the one presented in this chapter offer minimised computational expense compared with state-of-the-art numerical methods, all while remaining deterministic. Evidently, numerical methods have the potential to offer reliable solutions to the short FCG problem but are stifled by a lack of physical understanding and computer processing limitations. Recently, several articles have been published by Zhou et al. [281], [282], [283] proposing a generalised multi-scale methodology for prediction of fatigue lives and performance of a range of steels with and without hierarchical microstructures. Using the crack tip sliding displacement range (ΔCTSD) as the driving force for crack growth, analytical models were used in combination with finite element modelling, incorporating important aspects of

microstructure (e.g., grain size, morphology, local elastic properties) with high computational efficiency. The approach could capture microstructure-linked differences in fatigue life across a range of applied stresses. Similar work by Ito et al. [284] presented a model which could capture the influence of grain boundaries on the FCG driving force. However, the models have not been applied to explicitly predict aspects of microstructurally short FCG (i.e., path or rate), which is a key aim of the current work. In this chapter, a novel mechanistic analytical model is presented, adopting a SED fracture criterion, and a new crystallographic crack path prediction method, based on a local SIF term (for full-field implementation). Experimental FCG rates and paths in Zircaloy-4 samples are used as a case study for comparison. To capture the ‘starting’ FCG behaviour, a dislocation shielding model is also incorporated. Compared with classical elastic FCG driving forces, e.g., Griffith energy [28], SED offers much greater microstructure-sensitivity [74]. Furthermore, SED is non-singular at the crack tip [175], compared with stress and strain, which are singular [285]. SED is a measure of energy stored in inter-connected dislocation structure, i.e., lattice curvature, ahead of a crack tip for example. Based on findings from Chapter 4, the cyclic accumulation of this elastically recoverable energy is argued in this chapter to be the driving force for short FCG. Hence, when the energy ahead of a crack tip exceeds a critical value (sufficient to open a crack), crack growth will occur.

5.2 Model development and implementation

5.2.1 Development of an analytical model for cyclic stored energy density at a crack tip

The FCG rate model is developed here based on the SED fracture criterion, for which a critical value, G_c , is required to allow crack extension. The crack tip SED must therefore be calculated before determining growth rate. Xu et al. [175] derived a one dimensional (single slip line contribution) analytical solution for crack tip SED based on the accumulation of GNDs due to strong local lattice curvature. The expression was derived by integrating over shear plastic strain increments along a given blocked slip line. The total shear on the slip line is decomposed into elastic and plastic parts, $\gamma = \gamma^e + \gamma^p$ and assuming there is no slip step (between the crack tip dislocation free zone (DFZ) and end of the plastic zone), this gives $\gamma^p = -\gamma^e$. In the polar coordinate system (r, θ) , the crack tip elastic strain field is $\gamma^e = \frac{\tau}{\mu} = \frac{K_\theta}{\mu(2\pi)^{\frac{1}{2}}} r^{-\frac{1}{2}}$, where $K_\theta = \frac{1}{4} \left(\sin\left(\frac{\theta}{2}\right) + \sin\left(\frac{3\theta}{2}\right) \right) \cdot K^\infty$ is an orientation-dependent SIF. The plastic shear is then $\gamma^p =$

$-\frac{K_\theta}{\mu(2\pi)^{\frac{1}{2}}}r^{-\frac{1}{2}}$. An analytical solution for GND density can then be obtained from Ashby's plastic strain gradient equation [169],

$$\rho_{\text{GND}} = \frac{1}{b} \frac{\partial \gamma^p}{\partial r} = \frac{K_\theta}{2b\mu(2\pi)^{\frac{1}{2}}} r^{-\frac{3}{2}} \quad (5.1)$$

where b is the slip direction Burgers vector. The local single slip SED is then given by integrating over the slip line as follows, subject to the constraint $\tau = \tau_c$ upon slip activation,

$$G = \int_0^{l_s} \frac{\tau d\gamma^p}{\sqrt{\rho_{\text{GND}}}} = \int_0^{l_s} \frac{\tau_c \frac{K_\theta}{2\mu(2\pi)^{\frac{1}{2}}} r^{-\frac{3}{2}} dr}{\sqrt{\frac{K_\theta}{2b\mu(2\pi)^{\frac{1}{2}}} r^{-\frac{3}{2}}}} = \int_0^{l_s} \tau_c \left[\frac{K_\theta b}{2\mu(2\pi)^{\frac{1}{2}}} \right]^{\frac{1}{2}} r^{-\frac{3}{4}} dr \quad (5.2)$$

which yields the following expression when integrated.

$$G = \left[\frac{8\tau_c^2 b K_\theta l_s^{\frac{1}{2}}}{(2\pi)^{\frac{1}{2}}} \right]^{\frac{1}{2}} \quad (5.3)$$

For plane strain conditions, the crack tip plastic zone size from continuum level fracture mechanics is $r_p = \frac{(1-2\nu)}{2\pi} \left(\frac{K^\infty}{\sigma_y} \right)^2$, where σ_y is yield stress. The microstructural equivalent relationship for slip line length is $l_s = \frac{(1-2\nu)}{2\pi} \left(\frac{K_\theta}{\tau_c} \right)^2$, which when combined with Equation 5.3, gives the following single slip plane strain SED formulation,

$$G = \left[\frac{4\tau_c b (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} K_\theta \quad (5.4)$$

which unlike stress and strain, is a non-singular microstructure-sensitive solution for crack tip behaviour and is conveniently linked with classical fracture mechanics through K_θ . [67][175] K_θ is a shear component of the remote mode I crack tip SIF (which is a function of remote loading and projected crack length, a only), acting on their respective slip systems. However, the key shortcomings of this formulation are (i) that it accounts for single slip only at the crack tip (assuming that a single slip system contributes to crack tip SED), and (ii) its failure to capture the influence of mode II shear on K_θ . To address (i), multiple slip system contributions are

accounted for by summation. In addition, the fraction of total energy stored in dislocation structure [68], ξ , is incorporated as follows,

$$G = \xi \sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot K_{\theta}^i \quad (5.5)$$

where i is slip system number, τ_c is critical resolved shear stress (CRSS) of the slip system, b is Burgers vector magnitude, ν and μ are Poisson's ratio and shear modulus, respectively, and ΔK_{θ} is an orientation-specific shear SIF, linked with remote SIF by $\Delta K_{\theta} = \frac{1}{4} \left(\sin\left(\frac{\theta}{2}\right) + \sin\left(\frac{3\theta}{2}\right) \right) \cdot \Delta K^{\infty}$ (see Figure 56). M is the total number of active slip systems. The localisation of slip, and hence dislocations at the crack tip is represented schematically in Figure 56 (a) and (b). Based the 3D orientation of individual slip planes with respect to the remote loading direction (see Figure 56 (c)), a mode I local SIF, K_I may be estimated by calculating the component of remote SIF acting along slip plane normal via a simple Mohr's circle analysis. Similarly, the mode II component, K_{II} is assumed equivalent to the in-plane shear component of remote SIF. This is described in Figure 56 (c).

By energy conservation and classical fracture mechanics, it follows that Equation 5.5 is subject to the constraint $\sqrt{\sum_{i=1}^M K_{\theta}^i{}^2} \leq K^{\infty}$, since $K^{\infty 2} \propto G$ [258]. This root-mean-square formulation links the local crystallographic SIF terms with the remote applied SIF. Unlike CPFE modelling, the current method does not capture explicit levels of slip, and hence, the local stress state is not updated directly as dislocation slip activity occurs. Therefore, the constraint is an approximation which is essential for ensuring that the SED contributions of M slip systems are not over-predicted, and are relevant to the applied SIF, i.e., increasing the number of slip systems does not necessarily increase slip activity beyond a certain point.

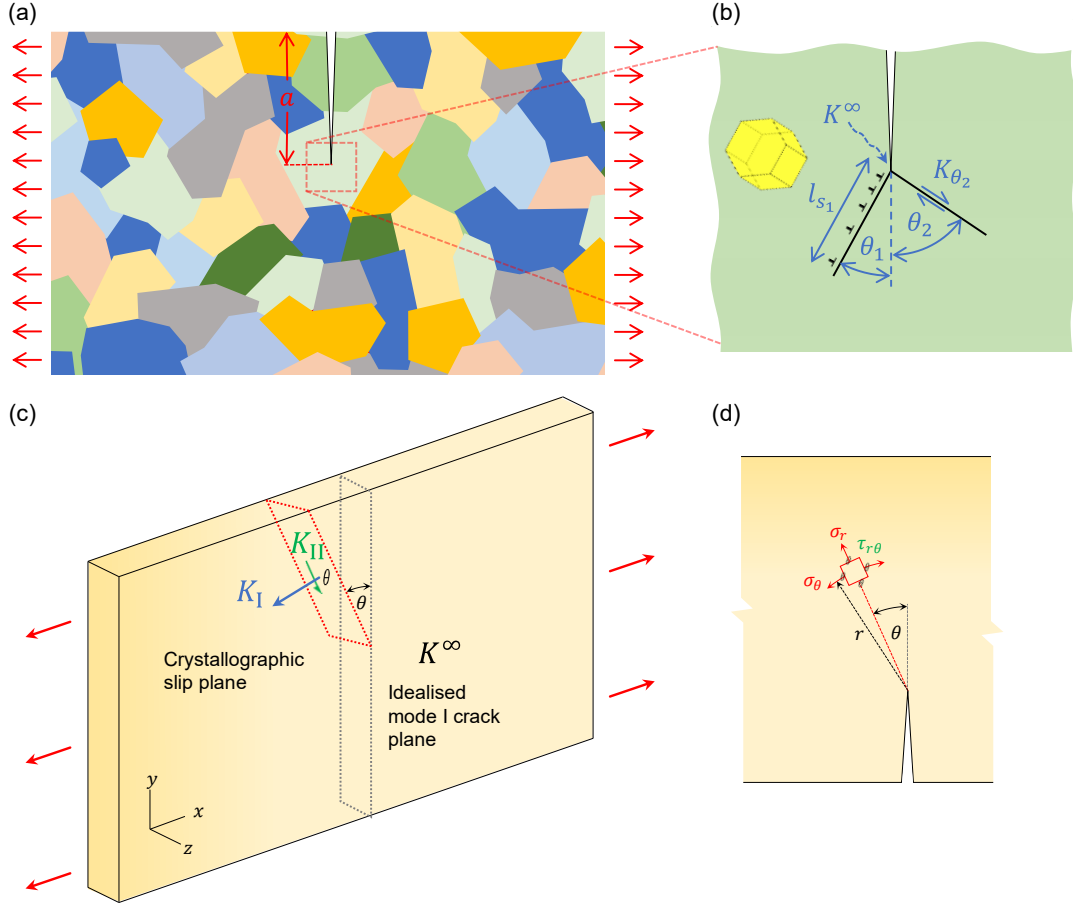


Figure 56: Overview of analytical modelling formulation within a polycrystal. (a) Illustration of a microstructurally short crack within a polycrystal. (b) Corresponding representation of crack tip within a single grain with sample slip line directions defined by ϑ_1 and ϑ_2 . (c) Illustration of mode I and II SIFs acting on a crystallographic slip plane with x , y , and z components. (d) Polar stress around the crack tip, adapted from Gdoutos [286].

In cases where the constraint is not met, Equation 5.5 is rewritten as follows.

$$G = \xi \left(\frac{K^\infty}{\sqrt{\sum_{i=1}^M K_\theta^i{}^2}} \right) \sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot K_\theta^i \quad (5.6)$$

Since the CRSS of the 2nd order $\langle c+a \rangle$ slip system is significantly larger than the other Zircaloy-4 slip systems, it is assumed that all crack tip deformation is accommodated by basal, prismatic, and 1st order pyramidal slip systems, i.e., $M = 24$. This constraint is also necessary to ensure SED is not over-estimated. To address (ii), the shear SIF term is modified to account for mode II contributions. Previously, it was assumed that only the remote mode I load contributed to shear along the slip line; hence, for a slip plane whose x - y trace lay parallel with the y -axis, there would be no shear SIF, and hence no SED. Here, the mode II contribution is incorporated by adding its effect to the polar shear stress, $\tau_{r\theta}$ (see Figure 56 (c) and (d)).

Assuming no slip step due to the presence of a DFZ ahead of the crack tip, the plastic shear strain along a slip line is given by $\gamma^p = -\gamma^e = -\frac{\tau_{r\theta}}{\mu}$. For mode I SIF on the slip plane, the polar shear stress contribution is given by $\tau_{r\theta,I} = \frac{K_I}{\sqrt{2\pi r}} \left(\frac{1}{4} \sin\left(\frac{\theta}{2}\right) + \frac{1}{4} \sin\left(\frac{3\theta}{2}\right) \right)$ [286]. For mode II, the contribution is $\tau_{r\theta,II} = \frac{K_{II}}{\sqrt{2\pi r}} \left(\frac{1}{4} \cos\left(\frac{\theta}{2}\right) + \frac{3}{4} \cos\left(\frac{3\theta}{2}\right) \right)$ [286]. Writing the shear stress, $\tau_{r\theta}$ in terms of K_θ gives $\tau_{r\theta} = \frac{K_\theta}{\sqrt{2\pi r}} = \tau_{r\theta,I} + \tau_{r\theta,II}$. Hence, the modified shear SIF, accounting for modes I and II on the i^{th} slip plane is given by Equation 5.7.

$$K_\theta^i = \frac{1}{4} \left[\left(\sin\left(\frac{\theta^i}{2}\right) + \sin\left(\frac{3\theta^i}{2}\right) \right) \cdot K_I^i + \left(\cos\left(\frac{\theta^i}{2}\right) + 3 \cos\left(\frac{3\theta^i}{2}\right) \right) \cdot K_{II}^i \right] \quad (5.7)$$

The purpose of the current model is not to capture the precise crack tip stress-state or slip levels, but rather to capture, more generally, the influence of crystallography (discrete slip system orientation, elastic anisotropy, etc.) on the short FCG driving force (argued here to be SED), and hence growth rate. The evolution of SED with fatigue cycles or cyclic SED is approximated in this work for loading ratios of $R \geq 0$ to avoid complications associated with fully reversed loading, including crack tip back-stresses, crack closure, etc.); hence, the static value for SIF in the static SED expression (Equation 5.6) is replaced by SIF range. Cyclic SED

is then written as $\frac{dG}{dN} = \xi \sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot \Delta K_\theta^i$.

5.2.2 Analytical solution for crack growth rate

To illustrate the behaviour of SED at a crack tip, a simple single crystal CPFE-XFEM simulation is performed, similar to that presented in Chapter 4 for single crystal orientations A and B. Figure 57 (a) shows the predicted crack tip SED field for the single crystal orientation shown (orientation A from before). Figure 57 (b) shows the crack tip SED evolution during $R = 0$ fatigue loading. The cyclic SED, $\frac{dG}{dN}$ is shown to increase with cycle number. For small numbers of cycles, SED increases approximately linearly until the critical value is reached. At this point, the crack extends by some increment, Δa (Figure 57 (a)). This gives a SED discontinuity as a new crack tip is created. However, it is clear (for the current Δa) that crack tip SED remains greater than zero after extension. Hence, there is SED which remains in the element directly ahead of the new crack tip, G_r , which is shown to vary with crack length. G_r is characterised here as a fraction of the critical value by $\chi = \frac{G_r}{G_c}$.

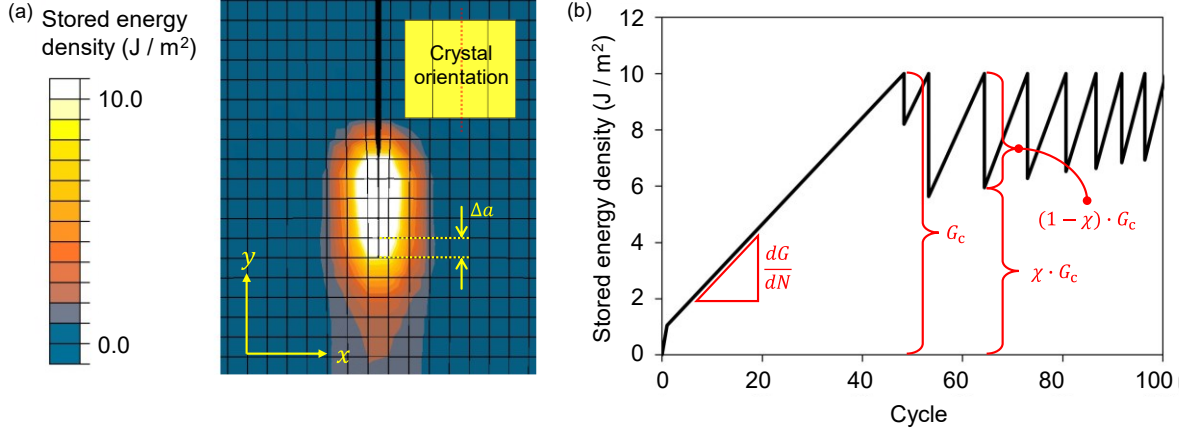


Figure 57: Mechanistic understanding of single crystal crack propagation from CPFE simulation. (a) CPFE-simulated SED field about a growing crack within a single crystal. The orientation selected for this enables crack growth perpendicular to the primary loading direction (x-direction). As shown, the crack path is straight and vertical, and propagates along prismatic slip planes perpendicular to their Burgers vector directions. The crack increment length, Δa is shown also, and is equal to the mesh size, 10 μm . The critical SED is arbitrarily selected to be 10 J/m². (b) Evolution of SED ahead of the growing crack tip with increasing cycle number, i.e., after the crack propagates, a new crack tip is defined. Concepts of cyclic SED and χ are introduced.

The FCG rate may be written in terms of crack length increment as $\frac{da}{dN} = \frac{\Delta a}{\Delta N}$, where ΔN is the number of cycles required to reach G_c . For stable FCG, the cumulative SED required to achieve this is $\Delta G = (1 - \chi) \cdot G_c$. For small numbers of cycles (small ΔN), cyclic SED is expressed as $\frac{dG}{dN} = \frac{\Delta G}{\Delta N}$. Therefore, $\Delta N = \frac{\Delta G}{dG/dN}$. FCG rate is thereby expressed as,

$$\frac{da}{dN} = \frac{\Delta a}{(1 - \chi) \cdot G_c} \cdot \frac{dG}{dN} \quad (5.8)$$

where Δa is an arbitrarily selected crack extension increment, and χ is dependent upon Δa . $\frac{dG}{dN}$ varies depending on loading conditions, crack length, and material. Incorporation of the expression for cyclic SED gives Equation 5.9.

$$\frac{da}{dN} = \frac{\Delta a}{(1 - \chi) \cdot G_c} \cdot \xi \sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1 - 2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot \Delta K_{\theta}^i \quad (5.9)$$

A drawback of the model in this form is that for some arbitrary Δa , the remaining SED fraction, χ , is unknown. Since SED accumulates under plastic deformation, it may be assumed that remaining SED equals zero at the plastic zone edge, such that $\Delta a = l_s$ (slip line length), i.e., where $\Delta a = l_s$, $G_r = \chi = 0$. Hence, the evolution of χ with increasing crack length can effectively be captured using slip line length evolution. An expression for slip line length can be derived from fracture mechanics methods for estimating plastic zone radius [287] as $l_s =$

$\frac{(1-2\nu)}{2\pi} \left(\frac{K_\theta}{\tau_c} \right)^2$ [175]. Hence, crack length increment, Δa , is given mechanistic basis, and χ can be reduced to zero at the end of the slip line. The value of l_s will depend on crack direction, which is confined to the set of M slip systems, where the selected slip system for crack path is $i = j$, i.e., j represents the slip system index number which controls crack propagation direction. Since the crack length considered for remote SIF calculation is projected length, the crack length increment is now written in terms of crack direction and slip line length as $\Delta a = l_s \cos(\theta)$ (see Figure 56 for θ meaning). FCG rate is now therefore dependent on crack propagation direction and is given by Equation 5.10.

$$\frac{da}{dN} = \frac{l_s^j \cos \theta^j}{G_c} \cdot \xi \sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot \Delta K_\theta^i \quad (5.10)$$

5.2.3 Incorporation of dislocation shielding

For predicting microstructure sensitive FCG rate, it is useful to link microstructural parameters with classical fracture mechanics approaches, i.e., via SIF. However, crack tip SIF alone describes only the elastic behaviour of the material, and therefore does not account for the effects of cyclic plasticity on the local stress field, i.e., the development of compressive residual stresses around the crack tip. Due to the vast number of cycles often required to propagate short or stage I fatigue cracks, this is an important consideration. Figure 58 illustrates the effects of cyclic plasticity on the (unloaded) local stress state.

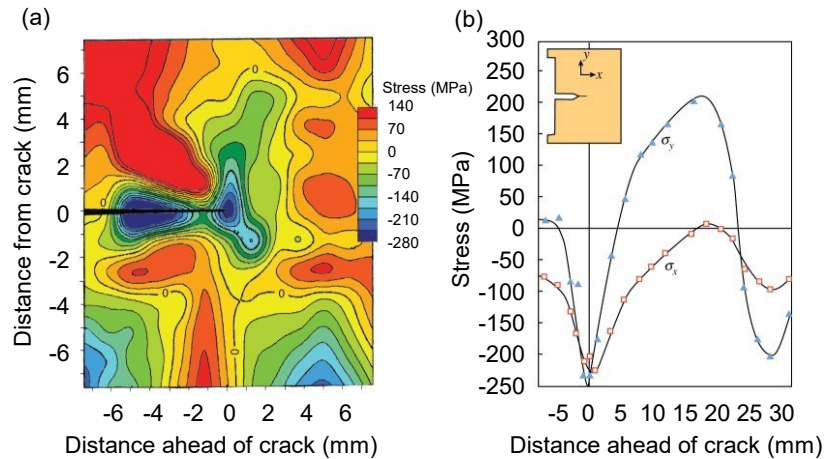


Figure 58: (a) Crack tip residual stress map from X-ray diffraction measurements in 1020 steel after fatigue cycling and crack growth at 16.9 MPa√m. The compressive stress wake leftward of the crack tip is illustrative of the diminished shielding effect as the crack advances [288]¹⁴. (b) Crack-tip residual stress field determined at mid-thickness of a 19 mm thick ferritic steel test piece from neutron diffraction measurements [289]¹⁵.

¹⁴ Reprinted from [288] under the Creative Commons Attribution 4.0 International Public License.

¹⁵ Reprinted from [289] with permission from Taylor & Francis.

At the microstructural level, this effect is referred to as dislocation shielding [290], [291], as individual dislocations contribute towards the diminution of maximum stress, locally. For the purposes of the current chapter, it is assumed that crack tip shielding is dominated by changes in the local stress state due to cyclic plasticity, and other known effects including crack closure, crack tip blunting/sharpening, and oxide bridging are negligible. Experimental observations of dislocation emission from crack tips using TEM show that shielding dislocations, which make up the plastic zone, first cross the DFZ [291], [292]; this is consistent with the theory of the current analytical model. While it may seem that dislocations have no shielding effect at the crack tip due to the DFZ, it is argued here that compared with the length scale over which SED is relevant to FCG (i.e., slip line length, micron-scale), the size and hence the effect of the DFZ (sub-micron scale) is trivial. Figure 59 (a) and (b) illustrate this point. Based on the early work of Rice and Thomson [227], Wang [240] presented a plane strain expression to quantify the impact of a single dislocation on local SIF, which is given by $K_{\theta}^{\perp} = -\frac{3\mu b}{2(1-\nu)} \left(2\pi r_{\text{avg}}^{-\frac{1}{2}} \right)$, where r_{avg} is an averaged distance from the crack tip for n emitted dislocations, approximated here as the slip line length, l_s . It is assumed that shielding dislocations only affect the SED contribution of slip system j due to the discrete nature of this model. The number of shielding dislocations, n is taken as an average of the number of dislocations emitted per cycle, such that the total dislocation shielding contribution for a given crack length is $\Delta K_{\theta}^{\perp, \text{total}} = \Delta N \cdot n \cdot K_{\theta}^{\perp}$, where $\Delta N = \Delta a \cdot \left(\frac{da}{dN} \right)^{-1}$. Accounting for dislocation shielding, Equation 5.11 is now rewritten to give a diminished cyclic SED and hence growth rate as follows.

$$\frac{da}{dN} = \frac{l_s^j \cos(\theta^j)}{G_c} \cdot \xi \left(\sum_{i=1}^M \left[\frac{4\tau_c^i b^i (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot \Delta K_{\theta}^i + \left[\frac{4\tau_c^j b^j (1-2\nu)}{\pi\mu} \right]^{\frac{1}{2}} \cdot \Delta K_{\theta}^{\perp, \text{total}} \right) \quad (5.11)$$

Since values for n correspond to the mean number of dislocations emitted from the crack tip per cycle, i.e., not accounting for pile-up effects, n is selected to maximise the cyclic dislocation shielding effect (for the initial crack length) in each case. Since the total shielding SIF is inversely proportional to FCG rate $\left(\Delta K_{\theta}^{\perp, \text{total}} \propto \left(\frac{da}{dN} \right)^{-1} \right)$, rearrangement of Equation 5.11 gives a quadratic relationship between the mean number of emitted dislocations, n and $\Delta K_{\theta}^{\perp, \text{total}}$. There are two roots of such a relationship which give (i) a positive and (ii) a negative gradient,

$\frac{\partial n}{\partial \Delta K_{\theta}^{\perp, \text{total}}}$. Naturally, a negative gradient is illogical since this suggests there is an increased shielding effect with decreasing n . Therefore, to maximise the shielding effect in a physically sensible manner, the inflection point is selected, for n , where $\frac{\partial n}{\partial \Delta K_{\theta}^{\perp, \text{total}}} = 0$, as illustrated in Figure 59 (c). Mean dislocation spacing is calculated as $l = \frac{l_s^j}{\Delta N_0 \cdot n}$, and is used to estimate corresponding crack tip dislocation density assuming uniform dislocation spacing in 2D. ΔN_0 is the initial number of cycles before the crack propagates. Dislocation density is given by $\rho_{\text{dis}} = \sqrt{1/l}$ for comparison with experimental crack tip measurements. Hence, the final model for FCG rate (Equation 5.11) is highly microstructure sensitive as it accounts for key microstructural material properties including crystal orientation, corresponding anisotropic elastic constants, slip properties, discrete slip directions, crack direction, and dislocation shielding effects.

As noted in the previous section, unlike CPFE modelling, the analytical model does not account for grain-grain or crack-GB interactions but is instead aimed at capturing FCG rate trends in polycrystals, based on local crystallography and crack length alone. In addition, local crack path oscillations within individual grains are not captured using the current model, since the local stress state is unknown. Hence the current model is limited to materials where grain-grain interactions are small compared to crack tip plasticity, and to crack paths which are reasonably straight within individual grains.

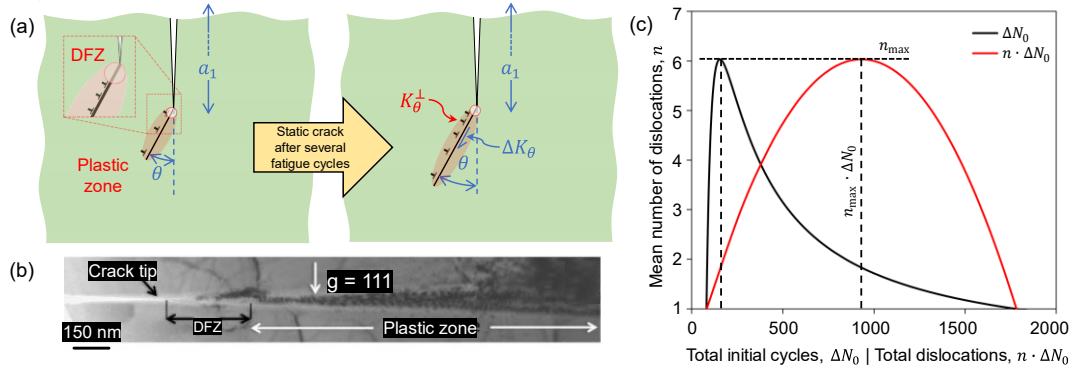


Figure 59: Depiction of dislocation shielding model with mean emitted crack tip dislocations strategy. (a) Schematic representation of cyclic dislocation shielding mechanism. After several fatigue cycles, dislocations accumulate ahead of the static crack tip, affecting the local shear SIF. The comparatively small DFZ is also shown. (b) TEM image showing crack tip dislocations and DFZ in a nickel-based super alloy [291], [293]¹⁶. This indicates that for a physically and microstructurally short tensile crack, the DFZ is of the order of 0.5 μm . (c) Sample dislocation number calculation showing relationship with initial number of cycles and total number of dislocations before crack extension. Maximum number of dislocations is selected such that dislocation shielding is maximised for the initial crack length, as shown.

¹⁶ Reprinted from [291] under the Creative Commons Attribution 4.0 International Public License.

In this chapter, it is assumed that the dominant factors affecting FCG rate are related to crystallography, and hence, that local crack path oscillations and interactions with microstructural features have little or no overall effect on the FCG rate trend. The critical SED for Zircaloy-4, which was estimated in Chapter 4 is used here.

5.2.4 A simplified method for crack path selection and full-field implementation

To utilise the current model for full-field (2D free surface) crack path and FCG rate prediction within polycrystalline materials, a computer program is developed, whereby grain structure is defined using an image-based method. It is assumed that 2D grain structure information is sufficient for prediction of short FCG, and hence that substructural morphological variation is negligible, since the experimental crack growth rates and paths considered here were measured at the sample free surfaces. This assumption is further reinforced by application to blocky alpha microstructures, which minimise substructural variations. Despite this assumption, full 3D grain orientations obtained from electron backscatter diffraction (EBSD) are used to account for crystallographic slip plane directions with respect to the loading conditions. In the experiments, notched beam samples measuring $3 \times 3 \times 12$ mm were loaded in cyclic 3-point bending. Hence, in the bulk of each sample, rates of crack propagation are likely to vary in 3D (since even blocky alpha grains measuring $100 - 300 \mu\text{m}$ are much smaller than total sample thickness). However, due to the strongly textured large-grain microstructures considered, it is reasonable to assume that substructural morphological variations are low (with respect to crack length), and hence, based on *St. Venant's principle*, that free-surface crack growth rates may be adequately described using 2D grain morphologies. The same assumptions apply to crack path prediction. Using CPFE modelling, criteria for short crack path are often controlled by maximum slip or some energy related term [211]. However, since slip levels are not quantified using the current method, three crystallographic SIF-based criteria are proposed: (i) maximum shear SIF, combining modes I and II, $\max(K_{\theta}^I)$, (ii) maximum mode I SIF, $\max(K_I^I)$, and (iii) maximum mode II SIF, $\max(K_{II}^I)$ (see section 5.2.1 for details). The implementation of the maximum SIF method(s) is described in Figure 60 (a). These new methods are compared in this chapter to find a solution which gives greatest agreement with experimental crack paths. Hence, prediction of crack paths within polycrystalline microstructures will enable full-field implementation of the crack growth rate model for direct comparison with experiments. As shown in Figure 60 (b), the method used here involves point-wise calculation of FCG rate within individual grains, tracking the spatial position of the crack

tip as it extends. A single crack propagation direction is defined for each crystal orientation (or grain). Hence the crack growth rate will be governed by projected crack length, and crystallographic quantities (orientation, discrete slip etc.) corresponding to the crack tip position until such a point that the crack is detected to have traversed a grain boundary (Figure 60 (c)).

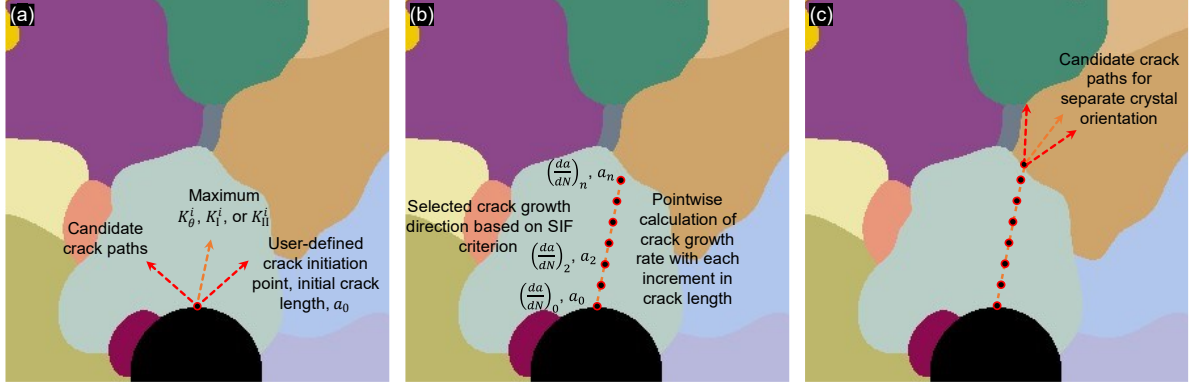


Figure 60: Method for full-field polycrystalline implementation of analytical solution for FCG rate. (a) Selection of crack path at the initiation stage based on (selected) crystallographic SIF-based criterion for short crack path. Since the crack path is simplified to be controlled by remote loading and crystallography alone, a single crack direction is assigned to each grain. (b) Progression of crack along pre-defined path; at equally spaced intervals, the growth rate is computed. (c) Upon traversing a grain boundary, a new crack direction is assigned and the process repeats.

Accordingly, grain-grain interactions and crack-grain-grain interactions are unaccounted for using the current method (which, of course are accounted for, using CPFE modelling, for example). While such interactions are likely to have some effect, they are considered negligible in large-grain, strongly textured microstructures, such as those presented in this chapter. Simulations using the analytical solution are carried out within commercial software, *Matlab*. The program computes relevant in-plane elastic properties (μ and ν) before the solver begins. This requires rotation of the 6×6 compliance matrix, $\mathbf{S}$ according to the Euler angles of individual grains, using $\mathbf{S}' = \mathbf{M}\mathbf{S}\mathbf{M}^T$, where $\mathbf{S}'$ is the rotated compliance matrix, and $\mathbf{M}$ is the 6×6 rotation matrix. For rotations about the a- and c-axis directions, $\mathbf{M}$ is given by Auld [294]. The solution for $\frac{da}{dN}$ in Equation 5.11 requires an explicit or iterative solving approach due to the self-dependence of input parameter, $\Delta K_{\theta}^{\perp, \text{total}}$ on crack growth rate. Newton-Raphson iterative solving is used for this purpose, as described in Figure 61.

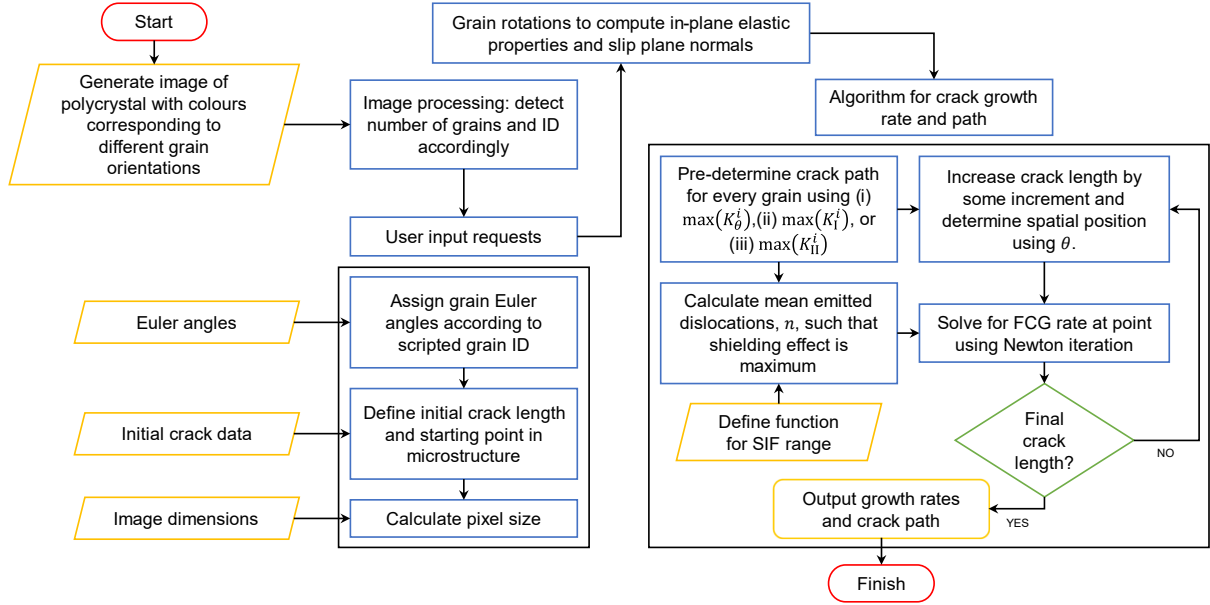


Figure 61: Flowchart describing algorithm and full-field implementation of analytical FCG rate solution with crack path prediction.

Lastly, for comparison with experiments, FCG is modelled in notched blocky alpha polycrystalline Zircaloy-4 samples which were loaded cyclically in 3-point bending (loading ratio, $R = 0$). Hence, the remote SIF range is calculated using the methodology described in Chapter 4 for 3-point bending.

5.3 Results and discussion

5.3.1 Case study: crack path prediction and comparison with experiments

In this section, three crack path prediction methods based on local slip plane SIF are compared. Specifically, results from maximum SIF crystallographic deformation mode (ΔK_{θ}^I , ΔK_I^I , and ΔK_{II}^I) criteria are compared with experiments and corresponding full-field CPFE-simulated crack paths (which use a maximum slip criterion) for selection of an optimal method which is subsequently used to compare FCG rates in polycrystalline Zircaloy-4. In Figure 62, a comparison is made between the three candidate methods and experiments and CPFE simulation results. Samples were loaded in three-point bending, under cyclic $R = 0$ loading conditions. Note that the CPFE contour fields presented in Figure 62 represent the distribution of SED about the crack. ΔK_I^I and ΔK_{II}^I represent local slip plane mode I and mode II SIF ranges, as illustrated in Figure 56.

The CPFE maximum-slip-based crack path results from Chapter 4 (samples AA and BB) and Wilson et al. [74] (samples CC and DD) in Figure 62 (b), (g), (l), (q), demonstrate overall strong agreement with experiments. One exception to this rule, however, is for sample BB,

shown in Figure 62 (g), where the maximum slip criterion predicts a very different crack path from the experiment (Figure 62 (f)). While the maximum slip criterion (or indeed other mechanistic criteria, as presented in Chapter 4) cannot be replicated within the new method (since the full-field stress state is unknown), the local SIF criterion which is most closely related to the maximum slip criterion is maximum ΔK_{θ}^i . This term accounts for both mode I and II SIF on the i^{th} slip plane and is a measure of SIF along a slip line, constrained to lie on the free surface plane. For samples BB and DD, there is reasonable agreement between this method (Figure 62 (h), (r)) and the maximum slip criterion Figure 62 (g), (q)), but much less so for samples AA and CC. This is because the Burgers vector directions of the dominant slip planes in samples BB and DD lie approximately within the free surface plane. Clearly, this is not the case for samples AA and CC, since the CPFE-predicted crack paths are almost perpendicular to the remote loading direction. The maximum ΔK_{θ}^i criterion also tends to favour planes whose traces give $\theta \sim 45^\circ$; this inadvertently results in grain boundary (GB) crack path oscillations in sample CC (Figure 62 (m)). The maximum mode I SIF (ΔK_{I}^i) criterion for crack path demonstrates strong agreement with experiments. Indeed, this method gives more precise predictions than CPFE for samples BB and CC, but of course fails to capture clear crack path tortuosity within a single grain in sample DD. Lastly, the maximum mode II SIF (ΔK_{II}^i) predictions are shown in Figure 62 (e), (j), (o), (t), and compare poorly with experimental paths. In some regards, ΔK_{II}^i performs better than ΔK_{θ}^i (e.g., grain 4 of sample AA in Figure 62 (e)) but is clearly not the best available driver for crack path.

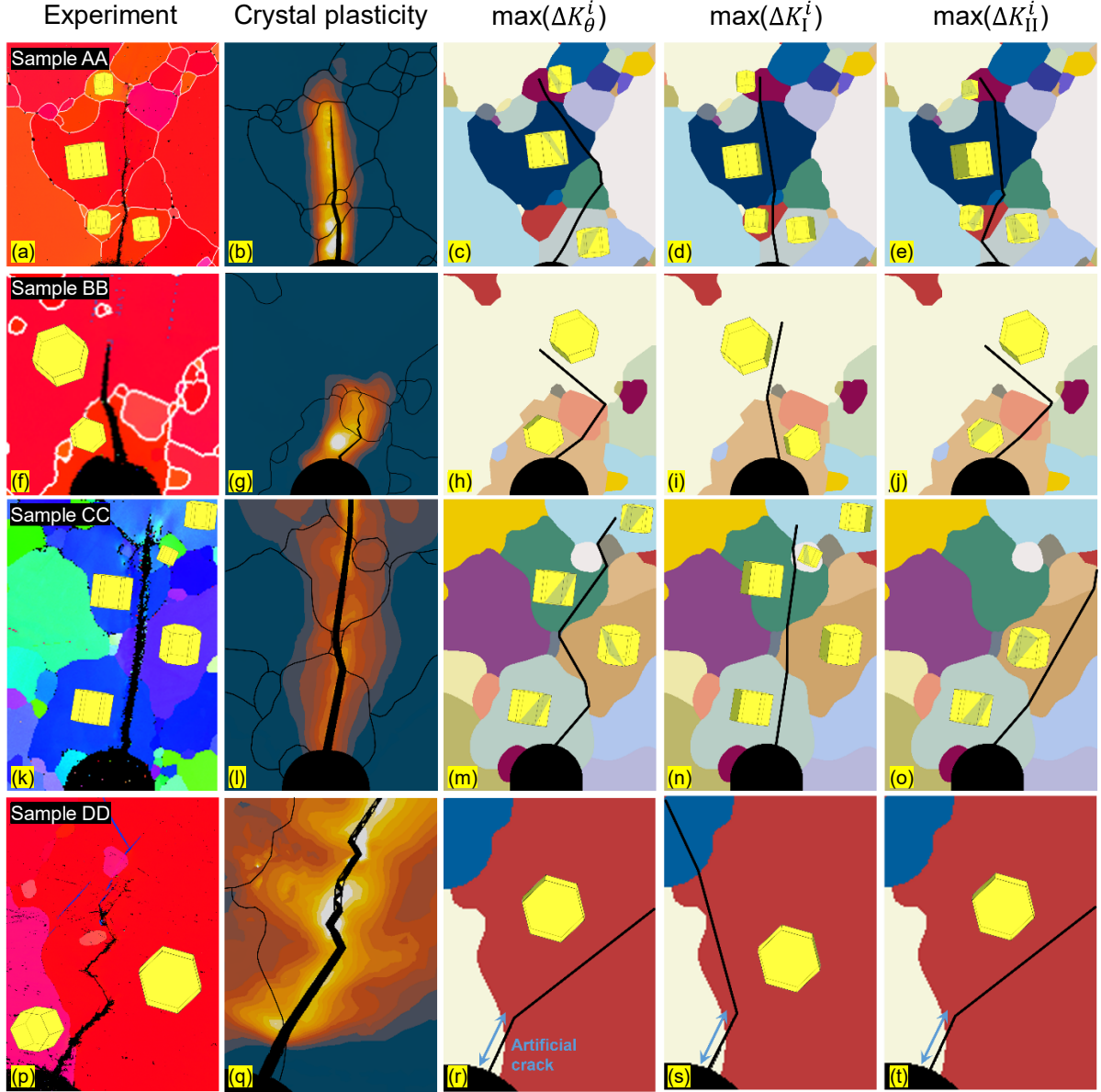


Figure 62: Comparison of experimental crack path and CPFE simulation results with local SIF crack path prediction methods for (a) – (e) Sample AA. (f) – (j), Sample BB. (k) – (o), Sample CC. (p) – (t), Sample DD. Maximum SIF criteria from left to right are maximum in-plane shear SIF, maximum mode I SIF, and maximum mode II SIF. CPFE simulations were carried out in previous work for Samples AA and BB (Chapter 4) and CC and DD [74]. Local SIF criteria crack paths are presented alongside corresponding slip plane for each relevant grain.

Hence, the maximum ΔK_I^I criterion will be used for subsequent crack path and corresponding FCG rate predictions in this chapter. Unsurprisingly, the maximum ΔK_I^I criterion is similar to the new maximum normal SED criterion presented in Chapter 4.

5.3.2 Case study: simulated crack growth rates and comparison with experiments

The analytical model for FCG rate, given by Equation 5.11, is applied to samples AA-DD for comparison with experiments, using the maximum ΔK_I^I criterion for crack path. Figure 63 (a)

shows that due to the strongly differing textures in samples AA and BB, there are vast differences in FCG rate. Reasons for this are discussed in the next section. The model effectively captures these differences. For sample AA, there are also substantial growth rate fluctuations as the crack interacts with and crosses various microstructural features GB2 and GB3. These fluctuations are reflected by the analytical model, but rather subtly (compared with experiment), since the effect of the GB is not explicitly captured, as discussed previously. While it is clear that grain boundaries have a local effect (in the context of the current blocky alpha microstructures), the overall crack growth rate trends are reasonably well captured, which is the aim of the current work. Incorporation of grain-grain interactions is non-trivial, as there are morphological and crystallographic effects, as well as cyclic plasticity to consider. For example, depending on the orientation of one grain, i.e., set of crystallographic slip systems, with respect to another, slip blockage or transmission may occur. The Luster and Morris slip transfer parameter was used by Sudha et al. [295] to identify common slip transfer pairs between neighbouring grains based on TEM data after static tensile testing in a titanium alloy. Some such method could be applied here to capture microstructure interaction effects but would require sufficient statistical data for a given alloy, and hence, the model would no longer be deterministic. Moreover, the growth rate transition across a GB is coarser than in the experiment since the model has no knowledge of the GB until it is crossed. In the final grain of sample AA, a substantial drop in FCG rate is predicted, which reflects the experimental data, though not entirely for reasons outlined above. While the link between FCG rate and GBs is less clear for sample BB, the growth rate trend is effectively captured using the model, despite under-predicting it somewhat.

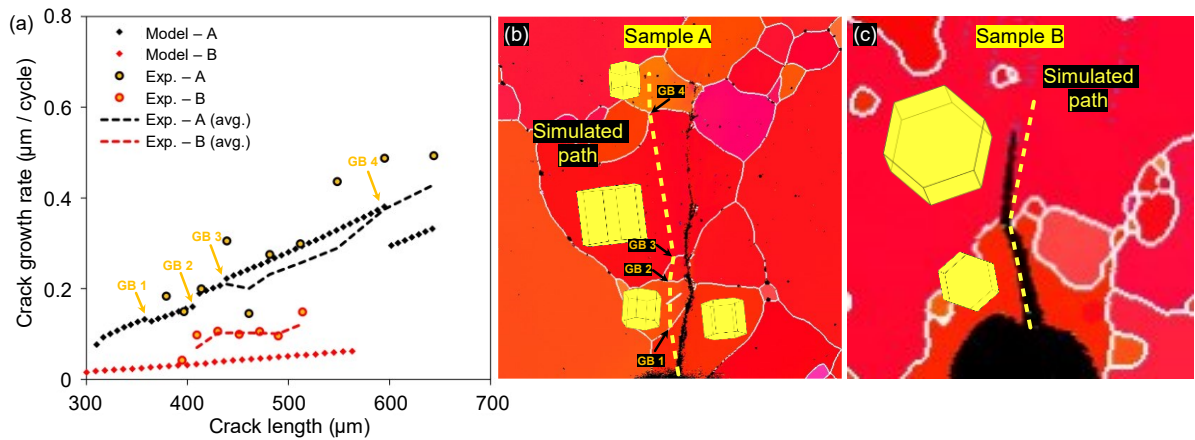


Figure 63: Assessment of microstructural sensitivity of analytical solution. (a) Comparison of analytical solution for FCG rate and experimental data for samples AA and BB. Comparison of predicted crack path using maximum mode I SIF criterion with experiment in (b) sample AA and (c) sample BB.

In terms of texture, samples CC and DD are very similar to samples AA and BB, respectively, and hence, rates of growth are significantly higher in CC than DD, as shown in Figure 64 (a). As before, the analytical model captures the general trend for each sample but does not capture substantial FCG rate fluctuations. The larger fluctuations here are likely due to greater anisotropy between neighbouring grains in sample CC, than in AA. Interestingly, after the crack crosses GB3 of sample CC, there is a substantial increase in rate due to the orientation of the fourth grain, increasing to values comparable with the experiment. FCG rates are partially under-predicted by the model for each of these samples. This could be corrected by adjusting the critical SED, G_c , but is not, in order to maintain consistency with previous CPFE modelling (Chapter 4). In any case, adjusting G_c is unlikely to reflect the true mechanistic driver of these differences, since GB localisation is anticipated to be the dominant factor.

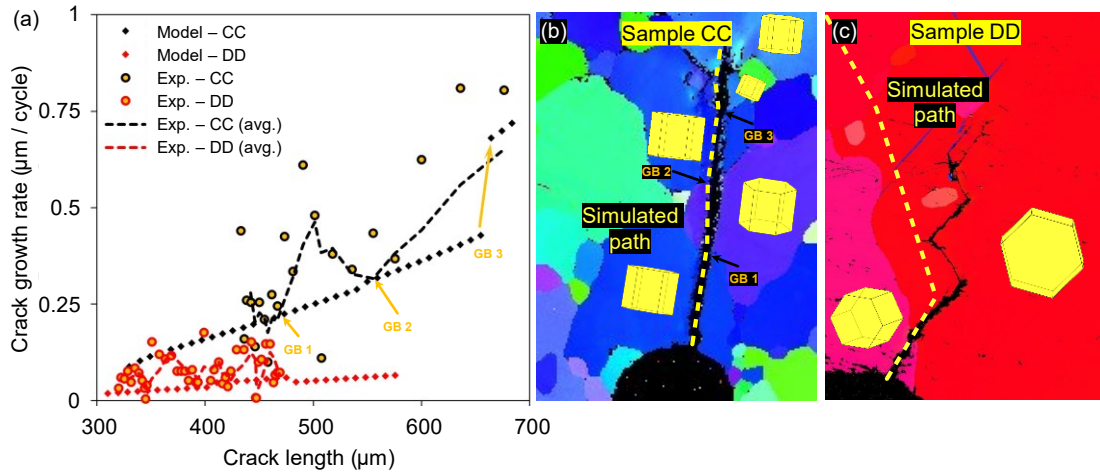


Figure 64: Assessment of microstructural sensitivity of analytical solution. (a) Comparison of analytical solution for FCG rate and experimental data for samples CC and DD. Comparison of predicted crack path using maximum mode I SIF criterion with experiment in (b) sample CC and (c) sample DD.

The overall growth rate magnitude in sample DD is captured by the model, but as with previous samples, fluctuations are not. In this case, crack path tortuosity is likely to be the driver of these fluctuations, which cannot be captured using the current criterion for crack path. An important factor to consider in these results, as with the CPFE results, is that all crack path and crack growth rate results are determined using free surface EBSD data. Hence, there are potentially 3D, i.e., substructure effects, which contribute towards differences between experiment and model. Despite this, the 2D approximation is valid at the free surface and enables reasonably accurate predictions of short crack growth rate and path.

For samples AA and BB, CPFE simulated crack paths and rates from Chapter 4 are compared with the analytical model. Figure 65 (a) shows strong agreement between both simulation

methods in terms of rate for both samples, despite differences in crack path, which are shown clearly in Figure 65 (b) and (c).

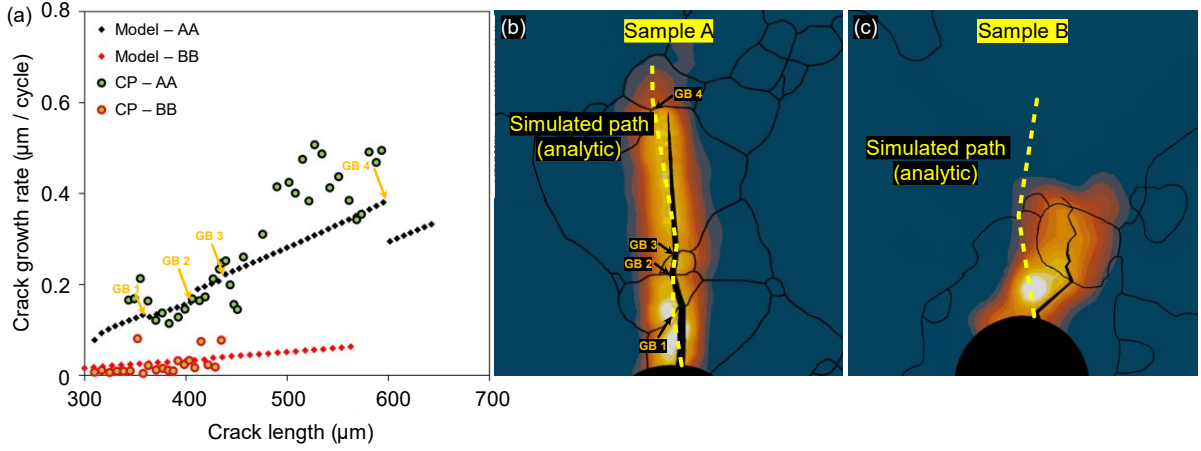


Figure 65: Comparison of analytical solution for FCG rate and CPFE simulated data for samples AA and BB. (a) Comparison of predicted crack path using maximum mode I SIF criterion with CPFE maximum slip criterion in (b) sample AA and (c) sample BB. (b) and (c) also display corresponding SED fields.

Given the simplicity of the current crack path prediction method, it is surprising that it achieves much greater agreement with experiment than the maximum-slip-based CPFE result. However, full-field CPFE predictions are more sensitive than the current method to microstructure interactions, enabling more precise growth rate predictions (where the crack path is successfully captured). Furthermore, it is important to note that to generate the CPFE data presented, it took between 5,000-10,000 CPU hours, compared with mere seconds for the analytical model. Both numerical and analytical simulations were carried out using Intel Gold 6152 processors at 2.10 GHz. 16 cores and 8 GB of RAM were assigned to each job.

5.3.3 Influence of crystal orientation and dislocation shielding with application to stage II cracks

The model is applied to Zircaloy-4 single crystals to study the influence of orientation on FCG rate. Five orientations are selected to broadly capture material behaviour. The influence of dislocation shielding is studied by comparing rates of FCG with and without shielding, i.e., where $K_{\theta}^{\perp} = 0$, in the SIF range 10-15 MPa√m. A comparison of these results is presented in Figure 66. Figure 66 (a) shows that the model predicts very strong microstructural sensitivity in terms of growth rate, with rates in orientation 3 greater than orientation 5 by more than a factor of 6. Growth rate is highest in orientation 3 due to its elastic properties contributing to highest cyclic SED and favourable orientation and length of the dominant slip line. While the

in-plane elastic properties are the same for orientation 5, growth rate is compromised due to there being no basal or prismatic contribution to SED since basal planes lie perpendicular to the remote loading direction, and prismatic planes lie parallel. This leaves highly inclined pyramidal planes to control crack direction, meaning that the projected crack extension (Δa) is relatively low. FCG rates for orientation 1 are comparably low due to elastic properties giving low cyclic SED as well as high prismatic slip plane inclination. Orientations 2 and 4 are predicted to have similar growth rate trends due to a combination of the factors discussed. Results also highlight the influence of dislocation shielding on FCG rate. Within the SIF range studied, dislocation shielding reduces growth rate by approximately 20%. More importantly, the model captures that shielding has the greatest influence initially (at lowest SIF). Values for mean number of dislocations emitted per cycle range between 6 to 42. Equivalent dislocation densities are estimated to be between 4000 – 5000 μm^{-2} . These estimates are consistent with experimental X-ray diffraction (XRD) measurements of dislocation densities at crack tips [296] (around 2500 μm^{-2}).

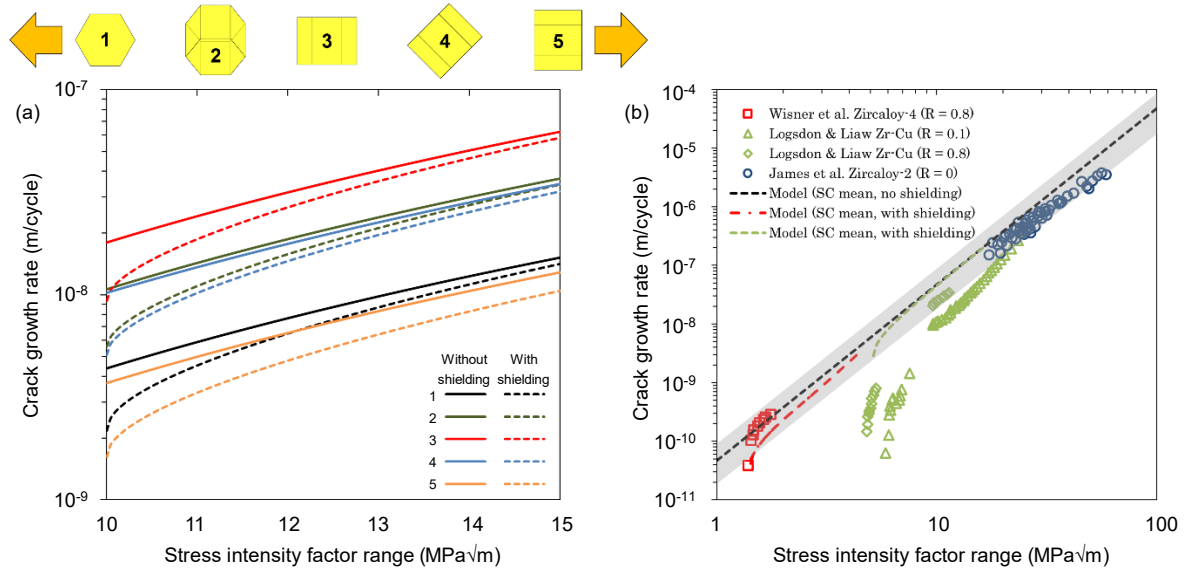


Figure 66: Single crystal simulation results. (a) Influence of crystal orientation and dislocation shielding model on SIF range-FCG rate relationship. Dashed lines correspond to the model with shielding included. Loading direction is indicated by arrows. Projected crack growth direction is vertical, relative to these crystal orientations. The vertical axis is presented in logarithmic scale. (b) Comparison of single crystal (denoted SC) analytical model (with and without shielding) with experimental FCG rate data from literature [297], [298], [299]. Single crystal simulation results are based on the mean growth rates for orientations 1 – 5. Grey shaded region covers the total range of growth rates predicted for orientations 1 – 5 without dislocation shielding. Both axes are in logarithmic scale.

To understand the influence of the various HCP slip systems in Zircaloy-4, their contributions to cyclic SED are presented in Table 11, as fractions of total cyclic SED for a given crack

length and loading conditions. The variation of contributions is compared across the five single crystal orientations considered in Figure 66.

Table 11: Contributions of slip system groups to cyclic SED for a static crack with a SIF of 10 MPa $\sqrt{\text{m}}$ under $R = 0$ loading (without dislocation shielding). Contributions are represented as fractions of the total cyclic SED for each crystal orientation, i.e., as $\hat{G}_{\text{sys}}/\hat{G}_{\text{total}}$. Five single crystal orientations are considered with numbering and loading direction corresponding to those outlined in Figure 66.

Orientation	Prismatic	Basal	Pyramidal $\langle a \rangle$	Pyramidal $\langle c+a \rangle$
1	0.2513	0	0.4754	0.2732
2	0.2608	0	0.4694	0.2698
3	0.1549	0	0.5367	0.3084
4	0.2053	0.2708	0.3327	0.1912
5	0	0	0.6350	0.3650

Table 11 shows that on average, according to the current model, the greatest contributors to cyclic SED in HCP Zircaloy-4 are the $\langle a \rangle$ pyramidal slip systems, followed by $\langle c+a \rangle$ pyramidal, prismatic, and basal. The pyramidal contributors are largest due (primarily) to there being 6 $\langle a \rangle$ and 12 $\langle c+a \rangle$ systems, while there are just 3 prismatic and basal systems, respectively. Hence, normalised results would indicate that on average, prismatic systems make the greatest individual contributions to cyclic SED, i.e., undergo maximum slip activity, which is consistent with experiments [161] and numerical modelling [175]. The relatively low combined contribution of $\langle c+a \rangle$ slip systems to cyclic SED is due to the comparably high CRSS.

The universality of the model is assessed through comparison with experimental FCG rate data over a wide SIF range. Wisner et al. [298] tested the fatigue behaviour of Zircaloy-4 at low SIF values (in the range 1-2 MPa $\sqrt{\text{m}}$). Precise crystallographic details of samples are unavailable, but Kearns factors [300] were of the order of 0.7. Logsdon and Liaw [299] tested a Zr-Cu alloy (Zr content >99%) in the SIF range 5-30 MPa $\sqrt{\text{m}}$ with loading ratios 0.1 and 0.8. James [297] carried out room temperature tests on Zircaloy-2 in the range 10-60 MPa $\sqrt{\text{m}}$ in samples where the crack direction was either orthogonal to or parallel to the sheet rolling direction. Minimal growth rate variation was observed between samples, particularly for high SIFs, indicating low microstructure-sensitivity. A comparison of the experimental FCG data mentioned above with the analytical model (with and without shielding) is presented in Figure 66 (b), using generalised mean single crystal results. The comparison shows strong agreement between the model and experiments, despite obvious differences in microstructure and

composition. Unsurprisingly, strongest agreement is attained with Zircaloy-4 data from Wisner et al. [298], excluding for lower SIF ranges. However, application of the dislocation shielding model is shown to capture some of the experimental ‘starting’ FCG behaviour, while somewhat under-predicting magnitude. Similarly, Zr-Cu FCG rates are over-predicted, but application of the shielding model also captures the transition from ‘starting’ to Paris’ law [20] behaviour. This crack growth starting behaviour is captured for a range of initial SIF values and is akin to the asymptotic thresholding behaviour observed in many materials, whereby for decreasing values of SIF range, cracks cease to propagate [301]; the threshold SIF range is denoted ΔK_{th} . Figure 66 (b) demonstrates that as the initial or starting value for SIF range is decreased, the shielding behaviour becomes more dramatic. However, this does not indicate that there is some threshold value which can be predicted using the current model; rather, it captures the phenomenon to an extent, as predicted growth rates reach as low as single atomic lattice spacings per cycle. Incorporation of additional physics-based understanding of dislocation emission from the crack tip, rather than the mean approach incorporated here, may however allow for this behaviour to be captured more precisely.

In addition, the model appears to capture loading ratio effects, despite not accounting for loading ratio explicitly. Effectively, the SIF range helps to capture the cyclic behaviour, while the slip line length, l_s , accounts for the peak SIF magnitude. In combination, these factors represent a loading ratio effect. Lastly, Zircaloy-2 FCG data is shown to lie within the growth rate range (shaded grey region), even for high values of ΔK^∞ , demonstrating that the model has the potential to capture stage II FCG behaviour.

5.4 Conclusions

A deterministic analytical model has been developed, which is a prospective candidate for prediction of short crack growth rates in a range of ductile metallic materials. It is evident that SED, which forms the basis for the model, can effectively capture the effect of key microstructural characteristics (elastic anisotropy, discrete slip, etc.) on FCG rate. The development of a new crack path prediction tool based on maximum (slip plane-based) mode I SIF, in combination with the analytical solution was shown to capture overall FCG rate trends and crack paths in a range of Zircaloy-4 polycrystals, and to even out-perform CPFE (maximum slip-based) crack path predictions in some instances. Furthermore, a distinct advantage of the analytical model over numerical, e.g., CPFE calculations, is the substantial

reduction in computer processing requirements; solving times using the analytical approach are of the order of CPU-seconds, compared with thousands of CPU-hours required using numerical methods. Finally, application of the model to stage II FCG data from the literature demonstrates its versatility, as it showed strong agreement with the data within a broad range of ΔK values. The comparison also demonstrates the ability of the dislocation shielding term to capture the transition from fatigue ‘starting’ behaviour through to stage I or II crack growth.

6 Synergistic Coupling of Thermomechanical Loads and Irradiation Damage

Some of the results presented in this chapter were produced by Dr Yang Liu (YL) at Imperial College London. As this work is the result of a collaborative effort, and is intended for publication, all results have been maintained to ensure a coherent story is told. Specifically, the implementation of new models which were generated for this chapter were implemented within user-material subroutine codes by YL. Accordingly, YL also ran the CPFE simulations and produced Figure 73, Figure 74, and Figure 75 from post-processed results.

6.1 Introduction

The impact of the synergistic evolution of irradiation damage and thermomechanical loading has not yet been simulated for any material. This chapter addresses this systematically using new phenomenological models for temperature and damage-dependant evolution of dislocation loop density in Zircaloy-4, within a CPFE framework. Zircaloy-4 is designed to act as a barrier between radioactive fuel pellets and water in a pressurised water reactor; it is therefore vital that factors affecting its structural integrity are fully understood. Recently, several studies [302], [303], [304] addressed the effect of irradiation on dislocation loop density using TEM and advanced XRD techniques. Topping et al. [304] demonstrated the thermal instability, and hence recovery of dislocation loops in zirconium alloys across a temperature range relevant to civil nuclear power. Torimaru et al. [305] showed that after just 15 s of heat treatment at 600 – 700 °C, the ultimate tensile strength and fatigue life of neutron irradiated (hardened) zirconium cladding tubes were restored to near unirradiated values. Similarly, early Zircaloy-2 data from Lowe and Thomas [306] showed increased softening with increasing annealing temperature. Moreover, the flow stress vs. annealing temperature trend from Lowe and Thomas [306] closely resembles the dislocation loop density vs. annealing temperature trend presented by Topping et al. [304], highlighting the important link between irradiation damage, thermal history, and mechanical response. As discussed in Chapter 2, dislocation loops formed during irradiation lead to both significant hardening and post-yield strain softening behaviour in cladding materials [90], due to the initial pinning and subsequent annihilation of dislocation loops, resulting in channel clearing [307]. The annihilation of an irradiation loop occurs when it interacts with a moving dislocation. Under sufficient stress, the moving dislocation, initially pinned by the loop, can drag and/or annihilate the loop, as described by various authors [308],

[309], [310], [311]. This results in the formation of so-called dislocation channels, within which plastic deformation is localised. These features are analogous to persistent slip bands (PSBs) which arise during fatigue of metals [312]. Hence, a fully coupled model requires fundamental understanding to describe the evolution of dislocation loop density with respect to plastic strain, irradiation dose, and temperature. Recent CPFE modelling advances [111], [313], [314], [315], underpinned by in-situ high resolution digital image correlation (DIC) [91], have offered improved mechanistic understanding of microscale plastic deformation in irradiated materials. However, for investigation of synergistic irradiated loading, experiments reach a practical limit. Ideally, experiments would involve thermomechanical testing with in-situ neutron irradiation. However, the cost, time, and radioactivity issues associated with neutron irradiation render this approach infeasible in most cases. Xu et al. [316] carried out micro tensile testing of single crystal nickel with in-situ helium ion irradiation, which has less safety implications and is less expensive than neutron irradiation. While this approach is attractive for these reasons, the damage mechanisms associated with helium ion irradiation are fundamentally different and the damage is concentrated at the surface [317], thus the effect of irradiation on bulk properties is unattainable. The major literature gap is in understanding material behaviour under concurrent irradiation damage and thermomechanical loads; while comprehensive, published studies thus far rely on mechanical test data from post-irradiated samples. This chapter aims to (i) determine if synergistic effects have any major implications in terms of structural integrity compared with conventional post-irradiation testing, (ii) develop understanding of how irradiation-linked damage mechanisms (e.g., channel clearing) are affected at elevated temperatures, and (iii) assess the influence of irradiation damage rate on microstructural damage evolution. Hence, this chapter provides an essential path towards total process-structure-property characterisation of safety-critical components for civil nuclear power (both for fission components, e.g., cladding and reactor pressure vessels, and fusion in-vessel components). Theoretical insights are provided for industrially relevant case studies.

6.2 Methodology

6.2.1 Adaptation of existing crystal plasticity formulation for irradiation hardening and softening

To capture both hardening and softening effects due to irradiation damage in Zircaloy-4, Hardie et al. [111] developed a mechanistic CPFE-dislocation-based model. Central to this is the incorporation of an irradiation hardening term (a function of dislocation loop density)

within the classical Taylor model. Here, a modified isotropic linear hardening (ISO-LH) model [111] is incorporated, whereby the dislocation loop density is treated as equivalent to other dislocation types. The CRSS is then given by,

$$\tau_c^i = \tau_0^i + \mu b \sqrt{\rho_{\text{SSD}} + \rho_{\text{GND}} + \rho_{\text{loop}}} \quad (6.1)$$

where τ_0^i is the intrinsic material CRSS (measured experimentally [164]), μ is shear modulus, b is Burgers vector magnitude, ρ_{SSD} is the density of statistically stored dislocations (SSDs), whose evolution is linear with plastic strain [318], ρ_{GND} is the density of geometrically necessary dislocations (GNDs) [319], and ρ_{loop} is the total density of dislocation loops, which form during irradiation. Temperature-dependant elastic-plastic properties [318] are also incorporated. SSD density evolution is controlled by $\rho_{\text{SSD}} = \gamma^{\text{st}} p$, where p is cumulative plastic strain and γ^{st} is a material constant. ρ_{GND} is calculated based on gradients of plastic strain [169], the implementation of which has been described by Xu et al. [175]. Note that for industrially relevant irradiation doses, the $\langle c \rangle$ dislocation loop density is negligible [320], since experimental TEM observations have identified that $\langle c \rangle$ loops only form at doses > 10 dpa [321]. Hence, only $\langle a \rangle$ loops are considered here. In the ISO-LH model, the density of dislocation loops varies equally on each $\langle a \rangle$ plane as a function of plastic strain, i.e., the softening behaviour is controlled by the annihilation of dislocation loops. The annihilation rate (due to slip) of total $\langle a \rangle$ loop density is given in differential form in Equation 6.2 [111],

$$\left[\frac{\partial \rho_{\text{loop}}}{\partial t} \right]_{\dot{\gamma}} = -N_{\langle a \rangle} \sum_{i=1}^{N_{\text{sys}}} \psi^i \frac{|\dot{\gamma}^i| \sqrt{\frac{\rho_{\text{loop},t}}{N_{\langle a \rangle}}}}{b^i} \quad (6.2)$$

where t is time, $N_{\langle a \rangle} = 3$ is the number of $\langle a \rangle$ directions, N_{sys} is the total number of HCP slip systems, and ψ^i is the annihilation probability for slip system i . Key properties are provided in Table 12. As shown in Figure 67 (a) and (b), the ISO-LH model is applied to a polycrystalline microstructure to capture room temperature macroscale stress-strain responses in Zircaloy-4 at different irradiation doses (from 0 to 0.8 dpa). There is strong agreement between model and experiment at each level of irradiation, using a strong basal texture (c-axes predominantly perpendicular to the loading direction) to best represent samples cut from a rolled sheet [90]; the texture was randomly generated with c-axes constrained to remain within 15 degrees of orthogonal to the remote loading direction, i.e., *soft* texture. Initial values for ρ_{loop} were selected for each dose to capture the tensile behaviour. Figure 67 (b) shows the

plastic strain distribution after post-irradiation (0.8 dpa) room temperature loading of the polycrystal model. Highly localised plasticity in the form of shear bands is shown, which leads to the softening behaviour captured in Figure 67 (a).

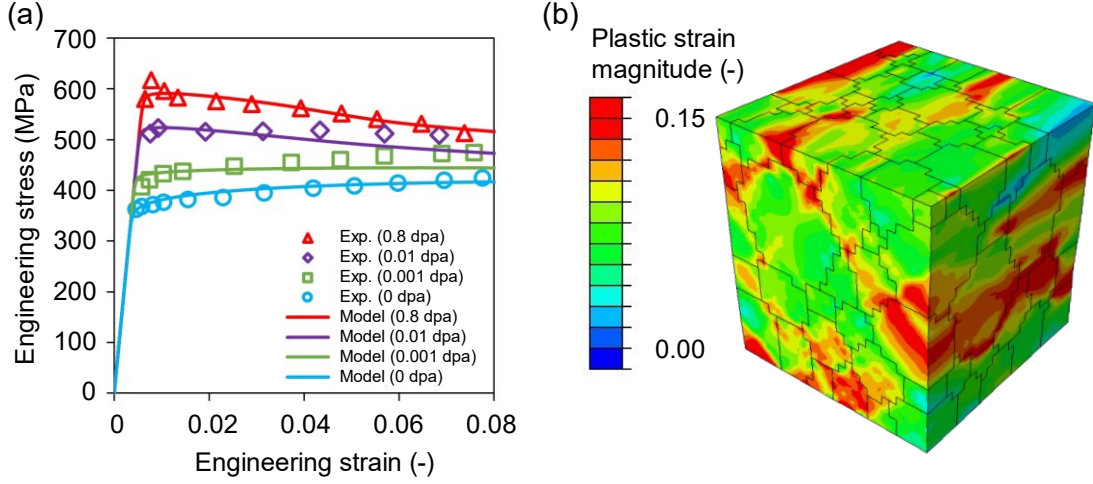


Figure 67: Capturing the polycrystalline response of Zircaloy-4 at different levels of irradiation using the modified isotropic linear hardening model from Hardie et al. [111]. Experimental data are from Farrell et al. [90]. (b) Distribution of cumulative plastic strain across 200 grain polycrystal model (0.8 dpa) at a mean applied strain of 8%.

6.2.2 Development of a simplified model for loop density evolution during irradiation

Under synergistic loading, there is competition between the formation of dislocation loops due to irradiation, their thermal annihilation, and their mechanical annihilation, as governed by Equation 6.2. New phenomenological models are proposed to fully describe the evolution of dislocation loops and to link irradiation dose with loop density. The number of atomic displacements per unit volume is given by multiplying the irradiation dose, φ (with units dpa) by the atomic number density, N (with units m^{-3}) [322]. The mean spacing between defects is therefore given by $l_d = \frac{1}{\sqrt[3]{\varphi N}}$. Since dislocation loops are formed by the coalescence of defects, i.e., displacements, the mean loop spacing is determined by multiplying the defect spacing by the ratio of defects to dislocation loops. For a planar circular dislocation loop, this is given by the ratio of dislocation loop to atomic defect areas. Hence, mean dislocation loop spacing is $l_{\text{loop}} = l_d \cdot \frac{\bar{A}_{\text{loop}}}{N^{-1/a}}$, where $\bar{A}_{\text{loop}}$ represents the mean planar area of dislocation loops and a is atomic spacing; note that in this simple preliminary model, the mean dislocation loop area is assumed constant, and is consistent with TEM-based dislocation loop diameter measurements in zirconium alloys [90] (see Table 12). If all defects contribute to the formation of dislocation loops, the theoretical maximum areal dislocation density is simply $\rho_{\text{loop}} = l_{\text{loop}}^{-2}$. However, due

to replacement collisions and recombination during the thermal spike of the collision cascade, only a small fraction of these displacements remain within the lattice as primary defects [323], [324], [325]. Additionally, many defects are lost over larger timeframes during irradiation due to recombination [326] or migration to sinks [327], the probability of which increases with dose (this is evident from CRA modelling of irradiation damage which predicts that both vacancy and interstitial concentrations saturate with increasing dose, and that the interstitial concentration can undergo a period of substantial decline with increasing damage [328]). Finally, only a fraction of surviving defects are likely to agglomerate forming dislocation loops. Due to the physical processes described above, the actual dislocation loop density is calculated by multiplying the theoretical maximum dislocation loop density by an efficiency term, n , which is assumed proportional to defect spacing as follows,

$$n = \eta l_d = \frac{\eta}{\sqrt[3]{\varphi N}} \quad (6.3)$$

where η is a material-specific empirical constant (with units m^{-1}) used for fitting, i.e., the efficiency of loop formation, n , decreases with increasing irradiation dose. The defects which are lost during irradiation are accounted for by the efficiency factor, giving $\rho_{\text{loop}} = n l_{\text{loop}}^{-2}$ [327]. Hence, the dislocation loop density is given by Equation 6.4.

$$\rho_{\text{loop}} = \frac{\eta(\varphi N)^{\frac{1}{3}}}{a^2 \bar{A}_{\text{loop}}^2 N^2} \quad (6.4)$$

Using the property values in Table 12, an estimate for η is enabled based on TEM measurements of dislocation loop density [90], [303], [304] at various irradiation doses. The influence of the loop density formation efficiency term, n is presented in Figure 68 using these properties. It is shown that for irradiation damage levels beyond 3 dpa, the efficiency is below 10%, which is consistent with results from a detailed numerical analysis of damage accumulation by Stoller and Mansur [329].

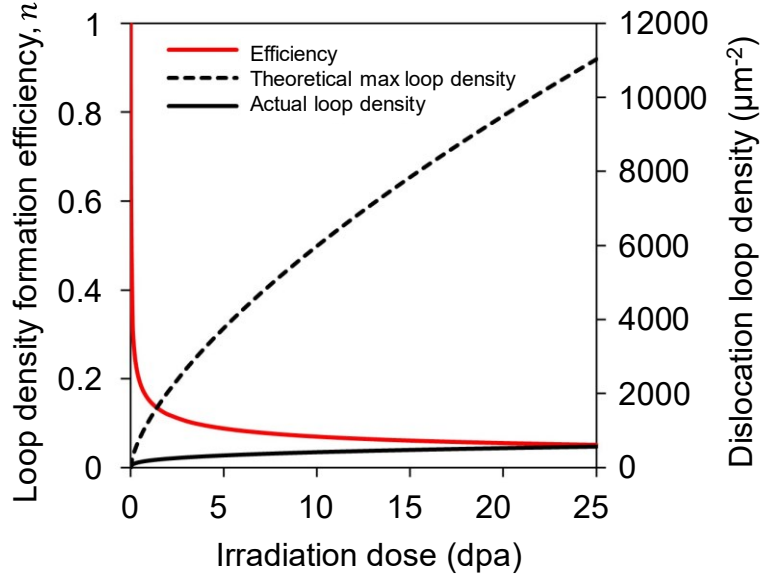


Figure 68: Evolution of loop density efficiency, and corresponding theoretical maximum and actual (fitted model) dislocation loop densities with increasing irradiation dose.

As shown in Figure 69 (a), the overall experimental data trend is captured reasonably well by the model from 0 to 25 dpa, including the aforementioned dislocation densities from the ISO-LH model. It is important to note that Equation 6.4 suggests that as the irradiation damage level approaches infinity, so too will dislocation loop density, while it has been reported to saturate experimentally [302], [330], [331]. The preliminary loop density-damage model does not entirely capture the phenomena responsible for saturation at higher doses and is therefore only applicable within the irradiation dose range presented in Figure 69 (a), i.e., $0 < \varphi < 25$ dpa.

6.2.3 Development of a model for dislocation loop annihilation at elevated temperatures

Recent experiments [304] demonstrated that heat treatment of irradiated zirconium alloys can lead to significant thermal annihilation of dislocation loops. This effect is incorporated into the current framework based on the bulk diffusion mechanism proposed by Eyre and Maher [332]. This mechanism suggests that the growth and shrinkage, and hence annihilation of dislocation loops is controlled by the diffusion of point defects to and from dislocation loops. Further, it is assumed that interstitial diffusion is dominant (despite the high formation energy compared with vacancies [333], interstitials arise due to the formation of Frenkel pairs [334]), as the migration energy of interstitials is much lower than that of vacancies [335], and so the interstitials migrate rapidly to the various sinks, including grain boundaries, interstitial loops, vacancy loops, dislocations, surfaces, etc. For a perfectly circular dislocation loop, the line

tension is given by Hirth and Lothe [336] as $\tau_{\text{loop}} = \frac{\mu b^2}{4(1-\nu)} \ln\left(\frac{4r_{\text{loop}}}{b}\right)$, where ν is Poisson's ratio.

The elastic energy released per vacancy emission is given by $w = \frac{\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b}$ [337] where V_{at} is the characteristic atomic volume ($V_{\text{at}} = N^{-1}$). From statistical mechanics, the concentration of interstitials in equilibrium with a loop is given by $c_{\text{I}} = c_0 \exp\left(-\frac{w}{k_{\text{B}} T}\right) = c_0 \exp\left(-\frac{\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b k_{\text{B}} T}\right)$ [337], where c_0 is the thermal equilibrium concentration of interstitials in the bulk. Conversely, for vacancy loops, interstitial emission results in a decrease in elastic energy, and hence, the local vacancy concentration in equilibrium with a loop is $c_{\text{V}} = c_0 \exp\left(\frac{\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b k_{\text{B}} T}\right)$. For a given irradiation dose, an initial dislocation loop density, $\rho_{\text{loop},0}$ is defined, corresponding to values measured using TEM at room temperature [304]. The temperature-dependant loop density is the difference between the initial value and the density of annihilated dislocations, $\rho_{\text{loop}} = \rho_{\text{loop},0} - \rho_{\text{an}} = \frac{\rho_{\text{loop},0}}{1 + \frac{\rho_{\text{an}}}{\rho_{\text{loop}}}}$. Assuming an annihilation event occurs for every vacancy and interstitial

interaction, there is a positive linear relationship between their concentrations. Since the diffusion of an interstitial defect can lead to annihilation of both vacancy and interstitial loops, there is also a direct correlation between dislocation density and interstitial concentration. Hence, it is reasonable to assume that the ratio, $\frac{\rho_{\text{an}}}{\rho_{\text{loop}}}$ is proportional to the ratio of interstitial to vacancy concentrations, giving $\frac{\rho_{\text{an}}}{\rho_{\text{loop}}} = \alpha \frac{c_{\text{I}}}{c_{\text{V}}}$, where α is a constant, potentially dependant on composition and microstructure. Applying the temperature dependent equations for vacancy and interstitial concentration gives $\frac{\rho_{\text{an}}}{\rho_{\text{loop}}} = \alpha \exp\left(-\frac{2\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b k_{\text{B}} T}\right)$. At temperature $T_{0.5}$, the initial dislocation loop density is reduced by half, $\rho_{\text{loop}} = \frac{\rho_{\text{loop},0}}{2}$, such that $\frac{\rho_{\text{an}}}{\rho_{\text{loop}}} = 1$. This gives $\alpha =$

$\frac{1}{\exp\left(-\frac{2\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b k_{\text{B}} T_{0.5}}\right)}$. Dislocation loop density is then given by Equation 6.5.

$$\rho_{\text{loop}} = \frac{\rho_{\text{loop},0}}{1 + \exp\left(\frac{2\tau_{\text{loop}} V_{\text{at}}}{r_{\text{loop}} b k_{\text{B}} T} \left(\frac{1}{T_{0.5}} - \frac{1}{T}\right)\right)} \quad (6.5)$$

As shown in Figure 69 (b), this new model captures experimental TEM measurements [304] [338] at different irradiation doses, i.e., two datasets representing different initial dislocation densities at room temperature.

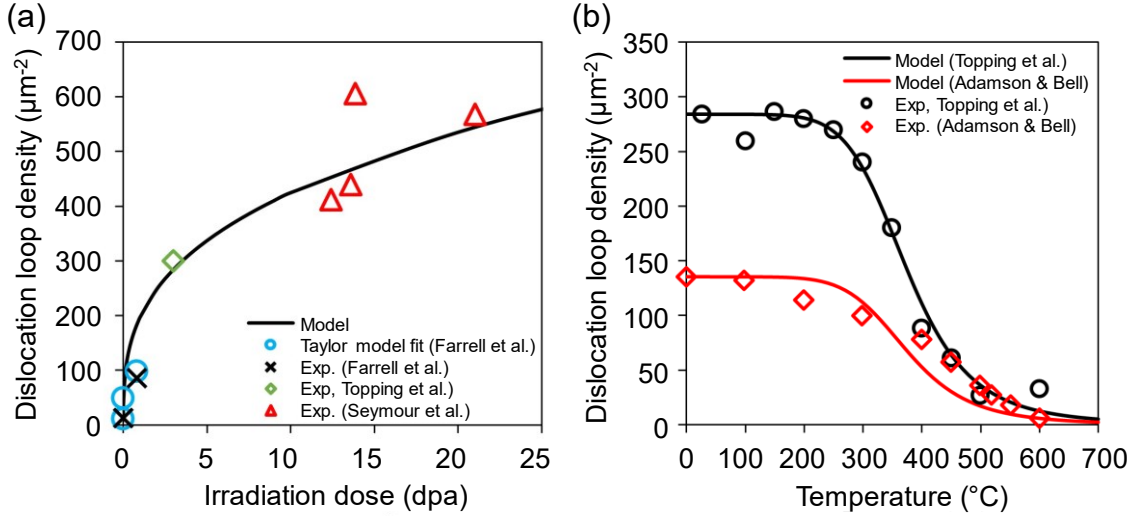


Figure 69: (a) Comparison of the new model for dislocation loop density as a function of irradiation dose with stress-parameterised values, and experimental TEM measurements from Farrell et al. [90], Topping et al. [304], and Seymour et al. [303]. (b) Comparison of model for steady-state annealing of dislocation loops with experimental data from Topping et al. [304] and Adamson and Bell [338].

With the dislocation loop density now described as a function of plastic strain, irradiation dose, and temperature, its fully coupled time-dependant evolution is written in differential form as,

$$\frac{d\rho_{\text{loop}}}{dt} = \left[\frac{\partial \rho_{\text{loop}}}{\partial t} \right]_{\dot{\gamma}} + \frac{\partial \rho_{\text{loop}}}{\partial \phi} \cdot \frac{\partial \phi}{\partial t} + \frac{\partial \rho_{\text{loop}}}{\partial T} \cdot \frac{\partial T}{\partial t} \quad (6.6)$$

where $\frac{\partial \rho_{\text{loop}}}{\partial \phi}$ and $\frac{\partial \rho_{\text{loop}}}{\partial T}$ are given by differentiating Equations 6.4 and 6.5, respectively, and $\frac{\partial \phi}{\partial t}$ and $\frac{\partial T}{\partial t}$ are loading conditions. Note that the temperature dependant evolution of loop density is subject to the constraint $\frac{\partial \rho_{\text{loop}}}{\partial T} = 0$ where $\partial T \leq 0$, i.e., decreasing the temperature has no direct effect on loop density. Moreover, it is assumed that the timescales considered in this chapter (several hours) are sufficient to apply the steady state model in Equation 6.5, i.e., transient effects are negligible.

Table 12: Important material properties. Dislocation loop radius is consistent with the range of experimentally measured values [90], [332], [337]. The critical annihilation temperature and annihilation probability are estimated in the current work.

Parameter	Symbol	Value	Unit
Annihilation probability (ISO-LH)	ψ^{1-30}	0.005	-
Loop formation probability constant	η	5.30×10^8	m^{-1}
Atomic number density	N	4.29×10^{28}	m^{-3}
Mean dislocation loop area	$\bar{A}_{\text{loop}}$	7.1×10^{-6}	μm^2
Dislocation loop radius	r_{loop}	1.5×10^{-3}	μm
Critical annihilation temperature	$T_{0.5}$	648	K

6.2.4 Case study: a global cladding model for representative loading conditions

6.2.4.1 Cladding stress due to internal pressure

Of the many sources of cladding stress, the build-up of internal pressure due to fission gas release is probably most severe. Unlike stresses associated with longitudinal irradiation growth, positive internal cladding pressure will always promote a tensile (circumferential) stress, which is likely to vary cyclically in parallel with temperature fluctuations. Moreover, cyclic tensile stresses have the potential to be highly damaging, particularly due to water-side environmental degradation processes, which can drive stress concentrations due to the formation of geometric inhomogeneities. In this section, a simple finite element model is used to estimate cladding stresses, based on a worst-case scenario internal pressure which is commensurate with high levels of burn-up. Based on analyses by Eidelpes et al. [339], a 20 MPa pressure is applied to the inner wall of an axisymmetric finite element based cladding model (using isotropic elastic-plastic properties for Zircaloy-4 at room temperature) as illustrated in Figure 70 (a). Cladding dimensions are approximate. Figure 70 (b) shows that the circumferential stress is greatest at the inner wall.

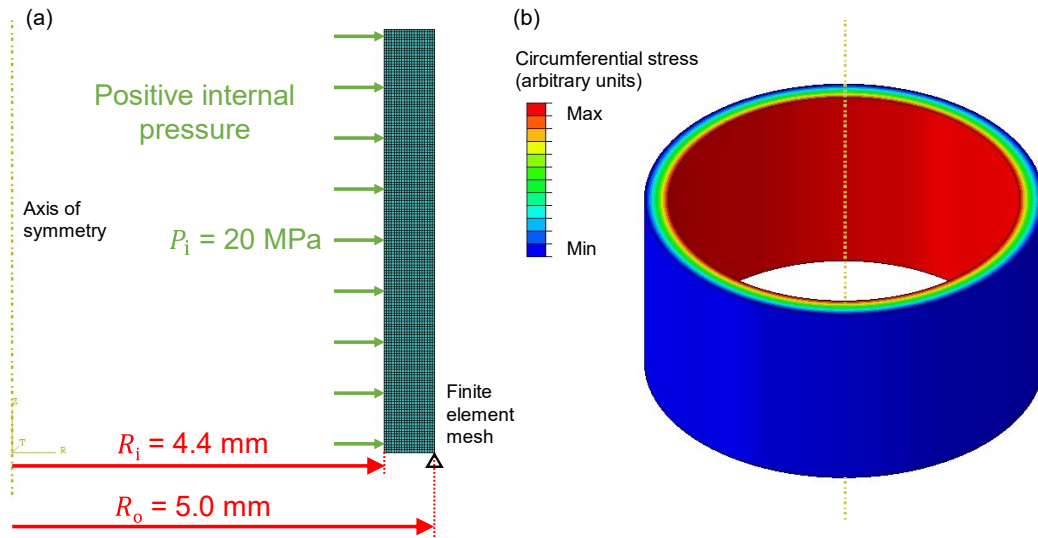


Figure 70: Isotropic elastic-plastic model to estimate stress distributions based on worst-case-scenario cladding internal pressure. (a) Axisymmetric (plane strain) finite element representation of section of cladding with internal pressure 20 MPa. (b) Representative circumferential stress distribution.

Figure 71 shows the resultant distributions of circumferential, radial, and longitudinal stress from the inner to outer cladding wall. Naturally, using simply supported boundary conditions in the axial direction, the longitudinal stress is always zero; in practise, this is not true, but longitudinal effects are assumed negligible for the purposes of this study.

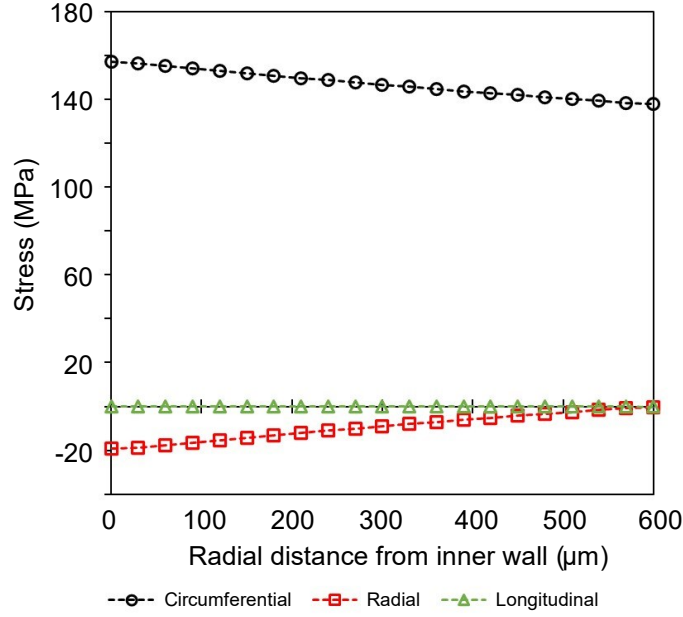


Figure 71: Circumferential, radial, and longitudinal (zero constraint) stress distributions in section of cladding, based on unirradiated Zircaloy-4 material properties from Figure 67 (a).

The circumferential and radial stress distributions are shown to vary almost linearly from the inner to outer cladding wall, with radial stresses corresponding directly to internal-external pressure differences. As anticipated, circumferential stresses are tensile and greatest in magnitude, with highest values at around 160 MPa at the inner wall. Based on the isotropic elastic-plastic material properties for unirradiated Zircaloy-4 (Figure 67 (a)), this worst-case-scenario will not lead to the onset of plasticity, and hence, synergistic irradiation loading will have no real consequences for structural integrity. For a more interesting case study therefore, a notch, i.e., stress concentration feature, is introduced to a section of cladding with similar pressure-based loading conditions in the next subsection.

6.2.4.2 *Introducing a notch: extraction of crystal plasticity sub-model boundary conditions*

In this chapter, industrially relevant stress-controlled boundary conditions are imposed onto a $50 \times 50 \mu\text{m}$ microstructure section from Thomas et al. [91], as shown in Figure 72 (a). To simulate stress levels sufficient to initiate channel clearing phenomena in irradiated fuel cladding, a global model is constructed with an internal pressure equivalent to that from high levels of burn-up [339], and is notched at the outer surface (representing a surface defect due to e.g., interfacial cracking with the oxide layer [340], hydride blistering [341], etc.). Bulk isotropic elastic-plastic material properties are used in the global model. Figure 72 (a) shows that both normal and shear stress boundary conditions are extracted from the global model and applied to the sub-model. Plane strain boundary conditions are imposed in each case,

which are representative of a pipe. Figure 72 (b) shows the thermomechanical loading conditions applied to the sub-model; three repeating thermomechanical cycles [318] are used to study cyclic plasticity. One case of irradiation damage evolution up to 6 dpa is also presented in the figure, which shows the synergistic coupling of damage and thermomechanical loading.

The four cases presented here are summarised as follows:

- i. Unirradiated. Thermomechanical loads are applied to unirradiated material and hence, $\rho_{\text{loop}} = \frac{d\rho_{\text{loop}}}{dt} = 0$ throughout.
- ii. Full synergistic coupling. Thermomechanical loads are applied synergistically with linearly increasing irradiation damage, corresponding to a very high constant neutron flux of $1.0 \times 10^{20} \text{ n m}^{-2} \text{ s}^{-1}$ [342], which gives a damage rate, $\frac{\partial \phi}{\partial t} = 3.7 \times 10^{-5} \text{ dpa s}^{-1}$. The equivalent irradiation damage level after three cycles is 6 dpa.
- iii. Partial synergistic coupling. Irradiation dose evolution remains coupled with thermomechanical loads, while thermal annihilation of dislocation loops is unaccounted for, i.e., $\frac{\partial \rho_{\text{loop}}}{\partial T} = 0$.
- iv. Post-irradiated. The material is pre-irradiated to 6 dpa of damage (consistent with coupled cases) prior to loading. Initial dislocation loop density is estimated using Equation 6.4. Neutron flux is set to 0, i.e., $\frac{\partial \phi}{\partial t} = 0$. Unlike previous studies [111], the thermal recovery of dislocation loops is accounted for here.

An additional set of results with a reduced pre-irradiation damage level (0.6 dpa) is also presented for contrast. For cases of full and partial coupling, this corresponds to a damage rate of $3.7 \times 10^{-6} \text{ dpa s}^{-1}$, which is more comparable to e.g., a realistic pressurised water reactor environment (typically in the range $10^{-7} - 10^{-6} \text{ dpa s}^{-1}$ for reactor core components [343], [344]); the implications of artificially increasing the irradiation damage rate this for structural integrity are perused somewhat at the end of this chapter.

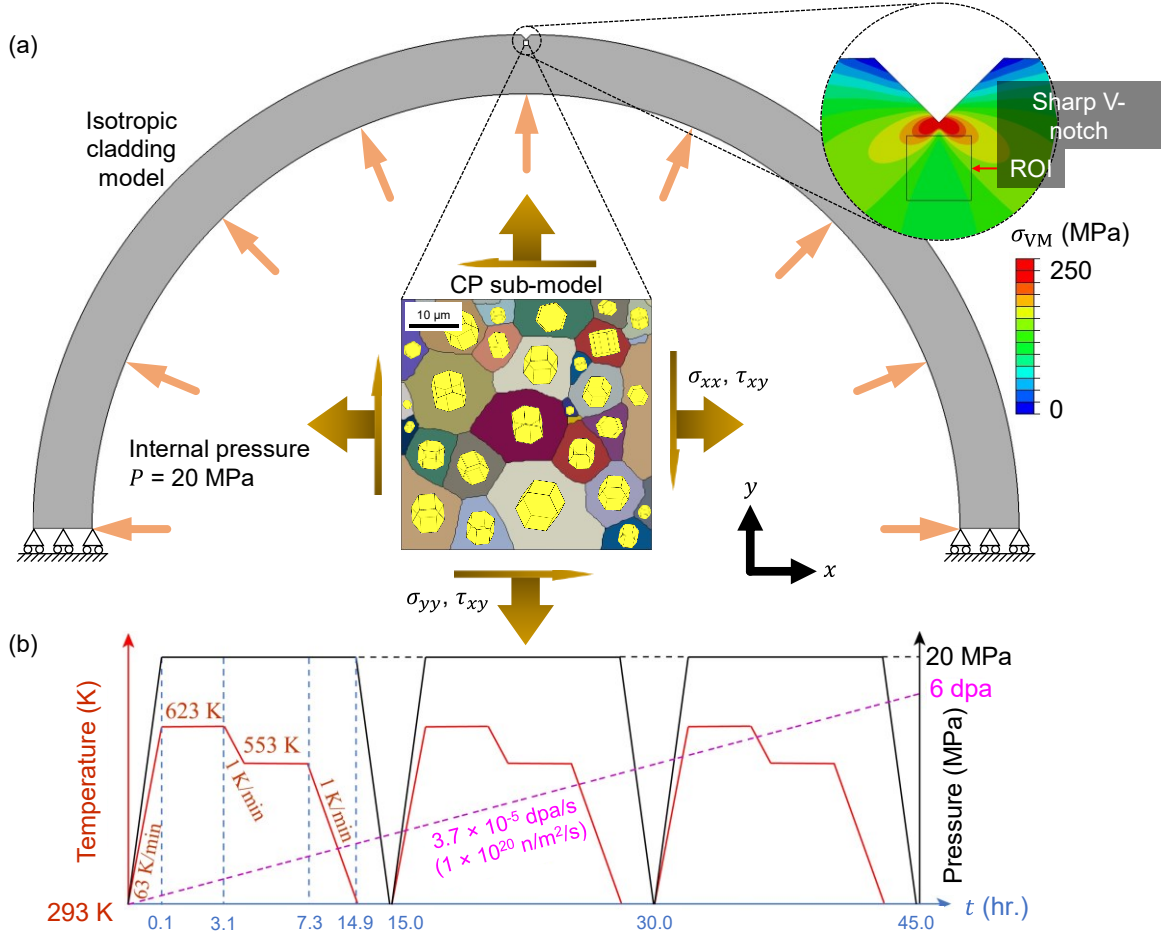


Figure 72: (a) Schematic illustration of notched cladding model and CPFE sub-model. The notched cladding model is used to extract normal and shear stress distributions around the region of interest, for application to the sub-model. The Von Mises stress distribution, in relation to the ROI is shown. Stresses are extracted at the beginning and end of the first cycle and are assumed unchanged for subsequent cycles. Internal and external cladding diameters are 8.8 and 10 mm, respectively. Notch depth is $50\ \mu\text{m}$. (b) Thermomechanical cycles used for each simulation from Liu et al. [318]. The selected maximum internal pressure is increased at the beginning of each cycle, held constant, and decreased, while there are two separate dwell periods for temperature. A single example of irradiation dose evolution is shown, whereby the equivalent level of damage increases linearly to 6 dpa over three thermomechanical cycles. This is a simplification for the purpose of this preliminary study. For demonstration purposes, a large dose rate is selected to reach 6 dpa within just a few cycles.

Nevertheless, results from high damage rate studies enable qualitative and semi-quantitative assessment of the long-term synergistic effects of irradiation damage and thermomechanical loading, while minimising computational cost.

6.3 Results and discussion

6.3.1 Influence of loading on key damage variables

The results of cases i. – iii. are presented in Figure 73 after three cycles, showing distributions of plastic strain, dislocation loop density, GND density, maximum slip, and stress. Contour plot results for the post-irradiated case have been omitted from Figure 73 as irradiation

hardening was predicted to prevent the onset of plastic deformation in the simulation (under the current loading conditions), even after three cycles. Figure 73 (a) – (c) also highlight that irradiation hardening effects under full and partial coupling somewhat inhibit plastic deformation compared with the unirradiated case. Specifically, most plastic deformation under full and partial coupling occurs within the first cycle due to progressive irradiation hardening, while there is a greater cyclic evolution in the unirradiated microstructure. Plastic strain fields from partial coupling demonstrate the importance of accounting for the thermal annihilation of dislocation loops, as the localisation of plasticity is intensified in the fully coupled case, which promotes channel clearing. Channel clearing is particularly evident in the grain highlighted in Figure 73 (b); parallel bands containing negligible dislocation loop density (which correlate with bands of high plastic strain) spanning the grain are shown in Figure 73 (d). Loop density line profiles from full and partial coupling are presented in Figure 73 (h) and show that overall loop densities are higher with partial coupling, while local variations are much larger in the fully coupled case, particularly between the first and second (from left to right) channel clearing bands. Curiously, the maximum loop density magnitudes for full and partial coupling along A-A' are close, despite there being large differences otherwise. This indicates that within the grain of interest, channel clearing is the dominant softening mechanism for both cases. Channel clearing is also shown to have a profound effect on the local GND density in Figure 73 (e) as there is significant localisation parallel to the channels, with a clear spatial correlation. Figure 73 (i) shows that for full coupling, GND localisation, which is driven by large strain gradients coincides with the largest loop density undulation. By contrast, GND density is shown to be more evenly distributed in the partially coupled model. Severe grain boundary driven strain localisation in the unirradiated material is shown to derive large GND densities, the magnitudes of which are comparable with peak values in the fully coupled case. Maximum shear strain $\left(\gamma_{\max} = \sqrt{\frac{1}{2}(\epsilon_{11} - \epsilon_{22})^2 + \epsilon_{12}^2}\right)$ is plotted in Figure 73 (f) showing bands of high shear strain localisation. However, Figure 73 (j) shows that these bands also exist in the unirradiated material, with approximately twice the magnitude. Even lower strain levels with minimal variation are shown with partial coupling. Lastly, Figure 73 (g) shows that there is some correlation between GND density and the x-direction stress, particularly along the inclined band. However, it is clear from Figure 73 (k) that due to interactions between the *soft* grain (c-axis $\perp$ x-axis major load) of interest and

neighbouring *hard* grains (c-axis $\parallel$ x-axis major load), stress shielding is occurring; stresses and strains are re-distributed such that strain localises in the *soft* grain, and stress is localised in *hard* grains. The effect is strongest in the fully coupled case, which also experiences the largest overall stress between the first and second channels, corresponding to the region with maximum GND density. A similar trend is shown in the unirradiated case, but with lower peak stresses and reduced stress re-distribution.

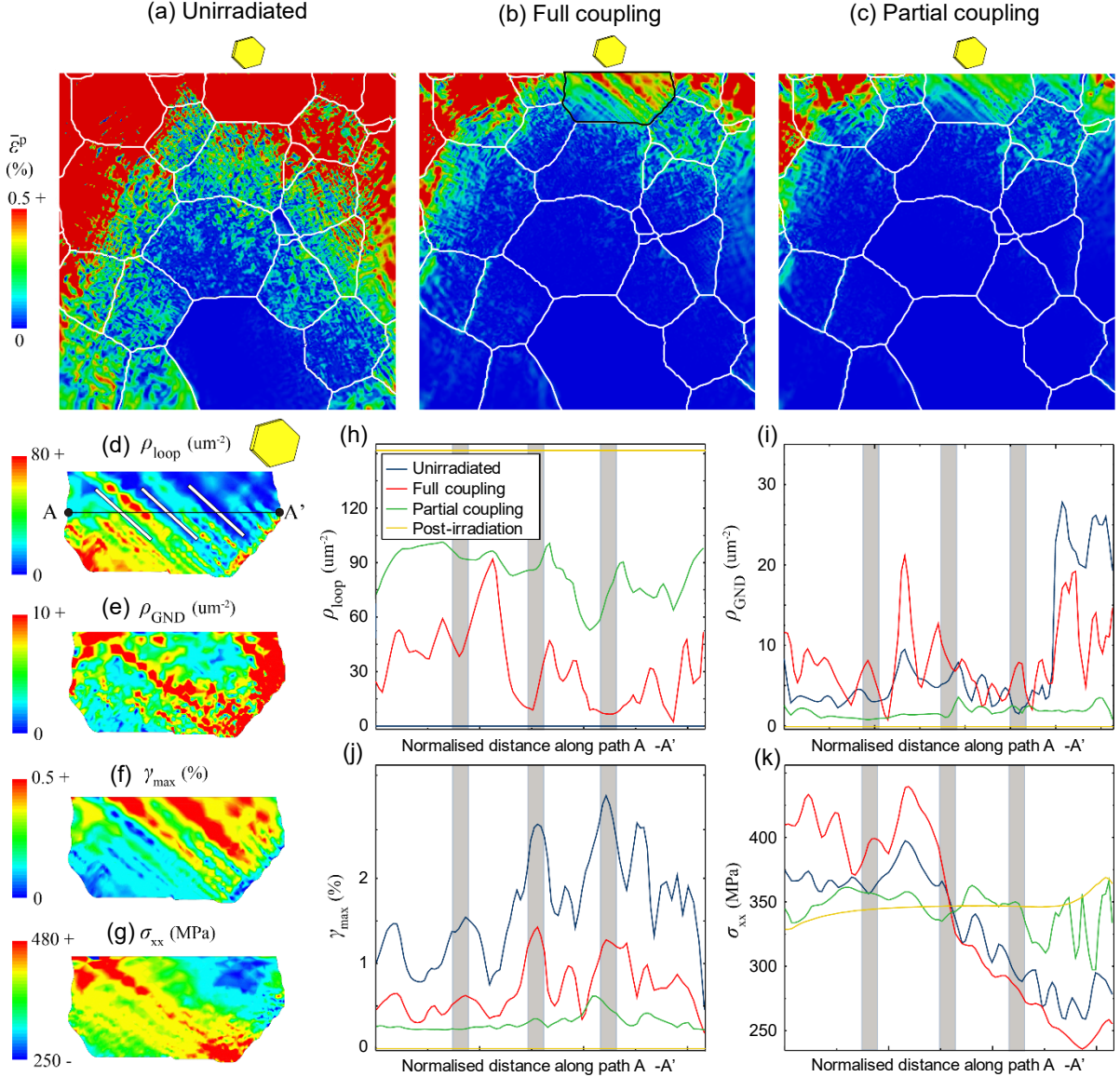


Figure 73: Results of unirradiated, partially coupled, and fully coupled CPFE simulations after three cycles: (a) – (c) plastic strain distributions. For full coupling, within the highlighted grain, (d) – (g) show the dislocation loop density, GND density, maximum shear strain, and x-direction stress distributions, respectively. Line A-A' is shown in (d), which traverses several ‘channel clearing’ regions, corresponding to drastically reduced dislocation loop density (as indicated by parallel white lines). Line profiles of these variables along line A-A' are shown for each case in (h) – (k), respectively. Channel clearing regions are indicated by grey shading.

Partial coupling shows comparably low stress re-distribution, and roughly follows the elastic, post-irradiated trend. Hence, despite the synergistic coupling of irradiation damage and loading leading to reduced strain localisation (compared with unirradiated material), channel clearing effects are shown to drive significant stress concentrations and localisation of GNDs, both of which have been linked with the nucleation of facets and crack initiation [172].

6.3.2 Dislocation loop density evolution

Figure 74 shows the mean dislocation loop density at the end of each cycle for every high irradiation dose rate case. Full coupling results in a progressive non-linear evolution, which reaches less than half of the initial post-irradiated value ($285 \mu\text{m}^{-2}$) by the end of the third cycle. Without thermal annihilation, partial coupling shows a more rapid evolution which reaches within 15% of the maximum possible value; this difference is due to plasticity-driven annihilation, while the fully coupled annihilation is due to both thermal and thermally assisted plasticity-driven annihilation. Loop density evolution in the fully elastic post-irradiated material is driven by thermal annihilation only, the rate of which is shown to decrease as loop density magnitude decreases.

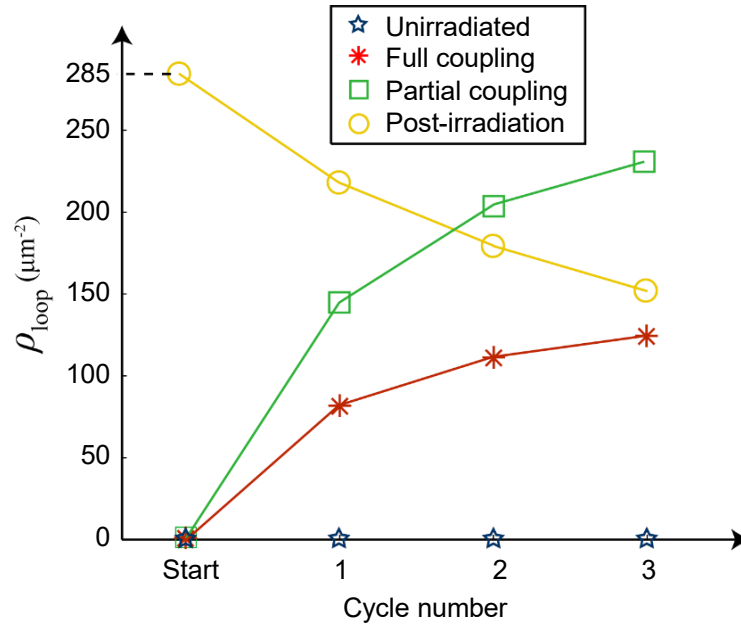


Figure 74: Cyclic evolution of mean dislocation loop density (averaged across entire microstructure) for each case of high irradiation dose investigated.

Results from the low irradiation dose rate study are presented in Figure 75. As with the high dose rate study, the model predicts a progressive non-linear evolution of dislocation loop density with increasing cycle number for full and partial coupling, despite dislocation loop

density magnitudes being substantially lower. As shown in the 'Full coupling' grain subfigure, channel clearing is also captured here; however, after just three cycles, the range of dislocation loop density is not significant enough to highlight key distinctions with e.g., the unirradiated case.

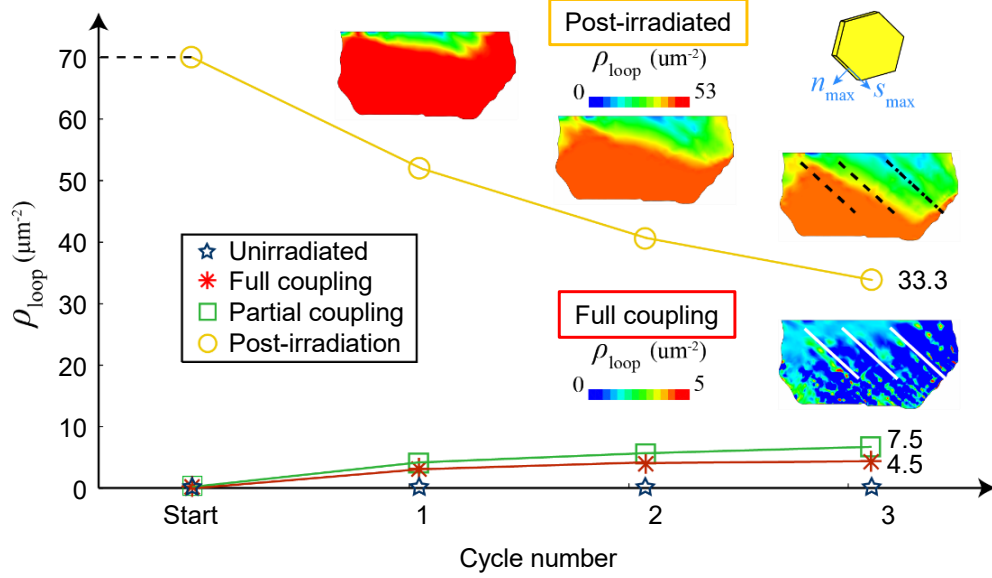


Figure 75: Cyclic evolution of mean dislocation loop density (averaged across entire microstructure) for each case of low irradiation dose investigated, with corresponding dislocation loop density fields in post-irradiated and fully coupled cases.

This demonstrates the utility of high irradiation dose rates from a computational cost perspective since important mechanistic insights are gained without exhausting resources. Interestingly, the comparably low initial dislocation loop density in the post-irradiated case results in some plasticity-driven dislocation loop annihilation, while virtually none was observed in the high irradiation dose rate due to increased hardening. However, it is clear from the grain subfigures that the resulting loop density distributions are entirely different (to those observed when using full synergistic coupling), and hence, failure mechanisms are likely to differ. More specifically, post-irradiated softening is initially dominated by the thermal annihilation of dislocation loops, which gradually leads to plasticity and diffuse loop annihilation (rather than the formation of discrete localised bands). By contrast, the synergistic coupling of irradiation damage is shown to lead to early plastic deformation, the localisation of which is increased due to channel clearing, as compared with no irradiation. This observation underpins the motivation for this chapter and brings to the forefront the importance of the synergistic coupling of irradiation damage, temperature, and mechanical load. Hence, there is a need for further research in this area, which will involve greater depth and improved

modelling, incorporating e.g., transient effects and loop density saturation using physically based models which are compatible with crystal plasticity, e.g., cluster dynamics [345]. Specifically, models accounting for full temperature-dependant damage evolution and annealing history are required (at present, only temperature dependant loop annihilation is considered). Recent work by Li et al. [346] showed that cluster dynamics modelling could be coupled with discrete dislocation plasticity to capture the concurrent effects of stress, plasticity, and irradiation damage in zirconium. It is envisaged that upon demonstrating the significance of synergistic effects in this chapter, a similar approach could be taken to bridge nano-scale irradiation damage with localisation of plasticity at microstructural length-scales in a more holistic manner.

6.3.3 Sub-study of transient behaviour at various irradiation dose rates at 293 K

In this chapter, a high irradiation dose rate (by industry standards [343]) is initially selected to minimise computational expense through reduced simulation time. However, this may bear implications on steady state slip resistance and strain accumulation, and ultimately, structural integrity. Hence, the results presented are contextualised in this section using a simple 1D (single slip) analysis of the evolution of dislocation loop density during irradiation and mechanical load at constant temperature; transient temperature effects are ignored here as they are not directly linked with the highly damaging channel clearing phenomenon. From Equations 6.2, 6.4, and 6.6, the time-dependent dislocation loop density evolution law is as follows,

$$\frac{\partial \rho_{\text{loop}}}{\partial t} = \psi^k \frac{|\dot{\gamma}^i| \sqrt{\frac{\rho_{\text{loop},t}}{N_{<a>}}}}{b^i} + \frac{\eta \phi^{-\frac{2}{3}}}{3a^2 \bar{A}_{\text{loop}}^2 N^{\frac{5}{3}}} \quad (6.7)$$

where i corresponds to a single slip system number. The total dislocation loop density is given by numerical integration over small time increments. Using Equation 3.4, the slip rate is calculated based on a linearly increasing resolved shear stress, τ^i , over a one-hour period. Similarly, three linear irradiation dose rates are selected which cover those considered in this chapter as well as more industrially relevant rates (1×10^{-7} dpa s⁻¹, 1×10^{-6} dpa s⁻¹, and 1×10^{-5} dpa s⁻¹). Hardening is accounted for using Equation 6.1 with the simplifying assumption that GND density is zero. Two stress rates, $\dot{\tau}^i$, and two initial dislocation loop densities are selected in this analysis, giving the following list of cases:

- (a) $\dot{\tau}^i = 1/12 \text{ MPa s}^{-1}$ from 0 to 300 MPa, initial dislocation loop density, $\rho_{\text{loop}} = 0 \text{ }\mu\text{m}^{-2}$.
- (b) $\dot{\tau}^i = 1/24 \text{ MPa s}^{-1}$ from 150 to 300 MPa, initial dislocation loop density, $\rho_{\text{loop}} = 0 \text{ }\mu\text{m}^{-2}$.
- (c) $\dot{\tau}^i = 1/12 \text{ MPa s}^{-1}$ from 0 to 300 MPa, initial dislocation loop density, $\rho_{\text{loop}} = 20 \text{ }\mu\text{m}^{-2}$.
- (d) $\dot{\tau}^i = 1/24 \text{ MPa s}^{-1}$ from 150 to 300 MPa, initial dislocation loop density, $\rho_{\text{loop}} = 20 \text{ }\mu\text{m}^{-2}$.

Figure 76 shows the evolution of dislocation loop density over time at each irradiation dose rate and for each case (a) to (d). The point at which the loop density evolution rate is zero (equilibrium point) is marked on each curve. Emphasis is placed on the equilibrium point as it is an indicator for the onset of channel clearing. Figure 76 (a) highlights that while the irradiation damage rates selected result in significant differences in dislocation loop density evolution, the time-points at which channel clearing is likely to begin or end are less affected.

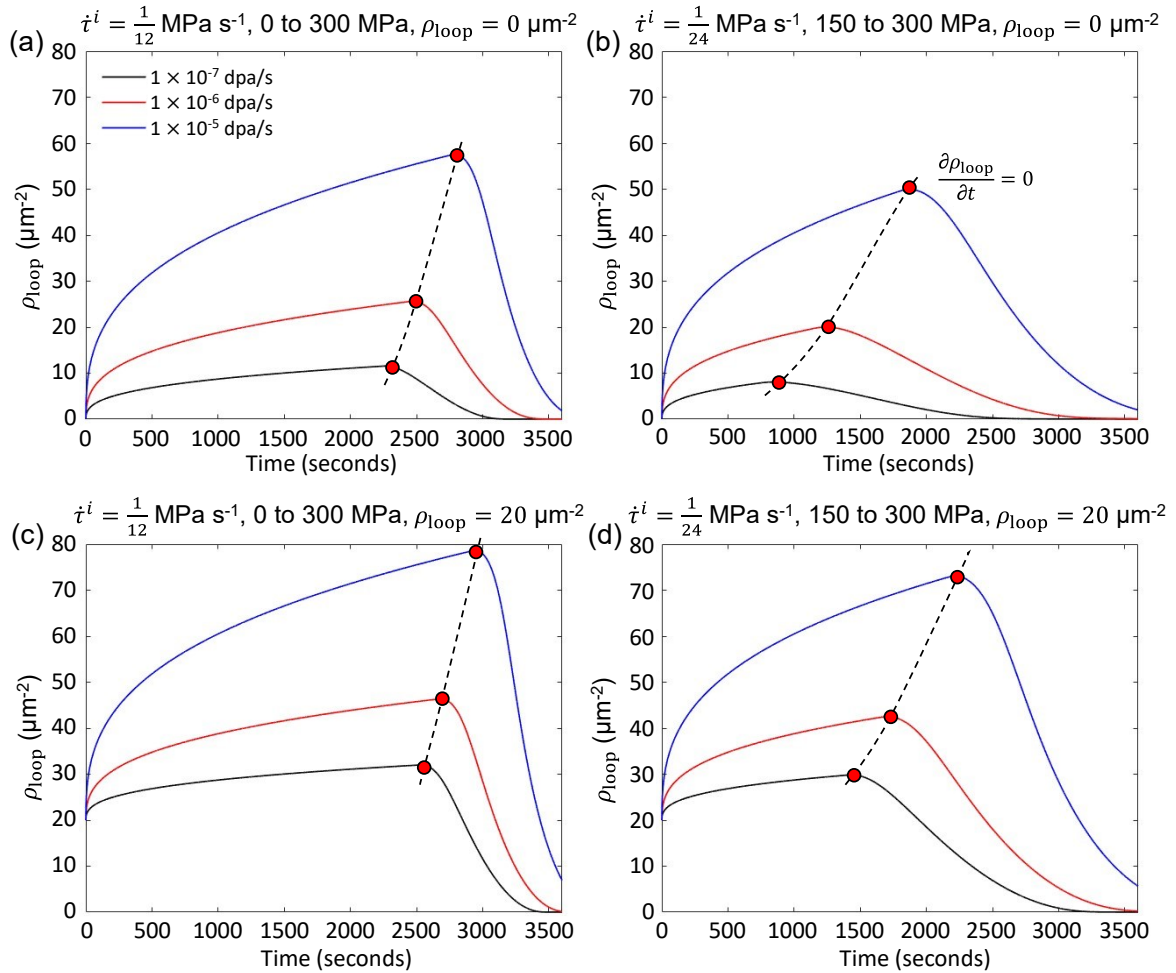


Figure 76: 1D representation of influence of irradiation damage rate on dislocation loop density evolution during synergistic mechanical load and irradiation damage at constant temperature (293 K) for cases (a) to (d).

Figure 76 (b) shows that the damage rate effect is heightened due to a reduced stress rate ($1/24 \text{ MPa s}^{-1}$); although the stress range considered here is conservatively large (since the

onset of plasticity leads to stress redistribution and a reduction in stress locally), this indicates that the combined selection of irradiation dose rate and stress rate can have important implications. Furthermore, Figure 76 (c) and (d) highlight that hardening due to prior irradiation via an initial dislocation loop density (or e.g., GNDs), the effect of irradiation dose rate is somewhat diminished. More importantly, the resolved shear stress values at the onset of channel clearing are presented for each case in Figure 77. Despite some apparently large variations in time at the onset of channel clearing, Figure 77 shows that the large variations are not reflected by the resolved shear stress. The largest stress increase as a result of increased irradiation damage rate (10^{-7} dpa s^{-1} to 10^{-5} dpa s^{-1}) is 20% for case (b). Hence, the implications for selecting higher dose rates are that (i) depending on the applied stress during synergistic irradiation and mechanical load, the onset of channel clearing may not be predicted where it otherwise would be within a 20% range, and contrastingly (ii) if stresses are sufficient to initiate channel clearing, the high stress and localised plasticity are likely to represent a worst case when considering structural integrity.

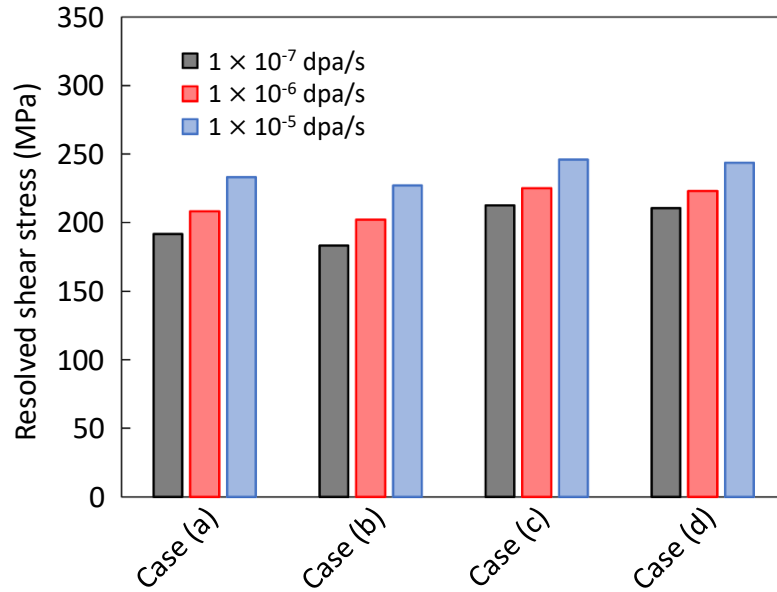


Figure 77: Resolved shear stress magnitude at transition point ($\frac{\partial \rho_{\text{loop}}}{\partial t} = 0$) for each irradiation dose rate. Cases (a) to (d) correspond to the sub-figure labels of Figure 76.

6.4 Conclusions

New models describing the formation and annihilation of dislocation loops were developed and implemented in a CPFE modelling framework. The role of synergistically coupling irradiation damage, thermal variations, and mechanical loads has been investigated in the context of a Zircaloy-4 microstructure. Simulation results have shown the significance of synergistic

coupling on the mechanisms involved in the initiation and localisation of plasticity, which is due to both thermal annihilation of dislocation loops, leading to softening, and the early onset of plasticity at low irradiation doses, leading to channel clearing. Thermal annihilation of dislocation loops is shown to be an important consideration, as it ultimately has a major effect on the distribution and magnitude of key damage quantities including GND density and stress. Crucially, partial coupling alone may indicate that irradiation can lead to beneficial reductions of these key quantities (compared with unirradiated material), while full coupling highlights the opposite effect.

7 Delta Hydrides: Influence on Fatigue Crack Growth and Localisation under Thermomechanical Loads

7.1 Introduction

Most studies of structural integrity involving hydrides in reactor fuel cladding materials are centred around DHC. Hence, DHC, which is most relevant to dry storage of spent fuel rods, is already reasonably well understood and is not dealt with within the current chapter. There is, however, a major literature gap in understanding how hydrides influence FCG, particularly within the stage I regime. Moreover, the issue of strain ratcheting in hydrided Zircalloys under reactor-relevant thermomechanical cyclic loading has been reported, but it is not yet known how this affects performance. Hence, to enable predictive capabilities for structural integrity, it is first necessary to produce baseline experimental data to address these knowledge gaps.

Early work by Wanhill et al. [347] sought to study the influence of zirconium hydrides on fatigue in Zircaloy-2 at room temperature and at 300 °C under variable-tension and reversed torsion loading conditions. At room temperature and 300 °C, for samples containing various hydrogen contents, the authors found no statistical differences in fatigue life owing to their inability to measure FCG in the stage I regime, i.e., only stage II cracking was considered, which was also immeasurable due to rates of growth being too high. Nevertheless, post-deformation analyses of fracture surfaces highlighted substantial differences in behaviour at different temperatures and for different hydrogen concentrations. At room temperature, it was concluded that hydrogen content could have a significant effect on fatigue damage mechanisms, as samples containing 600 wppm hydrogen exhibited up to 50% less (ductile) fatigue damage than non-hydrided specimens, despite having completely fractured due to e.g., cleavage and decohesion mechanisms within hydrides and at hydride-matrix interfaces, respectively. At 300 °C however, no samples fractured completely, and similar cracking mechanisms were observed for hydrided and non-hydrided materials; fracture toughness was shown to be virtually unaffected by hydrogen content, which was attributed to enhanced matrix ductility and the ability of the material to cross-slip. In some instances, hydride particles were observed to accommodate matrix shear by undergoing plastic deformation. Recently, this observation was buttressed by CPFE modelling and HRDIC work presented by Liu et al. [155], which showed multiple examples of cross-hydride slip transfer in Zircaloy-4 under monotonic tensile loading. Extruded 3D crystal plasticity representations based on 2D surface measurements of

microstructure regions containing delta hydride packets were shown to capture slip transfer events reasonably well, and provided new insights into the mechanisms which promote these events, based on crystallographic alignment, etc. Wanhill et al. [347] showed that while dislocation slip activity accommodated most deformation during fatigue, even at room temperature, hydride cleavage and void formation between hydride-matrix and hydride-twin or matrix-twin interfaces were also important factors, particularly for crack nucleation. Twins were implicated in several instances of crack nucleation, though the authors failed to declare the origin(s) of the twin features or whether there were local hydride interactions. However, it is reasonable to assume that since void formation at matrix-twin interfaces has not been reported for non-hydrided zirconium alloys, only twin-induced hydride precipitation can lead to such effects.

Using in-situ synchrotron X-ray diffraction to capture strain evolution in hydrides (in Zircaloy-2), Kerr et al. [348] showed that in some instances, there are three key deformation regimes: (i) fully elastic behaviour, (ii) matrix post-yield elastic behaviour (corresponding to a reduced modulus due to plasticity in the matrix and load transfer to the hydride), and (iii) strain saturation in the hydride due to either hydride fracture, interface failure, i.e., decohesion, or plasticity within the hydride. Based on the work presented by Liu et al. [155], and depending on the hydride crystal orientation with respect to the remote loading direction, it is possible that (iii) is linked with both hydride plasticity and successive fracture or decohesion. The implications of this for fatigue are clear; where the applied load is sufficient to initiate plasticity in the matrix and the onset of regime (ii), the cyclic accumulation of plastic strain local to the hydride may generate stresses sufficient to initiate regime (iii), beginning with plasticity within the hydride, ultimately leading to fracture. This mechanism can aid in understanding the increased levels of brittle fracture in hydrided samples [347]. Garlea et al. [349] showed that in $R = 0$ fatigue testing of as-received and hydrided Zircaloy-4, FCG rates were substantially higher in the stage II regime, leading to premature failure (compared with steady Paris regime behaviour). Moreover, the fatigue ‘starting’ behaviour for hydrided samples showed that the SIF necessary to initiate fatigue cracking was almost an order of magnitude lower than that for non-hydrided Zircaloy-4; this is attributed to matrix-hydride interactions leading to localised matrix plasticity and hydride-induced crack initiation. Hydride precipitation within the as-received alpha ‘Widmanstätten’ microstructures was predominantly intergranular. The authors reported that the corresponding fracture paths were controlled by this, unlike FCG in

non-hydrided Zircaloy-4, which is mostly transgranular [252]. Li et al. [350] also studied crack initiation and FCG in hydrided zirconium alloys. Without reinforcement from corresponding FCG rate data, results indicated that fatigue behaviour is strongly affected by the morphological orientation of hydrides with respect to the remote loading direction. Specifically, post-deformation electron micrographs of sample surfaces revealed that hydrides which are oriented orthogonally to the loading direction are particularly damaging due to (i) increased cracking, i.e., longitudinally along the hydride, and (ii) the increased potential for hydride-matrix decohesion. In a more recent study by the same authors [154], the fatigue life reduction associated with hydride-reorientation (20% radial, i.e., orthogonal to the loading direction) is quantified at around 33%, which highlights the increasingly detrimental effect of hydrides in materials subjected to combined thermal and mechanical loads.

In a recent publication by Taherijam et al. [351], EBSD and CPFE modelling were used to quantify the effects of hydride precipitation (and subsequent dissolution) in the absence of an external stress on the local rotational fields, i.e., GND density, and hydrostatic stress states. Precipitation was shown to produce networks and discrete bands of GNDs which interconnect hydrides, with densities of up to $1000 \mu\text{m}^{-2}$ (with highest values near hydride tips); this disaccords with EBSD-based measurements from Birch et al. [352], which show maximum GND densities of around $30 \mu\text{m}^{-2}$. The hydrostatic stress states which develop upon dissolution, as predicted by Taherijam et al. [351], were enormous and tensile within the former hydride region. This indicates that reprecipitation is highly likely to occur within the same region, while various locations surrounding the original hydride were shown to develop both tensile and compressive hydrostatic stresses. The authors note that the GND networks produced initially are also likely to result in additional precipitation locally.

While multiple publications highlight the effects of high temperature environments on fatigue in hydrided zirconium alloys [347], [353], [354] and others the various hydrogen and strain ratcheting effects associated with thermal cycling [355], [356], [357], no studies have yet quantified the local stress or strain states associated with multi-cycle hydride precipitation, which is crucial for mechanistic understanding of failure and prediction thereof at microstructure length scales. This final thesis chapter, which is broadly experimental, is aimed at (i) determining the various mechanisms (and the factors which control these mechanisms) involved in microstructurally short FCG in Zircaloy-4, (ii) assessing the effects of hydride

precipitates on local short FCG rates, aided by in-situ DIC, and (iii) quantifying local deformation levels due to hydride precipitation and subsequent ratcheting under cyclic thermomechanical loads.

7.2 Development of a heat treatment device for hydride redistribution

7.2.1 Introduction

In an experimental laboratory setting, it is difficult to reproduce the conditions which result in engineering component failure, primarily for pragmatic reasons, such as time constraints. As discussed in Chapter 3, notch defects are introduced artificially to represent naturally occurring surface defects due to e.g., manufacture, in-service oxidation, or hydride blistering. However, since notches were machined prior to the introduction of hydrogen, unwanted interactions between the surface of the notch (due to residual stress) and hydrogen in solution can occur. This effect was reported by Garlea et al. [349]. For isothermal FCG studies, this is not a major concern since the propagating crack quickly extends to a point at which its growth is independent of any such effects. For combined thermal and mechanical loading however, hydride-notch effects can be highly problematic, as illustrated in Figure 78. At the beginning of a trial thermomechanical fatigue test, while synchronously increasing the temperature and stress, it quickly became clear that the hydrogen content local to the notch was extremely high due to near-immediate hydride stress reorientation.

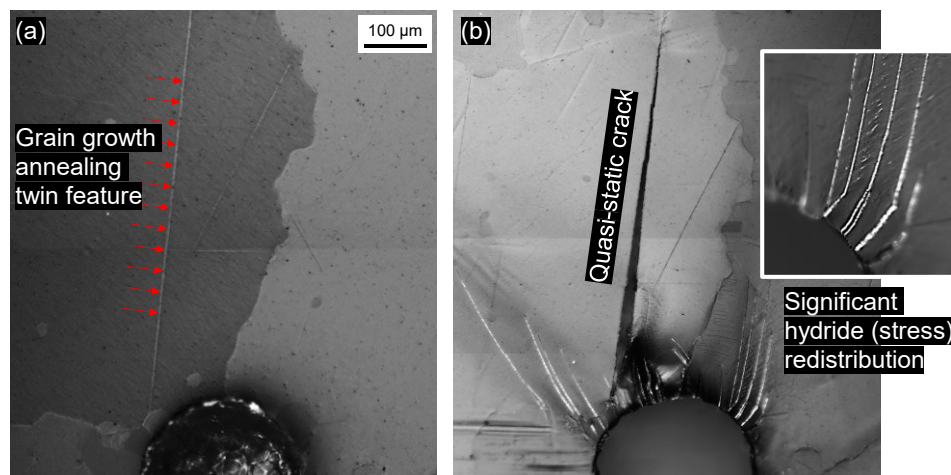


Figure 78: Detrimental effect of notch, annealing twin features, and hydride presence under monotonic thermomechanical loading. (a) Hydrided microstructure (sample H1) with large annealing twin prior to loading. (b) Same microstructure (H1) after synchronous thermal and mechanical monotonic loading, showing large quasi-static crack which was propagated along or through the annealing twin feature. Substantial hydride reorientation is also shown within the notch due to high initial hydrogen content local to the notch.

This was followed by substantial quasi-static cracking along the annealing twin feature, shown prior to testing in Figure 78 (a) (throughout which hydrides had precipitated). Hence, prior to achieving even a single thermomechanical fatigue cycle, the sample failed catastrophically due to unwanted and artificial hydride interactions, as shown in Figure 78 (b). In a previous attempt to counteract these effects, Honniball et al. [358] quoted a method by which the hydride distribution throughout a sample can be altered prior to testing, while ensuring that meaningful results are maintained. The method involves applying a thermal gradient heat treatment across notched and hydrided Zircaloy samples (like those used here). Having been originally developed by Canadian Nuclear Laboratories, details of the equipment used were not provided by Honniball et al. [358]; only process details were given, i.e., set temperatures at either end of the sample and heat treatment time. Hence, Section 7.2.2 describes the design of a thermal gradient heat treatment device for this purpose.

7.2.2 Design and manufacture of a device for thermal gradient heat treatments

The Soret effect (thermophoresis) is exploited by application of a thermal gradient across a hydrided Zircaloy-4 sample to ‘drive’ hydrogen away from the notch (for hydrogen in a zirconium lattice, it is driven from high to low temperature regions). Experimentalists at Canadian Nuclear Laboratories found that the optimal high and low temperature boundary conditions (surface temperatures) were 340 °C and 20 – 30 °C, respectively (note that samples were approximately 70% larger than those used in this chapter). Hence, the heat treatment device developed for the current studies utilises a cartridge heater (mounted within an aluminium block which is attached to a square aluminium contact plate) to heat one face of a sample, while the opposing face is cooled continuously via direct contact with a water-cooled heat sink (see Figure 79). Contact is maintained between the sample faces and the hot and cold ends using M3 × 40 bolts at each corner of the upper and lower aluminium plates. To mitigate against thermal stresses, springs are added to each bolt, allowing for thermal expansion, as shown in Figure 79 (c). A 200 W cartridge heater, which can reach temperatures of up to 400 °C is selected to ensure that the high surface temperature can be maintained given the required substantial thermal gradient (approx. 100 °C/mm), i.e., heat transfer rate.

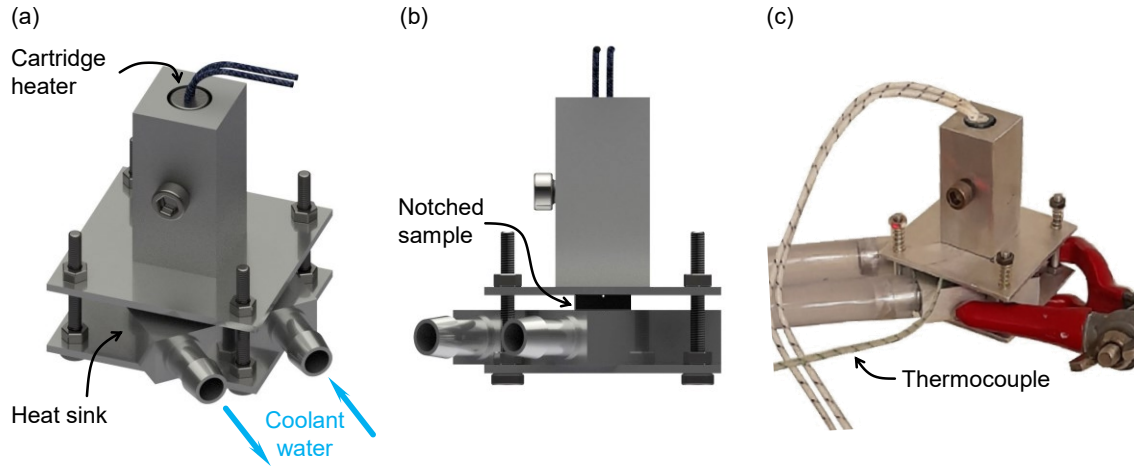


Figure 79: Design and implementation of thermal gradient heat treatment device. (a) Isometric view of design showing water-cooled heat sink, which is housed between two aluminium plates. The top plate is bolted to an aluminium block, which houses a 200 W cartridge heater (heating element). (b) Side elevation of device showing placement of notched beam sample. The notched end (face) is held in contact with the hot side, while the opposing face is in contact with the heat sink. (c) Actual image of heat treatment unit. Springs are added to the connecting bolts on the hot side to allow for thermal expansion while maintaining sample surface contact. A thermocouple is connected to the hot side aluminium plate, near the sample for temperature control.

For precise temperature control at the hot side, a *Eurotherm 2416* controller is used to control the cartridge heater power via a simple switching algorithm, based on live temperature measurements from a thermocouple mounted to the plate. Due to the substantial cooling rate however, the system takes several minutes to achieve temperature stability, at which point the switching frequency remains very high, resulting in cartridge heater damage and failure (on average, a new heating element was required after heat treatment of every second sample). It is therefore recommended that for future designs of such devices, a custom power control unit be included for constant power steady-state temperature control. At the time of testing, the mains water supply used to cool the heat sink was at around 15 °C. During steady-state operation, the surface temperature of the heat sink (close to the sample) was measured using a separate thermocouple at between 26 and 28 °C, which is within Honniball et al.'s recommended range [358].

7.2.3 Analytical modelling for spatial and temporal hydrogen redistribution

Despite using the same temperature boundary conditions as those previously reported [358], it is anticipated that the required heat treatment time will be reduced due to a reduced sample size. To generate a hydride denuded region spanning 30% of the sample thickness, Honniball et al. [358] applied the thermal gradient heat treatment for 150 hours. To estimate the heat treatment time required for the current work, a simple 1D spatial and transient analysis of

hydrogen transport is carried out based on the competition of temperature gradient (Soret) and concentration gradient (Fick) effects. Note that hydride dissolution and precipitation are ignored in the current analysis, since it is assumed that with the applied temperature gradient, most hydrogen within the region of interest is in solution. Hence, the constitutive equation used to predict the pointwise transient evolution of hydrogen concentration, C , is given by the spatial gradient of hydrogen flux as,

$$\frac{\partial C}{\partial t} = -\nabla(J_{\text{Fick}} + J_{\text{Soret}}) \quad (7.1)$$

where J_{Fick} and J_{Soret} are the fluxes due to concentration gradient and temperature gradient, respectively. It is important to note that the current model is only used to estimate an appropriate heat treatment time based on hydrogen transport, and disregards the effects of hydride precipitation and dissolution, etc. Hence the resultant through-thickness hydrogen distributions will be indicative but inaccurate. Fick's law is a function of the diffusion coefficient, D , and concentration gradient as follows,

$$J_{\text{Fick}} = -D\nabla C = -D_0 \exp\left(\frac{Q_D}{RT}\right) \nabla C \quad (7.2)$$

where D_0 is the diffusion constant (taken here as $7.9 \times 10^{-7} \text{ m}^2/\text{s}$ [359]), and Q_D the energy for hydrogen transport in zirconium (taken here as $44,900 \text{ J/mol}$ [359]), and R the universal gas constant. The Soret flux is given by the following equation,

$$J_{\text{Soret}} = -\frac{Q^*DC}{RT^2} \nabla T \quad (7.3)$$

where Q^* is the ‘‘heat of transport’’. Based on experimental hydrogen pick-up measurements from Kammenzind et al. [360], this is taken as $31,000 \text{ J/mol}$. Hence, using Equation 7.1, the updated hydrogen content at position x (between the hot and cold surfaces) after some time increment, Δt is given by Equation 7.4.

$$C(x, t_{i+1}) = C(x, t_i) + \frac{\partial C(t_{i+1})}{\partial t} \Delta t \quad (7.4)$$

At the boundaries (hot and cold surfaces, which are represented here as points), the total hydrogen flux is set to zero throughout the simulation, as shown in Figure 80. Since the hydrogen flux is dependent upon temperature and concentration gradients, and the hydrogen concentration dependent upon flux gradients, it is necessary to embed at least two layers of

discretisation, i.e., fluxes are calculated at uniform intervals between either sample face, and hydrogen concentrations and temperatures are calculated and defined at uniform intervals between neighbouring flux calculation points. This is illustrated more clearly in Figure 80.

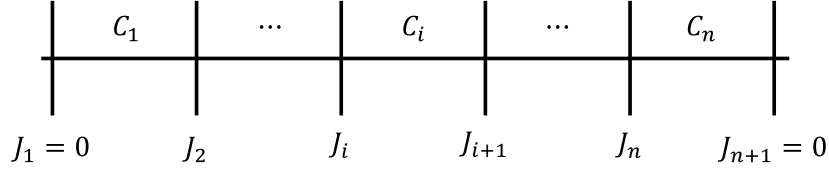


Figure 80: Illustration showing boundary conditions and method used to calculate hydrogen concentration distribution throughout sample, i.e., since concentration is a function of flux gradients, gradients are easily established between two points by linear interpolation.

Assuming immediate stabilisation of the predefined temperature gradient, the transient and spatial evolution of hydrogen is modelled using a custom *Matlab* script. The results of this are presented for a 3 mm thick sample with a constant thermal gradient for up to 200 hours in Figure 81. Figure 81 (a) shows the predicted spatial distributions of hydrogen at various stages of heat treatment. At the hot end, most hydrogen redistribution is predicted to occur within the first 10 hours of heat treatment; this is most clear in Figure 81 (b), which shows effective saturation after about 100 hours.

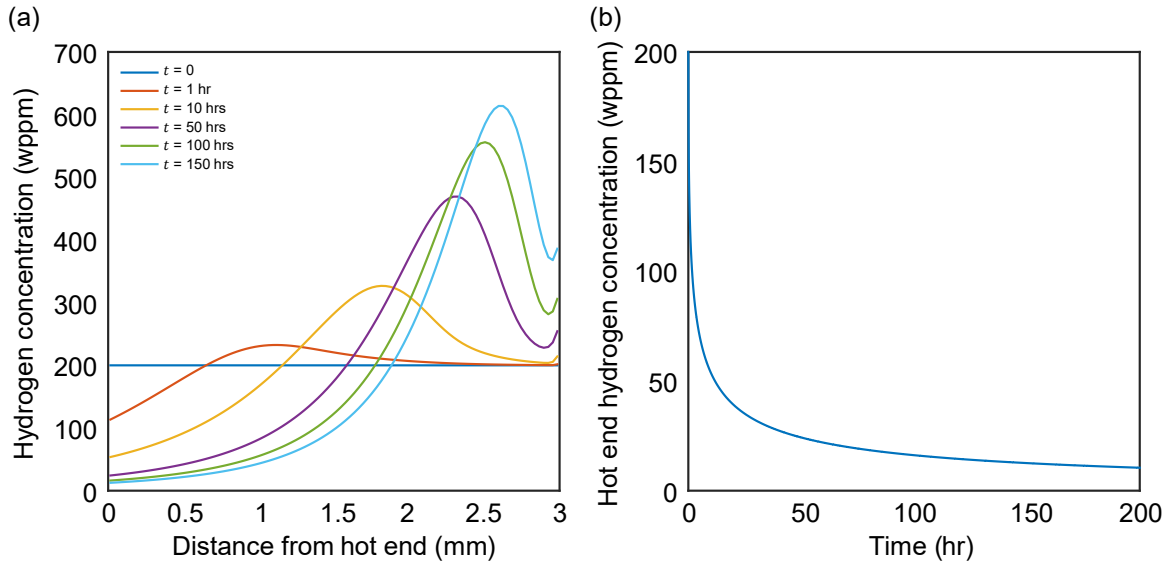


Figure 81: Influence of heat treatment time on hydrogen distribution. (a) Predicted distribution of hydrogen (at various points in time) between the hot and cold ends of a sample. (b) Predicted evolution of hot end hydrogen content over time.

Figure 81 (a) also shows that after 100 hours of heat treatment, there is a reduced level of hydrogen variation, spatially, between the hot end and the notch tip (which is approx. 300 μm from the hot end). Hence to reduce sample preparation time, a 100-hour heat treatment time is selected for this application.

7.2.4 Optical assessment of spatial redistribution of hydrides

Upon application of the thermal gradient heat treatment to an otherwise fully prepared sample (sample H2, which underwent a grain growth heat treatment, electrolytic hydrogen charging, and subsequent homogenisation heat treatment, as described in Chapter 3), it is necessary to evaluate its effectiveness prior to thermomechanical testing. Figure 82 shows optical micrographs throughout sample H2 prior to and after the heat treatment. Figure 82 (a) shows that prior to heat treatment, there is a reasonably uniform distribution of hydrides, with clear evidence of intragranular, intergranular, and twin boundary hydride precipitation. At the upper left-hand edge of the notch, there also appears to be substantial hydride precipitation.

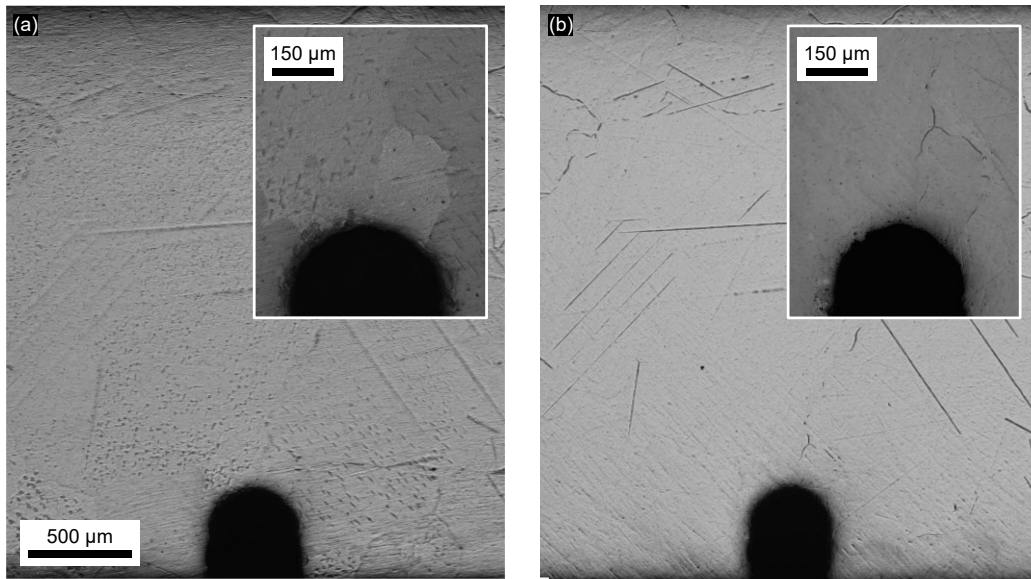


Figure 82: Hydride distribution throughout sample H2 (a) prior to and (b) after application of thermal gradient heat treatment.

Figure 82 (b) shows that after the 100-hour thermal gradient heat treatment, there is significant redistribution of hydrides. Firstly, it is immediately clear that throughout the sample, there is a substantial reduction of intragranular hydride fraction, which appears to have resulted in increased precipitation along grain and twin boundaries, as indicated by darkening of these regions. This is unsurprising, since the relatively long-term heat treatment (> 10 times longer than the hydride homogenisation heat treatment) allows ample time for hydrogen transport to these more energetically favourable sites [361]. More importantly however, the desired non-uniform hydride distribution is attained; specifically, between the lower (hot) end of the sample and the notch tip, virtually no hydrides are visible. Moreover, the hydrides which were observed to precipitate around the upper left-hand edge of the notch are shown to have been removed from this region. Immediately ahead of the notch, there is

still clearly grain boundary hydride precipitation, but this is unlikely to produce any unwanted effects during thermomechanical testing.

7.3 Results and discussion

7.3.1 Room temperature short fatigue crack growth

7.3.1.1 Crack propagation mechanisms

In this section, the results of isothermal (room temperature) FCG testing are presented; as discussed previously, the thermal gradient heat treatment is not necessary for such cases. Figure 83 (a) is a polarised light micrograph showing the microstructure (sample H3) used in this study. Number annotations, 1 – 3, point to intergranular, twin, and intragranular hydride features, respectively. As per simulation studies of FCG in samples under 3-point bending in Chapters 4 and 5, cyclic $R = 0$ loading is applied here, with loading frequency 0.1 Hz, ensuring sufficient time to capture images for DIC. Figure 83 (b) shows the resultant crack path through sample H3 during testing (i.e., an image from the long-range optical microscope) after several hundred fatigue cycles. The primary Zircaloy-4 grain and hydride boundaries are overlaid with black and yellow lines, respectively.

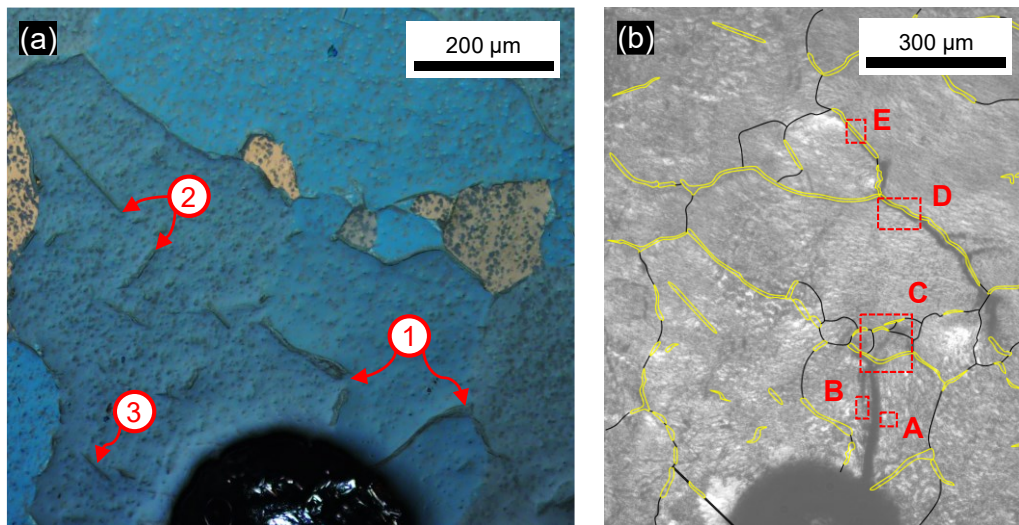


Figure 83: (a) Microstructure H3 with various hydride features: (1) grain boundary (intergranular) hydrides, (2) intragranular hydrides, and (3) twin boundary hydrides. (b) Snapshot from DIC microscope of sample H3 after several hundred $R = 0$ fatigue cycles and substantial crack propagation; a fatigue crack is shown to emanate from the notch, while a separate grain boundary hydride driven crack is also shown to have propagated. Regular grain and twin boundaries are shown using black lines, while yellow lines indicate hydride-matrix boundaries. Regions A – E will be shown subsequently in greater detail.

The crack path shown in Figure 83 (b) is unlike any published crack path in non-hydrided zirconium alloys in that it has two main sections: (i) that which nucleated at the notch and

propagated through the microstructure (in the usual manner) and (ii) the double-ended crack which nucleated near-instantaneously (within the first 5 fatigue cycles) along a grain boundary hydride feature at region D, despite it being reasonably far removed from the notch. Throughout the test, cracks (i) and (ii) propagate independently of one another, before eventually linking together, leading to detrimental sample failure. Surprisingly, the crack which nucleated at the notch did so within the middle of a grain which is bounded by hydrides at either end – there may be substructural reasons for this, particularly since grains at notches and other surfaces are often much smaller than those within the centre of each sample. The seemingly random nucleation of a crack at region D highlights the undesirable effect of uncertainty in structural integrity due to the presence of brittle hydride features.

Figure 84 to Figure 86 present post-deformation electron micrographs of subregions A to E (see Figure 83) to aid in understanding the various fatigue (and static) crack propagation mechanisms in Zircaloy-4. Figure 84 shows details of crack (i) within the first grain before (region A) and after (region B) a change in crack direction.

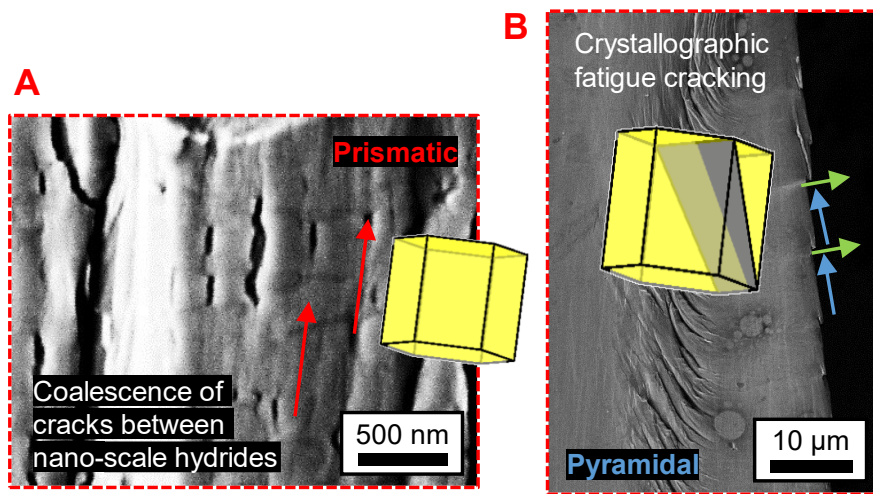


Figure 84: Regions A and B from sample H3. Region A is adjacent to the primary crack and shows multiple prismatic microcracks which are parallel to the primary crack growth direction. Cracks are shown to form between horizontal (basal) hydride platelets and appear to coalesce. Region B is within the same grain, after a slight crack path direction change (which also appears crystallographic).

The electron micrograph of region A in Figure 84 shows multiple parallel sub-micron prismatic plane cracks (which are also parallel with the primary crack growth direction this region). The shortest crack sections are observed to nucleate between hydride platelets (darker, near-horizontal features), which indicates that the nano to sub-micron hydride platelets are responsible for driving prismatic slip localisation and hence, the nucleation of crystallographic

facets. There are also examples of these facets extending through and beyond the brittle hydride platelets, which is likely to affect FCG rate. Moreover, region B, shown in Figure 84, highlights that after a change of crack direction, the crack path remains dominated by crystallography, i.e., slip plane orientation, as indicated by blue arrows, and pyramidal slip plane shown. The short crack segments corresponding to green arrows however appear non-crystallographic. Nevertheless, it is important to note that observations of intragranular FCG along prismatic and pyramidal planes within a grain of this orientation are consistent with those within non-hydrided Zircaloy-4 counterparts (despite interactions with intragranular hydrides, which are shown here).

Region C is microstructurally complex. It consists of several interconnected sections of grain boundary and triple junction hydrides around multiple matrix grains. The snapshot to the left of Figure 85 shows that once the intragranular notch crack (regions A and B) reaches region C, the crack path becomes dominated by grain boundaries, which is entirely atypical for non-hydrided zirconium alloys [252], [362]. This is however consistent with Garlea et al.'s observations [349]. It is shown to first propagate vertically along a grain boundary hydride between two triple junctions, before veering rightward along subsequent grain boundary hydrides; an increased magnification micrograph of one such region is shown in the right-hand side of Figure 85.

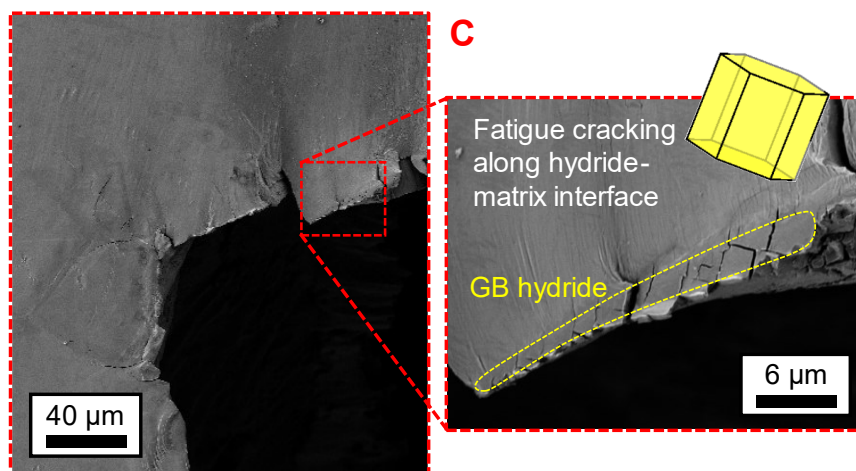


Figure 85: Region C from microstructure H3. A magnified view shows that the crack has propagated along grain boundary hydrides. Parallel microcracks are visible throughout the grain boundary hydride shown.

Here, the primary crack path is shown to have propagated non-uniformly to one side of the grain boundary hydride (which is outlined in yellow). The various parallel, cleavage-like sub-cracks throughout the grain boundary hydride appear to have strong interactions with the

primary crack and indicate that the overall crack propagation direction is heavily influenced by this sub-cracking. The crack propagation mechanism, therefore, is highly complex (as compared with intragranular crystallographic fatigue cracking or matrix-hydride decohesion). Furthermore, however, evidence of hydride-matrix decohesion can be found in sample H3, examples of which are shown in Figure 86. As discussed previously, region D corresponds to a location for premature cracking (within the first 5 cycles). Post-deformation examination of the crack path in Figure 86 (region D) indicates that interface decohesion is likely to have had a role in this. However, due to non-availability of the opposing crack face (due to severe deformation), it is not clear whether hydride-hydride interfaces or hydride-matrix interfaces are implicated.

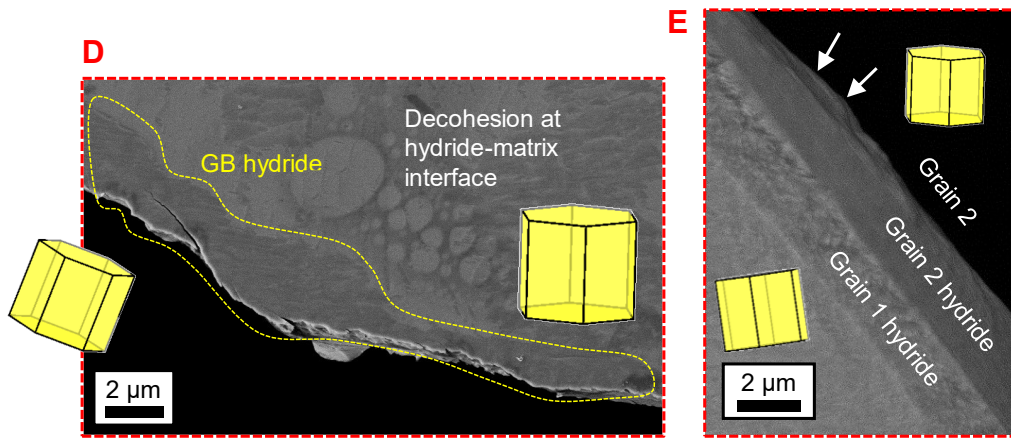


Figure 86: Regions D and E from microstructure H3. Both regions are far removed from the primary notch crack and show examples of grain boundary hydride cracking which are apparently interfacial. In region E, due to differences in contrast, it appears that there are two different hydride orientations present, corresponding to different matrix orientations.

For reasons which will subsequently be discussed in the context of region E, it is reasonable to assume the former, since (i) different grain orientations result in different hydride variant orientations which exhibit detectable differences in contrast using electron microscopy, i.e., since only one matrix hydride type is distinguishable in Figure 86, it is likely that the neighbouring grain's hydride remains attached to it, and (ii) the somewhat jagged crack face in region D may be indicative of tortuous decohesion between up to four hydride twin variants [146] at the original matrix boundary. A more distinct example of decohesion is shown at region E (see Figure 86). Region E corresponds to an upper section of the crack emanating from region D and is along a grain boundary hydride feature. Here, it is easy to distinguish between the original grain (Grain 1), and the hydride variants corresponding to Grains 1 and

2, as annotated. Unlike the crack path in region D, this is a clear case of decohesion along the interface of Grain 2 and its corresponding boundary hydride in region E, producing a comparably uniform crack surface. Examination of Grain 2's crack surface revealed no boundary hydride presence.

7.3.1.2 Crack growth rates

Having first discussed the various cracking mechanisms in sample H3, it is now appropriate to study FCG rates. Figure 87 shows how the rate of crack growth (for the notch crack only) evolves with increasing crack length. Above the graph, the corresponding 350 μm region of microstructure is presented with primary crack path(s) overlaid in purple. As before, matrix-matrix and hydride-matrix boundaries are also overlaid using black and yellow lines, respectively. Vertical red lines correspond to points at which sudden changes in crack propagation rate and important microstructure interactions occur.

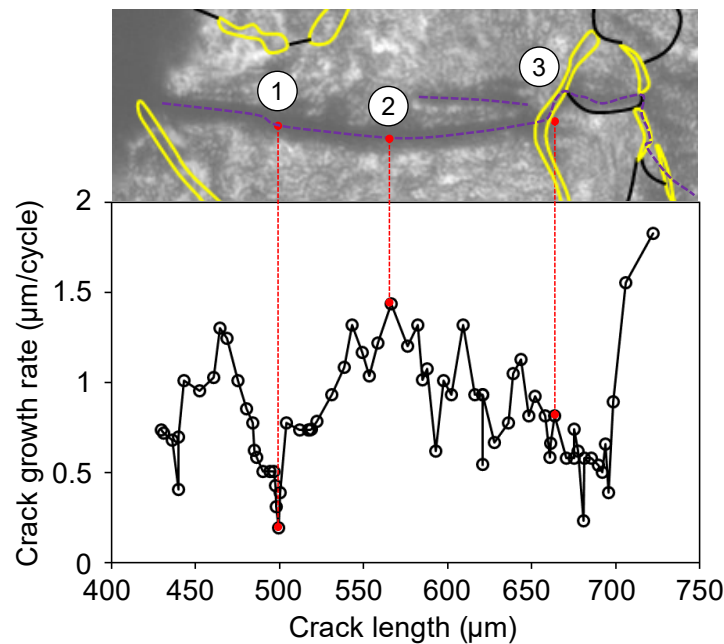


Figure 87: Projected crack length versus FCG rate of primary (notch) crack in sample H3. The primary crack path is shown above and is highlighted using a purple line. Vertical red lines correspond to important points (1 – 3) of crack growth including sudden changes in FCG rate, changes in crack growth direction, and interactions with microstructural features.

Figure 87 highlights the substantial crack growth rate variability through sample H3. Initially, ahead of the notch, there is a rapid growth rate fluctuation (rise and fall between 425 and 500 μm). As the growth rate reaches a minimum at a crack length of 500 μm , the red line shows that this point corresponds to the end of at least two dramatic changes in crack propagation direction. Chapter 4 (which relates to non-hydrated materials) showed that sudden changes in

crack propagation direction can lead to substantial FCG rate reductions, which could therefore help to explain this sudden fluctuation. The various in-plane DIC-based strain fields are shown at this stage of crack growth (Location 1) at the left of Figure 88. Beyond this point (within the zone corresponding to region A from before), the rate of FCG increases sharply (from 0.2 to 0.75 $\mu\text{m}/\text{cycle}$), and then continues to increase steadily to a local maximum value of around 1.5 $\mu\text{m}/\text{cycle}$ at Location 2. The sudden initial increase in growth rate could be attributed to a restoration to a level commensurate with the applied SIF following the period of tortuosity. Given the relatively straight crack path and lack of microstructural obstacles in the region thereafter, it is unsurprising that the FCG rate continues to rise. Moreover, the steep rate at which FCG rate rises could be attributed to (i) coalescence of a series of hydride platelet fractures driven by enhanced prismatic slip localisation, and (ii) an increasing level of interaction with the separately growing upper grain boundary hydride crack, i.e. crack which nucleated at region D. Evidence for (ii) is contained within the central set of DIC strain maps (particularly for ϵ_{xx}) in Figure 88.

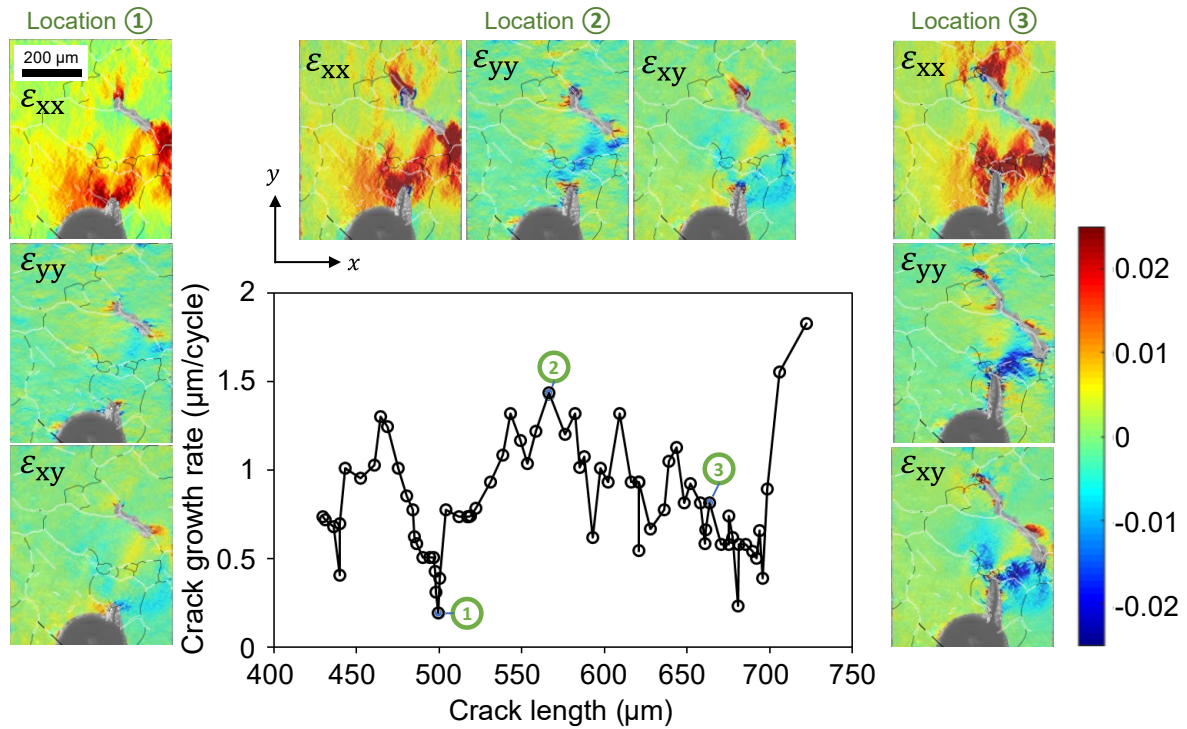


Figure 88: DIC total strain maps at key Locations (1, 2, 3) of FCG. All maps shown correspond to the peak load of a given fatigue cycle. Normal and shear strain plots are shown.

Between Locations 2 and 3 of crack growth, there is a steady FCG rate decline, with several minor undulations between. While the previous subsection of this chapter showed that between regions A and B, there is some shift in the overall crack propagation direction, this is not sufficient to explain this progressive FCG rate decline. However, the crack path outlined in

Figure 87 shows that between Locations 2 and 3, there is the seemingly random formation of an additional crack, which propagates almost parallel-wise with the primary crack. It is assumed that this crack originates from the facet coalescence mechanism discussed earlier, i.e., the localisation of strain at hydride platelet interfaces gives rise to the nucleation of new cracks. Hence, the reduction in FCG rate can at least partly be explained by the dispersion/dissipation of the primary crack tip driving force due to the formation of these new cracks. Moreover, beyond Location 3, as the crack propagates through region C (along grain boundaries etc.) the FCG rate declines even further (to a value commensurate with a crack length which is 200 μm shorter). Following from this proposed energy dissipation mechanism, it is assumed that the observed substantial crack branch formation associated with grain boundary hydride cracking (i.e., within region C) is also responsible for strain energy dissipation, and hence FCG rate reduction. The maximum shear strain plots shown in Figure 89 reinforce this theory. Specifically, when FCG rate is maximised (Figure 89 (b)), there is a direct and strong interaction between the upper and lower crack tips, whereas when the notch crack propagates into region C (Figure 89 (c)), the strain field is far more diffuse and effectively orthogonal to the primary crack growth direction. An electron micrograph of region C is also presented in Figure 89 (c) which shows significant crack branching ahead of the crack tip at this point. Upon traversing region C, crack growth accelerates enormously due to its proximity to the hydride boundary crack; beyond this, the sample failed completely.

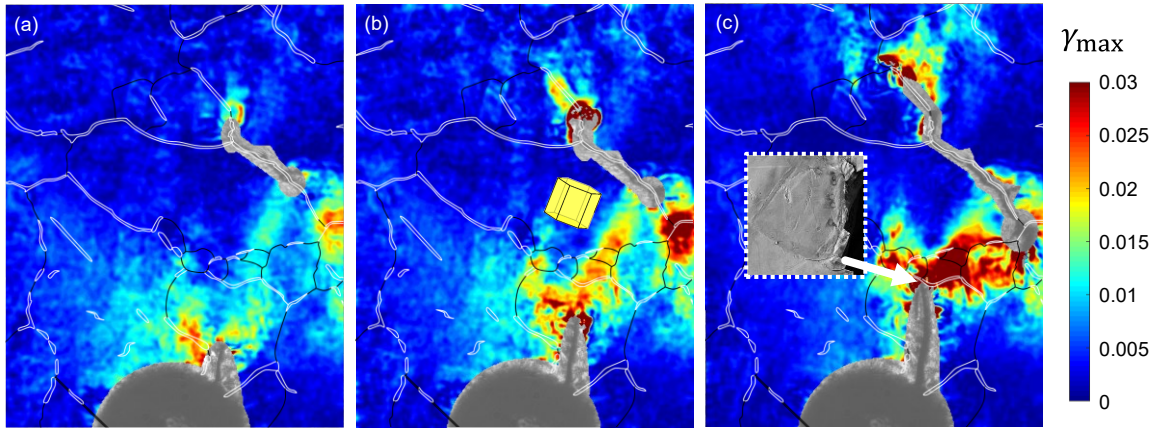


Figure 89: DIC maximum shear strain plots at key points ((a) 1, (b) 2, (c) 3) of FCG. Subfigure (c) shows an SEM image of the region of microstructure ahead of the crack tip, post-deformation showing crack branching throughout grain boundary hydrides.

Lastly, the FCG rate data from this study is compared with published experimental data from a non-hydrided Zircaloy-4 sample with similar underlying texture [76] in Figure 90. SIF range is used here for comparison to account for differences in applied load and sample geometry,

etc. While the SIF range applied to sample H3 is mostly higher than that of its non-hydrided counterpart, there is some overlap in the range 17 – 21 MPa√m. The data show that on average, FCG rates are twice as high in the hydrided case, despite also showing the greatest degree of rate variability, for reasons outlined earlier. At one point, FCG rate in the hydrided sample drops to a point which is twice as low as that of the non-hydrided case (corresponding to Location 1 from before) due to crack path oscillations. At around 22 MPa√m, the hydrided case FCG rate is also reduced significantly due to energy dissipation linked with additional crack nucleation and crack branching at grain boundary hydrides. In general, it is observed that intragranular and intergranular hydrides contribute to the respective acceleration and deceleration of FCG rates.

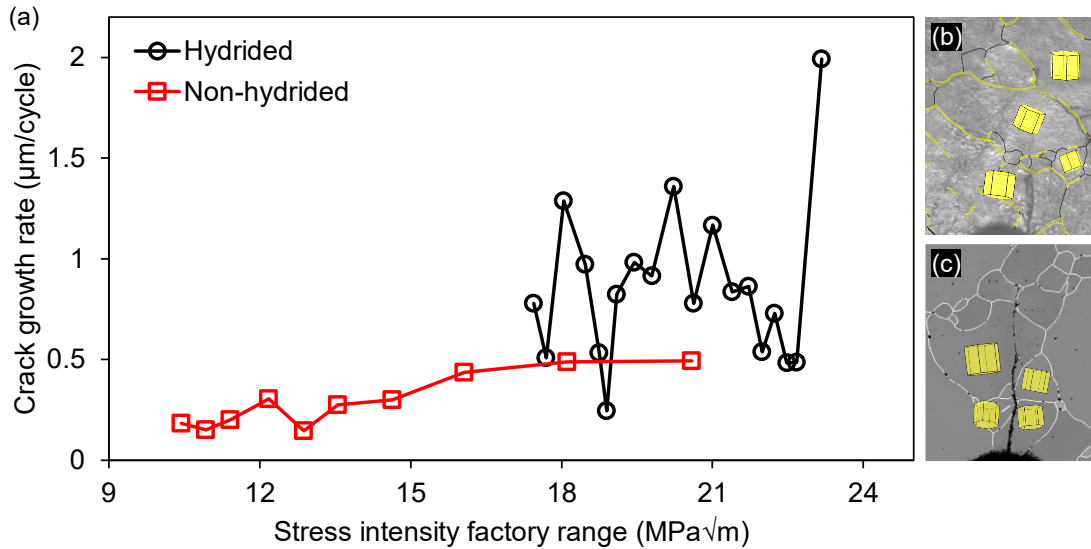


Figure 90: Comparison of FCG rates in hydrided (sample H3) and non-hydrided [76] Zircaloy-4 samples via SIF range (crack length is arbitrary when considering differences in applied load and geometry). For the non-hydrided case, the 4-point moving average of the raw data is used for clarity. (b) Sample H3 microstructure. (c) Non-hydrided sample microstructure [76].

It is entirely conceivable therefore that in as-received microstructures (consisting of grains of around 10 μm diameter), hydrides may provide the beneficial effect of an overall FCG rate reduction due to increased grain boundary hydride precipitation [136], [144].

7.3.2 Influence of annealing twin features on hydride precipitation and fatigue

To gather supplementary short FCG data across lower SIF ranges (beginning at 10 MPa√m), a repeat experiment is carried out using sample H4, which is shown in Figure 91 (a). An interesting difference between samples H4 and H3 is the notably increased proportion of annealing twin features in sample H4 and hydride precipitation therein. This is due to the

mean grain size in sample H4 being substantially larger than that of sample H3 (since increased grain growth corresponds to increased grain boundary migration, which promotes the diffusional growth of annealing twins [363]). After approximately 890 fatigue cycles with an applied notch (crack) tip SIF range of 10 MPa $\sqrt{m}$, the sudden nucleation and extension of near-mm length cracks through several of these hydrided twin features is observed, as shown in Figure 91 (b). First, the leftward extension of the left-hand crack through the first hydrided twin from near the notch led directly to another such feature in a neighbouring grain through which it continued to propagate (until reaching the end of the twin), all within a single cycle. After 23 subsequent fatigue cycles, a virtually symmetric event occurred through an additional set of hydrided twin features. The red boxes labelled 1 – 5 in Figure 91 (b), which track the crack paths, are shown in greater detail in optical micrographs presented in subfigures (c) to (g).

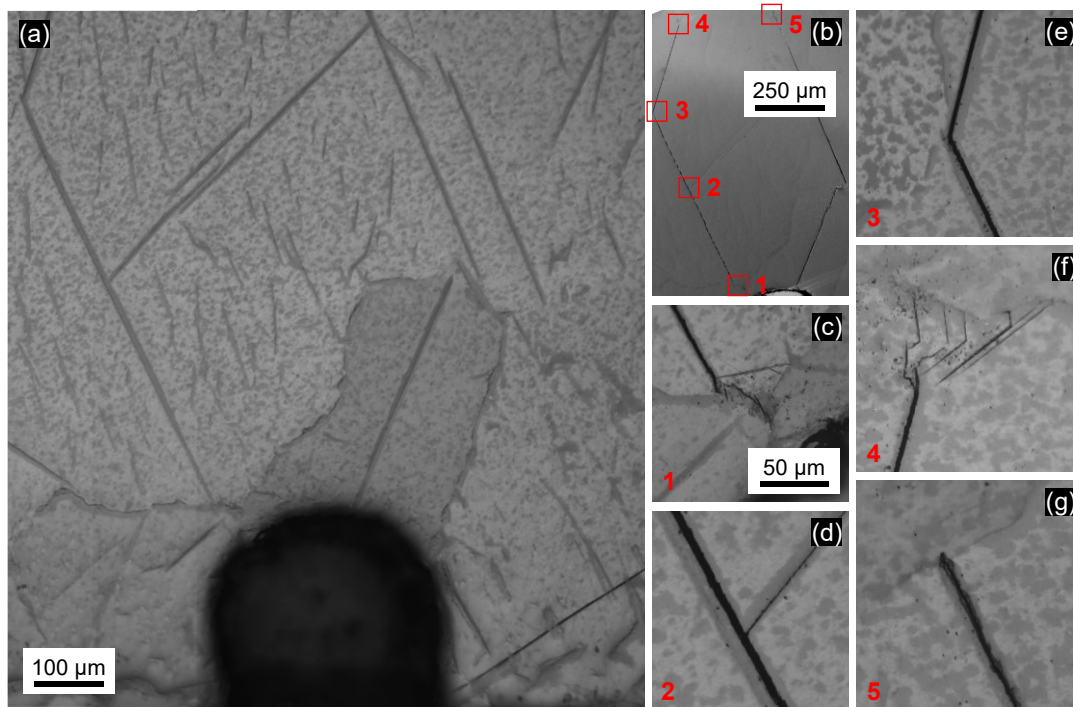


Figure 91: (a) Sample H4 microstructure prior to room temperature fatigue testing and (b) after approximately 890 fatigue cycles, showing mm-scale cracks which suddenly traversed the sample along twin hydrides. Subfigures (c) to (g) are increased magnification images of the cracks, corresponding to locations 1 – 5. The scale bar in (c) also applies to subfigures (d) to (g).

Figure 91 (c) shows the nucleation site (region 1) of the first (left-hand) crack. The crack nucleated and propagated within and through a grain boundary hydride near the notch root before reaching the twin, at which point it is assumed to have advanced irreparably through the entire feature. The initial grain boundary hydride crack is consistent with published

observations of fatigue cracking in zirconium hydrides [154]. The right-hand crack nucleated within the hydrided twin at the notch. However, it is interesting to note that this occurred after the left-hand crack had propagated, which is likely to have had a substantial effect on the stress state. Hence, the evidence suggests that while hydride twin features are detrimental to structural integrity, they appear to require external stress concentration features (i.e., cracks nucleating in a more conventional manner) before manifesting themselves as such. Figure 91 (d) reveals that at location 2, an interconnecting hydrided twin feature was also fractured, though not to the same extent, as it terminated shortly thereafter. Similarly, Figure 91 (e) shows the sharp crack path transition between one hydrided twin feature and another. Figure 91 (f) and (g) show the ends of the left-hand and right-hand cracks, respectively. In each case, the cracks are shown to terminate exactly at the end of the respective twin features. The left-hand crack resulted in the formation of several slip traces and cross-slip at its tip. The right-hand crack tip also coincides with where the twin connects with a grain boundary. Overall, an important observation is that each hydrided twin crack is shown to propagate through the twinned region, as opposed to cracking facilitated by matrix-hydride decohesion. For improved understanding of this detrimental failure mechanism, which was not previously reported, post-deformation EBSD analysis is used in two locations (shown here in Figure 92 and Figure 93) to reveal the crack location with respect to the underlying microstructure.

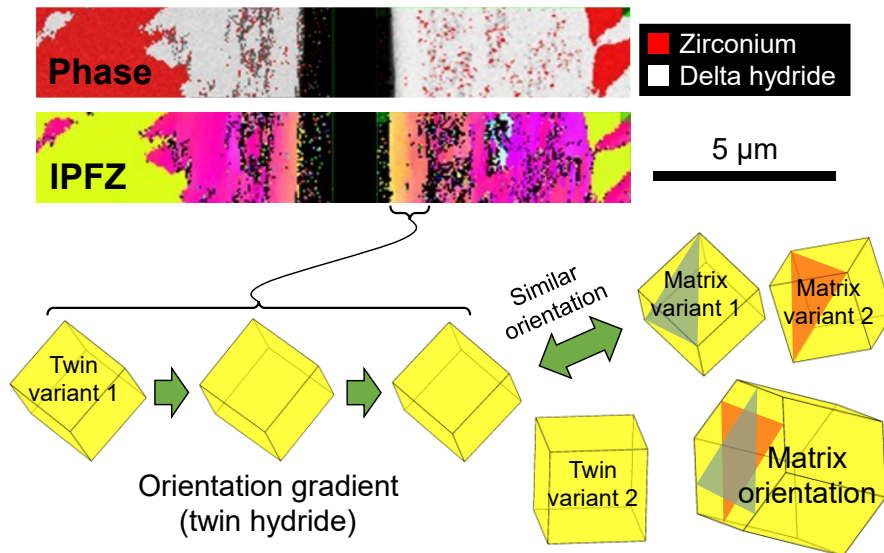


Figure 92: EBSD phase and orientation data around a crack (between locations 2 and 3 from Figure 91 (b)) in sample H4. The annealing twin region is shown to have been completely subsumed by hydride precipitation. Four hydride orientations have been identified: two primary grain (matrix) variants and two variants corresponding to the annealing twin (for which orientation data is unavailable).

Figure 92 presents phase and orientation maps of a section of the crack between regions 2 and 3 (referring to Figure 91 (b)). The phase map shows that the crack effectively only traverses

hydrides, i.e., has no interaction with the zirconium matrix. Interestingly, since only the matrix grain orientation is detected, this indicates that the annealing twin has been entirely subsumed by hydride precipitation. The same is true of the EBSD data presented in Figure 93 (which is of the right-hand crack, marginally below region 5). In terms of delta hydrides, the data in Figure 92 show that the full set of four hydride orientation variants are detected (two sets of two variants each corresponding to the matrix grain and matrix twin orientations). Moreover, this important data reveals that both crack interfaces (either side of the crack) are lined by twin hydride variants, i.e., the crack is therefore propagating through the centre of the former twin region, rather than at the twin hydride-matrix hydride interface. This is also shown by the second EBSD dataset in Figure 93, for which there is just a single detectable twin hydride orientation variant, which appears as turquoise in the x-direction inverse pole figure (IPF) orientation maps. The matrix hydride orientation variants are represented by green and pink in the IPFX maps, which show that both variants are roughly equally represented in the typical boundary hydride manner [146]. Interestingly, the IPFZ (z-direction) map in Figure 92 shows that there is a very clear hydride orientation gradient at either side of the crack. The gradient varies from an apparent twin orientation at the crack face to an orientation which is close to one of the matrix variants, and is likely to have developed during precipitation. The orientation gradient may also be indicative of the release of energy originally stored in lattice curvature, i.e., GND structure.

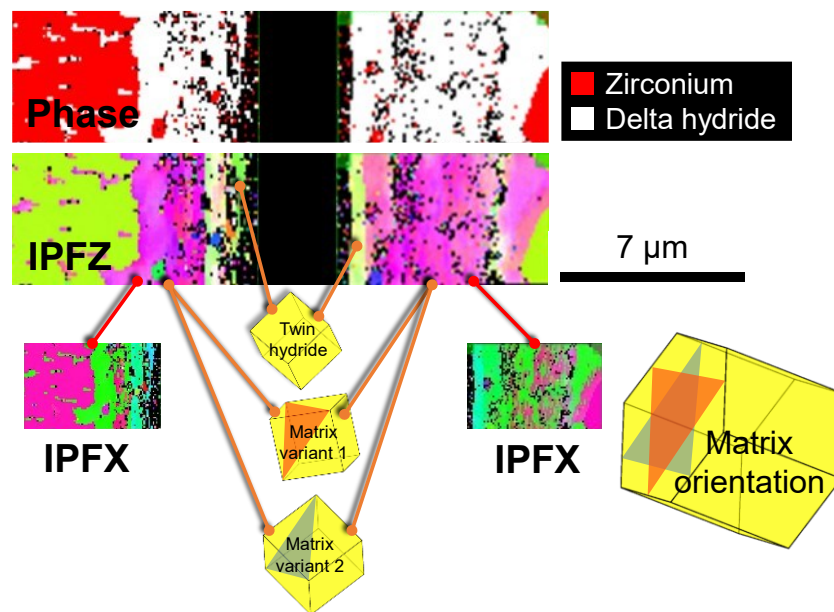


Figure 93: EBSD phase and orientation data around a crack (along crack path below location 5 from Figure 91 (b)) in sample H4. As in the previously indexed region, the twin orientation is unidentifiable due to being fully hydrided. Matrix hydride variants are identified, as well as a single twin hydride orientation.

The consistency and rate with which cracks are shown to propagate through multiple distinct hydrided annealing twin features indicate that there is some unifying mechanism responsible for this behaviour which has not yet been discussed in the literature. Hence, based on the data and observations discussed earlier, a hypothesis for the mechanism is proposed here. As outlined in Figure 94, for a section of material consisting of an annealing twin sandwiched between two sections of its matrix grain, the chemical potential along either twin-grain interface should be equal and be low enough to preferentially sequester mobile hydrogen atoms from the surrounding region at elevated temperatures. During cooling, the onset of hydride precipitation should therefore occur simultaneously along the opposing interfaces which contain the highest levels of hydrogen. As illustrated in Figure 94, hydride growth could then occur in up to four directions, i.e., along chemical potential gradients, resulting in inward twin hydride growth (variants δ_3 and/or δ_4) from either direction, and outward grain/matrix hydride growth (variants δ_1 and/or δ_2) in each direction.

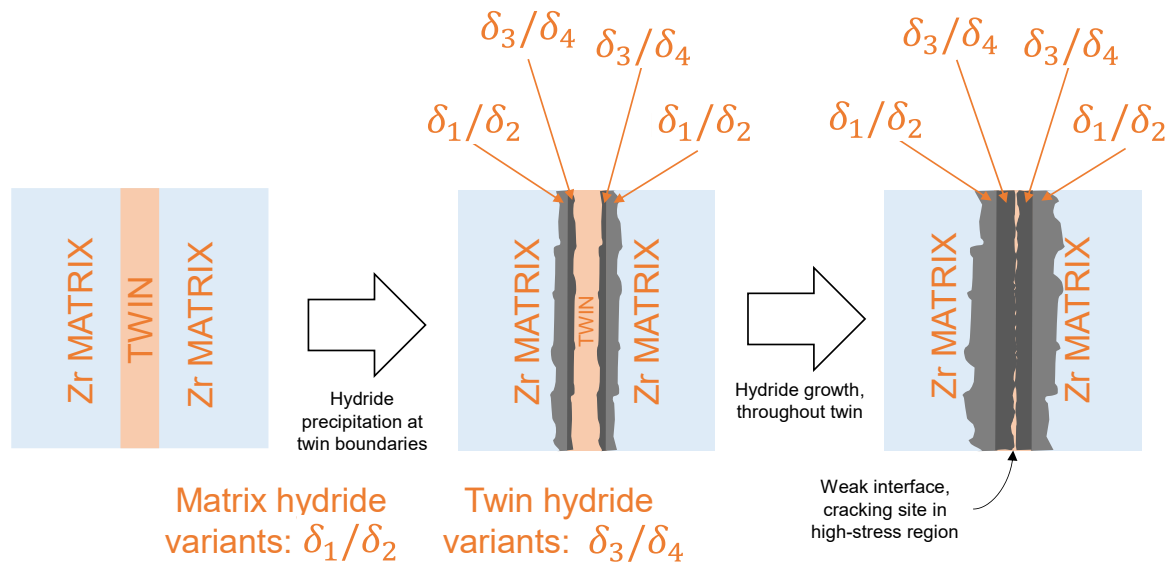


Figure 94: Hypothesis for crack propagation mechanism linked with formation of hydrides through annealing twin features.

Hence, the hypothesis proposed here is that at the centre of a twin which has undergone full hydride precipitation, there is some interface/region between hydrides which nucleated at either side of the twin. Owing to the non-uniform rate of hydride growth (as demonstrated by the undulating hydride-matrix interfaces in Figure 92 and Figure 93), the interface at the centre of the twin is also likely to be non-uniform, potentially consisting of packets of non-hydrided zirconium. This, in combination with the volumetric expansion associated with hydride precipitation is likely to lead to highly localised deformation (which is also suggested

by localised lattice rotation shown in Figure 92) and decohesion at the centre of the twin prior to the application of any external load, producing a severely weakened interface which is susceptible to cracking under the appropriate conditions. To verify this hypothesis, further work will be required to assess the post-precipitation deformation state using advanced characterisation.

7.3.3 Hydride ratcheting effects during thermomechanical fatigue

7.3.3.1 DIC measurement limitations

Because of the obfuscation of much of the free surface of sample H2 after thermal load (due to oxidation), DIC data is limited to regions in which the speckle pattern has remained detectable. Some image post-processing has been used to increase contrast and clarity for improved displacement measurement quality. Moreover, measurements of strain have been limited to the end of each thermomechanical cycle, i.e., residual strains, due to difficulty in obtaining subset pattern matches between the initial (room temperature) reference image and mid-test (high temperature) images because of intense illumination of the sample from the IR lamps. Consequently, total (elastic + plastic) strain and hence stress measurements are unattainable in the current study.

7.3.3.2 Hydride precipitation strains and orientation dependence

In this section, the issue of hydride strain ratcheting is investigated via thermomechanical loading of hydrided Zircaloy-4. Several thermomechanical cycles (the details of which will be discussed shortly) are applied to sample H2, the microstructure of which is shown in its pre-hydrided state in Figure 95 (a), with details of its crystallography (obtained using EBSD) given in Figure 95 (b).

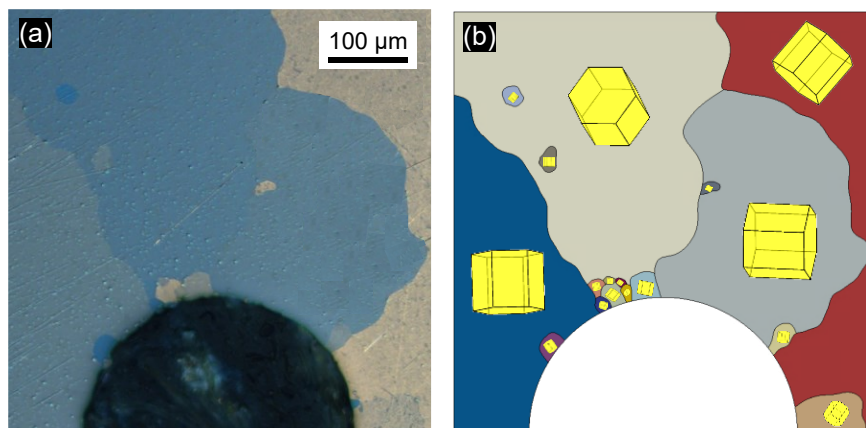


Figure 95: Sample H2 microstructure. (a) Polarised light micrograph around notch. (b) Graphical representation of region shown in (a) based on EBSD data.

Sample H2 was first prepared for testing using the thermal gradient heat treatment discussed at the beginning of this chapter. This first subsection is focused on interesting DIC-based strain measurements around an intragranular hydride stringer taken after the first thermomechanical cycle (in the unloaded state and at room temperature). The location of the hydride is shown in the micrograph at the left of Figure 96; it is reasonably far removed from the notch tip, and hence, any measurements of residual strain are assumed to be dominated by precipitation. Prior to the first cycle, there were no hydrides in this location. Focusing firstly on the set of in-plane strain distributions at the top of Figure 96, ϵ_{xx} , ϵ_{yy} , and ϵ_{xy} , hydride precipitation is shown to result in strain localisation at the centre of the hydride, the outline of which is overlaid on each image. Normal strains, ϵ_{xx} and ϵ_{yy} are shown to localise towards the top and bottom of the hydride, respectively, while the shear strain distribution is reasonably uniform. The maximum strain magnitude in any direction is around 2.5%.

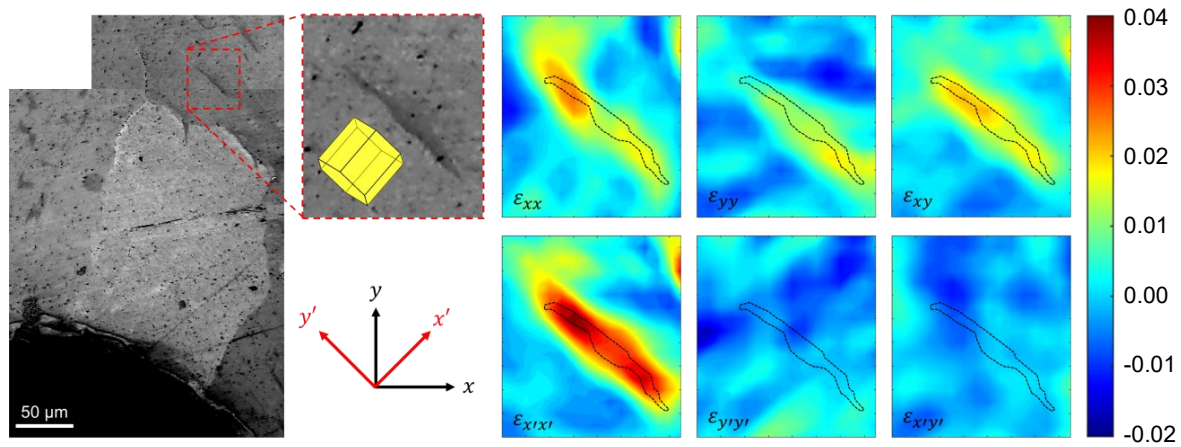


Figure 96: DIC-measured residual strain fields around a new hydride feature in sample H2 after a single thermomechanical cycle.

However, since delta hydride precipitation has a known morphological and crystal orientation dependence, the set of strain distributions shown at the bottom of Figure 96 correspond to the same dataset within a new frame of reference, $x' - y'$, such that the x' direction is orthogonal to the hydride stringer feature (i.e., approximately orthogonal to the basal plane), and the y' direction is parallel to it. The resultant strains, $\epsilon_{x'x'}$, $\epsilon_{y'y'}$, and $\epsilon_{x'y'}$ yield profoundly more interesting results, showing extreme localisation within the hydride (of up to 4%) in the x' direction, which is maximised near either hydride end. Strikingly, the $\epsilon_{y'y'}$ strain distribution shows virtually no correlation with the hydride location, despite the corresponding theoretical volumetric misfit strain for precipitation of 4.58% within the basal plane. Similarly, there is no clear correlation between the rotated shear strain field and hydride location, which is

unsurprising. Carpenter’s [364] theoretical volumetric misfit strain tensor for precipitation of a delta hydride platelet, i.e., with perfect stoichiometry, is given by Equation 7.5,

$$\boldsymbol{\varepsilon}_{\text{vm}} = \begin{bmatrix} 0.0458 & 0 & 0 \\ 0 & 0.0458 & 0 \\ 0 & 0 & 0.072 \end{bmatrix} \quad (7.5)$$

where the first and second normal components correspond to the $\langle a \rangle$ (basal plane) directions and the third normal component corresponds to the HCP c-axis direction. It is important to note that this strain tensor is based on a single hydride platelet, and hence is likely to differ somewhat from the strains which manifest from the precipitation of an entire hydride stringer (consisting of a series of platelets stacked in the *deck of cards* configuration).

7.3.3.3 Thermomechanical loading conditions and crystal plasticity finite element modelling

In this section, a CPFE-based modelling methodology is developed for quantitative assessment of the influence of hydrides on the cyclic accumulation of strain during thermomechanical loading. The finite element model, which is outlined in Figure 97, shows that ahead of the notch, a CPFE region of interest is defined containing the microstructure of sample H2 in its pre-hydrided state. Hence, in combination with experimental DIC strain measurements of the same microstructure in the hydrided state, subjecting the model to the same external environmental conditions will provide a like-for-like comparison, such that hydride strain ratcheting effects can be appropriately quantified. As with the CPFE modelling work presented in Chapter 4, the beam is represented by partial extrusion of its outer profile and notch microstructure. As indicated by Figure 97 (a), the region outside of the crystal plasticity region is modelled as fully elastic, with elastic properties corresponding to a random crystal orientation (since the microstructure in sample H2 is not very strongly textured). Figure 97 (c) shows a set of three thermomechanical cycles, which are applied to sample H2 (experimentally and via the model). The thermomechanical cycle used here is loosely based on real reactor operation conditions and was previously published by Liu et al. [318]. The applied load is selected to be sufficient to generate a stress of 500 MPa (at room temperature) at the notch root [358], which is first determined using the model. The load is initially increased linearly over time, assuming a linear stress rate of 1 MPa/s, followed by a dwell period, and unloading. The temperature cycle consists of a linear ramp through time, followed by two successive dwell and cooling periods. The maximum temperature of 340 °C is achieved over the same period taken to reach the maximum applied load, which is followed by a 3-hour dwell.

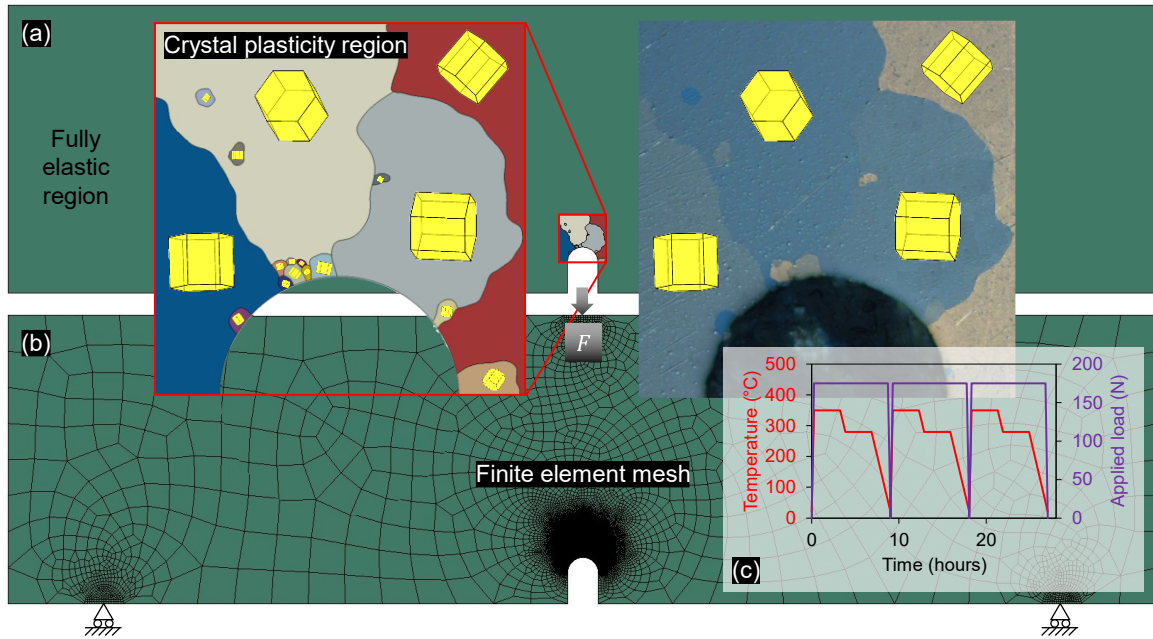


Figure 97: CPFE modelling representation of sample H2 in its non-hydrided state for isolation and quantification of hydride ratcheting effects through comparison with experiments.

At this temperature (and bulk hydrogen content of 200 wppm), most of the hydrogen is brought into solution. The temperature and load dwell periods allow for stress-driven hydrogen diffusion to occur. After the dwell, the first cooling stage occurs at 120 °C/hour until reaching 280 °C. Where stress-driven diffusion has occurred, the local hydrogen content in some locations, e.g., at grain boundaries, will be sufficient to precipitate hydrides, while in regions with lower hydrogen content, the hydrogen remains in solution. Based on the bulk hydrogen content, the 280 °C dwell temperature is between the temperature for TSSP and TSSD. The second 3-hour dwell period therefore facilitates additional steady-state stress-driven diffusion (of soluble hydrogen) and hydride precipitation. Lastly, the final cooling stage of each cycle will result in the precipitation of virtually all remaining hydrogen and is likely the most dominant source for intragranular hydride precipitation.

7.3.3.4 Simulated volumetric misfit strains

In this section, the CPFE model is utilised to gain mechanistic insights into the DIC strain data presented earlier. Specifically, the counterintuitive result in which the rotated strain maps show substantial localisation orthogonal to the intragranular hydride stringer, and virtually no localisation parallel-wise, is investigated. In the first instance, a simplistic representation of the stringer feature is generated via a line of finite element nodes at the same location as observed experimentally (see Figure 98 (a)). At each node, the (rotated) volumetric misfit tensor given by Equation 7.5 is applied incrementally and linearly over a precipitation period,

which is assumed here to coincide with the entire second cooling period of the first thermomechanical cycle. Within the user-material subroutine code, the volumetric misfit strain increment, $\Delta\epsilon_{t+\Delta t}^{\text{vm}}$, is incorporated within the trial stress as an inelastic term, in the same way as the thermal strain increment, $\Delta\epsilon_{t+\Delta t}^{\text{th}}$, as shown in Equation 7.6.

$$\sigma^{\text{tr}} = D(\epsilon_t^e + \Delta\epsilon_{t+\Delta t} - \Delta\epsilon_{t+\Delta t}^{\text{th}} - \Delta\epsilon_{t+\Delta t}^{\text{vm}}) \quad (7.6)$$

The predicted strain distributions, within the conventional and rotated frames of reference, are shown in Figure 98 (b). Due to morphological simplicity and absence of hydride mechanical properties in the model, the experimental strain distributions are not captured precisely. However, the model does capture the most important effect, which is maximised strain localisation in the x' direction.

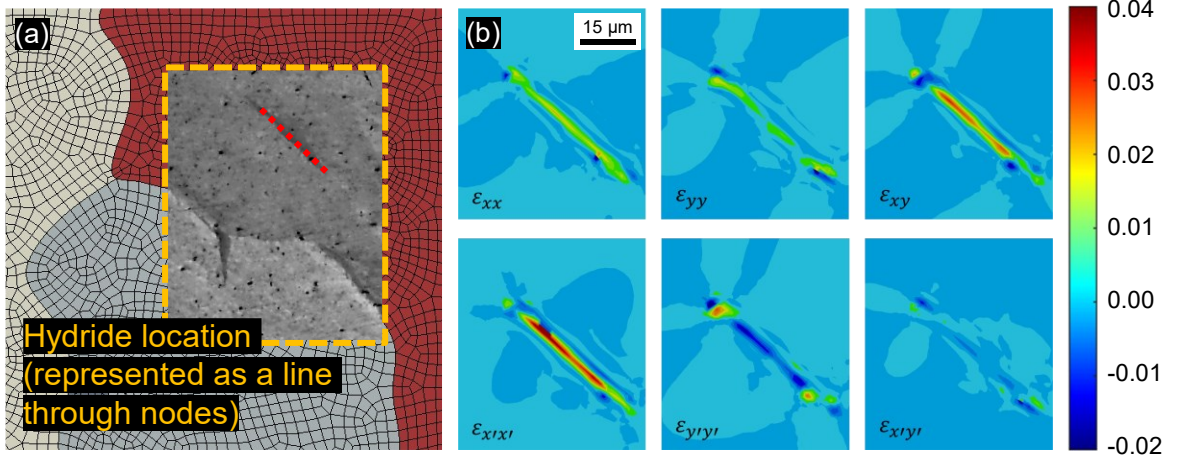


Figure 98: (a) Simple line-of-nodes representation of hydride of interest to understand the link between the combined hydride morphological and crystallographic (in terms of volumetric misfit only) orientation and the resultant precipitation strain fields. (b) Resultant strain distributions after single cycle precipitation. The volumetric misfit strain tensor is applied to each node in parallel, with strain increasing linearly over time from 0 at the beginning of the second cooling stage, and maximum at the end of the cycle.

Moreover, the model predicts an initially counterintuitive y' strain distribution, which is compressive throughout the hydride region, with compensatory tensile hot spots at either end. In this simplistic analysis, the key difference between strains in the x' and y' directions is that volumetric expansion in the x' is only (primarily) inhibited by the surrounding grain's resistance to deformation, while expansion in the y' direction is opposed by the simultaneous expansion of other 'points' along the hydride. Hence, the extreme case of y' expansion along the representative hydride line is counter effective and leads to compression (in this case) for purely morphological reasons. Hence, it can be deduced that for the less extreme example of

an actual hydride stringer, its morphology leads to the inhibition of volumetric expansion within the basal plane, with maximum straining along the HCP c-axis direction.

To study this effect in greater detail, the model is modified to account for the hydride's actual (2D) morphology and size. Figure 99 (a) shows this enhanced hydride representation, which is based on an optical micrograph of the hydride taken after the first thermomechanical cycle. In this unique and preliminary case study, the hydride is represented using only its anisotropic elastic properties, i.e., plastic deformation is unaccounted for during precipitation. Hence, prior to the hydride precipitation event, the region of material which is to be occupied by the hydride is initially prescribed the matrix orientation elastic properties (and is fully elastic). Based on post-test EBSD analysis, the measured hydride orientation (which satisfies the delta hydride orientation relationship) is utilised to calculate the resultant global compliance matrix, i.e., after precipitation. Hence, in parallel with the incremental application of the volumetric misfit strain over the entire hydride region, the compliance matrix is also set to evolve by linear interpolation between the temperature-sensitive Zircaloy-4 compliance matrix (no precipitation) and the room temperature hydride compliance matrix in the correct orientation, corresponding to full precipitation. Properties for the delta hydride compliance matrix were obtained from Olsson et al. [365].

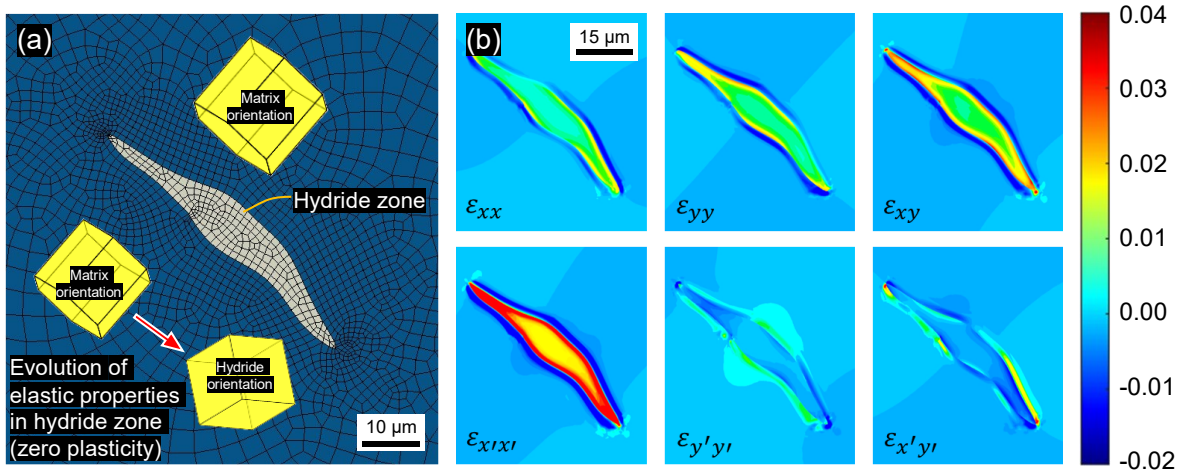


Figure 99: (a) Modified 3D finite element representation of hydride of interest, incorporating elastic properties which evolve linearly between those of the matrix orientation and of the hydride orientation, assuming the same precipitation period as above. (b) Resultant distributions of strain, with linear elastic hydride properties, and full matrix elastic-plastic deformation.

The strain field results from this model are presented in Figure 99 (b). Firstly, the results within the original $x - y$ reference frame show slightly under-predicted normal strains, while oddly capturing increased localisation of each component (x and y) at the bottom and top of

the hydride, respectively, as opposed to the experimentally observed localisation at the respective top and bottom. The shear (xy) component of strain is mostly localised near the hydride-matrix interface and at the ends of the hydride. In each case, substantial compressive strain localisation is predicted within the matrix, near the hydride-matrix interface. As shown by the rotated strain distributions, the interfacial matrix compressive strains are primarily along the x' direction. The x' strain distribution also shows that this is a countereffect of substantial tensile strain localisation within the hydride, near the interface. While the experimental x' strain distribution is not precisely captured by the model (which is likely due to not incorporating hydride plasticity), the important trends are captured, which include: (i) maximum strain localisation within the hydride in the x' direction, (ii) increased x' strain localisation near the hydride ends, (iii) virtually no discernible linkage between the y' and $x'y'$ strain distributions and the hydride itself, and (iv) asymmetric localisation of strain at opposing ends of the hydride in the x and y directions, respectively. The Von Mises stress distribution near the hydride tip, presented in Figure 100, can explain the discrepancies between experimental strain measurements and predicted strains based on a fully elastic hydride model.

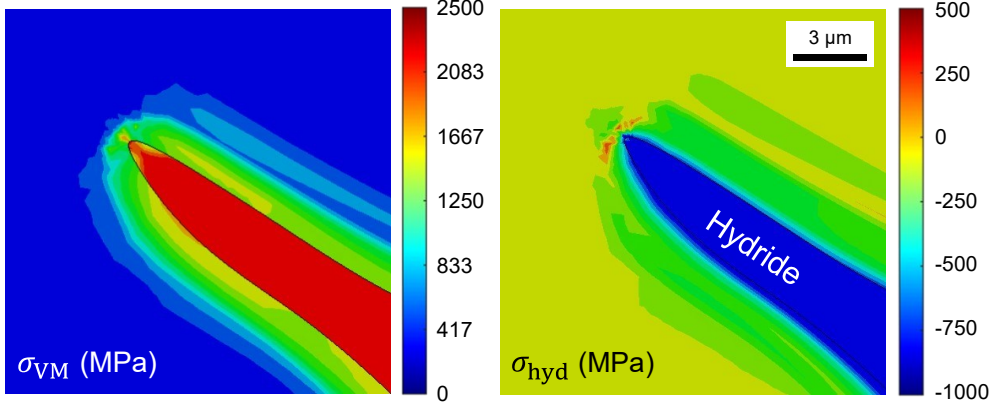


Figure 100: Von Mises (σ_{VM}) and hydrostatic (σ_{hyd}) stress distributions at hydride tip after precipitation. Due to volumetric expansion throughout the hydride, which is inelastic, the response of the surrounding matrix is to impose a compressive stress on the hydride [366]. Due to its purely elastic response in the current model, there is no stress relaxation within the hydride, resulting in extreme localisation of plastic strain within the surrounding matrix, and GPa-level compressive stresses within the hydride, as shown in Figure 100. Based on the uniform application of volumetric misfit (tensile) strain throughout the hydride, localisation of x' direction strain within the hydride, near the hydride-matrix interface, reflects the increased compressive elastic strain (and stress) towards the centre of the hydride, due to localised stress-

relaxation afforded by the matrix. Hence, with the incorporation of crystal-level hydride plasticity, this new modelling approach could potentially accurately capture the experimental DIC measurements and provide highly valuable insights into damage localisation during single and multi-cycle hydride precipitation. The hydride tip tensile hydrostatic stresses suggest that hydrogen redistribution and precipitation are likely to occur locally as a result of the initial precipitation.

7.3.3.5 *Cyclic accumulation of strain*

In this subsection, DIC data showing the cyclic accumulation of strain within sample M2 is compared quantitatively with CPFE-based predictions in the absence of hydrides. Figure 101 shows electron micrographs of a local region of interest ahead of the notch tip, consisting of several grain boundary hydride features after three thermomechanical cycles.

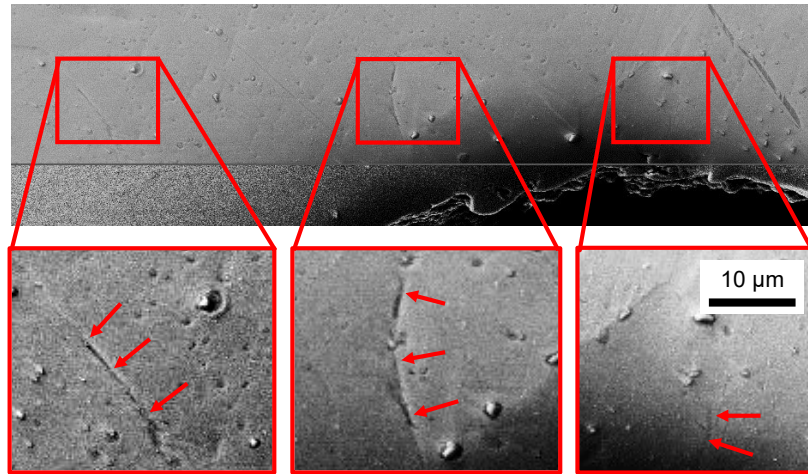


Figure 101: Electron micrographs showing grain boundary hydride precipitation at the notch after three thermomechanical cycles.

Path-based line plots of the residual strain distributions (with strain measured in the path direction) across this region of interest for the first three thermomechanical cycles are shown in red (with increasing transparency) in Figure 102 (a), with the corresponding measurement path shown above. Vertical red lines indicate the points of intersection between the measurement path and the three hydride locations shown in Figure 101. Figure 102 (a) also shows the equivalent non-hydrated CPFE-based strain distributions through the same region using black lines (with increased line transparency for increasing number of cycles). The measurement path is shown in the context of the CPFE-predicted residual strain distribution in Figure 102 (b), which also shows the underlying microstructure in this region in greater detail. From right to left along the measurement path, a very large grain is traversed before encountering strain localisation at the first grain boundary, followed by several much smaller

grains, within which, very little strain localisation is predicted, followed by crossing a slightly larger grain in which significant localisation is predicted, particularly near boundaries at either side, which is then followed by crossing the last grain boundary into another large grain.

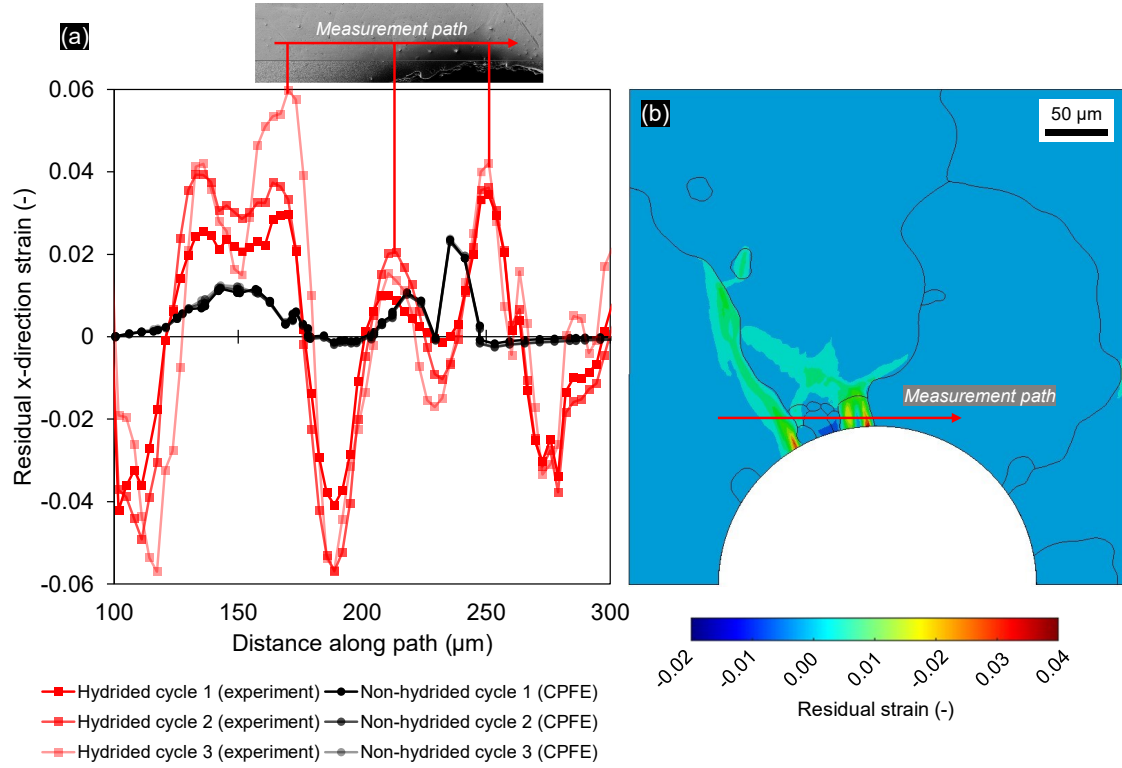


Figure 102: (a) Cyclic evolution of residual x -direction (horizontal) strain distribution along a path for three thermomechanical cycles, corresponding to the region of interest outlined by Figure 101 experimentally and from the CPFE model, which does not account for hydride precipitation effects. (b) Measurement path and resultant CPFE-predicted x -direction strain field after three cycles.

For the hydrided case, Figure 102 (a) shows that in regions where hydrides have been observed, residual strain is maximised (locally), and is shown to increase cyclically in the same locations, particularly in the left-hand most location, where the x -direction strain reaches up to 6% after the third cycle. Interestingly, the peaks at each of these three locations are rather close to strain peaks predicted by the non-hydrided CPFE model. A key difference, however, is that in the hydrided case, strain is maximised at the matrix boundaries (where hydride precipitation has occurred), while in the non-hydrided case, strain is maximised near the boundaries, within the softer grains, due to strain gradient (GND) hardening near the boundaries. Under the applied load (during the dwell period), most of the measurement path region is in tension, with tensile hydrostatic stress hot spots at grain boundaries, within ‘hard’ grains (hard referring to a grain with a greater c -axis component in the horizontal loading direction than its neighbouring grain). Since tensile hydrostatic stresses are known to promote hydrogen ingress, it follows that hydride precipitation would occur along each of the three grain

boundaries which are shown to promote the greatest levels of strain localisation. Another key difference between experimental and CPFE-predicted strain distributions are the substantial residual compressive strains which develop adjacent to regions showing highly localised tensile strains. This is a direct consequence of the volumetric expansion during precipitation, as compressive strains develop in the matrix to accommodate these changes. Hence, no such compressive residual strains are predicted by the CPFE model. This observation is consistent with the modelling results (for intragranular hydride precipitation) discussed earlier. Lastly, the experimental measurements of the cyclic accumulation of strain within tensile and compressive hot spots is not predicted by the CPFE model; only marginal strain accumulation is predicted. Hence, this helps to confirm that the strains accumulating within the hydrided sample are as a direct consequence of hydride dissolution and re-precipitation during each thermomechanical cycle. This is therefore the first known measurement of hydride strain ratcheting at the microstructural level.

The cyclic accumulation of x' direction strain and GNDs around the aforementioned intragranular hydride has also been measured and is presented in Figure 103 and (quantitatively) in Figure 104 for five thermomechanical cycles. Here, the GND density distributions are based entirely on gradients of plastic strain measured using DIC (as described in Chapter 3) and therefore, do not distinguish between volumetric expansion strains within the hydride and plastic strains within the hydride. Hence, measurements of GND density should only be considered from outside the hydride. In Figure 103, the outline of the initial intragranular hydride is shown for the first cycle, and in the fifth cycle, the final hydride is shown along with several smaller intragranular hydrides surrounding it. From Figure 103, a very clear cyclic strain ratcheting effect is shown. Initially, there is a band of tensile strain within the hydride, which is shown to increase cyclically, in parallel with the development of bands of compressive strain at either side of the hydride (which was previously predicted for a single cycle using the CPFE model). Using a measurement path crossing the centre of the hydride, Figure 104 (a) shows that the initial strain distribution is entirely tensile, with a maximum strain of around 4% at the centre of the hydride. Cyclically, this value is shown to increase by more than a factor of two after five cycles, at which point the strain ratcheting effects appear to saturate. The bands of compressive strain are shown to reach almost -6% within the surrounding matrix.

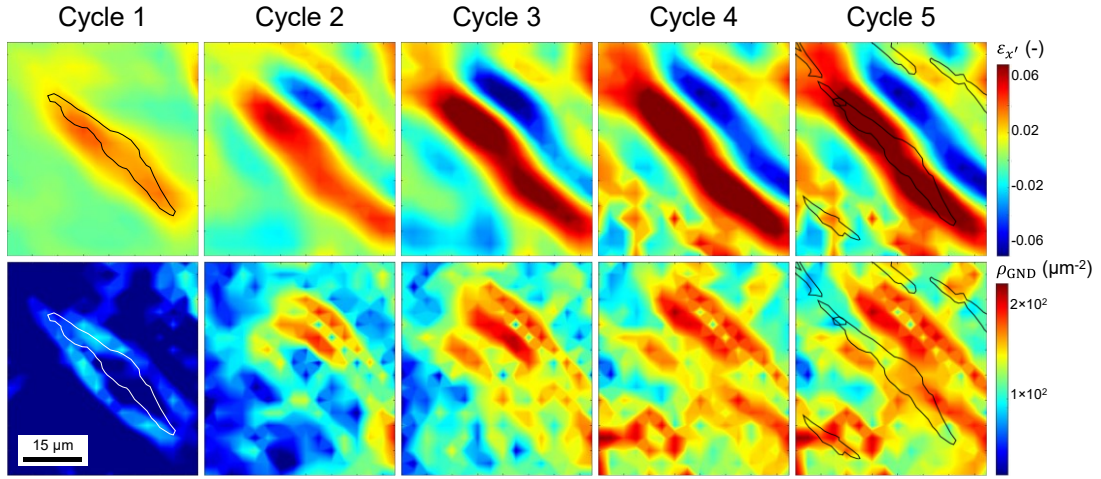


Figure 103: Cyclic evolution of $\epsilon_{x'}$ strain and GND density around the hydride of interest for five thermomechanical cycles.

For the first cycle, the distribution of GNDs is shown to surround the hydride, with maximum densities of up to $100 \mu\text{m}^{-2}$ (see Figure 103 and Figure 104 (b)). Subsequently however, the development of bands of compressive strain around the hydride is shown to generate strain gradients which make the GND distribution somewhat more perplexing. Nevertheless, the GND density is also shown to increase and (almost) achieve saturation after five thermomechanical cycles, with maximum values of around $200 \mu\text{m}^{-2}$. These levels are comparable with values predicted ahead of crack tips in Chapter 4, demonstrating the potentially highly damaging effects of hydride precipitation and ratcheting.

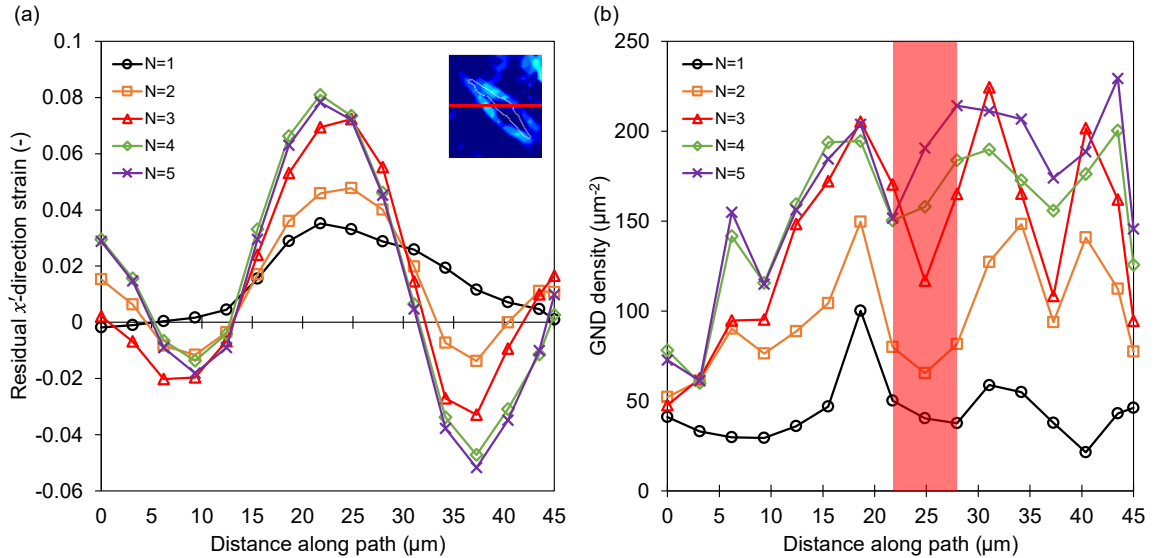


Figure 104: Path-based distributions of (a) $\epsilon_{x'}$ strain and (b) GND density over five thermomechanical cycles (hydride location marked in red). The path position is shown relative to the hydride in red in (a).

After the fifth cycle, the newly formed hydrides (which are overlaid in Figure 103) are potentially linked with the development of the GND network around the original hydride,

though there is no clear linkage between their locations and that of the GNDs. For future work, further CPFE model development, accounting for the development of plastic strains within the hydride, will be required to fully appreciate the new experimental data presented in this subsection.

7.4 Conclusions

Room temperature FCG and cyclic thermomechanical tests were carried out in notched and hydrided Zircaloy-4 samples with hydrogen contents of 200 wppm. FCG studies provided new mechanistic insights into how hydrides affect crack propagation mechanisms and ultimately, FCG rates. A variety of crack propagation mechanisms were observed at different length-scales: crystallographic slip-driven crack growth, fatigue cracking along matrix hydride interfaces, and decohesion at these interfaces. Crack growth rate trends are more stochastic in hydrided samples due to added complexity, but intragranular rates were found to be higher, which is potentially linked with facet formation and coalescence between nano-scale hydrides. It was demonstrated that grain boundary hydrides can reduce FCG rates due to strain field diffusion linked with crack branching, etc. It was also demonstrated that in samples containing large annealing twin features, hydride precipitation within and around the twins was common. Under cyclic loads, cracks which propagated into hydrided twins resulted in immediate and detrimental sample failure, leading to a new hypothesis for this previously unreported failure mechanism. For high temperature testing of notched and hydrided samples, a thermal gradient heat treatment device was developed to initially redistribute hydrides away from the notch, enabling gradual stress and concentration gradient driven hydrogen redistribution and precipitation under cyclic thermomechanical loads. Single cycle DIC-based residual strain measurements around a new intragranular hydride stringer revealed substantial precipitation strain orientation dependence. Specifically, the in-plane normal strain measured orthogonally to the stringer (along the HCP c-axis direction) was around 4% within the hydride, whereas virtually no strain was predicted in the direction parallel to the stringer. CPFE-based 1D and 2D representations of precipitation of the same hydride revealed that the hydride's morphology was responsible for this effect. Using subsequent thermomechanical cycling, hydride strain ratcheting effects were quantified for the first time using DIC, which also revealed the cyclic development of bands of substantial compressive strain within the surrounding matrix. This was shown to lead to the development of GND networks. Intragranular strain ratcheting was shown to saturate after five thermomechanical cycles.

Page would be left blank if not for this note.

8 Conclusions

This thesis has covered a comprehensive range of issues pertaining to the growth of short fatigue cracks and localisation of damage in Zircaloy-4. In the absence of common nuclear fuel cladding environmental damage factors (e.g., corrosion, irradiation, hydrides), Chapters 4 and 5 present new results and novel methods which provide key insights into the factors controlling microstructurally short FCG. These initial chapters have scope far beyond the nuclear fuel cladding and reactor structural integrity research communities. Firstly, the SED fracture criterion was assessed as a FCG driving force using a CPFE-XFEM modelling framework. It was shown that despite the inadequacies of the original maximum slip-based model for crack path, SED could reliably capture experimentally observed microstructure effects on short FCG rate including texture sensitivity and grain boundary interactions. This led to the development of two new criteria for crack path ((i) maximum energy release rate and (ii) maximum normal SED). The maximum normal SED criterion was shown to vastly outperform the conventional maximum slip criterion when compared with experiments. Moreover, with an improved understanding of the determinants of short crack path and propagation rate, an analytical solution for FCG rate (which requires no fitting parameters) was developed. The model was shown to capture experimental FCG rate trends to a level comparable with CPFE modelling, despite demanding just a small fraction of the computational resources. In Chapter 6, the effects of neutron irradiation damage in fuel cladding were considered in an entirely new, yet industrially relevant sense. By comparison with the widely used post-irradiation mechanical testing approach, new models describing the temperature-sensitive synergistic evolution of material behaviour during irradiation and loading showed that synergistic effects can yield vastly different results in terms of the localisation of key variables responsible for the nucleation of fatigue cracks. Due to practical limitations in testing synergistic conditions, this new modelling approach could have significant implications for future work in this area. Lastly, in Chapter 7, experimental and numerical analyses of short FCG and strain ratcheting behaviour in hydrided Zircaloy-4 were presented. FCG studies presented previously unreported crack propagation mechanisms and associated rate effects. DIC results showed that strain fields induced by hydride precipitation exhibit strong directionality with respect to the morphological orientation, which were reinforced by CPFE models. Overall, the crack propagation and synergistic irradiation damage modelling work, combined with new mechanistic insights from experiments have contributed to the foundation of a holistic modelling framework for structural integrity of reactor fuel cladding, incorporating the intricacies of short FCG, irradiation damage, and hydride precipitation.

Page would be left blank if not for this note.

9 Future Work: Irradiation and Fatigue Crack Growth

9.1 Introduction

While the properties of structural materials are known to change significantly due to irradiation, there is currently very little data, experimental or otherwise to describe the impact of irradiation damage on fatigue behaviour in nuclear fuel cladding materials. The modelling work proposed in this chapter is aimed at predicting how the initiation and propagation of short fatigue cracks in Zircaloy-4 might be affected by realistic irradiation doses.

Though it is useful to characterise the tensile behaviour of irradiated zirconium alloys at both component and microstructural length-scales, it is more industrially impactful to consider the effects of small-strain cyclic loading in a reactor due to e.g., pressure and temperature variations [260], [339] driven by power fluctuations, vibrations due to complex thermal hydraulics [367], etc. Most fatigue studies of irradiated zirconium alloys were carried out in the 1990s [298], [368], [369] and were primarily focused on generating bulk fatigue life data at different levels of irradiation and at different temperatures. Wisner et al. [298] showed that under fully reversed strain-controlled cyclic loading, irradiation would apparently have very little effect on the overall fatigue life of pure zirconium and Zircaloy-2. However, due to significant differences between irradiated and unirradiated stress-strain behaviour, the plastic strain range in irradiated materials was substantially lower, and hence, fatigue life predictions based on plastic strain amplitude were lower by almost an order of magnitude. With a similar test programme, Nakatsuka et al. [368] showed that neutron irradiation would lead to an increased fatigue life for total strain amplitudes above 0.3% and a decreased fatigue life for strain amplitudes below this value; though without evidence, the increase in fatigue life was attributed to irradiation hardening, while the decrease was attributed to increased localisation. These results disagree somewhat with those of Wisner et al [298].

For a more comprehensive insight into the impact of irradiation damage on fatigue, it is necessary to account for the role of microstructure as it is strongly linked with the nucleation and propagation of fatigue cracks in various hexagonal close packed (HCP) materials [208], [246], [370], [371]. However, due to the intricacies of both micro-scale fatigue testing and working with irradiated materials, it is currently more pragmatic to address the problem using theoretical methods. Recent advances in CPFE modelling of irradiated materials [111], [372], [373], [374], [375] have provided a pathway for microstructural fatigue studies. Liu et al. [372]

presented a stochastic CPFE based framework for microstructure-linked continuum modelling of irradiated copper. Probabilistic Weibull distributions for dislocation slip were incorporated, which could capture shear band formation across individual grains due to irradiation. However, results were not reinforced directly by experimental data. Hardie et al. [111] developed dislocation-based constitutive equations to describe the hardening and softening behaviour of irradiated Zircaloy-4 under monotonic loading. Simulations of real microstructures were validated against HRDIC data [91] and could capture localisation and slip band formation due to channel clearing. Using the framework developed by Hardie et al. [111], this chapter proposes methods to investigate the interactions between microstructures with and without irradiation damage and microstructurally short FCG in single crystal and polycrystalline Zircaloy-4.

9.2 Modelling methodology for irradiation damage and short crack growth

A preliminary method has been developed to study the multifarious interactions between physically short fatigue cracks and irradiation damage phenomena in single crystal Zircaloy-4. Even in single crystals, there are innumerable factors which could be considered. The method proposed here could be used to gain fundamental insights into the influence of irradiation damage on FCG with dependencies including (i) irradiation dose, (ii) crack length, (iii) applied load / displacement, (iv) irradiation shear band formation, and (v) crystal orientation. The proposed CPFE modelling method is presented in Figure 105. Details of the irradiation damage modelling framework were presented in Chapter 3.

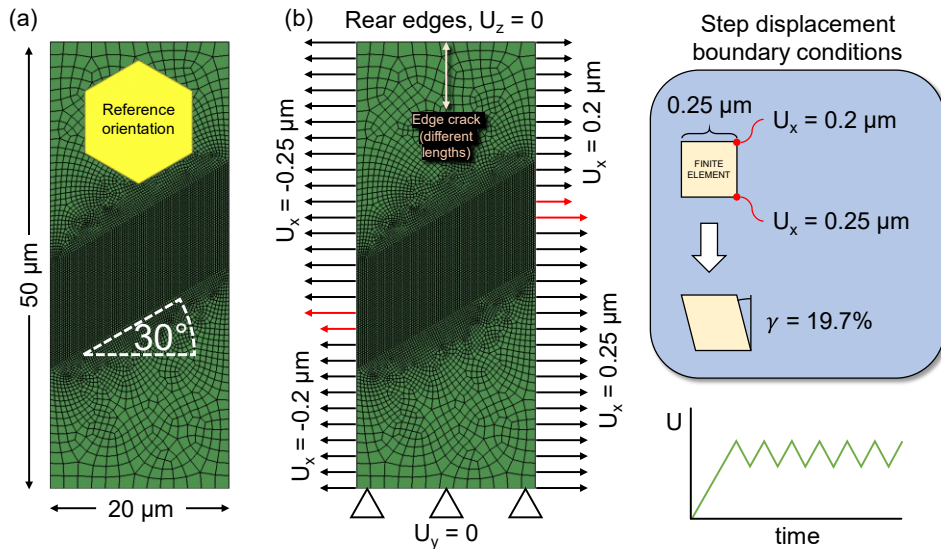


Figure 105: Proposed methodology for preliminary investigation of interactions between physically short static cracks and microstructure-driven localisation in irradiated and unirradiated materials. (a) $50 \times 20\ \mu\text{m}$ single crystal section with region containing refined mesh parallel with a prismatic slip plane. (b) Cyclic displacement-controlled (pseudo-stress-controlled) boundary conditions with initial overload, and step displacements (shear) at the boundaries representing microstructure interactions.

The model is designed to permit computationally efficient predictions of interactions between static fatigue cracks and irradiated (and unirradiated) single crystals, with crack lengths ranging between 5 and 25 μm . The elongated rectangular grain shape shown in in Figure 105 (a) enables the application of uniform fatigue boundary conditions, as well as the introduction of irradiation shear bands, i.e., channel clearing effects vial local discontinuities. Figure 105 (b) shows that along the outer edges of the model, step-displacement boundary conditions are applied to generate up to 20% shear strain, locally (which is commensurate with microstructure-induced localisation in irradiated materials [91]). In the absence of an edge crack, and with an initial mean applied strain of 1% in the horizontal direction, this approach can generate shear bands crossing the grain (interconnecting regions of localised shear) for various crystal orientations. Hence, with the introduction of edge cracks with progressively increasing length, their interactions with existing irradiation shear bands may be evaluated quantitatively. Preliminary static and fatigue results for irradiated and unirradiated Zircaloy-4 with a 20 μm edge crack are shown in Figure 106 (a) and (b).

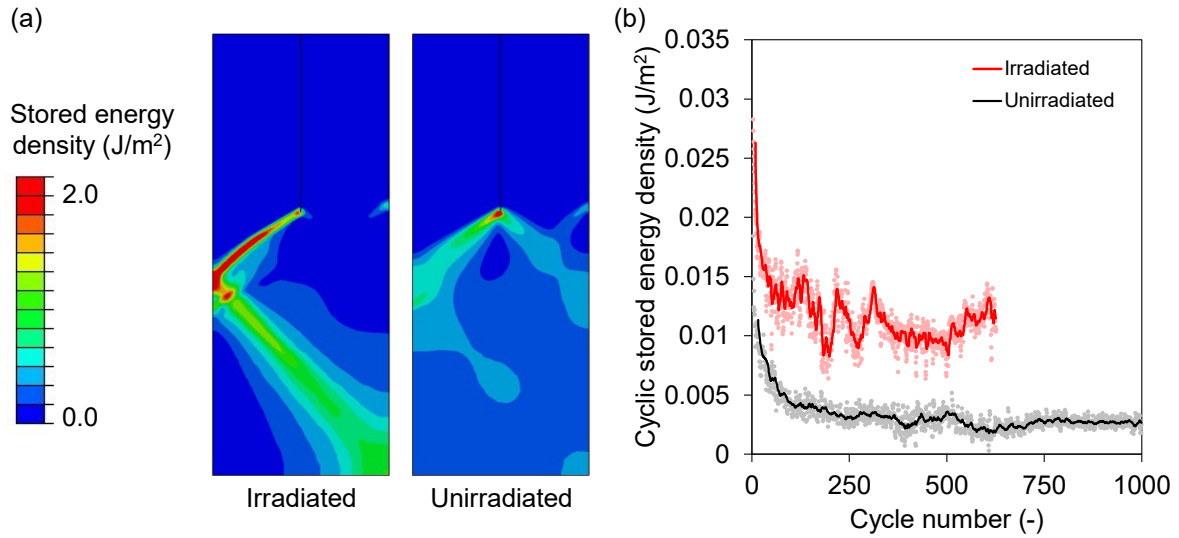


Figure 106: Preliminary results of a 20 μm static crack fatigue simulation in irradiated and unirradiated single crystals. (a) Distributions of SED in irradiated (0.8 dpa) and unirradiated material after one fatigue cycle. (b) The cyclic evolution of cyclic SED at crack tips in irradiated and unirradiated materials.

The single cycle SED fields presented in Figure 106 (a) also broadly capture the shear strain distributions, particularly for the irradiated case. The ‘irradiated’ SED field shows that for a 20 μm edge crack, the original prismatic shear band spanning the grain is removed due to increased interactions between highly localised shear at the boundary and the crack itself. The ‘unirradiated’ SED field also shows that there are interactions between the crack and the boundaries but to a lesser extent, as most SED is concentrated at the crack tip. These factors

could have important implications for crack path, FCG rate and ultimately structural integrity in irradiated materials. Preliminary crack tip cyclic SED results shown in Figure 106 (b) highlight a substantial difference in short FCG behaviour in irradiated Zircaloy-4, for the current loading conditions. Specifically, after around 300 fatigue cycles, the crack tip cyclic SED is shown to saturate for each case, with the irradiated case saturating at a level more than twice that of the unirradiated case. While this result is intriguing, it is highly sensitive to factors (i) – (v) raised earlier, which in combination with indeterminate data from the literature, highlights the necessity for a thorough study in this area.

Beyond single crystal analyses, it is envisaged that full microstructural studies of crack growth in irradiated and unirradiated Zircaloy-4 will be pursued for direct quantitative assessment of the impact of irradiation damage in real materials. Results from a preliminary microstructural study are presented here. The irradiated and unirradiated SED fields within an example microstructure containing a 5 μm facet after strain controlled monotonic loading up to 0.75% are shown in Figure 107.

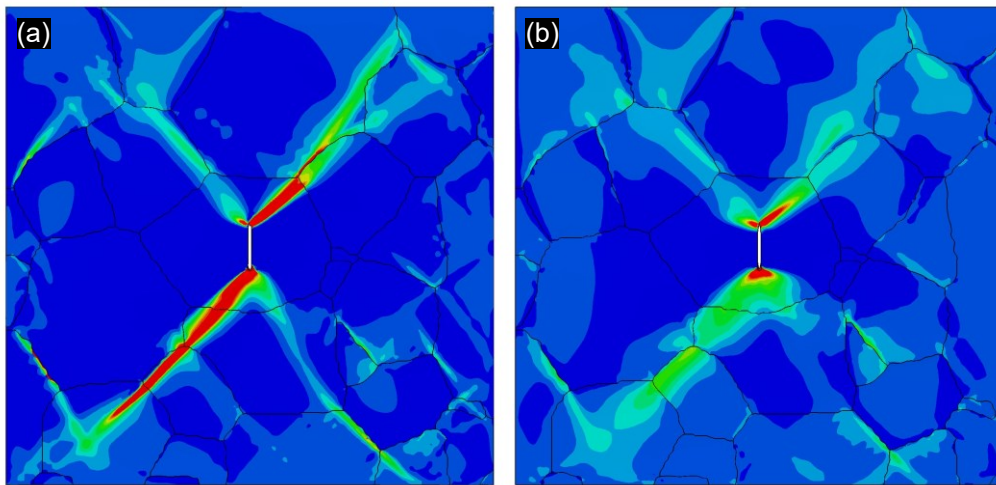


Figure 107: Distributions of SED after monotonic loading up to 0.75% strain in the horizontal direction in a cracked and (a) irradiated (0.8 dpa) and (b) unirradiated Zircaloy-4 polycrystal.

As anticipated, the interactions between the facet and the surrounding microstructure are much more pronounced in the irradiated case (Figure 107 (a)), with shear bands spanning several grains from their respective points of origin, compared with the diffuse interactions shown for the unirradiated case in Figure 107 (b). This indicates that detailed microstructural FCG studies in this area are likely to reveal substantial differences between irradiated and unirradiated materials.

10 References

- [1] “Energy Institute Statistical Review of World Energy,” 2023. Accessed: Nov. 22, 2023. [Online]. Available: <https://www.energyinst.org/statistical-review/>
- [2] B. Plackett, “Why France’s nuclear industry faces uncertainty,” *Nature*, Sep. 2022, doi: 10.1038/d41586-022-02817-2.
- [3] N. Higashi, “Japan’s Energy Policy and Nuclear Power Issues,” *Global Energy Law and Sustainability*, vol. 1, no. 1, pp. 122–128, Feb. 2020, doi: 10.3366/gels.2020.0011.
- [4] A. Einstein, “Ist die Trägheit eines Körpers von seinem Energieinhalt abhängig?,” *Ann Phys*, vol. 323, no. 13, pp. 639–641, Jan. 1905, doi: 10.1002/andp.19053231314.
- [5] B. R. T. Frost, *Nuclear Materials Part 2*. in Materials science and technology ; vol.10B. Weinheim: VCH Verlagsgesellschaft, 1994.
- [6] R. Adamson, F. Garzarolli, and C. Patterson, “In-Reactor Creep of Zirconium Alloys,” *Advanced Nuclear Technology International (Report)*, 2009.
- [7] ASTM, “Standard Specification for Zirconium and Zirconium Alloy Ingots for Nuclear Application (B350/B350M),” 2021.
- [8] E. Zuzek, J. P. Abriata, A. San-Martin, and F. D. Manchester, “The H-Zr (hydrogen-zirconium) system,” *Bulletin of Alloy Phase Diagrams*, vol. 11, no. 4, pp. 385–395, Aug. 1990, doi: 10.1007/BF02843318.
- [9] B. Cárdenas, R. Ibanez, J. Rouse, L. Swinfen-Styles, and S. Garvey, “The effect of a nuclear baseload in a zero-carbon electricity system: An analysis for the UK,” *Renew Energy*, vol. 205, pp. 256–272, Mar. 2023, doi: 10.1016/j.renene.2023.01.028.
- [10] G. E. Dieter, *Mechanical metallurgy*, 3rd ed. in McGraw-Hill series in materials science and engineering. New York ; McGraw-Hill, 1986.
- [11] L. F. Coffin, “Low cycle fatigue,” *Metals Engineering Quarterly*, vol. 3, pp. 15–24, 1963.
- [12] N. E. Prasad, A. G. Paradkar, G. Malakondaiah, and V. V Kutumbarao, “An analysis based on plastic strain energy for bilinearity in Coffin-Manson plots in an Al-Li alloy,” *Scripta Metallurgica et Materialia*, vol. 30, no. 12, pp. 1497–1502, 1994, doi: [https://doi.org/10.1016/0956-716X\(94\)90297-6](https://doi.org/10.1016/0956-716X(94)90297-6).

- [13] Y. Wang *et al.*, “Improving fatigue performance of Cr-coated zirconium alloy cladding by ultrasonic shot-peening proces,” *Journal of Materials Research and Technology*, vol. 27, pp. 6047–6057, 2023, doi: <https://doi.org/10.1016/j.jmrt.2023.11.102>.
- [14] A. Cruzado, S. Lucarini, J. LLorca, and J. Segurado, “Microstructure-based fatigue life model of metallic alloys with bilinear Coffin-Manson behavior,” *Int J Fatigue*, vol. 107, pp. 40–48, 2018, doi: <https://doi.org/10.1016/j.ijfatigue.2017.10.014>.
- [15] O. H. Basquin, “The Exponential Law of Endurance Tests,” in *American Society for Testing and Materials Proceedings*, 1910, pp. 625–630.
- [16] M. Ricotta, “Simple expressions to estimate the Manson–Coffin curves of ductile cast irons,” *Int J Fatigue*, vol. 78, pp. 38–45, 2015, doi: <https://doi.org/10.1016/j.ijfatigue.2015.03.025>.
- [17] S. R. Mediratta, V. Ramaswamy, and P. Rama Rao, “Two stage cyclic work hardening and two slope coffin-manson relationship in dual phase steels,” *Scripta Metallurgica*, vol. 20, no. 4, pp. 555–558, 1986, doi: [https://doi.org/10.1016/0036-9748\(86\)90253-X](https://doi.org/10.1016/0036-9748(86)90253-X).
- [18] A. Bhattacharyya, G. V. S. Sastry, and V. V. Kutumbarao, “On the dual slope Coffin-Manson relationship during low cycle fatigue of nickel-base alloy IN 718,” *Scr Mater*, vol. 36, pp. 411–415, 1997, doi: [https://doi.org/10.1016/S0142-1123\(98\)91088-X](https://doi.org/10.1016/S0142-1123(98)91088-X).
- [19] J. D. Morrow and D. F. Socie, “Review of contemporary approaches to fatigue damage analysis,” in *Risk and Failure Analysis for Improved Performance and Reliability*, J. J. Burke and V. Weiss, Eds., Plenum Publishing Corporation, 1980.
- [20] P. C. Paris, M. P. Gomez, and W. E. P. Anderson, “A Rational Analytic Theory of Fatigue,” *The Trend in Engineering*, vol. 13, Trend Eng., pp. 9–14, 1961. doi: <https://doi.org/10.11648/j.ijmea.s.2015030201.11>.
- [21] F. P. E. Dunne, A. J. Wilkinson, and R. Allen, “Experimental and computational studies of low cycle fatigue crack nucleation in a polycrystal,” *Int J Plast*, vol. 23, no. 2, pp. 273–295, 2007, doi: <https://doi.org/10.1016/j.ijplas.2006.07.001>.
- [22] D. L. McDowell, “An engineering model for propagation of small cracks in fatigue,” *Eng Fract Mech*, vol. 56, no. 3, pp. 357–377, 1997, doi: [https://doi.org/10.1016/S0013-7944\(96\)00057-4](https://doi.org/10.1016/S0013-7944(96)00057-4).

- [23] A. J. Wilkinson, S. G. Roberts, and P. B. Hirsch, “Modelling the threshold conditions for propagation of stage I fatigue cracks,” *Acta Mater*, vol. 46, no. 2, pp. 379–390, 1998, doi: [https://doi.org/10.1016/S1359-6454\(97\)00290-5](https://doi.org/10.1016/S1359-6454(97)00290-5).
- [24] P. Dai and Z. Li, “A plasticity-corrected stress intensity factor for fatigue crack growth in ductile materials,” *Acta Mater*, vol. 61, no. 16, pp. 5988–5995, 2013, doi: [10.1016/j.actamat.2013.06.039](https://doi.org/10.1016/j.actamat.2013.06.039).
- [25] K. Lu, J. Katsuyama, and Y. Li, “Plasticity Correction on Stress Intensity Factor Evaluation for Underclad Cracks in Reactor Pressure Vessels,” *J Press Vessel Technol*, vol. 142, no. 5, Oct. 2020, doi: [10.1115/1.4046977](https://doi.org/10.1115/1.4046977).
- [26] A. Camacho-Reyes, J. M. Vasco-Olmo, G. L. Gómez Gonzales, and F. A. Diaz, “Study of Effective Stress Intensity Factor through the CJP Model Using Full-Field Experimental Data,” *Materials*, vol. 16, no. 16, p. 5705, Aug. 2023, doi: [10.3390/ma16165705](https://doi.org/10.3390/ma16165705).
- [27] J. D. Eshelby, “The determination of the elastic field of an ellipsoidal inclusion, and related problems,” *Proc R Soc Lond A Math Phys Sci*, vol. 241, no. 1226, pp. 376–396, Aug. 1957, doi: [10.1098/rspa.1957.0133](https://doi.org/10.1098/rspa.1957.0133).
- [28] A. A. Griffith, “The phenomena of rupture and flow in solids,” *Royal Society*, vol. 221, no. 582–593, pp. 163–198, 1921, doi: <https://doi.org/10.1098/rsta.1921.0006>.
- [29] G. R. Irwin, “Fracture,” *Encyclopedia of Physics*, vol. 6, 1958.
- [30] G. R. Irwin, J. A. Kies, and H. L. Smith, “Fracture strength relative to onset and arrest of crack propagation,” *Proc. ASTM*, pp. 640–660, 1958.
- [31] J. R. Rice, “A path independent integral and the approximate analysis of strain concentration by notches and cracks,” *J Appl Mech*, vol. 35, pp. 379–386, 1968.
- [32] L. Banks-Sills and Y. Volpert, “Application of the cyclic J-integral to fatigue crack propagation of Al 2024-T351,” *Eng Fract Mech*, vol. 40, no. 2, pp. 355–370, 1991, doi: [10.1016/0013-7944\(91\)90270-B](https://doi.org/10.1016/0013-7944(91)90270-B).

- [33] H. Li, X. Luo, Y. Zhang, and R. Xu, “Stochastic fatigue damage in viscoelastic materials using probabilistic pseudo J-integral Paris’ law,” *Eng Fract Mech*, vol. 245, p. 107566, Mar. 2021, doi: 10.1016/j.engfracmech.2021.107566.
- [34] D. Wilson, Z. Zheng, and F. P. E. Dunne, “A microstructure-sensitive driving force for crack growth,” *J Mech Phys Solids*, vol. 121, pp. 147–174, 2018, doi: 10.1016/j.jmps.2018.07.005.
- [35] K. Stopka, G. Whelan, and D. McDowell, “Microstructure-Sensitive ICME Workflows for Fatigue Critical Applications,” in *SAMPE 2019 - Charlotte, NC*, SAMPE, Apr. 2019. doi: 10.33599/nasampe/s.19.1414.
- [36] M. W. Brown and K. J. Miller, “A Theory for Fatigue Failure under Multiaxial Stress-Strain Conditions,” *Proceedings of the Institution of Mechanical Engineers*, vol. 187, no. 1, pp. 745–755, Jun. 1973, doi: 10.1243/PIME_PROC_1973_187_161_02.
- [37] A. Fatemi and D. F. Socie, “a Critical Plane Approach To Multiaxial Fatigue Damage Including Out-of-Phase Loading,” *Fatigue Fract Eng Mater Struct*, vol. 11, no. 3, pp. 149–165, 1988, doi: 10.1111/j.1460-2695.1988.tb01169.x.
- [38] J. Gayda and R. V. Miner, “Fatigue crack initiation and propagation in several nickel-base superalloys at 650°C,” *Int J Fatigue*, vol. 5, no. 3, pp. 135–143, Jul. 1983, doi: 10.1016/0142-1123(83)90026-9.
- [39] Y. Xu *et al.*, “Predicting dwell fatigue life in titanium alloys using modelling and experiment,” *Nat Commun*, vol. 11, no. 1, p. 5868, Nov. 2020, doi: 10.1038/s41467-020-19470-w.
- [40] C. Lavogiez, C. Dureau, Y. Nadot, P. Villechaise, and S. Hémery, “Crack initiation mechanisms in Ti-6Al-4V subjected to cold dwell-fatigue, low-cycle fatigue and high-cycle fatigue loadings,” *Acta Mater*, vol. 244, p. 118560, Jan. 2023, doi: 10.1016/j.actamat.2022.118560.
- [41] A. Muth, R. John, A. Pilchak, S. R. Kalidindi, and D. L. McDowell, “Analysis of Fatigue Indicator Parameters for Ti-6Al-4V microstructures using extreme value statistics in the transition fatigue regime,” *Int J Fatigue*, vol. 153, Dec. 2021, doi: 10.1016/j.ijfatigue.2021.106441.

- [42] W. Sum, E. Williams, and S. Leen, “Finite element, critical-plane, fatigue life prediction of simple and complex contact configurations,” *Int J Fatigue*, vol. 27, no. 4, pp. 403–416, Apr. 2005, doi: 10.1016/j.ijfatigue.2004.08.001.
- [43] A. S. Cruces, A. Garcia-Gonzalez, B. Moreno, T. Itoh, and P. Lopez-Crespo, “Critical plane based method for multiaxial fatigue analysis of 316 stainless steel,” *Theoretical and Applied Fracture Mechanics*, vol. 118, p. 103273, Apr. 2022, doi: 10.1016/j.tafmec.2022.103273.
- [44] M. Zhang, D. L. McDowell, and R. W. Neu, “Microstructure sensitivity of fretting fatigue based on computational crystal plasticity,” *Tribol Int*, vol. 42, no. 9, pp. 1286–1296, Sep. 2009, doi: 10.1016/j.triboint.2009.04.036.
- [45] G. M. Castelluccio and D. L. McDowell, “Effect of annealing twins on crack initiation under high cycle fatigue conditions,” *J Mater Sci*, vol. 48, no. 6, pp. 2376–2387, Mar. 2013, doi: 10.1007/s10853-012-7021-y.
- [46] B. D. Smith, D. S. Shih, and D. L. McDowell, “Fatigue hot spot simulation for two Widmanstätten titanium microstructures,” *Int J Fatigue*, vol. 92, pp. 116–129, Nov. 2016, doi: 10.1016/j.ijfatigue.2016.05.002.
- [47] M. Yaghoobi, K. S. Stopka, A. Lakshmanan, V. Sundararaghavan, J. E. Allison, and D. L. McDowell, “PRISMS-Fatigue computational framework for fatigue analysis in polycrystalline metals and alloys,” *NPJ Comput Mater*, vol. 7, no. 1, Dec. 2021, doi: 10.1038/s41524-021-00506-8.
- [48] Z.-Y. Yu, S.-P. Zhu, Q. Liu, and Y. Liu, “Multiaxial Fatigue Damage Parameter and Life Prediction without Any Additional Material Constants,” *Materials*, vol. 10, no. 8, p. 923, Aug. 2017, doi: 10.3390/ma10080923.
- [49] Y. Charles, R. Estevez, Y. Bréchet, and E. Maire, “Modelling the competition between interface debonding and particle fracture using a plastic strain dependent cohesive zone,” *Eng Fract Mech*, vol. 77, no. 4, pp. 705–718, Mar. 2010, doi: 10.1016/j.engfracmech.2009.11.012.
- [50] A. Rovinelli, Y. Guilhem, H. Proudhon, R. A. Lebensohn, W. Ludwig, and M. D. Sangid, “Assessing reliability of fatigue indicator parameters for small crack growth via a

- probabilistic framework,” *Model Simul Mat Sci Eng*, vol. 25, no. 4, May 2017, doi: 10.1088/1361-651X/aa6c45.
- [51] Y. Liu, X. Zhang, and C. Oskay, “A comparative study on fatigue indicator parameters for near- α titanium alloys,” *Fatigue Fract Eng Mater Struct*, vol. 46, no. 1, pp. 271–294, Jan. 2023, doi: 10.1111/ffe.13862.
 - [52] S. Joseph, K. Joseph, T. C. Lindley, and D. Dye, “The role of dwell hold on the dislocation mechanisms of fatigue in a near alpha titanium alloy,” *Int J Plast*, vol. 131, p. 102743, Aug. 2020, doi: 10.1016/j.ijplas.2020.102743.
 - [53] J. Weertman, “Theory of rate of growth of fatigue cracks under combined static and cyclic stresses,” *International Journal of Fracture Mechanics*, vol. 5, no. 1, pp. 13–15, 1969, doi: 10.1007/BF00189934.
 - [54] J. Weertman, “Theory of fatigue crack growth based on a BCS crack theory with work hardening,” *Int J Fract*, vol. 9, no. 2, pp. 125–131, 1973, doi: 10.1007/BF00041854.
 - [55] K. Tanaka and T. Mura, “Dislocation Model for Fatigue Crack Initiation.,” *American Society of Mechanical Engineers (Paper)*, vol. 48, no. 81-APM-3, pp. 97–103, 1981.
 - [56] J. Zhou, R. A. Barrett, and S. B. Leen, “A physically-based method for predicting high temperature fatigue crack initiation in P91 welded steel,” *Int J Fatigue*, vol. 153, p. 106480, Dec. 2021, doi: 10.1016/j.ijfatigue.2021.106480.
 - [57] X. Liu and S. Lu, “A micro-crack initiation life simulation method by improving the Tanaka-Mura’s model of slip behavior,” *Int J Fatigue*, vol. 145, p. 106108, Apr. 2021, doi: 10.1016/j.ijfatigue.2020.106108.
 - [58] K. S. Chan, “A microstructure-based fatigue-crack-initiation model,” *Metallurgical and Materials Transactions A*, vol. 34, no. 1, pp. 43–58, Jan. 2003, doi: 10.1007/s11661-003-0207-9.
 - [59] E. Van der Giessen and A. Needleman, “Discrete dislocation plasticity: a simple planar model,” *Model Simul Mat Sci Eng*, vol. 3, no. 5, pp. 689–735, Sep. 1995, doi: 10.1088/0965-0393/3/5/008.

- [60] V. S. Deshpande, A. Needleman, and E. Van der Giessen, “Discrete dislocation modeling of fatigue crack propagation,” *Acta Mater*, vol. 50, no. 4, pp. 831–846, Feb. 2002, doi: 10.1016/S1359-6454(01)00377-9.
- [61] V. S. Deshpande, A. Needleman, and E. Van der Giessen, “Discrete dislocation plasticity modeling of short cracks in single crystals,” *Acta Mater*, vol. 51, no. 1, pp. 1–15, Jan. 2003, doi: 10.1016/S1359-6454(02)00401-9.
- [62] B. Gurrutxaga-Lerma, D. S. Balint, D. Dini, D. E. Eakins, and A. P. Sutton, “A dynamic discrete dislocation plasticity method for the simulation of plastic relaxation under shock loading,” *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 469, no. 2156, p. 20130141, Aug. 2013, doi: 10.1098/rspa.2013.0141.
- [63] C. Déprés, Z. Yang, and C. Robertson, “Stage-II fatigue crack growth model from 3D dislocation dynamics simulations,” *Philosophical Magazine*, vol. 102, no. 6, pp. 504–521, Mar. 2022, doi: 10.1080/14786435.2021.2009135.
- [64] T. N. Tak, A. Prakash, I. Samajdar, A. A. Benzerga, and P. J. Guruprasad, “A discrete dislocation dynamics framework for modeling polycrystal plasticity with hardening,” *Int J Solids Struct*, vol. 281, p. 112442, Oct. 2023, doi: 10.1016/j.ijsolstr.2023.112442.
- [65] J. C. Stinville *et al.*, “On the origins of fatigue strength in crystalline metallic materials,” *Science* (1979), vol. 377, pp. 1065–1071, 2022, doi: <https://doi.org/10.1126/science.abn0392>.
- [66] S. Kumar and W. A. Curtin, “Crack interaction with microstructure,” *Materials Today*, vol. 10, no. 9, pp. 34–44, Sep. 2007, doi: 10.1016/S1369-7021(07)70207-9.
- [67] V. V. C. Wan, D. W. Maclachlan, and F. P. E. Dunne, “A stored energy criterion for fatigue crack nucleation in polycrystals,” *Int J Fatigue*, vol. 68, pp. 90–102, 2014, doi: 10.1016/j.ijfatigue.2014.06.001.
- [68] M. Čebren and F. Kosel, “Stored energy predictions from dislocation-based hardening models and hardness measurements for tensile-deformed commercial purity copper,” *Strojniski Vestnik/Journal of Mechanical Engineering*, vol. 60, no. 7–8, pp. 462–474, 2014, doi: 10.5545/sv-jme.2013.1569.

- [69] V. V. C. Wan, J. Jiang, D. W. MacLachlan, and F. P. E. Dunne, “Microstructure-sensitive fatigue crack nucleation in a polycrystalline Ni superalloy,” *Int J Fatigue*, vol. 90, pp. 181–190, Sep. 2016, doi: 10.1016/j.ijfatigue.2016.04.013.
- [70] B. Chen, J. Jiang, and F. P. E. Dunne, “Microstructurally-sensitive fatigue crack nucleation in Ni-based single and oligo crystals,” *J Mech Phys Solids*, vol. 106, pp. 15–33, Sep. 2017, doi: 10.1016/j.jmps.2017.05.012.
- [71] B. Chen, J. Jiang, and F. P. E. Dunne, “Is stored energy density the primary meso-scale mechanistic driver for fatigue crack nucleation?,” *Int J Plast*, vol. 101, pp. 213–229, Feb. 2018, doi: 10.1016/j.ijsplas.2017.11.005.
- [72] Z. Zheng, N. G. Prastiti, D. S. Balint, and F. P. E. Dunne, “The dislocation configurational energy density in discrete dislocation plasticity,” *J Mech Phys Solids*, vol. 129, pp. 39–60, Aug. 2019, doi: 10.1016/j.jmps.2019.04.015.
- [73] D. Wilson and F. P. E. Dunne, “A mechanistic modelling methodology for microstructure-sensitive fatigue crack growth,” *J Mech Phys Solids*, vol. 124, pp. 827–848, 2019, doi: 10.1016/j.jmps.2018.11.023.
- [74] D. Wilson, W. Wan, and F. P. E. Dunne, “Microstructurally-sensitive fatigue crack growth in HCP, BCC and FCC polycrystals,” *J Mech Phys Solids*, vol. 126, pp. 204–225, 2019, doi: 10.1016/j.jmps.2019.02.012.
- [75] K. Tokaji, T. Ogawa, and K. Ohya, “The effect of grain size on small fatigue crack growth in pure titanium,” *Int J Fatigue*, vol. 16, no. 8, pp. 571–578, 1994, doi: 10.1016/0142-1123(94)90483-9.
- [76] W. Wan, “Microstructurally-sensitive short crack growth in Zircaloy-4,” PhD, Imperial College London, 2020. doi: <https://doi.org/10.25560/90249>.
- [77] J. A. Brinkman, “Production of Atomic Displacements by High-Energy Particles,” *Am J Phys*, vol. 24, no. 4, pp. 246–267, Apr. 1956, doi: 10.1119/1.1934201.
- [78] W. R. Martin and J. R. Weir, “The effect of irradiation temperature on the post-irradiation stress-strain behaviour of stainless steel,” *ASTM*, pp. 251–268, 1965, [Online]. Available: www.astm.org

- [79] J. A. Brinkman, “On the nature of radiation damage in metals,” *J Appl Phys*, vol. 25, no. 8, pp. 961–970, 1954, doi: 10.1063/1.1721810.
- [80] G. J. Dienes, “Radiation effects in solids,” *Annual Reviews in Nuclear Science*, vol. 2, pp. 187–20, 1953, [Online]. Available: www.annualreviews.org
- [81] F. W. Kunz and A. N. Holdent, “The effect of short-time moderate flux neutron irradiations on the mechanical properties of some metals,” *Acta Metallurgica*, vol. 2, pp. 816–822, 1954.
- [82] J. W. Zhang, S. M. Liu, and W. Z. Han, “Interfaces Reduce Dislocation Loop Formation in Irradiated Nanolayered Zr-2.5Nb,” *Scr Mater*, vol. 200, Jul. 2021, doi: 10.1016/j.scriptamat.2021.113902.
- [83] P. M. Kelly and R. G. Blake, “The characterization of dislocation loops in neutron irradiated zirconium,” *Philosophical Magazine*, vol. 28, no. 2, pp. 415–426, 1973, doi: 10.1080/14786437308217463.
- [84] S. M. Liu, I. J. Beyerlein, and W. Z. Han, “Two-dimensional vacancy platelets as precursors for basal dislocation loops in hexagonal zirconium,” *Nat Commun*, vol. 11, no. 1, Dec. 2020, doi: 10.1038/s41467-020-19629-5.
- [85] G. J. C. Carpenter and J. F. Watters, “A study of electron irradiation damage in zirconium using a high voltage electron microscope,” *Journal of Nuclear Materials*, vol. 96, pp. 213–226, 1981.
- [86] H. Yu, Z. Yao, Y. Idrees, H. K. Zhang, M. A. Kirk, and M. R. Daymond, “Accumulation of dislocation loops in the α phase of Zr Excel alloy under heavy ion irradiation,” *Journal of Nuclear Materials*, vol. 491, pp. 232–241, Aug. 2017, doi: 10.1016/j.jnucmat.2017.04.038.
- [87] M. Griffiths, R. W. Gilbert, and C. E. Coleman, “Grain boundary sinks in neutron-irradiated Zr and Zr-alloys,” *Journal of Nuclear Materials*, vol. 159, pp. 405–416, Oct. 1988, doi: 10.1016/0022-3115(88)90107-9.
- [88] T. Fujita, J. Hirabayashi, Y. Katayama, F. Kano, and H. Watanabe, “Contribution of dislocation loop to radiation-hardening of RPV steels studied by STEM/EDS with

- surveillance test pieces,” *Journal of Nuclear Materials*, vol. 572, Dec. 2022, doi: 10.1016/j.jnucmat.2022.154055.
- [89] F. Onimus, I. Monnet, J. L. Béchade, C. Prioul, and P. Pilvin, “A statistical TEM investigation of dislocation channeling mechanism in neutron irradiated zirconium alloys,” *Journal of Nuclear Materials*, vol. 328, no. 2–3, pp. 165–179, Jul. 2004, doi: 10.1016/j.jnucmat.2004.04.337.
 - [90] K. Farrell, T. S. Byun, and N. Hashimoto, “Deformation mode maps for tensile deformation of neutron-irradiated structural alloys,” *Journal of Nuclear Materials*, vol. 335, no. 3, pp. 471–486, Dec. 2004, doi: 10.1016/j.jnucmat.2004.08.006.
 - [91] R. Thomas *et al.*, “Characterisation of irradiation enhanced strain localisation in a zirconium alloy,” *Materialia (Oxf)*, vol. 5, Mar. 2019, doi: 10.1016/j.mtla.2019.100248.
 - [92] V. Fidleris, “The irradiation creep and growth phenomena,” *Journal of Nuclear Materials*, vol. 159, pp. 22–42, Oct. 1988, doi: 10.1016/0022-3115(88)90083-9.
 - [93] C. Liu *et al.*, “Texture and yielding anisotropy of Zircaloy-4 alloy cladding tube produced by cold Pilger rolling and annealing,” *Materials Science and Engineering: A*, vol. 719, pp. 147–154, Mar. 2018, doi: 10.1016/j.msea.2018.02.043.
 - [94] S. L. Dudarev, “Density Functional Theory Models for Radiation Damage,” *Annu Rev Mater Res*, vol. 43, no. 1, pp. 35–61, Jul. 2013, doi: 10.1146/annurev-matsci-071312-121626.
 - [95] M. Fyta, “Quantum-mechanical methods,” in *Computational Approaches in Physics*, Morgan & Claypool Publishers, 2016, pp. 2-1-2–30. doi: 10.1088/978-1-6817-4417-9ch2.
 - [96] C. Chang, V. L. Deringer, K. S. Katti, V. Van Speybroeck, and C. M. Wolverson, “Simulations in the era of exascale computing,” *Nat Rev Mater*, vol. 8, no. 5, pp. 309–313, Mar. 2023, doi: 10.1038/s41578-023-00540-6.
 - [97] C. P. Race, D. R. Mason, M. W. Finnis, W. M. C. Foulkes, A. P. Horsfield, and A. P. Sutton, “The treatment of electronic excitations in atomistic models of radiation damage in metals,” *Reports on Progress in Physics*, vol. 73, no. 11, p. 116501, Nov. 2010, doi: 10.1088/0034-4885/73/11/116501.

- [98] C. Björkas and K. Nordlund, “Comparative study of cascade damage in Fe simulated with recent potentials,” *Nucl Instrum Methods Phys Res B*, vol. 259, no. 2, pp. 853–860, Jun. 2007, doi: 10.1016/j.nimb.2007.03.076.
- [99] F. Granberg, J. Byggmästar, and K. Nordlund, “Molecular dynamics simulations of high-dose damage production and defect evolution in tungsten,” *Journal of Nuclear Materials*, vol. 556, p. 153158, Dec. 2021, doi: 10.1016/j.jnucmat.2021.153158.
- [100] C. S. Becquart and C. Domain, “Modeling Microstructure and Irradiation Effects,” *Metallurgical and Materials Transactions A*, vol. 42, no. 4, pp. 852–870, Apr. 2011, doi: 10.1007/s11661-010-0460-7.
- [101] M. I. Brand, E. G. Obbard, and P. A. Burr, “Spatial distribution of primary radiation damage in microstructures,” *Npj Mater Degrad*, vol. 7, no. 1, p. 23, Apr. 2023, doi: 10.1038/s41529-023-00337-6.
- [102] M. Boleininger, D. R. Mason, A. E. Sand, and S. L. Dudarev, “Microstructure of a heavily irradiated metal exposed to a spectrum of atomic recoils,” *Sci Rep*, vol. 13, no. 1, p. 9716, Jun. 2023, doi: 10.1038/s41598-023-36723-y.
- [103] J. D. Jenkins *et al.*, “The benefits of nuclear flexibility in power system operations with renewable energy,” *Appl Energy*, vol. 222, pp. 872–884, Jul. 2018, doi: 10.1016/j.apenergy.2018.03.002.
- [104] P. M. Derlet and S. L. Dudarev, “Microscopic structure of a heavily irradiated material,” *Phys Rev Mater*, vol. 4, no. 2, p. 023605, Feb. 2020, doi: 10.1103/PhysRevMaterials.4.023605.
- [105] A. R. Warwick *et al.*, “Dislocation density transients and saturation in irradiated zirconium,” *Int J Plast*, vol. 164, p. 103590, May 2023, doi: 10.1016/j.ijplas.2023.103590.
- [106] T. Ungár, P. Frankel, G. Ribárik, C. P. Race, and M. Preuss, “Size-distribution of irradiation-induced dislocation-loops in materials used in the nuclear industry,” *Journal of Nuclear Materials*, vol. 550, p. 152945, Jul. 2021, doi: 10.1016/j.jnucmat.2021.152945.
- [107] D. T. Gillespie, “A general method for numerically simulating the stochastic time evolution of coupled chemical reactions,” *J Comput Phys*, vol. 22, no. 4, pp. 403–434, Dec. 1976, doi: 10.1016/0021-9991(76)90041-3.

- [108] J. Marian and V. V. Bulatov, “Stochastic cluster dynamics method for simulations of multispecies irradiation damage accumulation,” *Journal of Nuclear Materials*, vol. 415, no. 1, pp. 84–95, Aug. 2011, doi: 10.1016/j.jnucmat.2011.05.045.
- [109] A. Y. Dunn, L. Capolungo, E. Martinez, and M. Cherkaoui, “Spatially resolved stochastic cluster dynamics for radiation damage evolution in nanostructured metals,” *Journal of Nuclear Materials*, vol. 443, no. 1–3, pp. 128–139, Nov. 2013, doi: 10.1016/j.jnucmat.2013.07.009.
- [110] R. E. Stoller, S. I. Golubov, C. Domain, and C. S. Becquart, “Mean field rate theory and object kinetic Monte Carlo: A comparison of kinetic models,” *Journal of Nuclear Materials*, vol. 382, no. 2–3, pp. 77–90, Dec. 2008, doi: 10.1016/j.jnucmat.2008.08.047.
- [111] C. Hardie, R. Thomas, Y. Liu, P. Frankel, and F. Dunne, “Simulation of crystal plasticity in irradiated metals: A case study on Zircaloy-4,” *Acta Mater*, vol. 241, Dec. 2022, doi: 10.1016/j.actamat.2022.118361.
- [112] A. Sato, T. Mifune, and M. Meshii, “Irradiation softening in pure iron single crystals,” *Physica Status Solidi (a)*, vol. 18, no. 2, pp. 699–709, Aug. 1973, doi: 10.1002/pssa.2210180232.
- [113] X. Xiao, D. Terentyev, L. Yu, D. Song, A. Bakaev, and H. Duan, “Modelling irradiation-induced softening in BCC iron by crystal plasticity approach,” *Journal of Nuclear Materials*, vol. 466, pp. 312–315, Nov. 2015, doi: 10.1016/j.jnucmat.2015.08.017.
- [114] X. Xiao, “Fundamental Mechanisms for Irradiation-Hardening and Embrittlement: A Review,” *Metals (Basel)*, vol. 9, no. 10, p. 1132, Oct. 2019, doi: 10.3390/met9101132.
- [115] L. A. S. Carno, J. J. Turner, and P. G. Martin, “Modelling ‘Type B’ ejecta formation reveals reactor Unit 1 conditions during the Fukushima Daiichi Nuclear Disaster,” *Sci Rep*, vol. 13, no. 1, p. 3686, Mar. 2023, doi: 10.1038/s41598-023-30903-6.
- [116] A. T. Motta and L.-Q. Chen, “Hydride Formation in Zirconium Alloys,” *JOM*, vol. 64, no. 12, pp. 1403–1408, Dec. 2012, doi: 10.1007/s11837-012-0479-x.
- [117] A. T. Motta *et al.*, “Hydrogen in zirconium alloys: A review,” *Journal of Nuclear Materials*, vol. 518, pp. 440–460, 2019, doi: 10.1016/j.jnucmat.2019.02.042.

- [118] J. J. Kearns, “Terminal solubility and partitioning of hydrogen in the alpha phase of zirconium, Zircaloy-2 and Zircaloy-4,” *Journal of Nuclear Materials*, vol. 22, no. 3, pp. 292–303, Jun. 1967, doi: 10.1016/0022-3115(67)90047-5.
- [119] A. McMinn, E. C. Darby, and J. S. Schofield, “The Terminal Solid Solubility of Hydrogen in Zirconium Alloys,” *ASTM Special Technical Publication*, no. 1354, pp. 173–195, 2000, [Online]. Available: <https://www.scopus.com/inward/record.uri?eid=2-s2.0-0033906312&partnerID=40&md5=044ba6771b790c2e0938555d03b3debb>
- [120] K. Une and S. Ishimoto, “Terminal Solid Solubility of Hydrogen in Unalloyed Zirconium by Differential Scanning Calorimetry,” *J Nucl Sci Technol*, vol. 41, no. 9, pp. 949–952, Sep. 2004, doi: 10.1080/18811248.2004.9715569.
- [121] Z. L. Pan, I. G. Ritchie, and M. P. Puls, “The terminal solid solubility of hydrogen and deuterium in Zr-2.5Nb alloys,” *Journal of Nuclear Materials*, vol. 228, no. 2, pp. 227–237, Mar. 1996, doi: 10.1016/S0022-3115(95)00217-0.
- [122] K. B. Colas *et al.*, “In situ study of hydride precipitation kinetics and re-orientation in Zircaloy using synchrotron radiation,” *Acta Mater*, vol. 58, no. 20, pp. 6575–6583, Dec. 2010, doi: 10.1016/j.actamat.2010.07.018.
- [123] P. Vizcaíno, A. D. Banchik, and J. P. Abriata, “Solubility of hydrogen in Zircaloy-4: irradiation induced increase and thermal recovery,” *Journal of Nuclear Materials*, vol. 304, no. 2–3, pp. 96–106, Aug. 2002, doi: 10.1016/S0022-3115(02)00883-8.
- [124] G. F. Slattery, “The Mechanical Properties of Zircaloy-2 Tubing Containing Circumferentially Aligned Hydride,” in *Applications-Related Phenomena in Zirconium and its Alloys*, 100 Barr Harbor Drive, PO Box C700, West Conshohocken, PA 19428-2959: ASTM International, 1968, pp. 95–110. doi: 10.1520/STP43822S.
- [125] F. Xu, R. A. Holt, and M. R. Daymond, “Modeling lattice strain evolution during uniaxial deformation of textured Zircaloy-2,” *Acta Mater*, vol. 56, no. 14, pp. 3672–3687, Aug. 2008, doi: 10.1016/j.actamat.2008.04.019.
- [126] S. Kang *et al.*, “Determination of the hydrogen heat of transport in Zircaloy-4,” *Journal of Nuclear Materials*, vol. 573, p. 154122, Jan. 2023, doi: 10.1016/j.jnucmat.2022.154122.

- [127] A. Fick, “Ueber Diffusion,” *Ann Phys*, vol. 170, no. 1, pp. 59–86, Jan. 1855, doi: 10.1002/andp.18551700105.
- [128] E. Elmukashfi, E. Tarleton, and A. C. F. Cocks, “A modelling framework for coupled hydrogen diffusion and mechanical behaviour of engineering components,” *Comput Mech*, vol. 66, no. 1, pp. 189–220, Jul. 2020, doi: 10.1007/s00466-020-01847-9.
- [129] R. S. Daum, Y. S. Chu, and A. T. Motta, “Identification and quantification of hydride phases in Zircaloy-4 cladding using synchrotron X-ray diffraction,” *Journal of Nuclear Materials*, vol. 392, no. 3, pp. 453–463, Aug. 2009, doi: 10.1016/j.jnucmat.2009.04.004.
- [130] D. L. Douglass, *The Metallurgy of Zirconium*, 1st ed. New York: IAEA, 1971.
- [131] J. P. Langeron and P. Lehr, “Préparation de gros cristaux de zirconium et détermination de l’orientation des précipités d’hydrure de zirconium,” *La Revue de Métallurgie*, vol. 55, pp. 901–906, 1958, doi: <https://doi.org/10.1051/metal/195855100901>.
- [132] D. G. Westlake, “The habit planes of zirconium hydride in zirconium and zircaloy,” *Journal of Nuclear Materials*, vol. 26, no. 2, pp. 208–216, 1968, doi: 10.1016/0022-3115(68)90072-X.
- [133] A. T. Motta and L. Q. Chen, “Hydride formation in zirconium alloys,” *Journal of Minerals, Metals and Materials Society*, vol. 64, no. 12, pp. 1403–1408, 2012, doi: 10.1007/s11837-012-0479-x.
- [134] V. Perovic, G. C. Weatherly, and C. J. Simpson, “Hydride precipitation in α/β zirconium alloys,” *Acta Metallurgica*, vol. 31, no. 9, pp. 1381–1391, 1983.
- [135] G. M. Han *et al.*, “Phase-field modeling of stacking structure formation and transition of δ -hydride precipitates in zirconium,” *Acta Mater*, vol. 165, pp. 528–546, Feb. 2019, doi: 10.1016/j.actamat.2018.12.009.
- [136] H. E. Weekes, N. G. Jones, T. C. Lindley, and D. Dye, “Hydride reorientation in Zircaloy-4 examined by in situ synchrotron X-ray diffraction,” *Journal of Nuclear Materials*, vol. 478, pp. 32–41, 2016, doi: 10.1016/j.jnucmat.2016.05.029.

- [137] K. B. Colas, A. T. Motta, M. R. Daymond, M. Kerr, and J. D. Almer, “Hydride platelet reorientation in zircaloy studied with synchrotron radiation diffraction,” *J ASTM Int*, vol. 8, no. 1, 2011, doi: 10.1520/JAI103033.
- [138] L. Reali, D. S. Balint, and M. R. Wenman, “Dislocation modelling of the plastic relaxation and thermal ratchetting induced by zirconium hydride precipitation,” *J Mech Phys Solids*, vol. 167, p. 104988, Oct. 2022, doi: 10.1016/j.jmps.2022.104988.
- [139] A. Tondro and H. Abdolvand, “Quantifying hydrogen concentration in the vicinity of zirconium hydrides and deformation twins,” *J Mech Phys Solids*, vol. 148, p. 104287, Mar. 2021, doi: 10.1016/j.jmps.2020.104287.
- [140] J. Cui, G. K. Shek, and Z. Wang, “Overload Fracture of Hydrided Region at Simulated Blunt Flaws in Zr-2.5Nb Pressure Tube Material,” *J Press Vessel Technol*, vol. 131, no. 4, Aug. 2009, doi: 10.1115/1.3147743.
- [141] A. V. Barashev, Q. Zhao, Q. Wang, Q. Yan, and F. Gao, “Cluster dynamics simulation of Zr hydrides formation on grain boundaries in Zr,” *Journal of Nuclear Materials*, vol. 561, p. 153521, Apr. 2022, doi: 10.1016/j.jnucmat.2022.153521.
- [142] C. M. Silva, F. Ibrahim, E. G. Lindquist, J. W. McMurray, and C. D. Bryan, “Brittle nature and the related effects of zirconium hydrides in Zircaloy-4,” *Materials Science and Engineering: A*, vol. 767, p. 138396, Nov. 2019, doi: 10.1016/j.msea.2019.138396.
- [143] D. Woo and Y. Lee, “Understanding the mechanical integrity of Zircaloy cladding with various radial and circumferential hydride morphologies via image analysis,” *Journal of Nuclear Materials*, vol. 584, Oct. 2023, doi: 10.1016/j.jnucmat.2023.154560.
- [144] R. Birch, S. Wang, V. S. Tong, and T. B. Britton, “The effect of cooling rate and grain size on hydride microstructure in Zircaloy-4,” *Journal of Nuclear Materials*, vol. 513, pp. 221–225, 2019, doi: 10.1016/j.jnucmat.2018.11.011.
- [145] W. Qin, N. A. P. Kiran Kumar, J. A. Szpunar, and J. Kozinski, “Intergranular δ -hydride nucleation and orientation in zirconium alloys,” *Acta Mater*, vol. 59, no. 18, pp. 7010–7021, Oct. 2011, doi: 10.1016/j.actamat.2011.07.054.
- [146] S. Wang, F. Giuliani, and T. Ben Britton, “Microstructure and formation mechanisms of δ -hydrides in variable grain size Zircaloy-4 studied by electron backscatter

- diffraction,” *Acta Mater*, vol. 169, pp. 76–87, May 2019, doi: 10.1016/j.actamat.2019.02.042.
- [147] T. M. Ahn, “Delayed-hydride cracking (DHC) of cladding materials: An analysis for storage and disposal of spent nuclear fuel,” *Nuclear Engineering and Design*, vol. 350, pp. 128–136, Aug. 2019, doi: 10.1016/j.nucengdes.2019.04.010.
- [148] A. W. Colldeweih, F. Fagnoni, P. Trtik, R. Zubler, M. A. Pouchon, and J. Bertsch, “Delayed hydride cracking in Zircaloy-2 with and without liner at various temperatures investigated by high-resolution neutron radiography,” *Journal of Nuclear Materials*, vol. 561, p. 153549, Apr. 2022, doi: 10.1016/j.jnucmat.2022.153549.
- [149] A. W. Colldeweih and J. Bertsch, “Effect of temperature and hydrogen concentration on the threshold stress intensity factor of radial delayed hydride cracking in fuel cladding,” *Journal of Nuclear Materials*, vol. 565, p. 153737, Jul. 2022, doi: 10.1016/j.jnucmat.2022.153737.
- [150] A. W. Colldeweih, M. G. Makowska, O. Tabai, D. F. Sanchez, and J. Bertsch, “Zirconium hydride phase mapping in Zircaloy-2 cladding after delayed hydride cracking,” *Materialia (Oxf)*, vol. 27, p. 101689, Mar. 2023, doi: 10.1016/j.mtla.2023.101689.
- [151] C. Pan *et al.*, “Effect of temperature on the delayed hydride cracking rate of Zr-2.5Nb alloy pressure tubes,” *Journal of Nuclear Materials*, vol. 588, p. 154778, Jan. 2024, doi: 10.1016/j.jnucmat.2023.154778.
- [152] D. Woo and Y. Lee, “Understanding the mechanical integrity of Zircaloy cladding with various radial and circumferential hydride morphologies via image analysis,” *Journal of Nuclear Materials*, vol. 584, p. 154560, Oct. 2023, doi: 10.1016/j.jnucmat.2023.154560.
- [153] J. Li *et al.*, “Effect of hydride precipitation on the fatigue cracking behavior in a zirconium alloy cladding tube,” *Int J Fatigue*, vol. 129, p. 105230, Dec. 2019, doi: 10.1016/j.ijfatigue.2019.105230.
- [154] J. Li *et al.*, “Uncovering the hydride orientation-mediated hoop fatigue mechanism in a zirconium alloy cladding tube,” *Int J Plast*, vol. 159, p. 103440, Dec. 2022, doi: 10.1016/j.ijplas.2022.103440.

- [155] Y. Liu, R. Thomas, C. D. Hardie, P. Frankel, and F. P. E. Dunne, “Exploring the hydride-slip interaction in zirconium alloys,” *Acta Mater*, vol. 261, p. 119388, Dec. 2023, doi: 10.1016/j.actamat.2023.119388.
- [156] L. Reali, D. S. Balint, and M. R. Wenman, “Dislocation modelling of the plastic relaxation and thermal ratchetting induced by zirconium hydride precipitation,” *J Mech Phys Solids*, vol. 167, p. 104988, Oct. 2022, doi: 10.1016/j.jmps.2022.104988.
- [157] F. P. E. Dunne, D. Rugg, and A. Walker, “Lengthscale-dependent, elastically anisotropic, physically-based hcp crystal plasticity: Application to cold-dwell fatigue in Ti alloys,” *Int J Plast*, vol. 23, no. 6, pp. 1061–1083, 2007, doi: 10.1016/j.ijplas.2006.10.013.
- [158] E. H. Lee and D. T. Liu, “Finite Strain Elastic-Plastic Theory,” in *Irreversible Aspects of Continuum Mechanics and Transfer of Physical Characteristics in Moving Fluids*, H. Parkus and L. I. Sedov, Eds., Vienna: Springer Vienna, 1968, pp. 213–222.
- [159] F. P. E. Dunne, A. Walker, and D. Rugg, “A systematic study of hcp crystal orientation and morphology effects in polycrystal deformation and fatigue,” *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 463, no. 2082, pp. 1467–1489, Jun. 2007, doi: 10.1098/rspa.2007.1833.
- [160] G. B. Gibbs, “The Thermodynamics of Creep Deformation,” *Physica Status Solidi (b)*, vol. 5, no. 3, pp. 693–696, 1964, doi: 10.1002/pssb.19640050323.
- [161] Y. Liu, W. Wan, S. El Chamaa, M. R. Wenman, C. M. Davies, and P. E. Fionn, “Creep and strain rate sensitivity in textured polycrystal Zr alloys”.
- [162] G. Laplanche, J. Bonneville, C. Varvenne, W. A. Curtin, and E. P. George, “Thermal activation parameters of plastic flow reveal deformation mechanisms in the CrMnFeCoNi high-entropy alloy,” *Acta Mater*, vol. 143, pp. 257–264, Jan. 2018, doi: 10.1016/j.actamat.2017.10.014.
- [163] N. A. Fleck and J. W. Hutchinson, “A phenomenological theory for strain gradient effects in plasticity,” *J Mech Phys Solids*, vol. 41, no. 12, pp. 1825–1857, 1993.
- [164] J. Gong, T. Benjamin Britton, M. A. Cuddihy, F. P. E. Dunne, and A. J. Wilkinson, “(a) Prismatic, (a) basal, and (c+a) slip strengths of commercially pure Zr by micro-

- cantilever tests,” *Acta Mater*, vol. 96, pp. 249–257, 2015, doi: 10.1016/j.actamat.2015.06.020.
- [165] G. I. Taylor, “The Mechanism of Plastic Deformation of Crystals. Part I. Theoretical,” *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, vol. 145, no. 855, pp. 362–387, 1934, [Online]. Available: <http://www.jstor.org/stable/2935509>
- [166] F. Hiura and M. Niewczas, “Latent hardening effect under self- and coplanar dislocation interactions in Mg single crystals,” *Scr Mater*, vol. 106, pp. 17–20, Sep. 2015, doi: 10.1016/j.scriptamat.2015.04.008.
- [167] T. Belytschko and T. Black, “Elastic crack growth in finite elements with minimal remeshing,” *Int J Numer Methods Eng*, vol. 45, no. 5, pp. 601–620, 1999, doi: 10.1002/(SICI)1097-0207(19990620)45:5<601::AID-NME598>3.0.CO;2-S.
- [168] A. Cottrell, *The Mechanical Properties of Matter*. in Wiley Series on the Science and Technology of Materials. Wiley, 1964. [Online]. Available: <https://books.google.co.uk/books?id=xCEHrV2pDjwC>
- [169] M. F. Ashby, “The deformation of plastically non-homogeneous materials,” *The Philosophical Magazine: A Journal of Theoretical Experimental and Applied Physics*, vol. 21, no. 170, pp. 399–424, Feb. 1970, doi: 10.1080/14786437008238426.
- [170] J. F. Nye, “Some geometrical relations in dislocated crystals,” *Acta Metallurgica*, vol. 1, pp. 153–162, 1953, doi: [https://doi.org/10.1016/0001-6160\(53\)90054-6](https://doi.org/10.1016/0001-6160(53)90054-6).
- [171] J. Cheng and S. Ghosh, “A crystal plasticity FE model for deformation with twin nucleation in magnesium alloys,” *Int J Plast*, vol. 67, pp. 148–170, 2015, doi: 10.1016/j.ijplas.2014.10.005.
- [172] B. Chen, J. Jiang, and F. P. E. Dunne, “Is stored energy density the primary meso-scale mechanistic driver for fatigue crack nucleation?,” *Int J Plast*, vol. 101, pp. 213–229, Feb. 2018, doi: 10.1016/j.ijplas.2017.11.005.
- [173] Z. Wang, C. Cochrane, T. Skippon, Q. Dong, and M. R. Daymond, “Dislocation evolution at a crack-tip in a hexagonal close packed metal under plane-stress conditions,” *Acta Mater*, vol. 164, pp. 25–38, 2019, doi: 10.1016/j.actamat.2018.10.022.

- [174] G. D. Nguyen, A. M. Korsunsky, and J. P. H. Belnoue, “A nonlocal coupled damage-plasticity model for the analysis of ductile failure,” *Int J Plast*, vol. 64, pp. 56–75, 2015, doi: 10.1016/j.ijplas.2014.08.001.
- [175] Y. Xu, W. Wan, and F. P. E. Dunne, “Microstructural fracture mechanics: Stored energy density at fatigue cracks,” *J Mech Phys Solids*, vol. 146, no. August 2020, p. 104209, 2021, doi: 10.1016/j.jmps.2020.104209.
- [176] P. Zhang, L. Zhang, K. P. Baxevanakis, S. Lu, L. G. Zhao, and C. Bullough, “Discrete crystal plasticity modelling of slip-controlled cyclic deformation and short crack growth under low cycle fatigue,” *Int J Fatigue*, vol. 145, Apr. 2021, doi: 10.1016/j.ijfatigue.2020.106095.
- [177] P. Zhang, L. Zhang, K. P. Baxevanakis, L. G. Zhao, and C. Bullough, “Modelling short crack propagation in a single crystal nickel-based superalloy using crystal plasticity and XFEM,” *Int J Fatigue*, vol. 136, Jul. 2020, doi: 10.1016/j.ijfatigue.2020.105594.
- [178] F. Dunne and N. Petrinic, *Introduction to Computational Plasticity*. Oxford University Press, 2007.
- [179] E. S. Fisher and C. J. Renken, “Single-Crystal Elastic Moduli and the hcp - bcc Transformation in Ti, Zr, and Hf,” *Physical Review*, vol. 135, no. 2A, pp. A482–A494, Jul. 1964, doi: 10.1103/PhysRev.135.A482.
- [180] Y. Liu, S. El Chamaa, M. R. Wenman, C. M. Davies, and F. P. E. Dunne, “Cyclic plasticity and hydrogen concentration in notched Zircaloy-4”.
- [181] J. L. Derep, S. Ibrahim, R. Roubey, and G. Fantozzi, “Deformation behaviour of Zircaloy-4 between 77 and 900 K,” *Acta Metallurgica*, vol. 28, no. 5, pp. 607–619, 1980, doi: [https://doi.org/10.1016/0001-6160\(80\)90127-3](https://doi.org/10.1016/0001-6160(80)90127-3).
- [182] A. Cuniberti and A. Picasso, “Activation volume measurement techniques: application to Zircaloy-4,” *Physica Status Solidi A Appl Res*, vol. 183, no. 2, pp. 373–379, Feb. 2001, doi: 10.1002/1521-396X(200102)183:2<373::AID-PSSA373>3.0.CO;2-N.
- [183] B. O. Poole, “Deformation mechanisms in iron-base hard facing alloys,” PhD, Imperial College London, London, 2020.

- [184] Research Inc., “Model 4150 Infrared Spot Heater Instruction Manual,” 1998
- [185] K. H. Ang, G. Chong, and Y. Li, “PID control system analysis, design, and technology,” *IEEE Transactions on Control Systems Technology*, vol. 13, no. 4, pp. 559–576, Jul. 2005, doi: 10.1109/TCST.2005.847331.
- [186] N. Quinlan, “The lumped capacitance model for transient heat transfer,” <https://www.youtube.com/watch?v=vzWxtfhndjo>.
- [187] I. Newton, “A scale of the degrees of heat,” *Philosophical Transactions of the Royal Society London*, vol. 4, pp. 572–575, 1809.
- [188] L. Boltzmann, “Ableitung des Stefanschen Gesetzes, Betreffend die Abhängigkeit der Wärmestrahlung von der Temperatur aus der Electro-magnetischen Lichttheorie,” *Annalen der Physik und Chemie*, vol. 258, pp. 291–294, 1884.
- [189] E. V. Murphy and F. Havelock, “Emissivity of zirconium alloys in air in the temperature range 100–400°C,” *Journal of Nuclear Materials*, vol. 60, no. 2, pp. 167–176, 1976, doi: [https://doi.org/10.1016/0022-3115\(76\)90163-X](https://doi.org/10.1016/0022-3115(76)90163-X).
- [190] L. R. Bunnell, G. B. Mellinger, J. L. Bates, and C. R. Hann, “High temperature properties of Zircaloy-Oxygen alloys,” Palo Alto, Mar. 1977.
- [191] T. Jailin, N. Tardif, J. Desquines, M. Coret, M.-C. Baietto, and V. Georgenthum, “Experimental study and modelling of the phase transformation of Zircaloy-4 alloy under high thermal transients,” *Mater Charact*, vol. 162, p. 110199, 2020, doi: <https://doi.org/10.1016/j.matchar.2020.110199>.
- [192] G. Ellis, “Four Types of Controllers (Chapter 6),” in *Control System Design Guide (Fourth Edition)*, G. Ellis, Ed., Boston: Butterworth-Heinemann, 2012, pp. 97–119. doi: <https://doi.org/10.1016/B978-0-12-385920-4.00006-0>.
- [193] J. S. Bradbrook, G. W. Lorimer, and N. Ridley, “The precipitation of zirconium hydride in zirconium and zircaloy-2,” *Journal of Nuclear Materials*, vol. 42, no. 2, pp. 142–160, 1972, doi: [https://doi.org/10.1016/0022-3115\(72\)90021-9](https://doi.org/10.1016/0022-3115(72)90021-9).
- [194] R. Bigger *et al.*, “A Good Practices Guide for Digital Image Correlation,” Oct. 2018. doi: 10.32720/idics/gpg.ed1.

- [195] D. Kim, J. H. Kang, and Y. Lee, “Accurate prediction of threshold stress for hydride reorientation in Zircaloy-4 with directly measured interface orientation relationship,” *Materialia (Oxf)*, vol. 21, Mar. 2022, doi: 10.1016/j.mtla.2021.101291.
- [196] W. H. Bragg, “The reflection of X-rays by crystals,” *Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character*, vol. 88, no. 605, pp. 428–438, Jul. 1913, doi: 10.1098/rspa.1913.0040.
- [197] T. Zhang, J. Jiang, B. A. Shollock, T. Ben Britton, and F. P. E. Dunne, “Slip localization and fatigue crack nucleation near a non-metallic inclusion in polycrystalline nickel-based superalloy,” *Materials Science and Engineering: A*, vol. 641, pp. 328–339, Aug. 2015, doi: 10.1016/j.msea.2015.06.070.
- [198] National Center for Biotechnology Information, “PubChem Compound Summary for CID 24247, Perchloric Acid,” National Center for Biotechnology Information.
- [199] N. Fang, R. Birch, and T. Ben Britton, “Optimizing broad ion beam polishing of zircaloy-4 for electron backscatter diffraction analysis,” *Micron*, vol. 159, p. 103268, 2022, doi: <https://doi.org/10.1016/j.micron.2022.103268>.
- [200] V. S. Tong and T. Ben Britton, “TrueEBSD: Correcting spatial distortions in electron backscatter diffraction maps,” *Ultramicroscopy*, vol. 221, p. 113130, 2021, doi: <https://doi.org/10.1016/j.ultramic.2020.113130>.
- [201] K. Elbachiri *et al.*, “Characterization of Local Strain Distribution in Zircaloy-4 and M5® Alloys,” *J ASTM Int*, vol. 5, no. 9, p. 101125, 2008, doi: 10.1520/JAI101125.
- [202] V. S. Tong, “EBSD characterisation of heterogeneous microstructures in zirconium alloys,” PhD, Imperial College London, London, 2016.
- [203] A. R. Massih and L. O. Jernkvist, “Solid state phase transformation kinetics in Zr-base alloys,” *Sci Rep*, vol. 11, no. 1, p. 7022, Mar. 2021, doi: 10.1038/s41598-021-86308-w.
- [204] A. D. Lepage, W. A. Ferris, and G. A. Ledoux, “Procedure for Adding Hydrogen to Small Sections of Zirconium Alloys,” 1998.

- [205] Z. Wang, Q. Dong, N. Guo, and M. R. Daymond, “Crack propagation path selection and plastic deformation at a crack tip in zirconium,” *Materials Science and Engineering A*, vol. 779, no. February, p. 139143, 2020, doi: 10.1016/j.msea.2020.139143.
- [206] A. I. Saville *et al.*, “Schmid factor crack propagation and tracking crystallographic texture markers of microstructural condition in direct energy deposition additive manufacturing of Ti-6Al-4V,” *Addit Manuf*, vol. 58, no. June, p. 103023, 2022, doi: 10.1016/j.addma.2022.103023.
- [207] R. Shi, J. Zheng, T. Li, H. Shou, D. Yin, and J. Rao, “Quantitative analysis of the deformation modes and cracking modes during low-cycle fatigue of a rolled AZ31B magnesium alloy: The influence of texture,” *Materials Science and Engineering A*, vol. 844, no. March, p. 143103, 2022, doi: 10.1016/j.msea.2022.143103.
- [208] X. Zeng, C. Zhang, W. Zhu, M. Zhu, Y. Wang, and X. Zhang, “Quantitative characterization of short fatigue crack and grain boundary interaction behavior in zirconium,” *Int J Fatigue*, vol. 161, no. February, p. 106894, 2022, doi: 10.1016/j.ijfatigue.2022.106894.
- [209] T. Zhai, A. J. Wilkinson, and J. W. Martin, “A crystallographic mechanism for fatigue crack propagation through grain boundaries,” vol. 48, pp. 4917–4927, 2000.
- [210] W. Wen, P. Cai, A. H. W. Ngan, and T. Zhai, “An experimental methodology to quantify the resistance of grain boundaries to fatigue crack growth in an AA2024 T351 Al-Cu Alloy,” *Materials Science and Engineering A*, vol. 666, pp. 288–296, 2016, doi: 10.1016/j.msea.2016.04.071.
- [211] A. Rovinelli, R. A. Lebensohn, and M. D. Sangid, “Influence of microstructure variability on short crack behavior through postulated micromechanical short crack driving force metrics,” *Eng Fract Mech*, vol. 138, pp. 265–288, 2015, doi: 10.1016/j.engfracmech.2015.03.001.
- [212] A. Rovinelli, M. D. Sangid, H. Proudhon, and W. Ludwig, “Using machine learning and a data-driven approach to identify the small fatigue crack driving force in polycrystalline materials,” *NPJ Comput Mater*, vol. 4, no. 1, pp. 1–10, 2018, doi: 10.1038/s41524-018-0094-7.

- [213] C. Laird, “The Influence of Metallurgical Structure on the Mechanisms of Fatigue Crack Propagation,” *ASTM International*, vol. STP415-EB, pp. 131–150, 1967.
- [214] S. S. Youssef *et al.*, “Effects of Al content and α_2 precipitation on the fatigue crack growth behaviors of binary Ti–Al alloys,” *Materials Science and Engineering A*, vol. 819, no. October 2020, p. 141513, 2021, doi: 10.1016/j.msea.2021.141513.
- [215] R. W. Hertzberg, *Deformation and fracture mechanics of engineering materials*. New York: Wiley, 1997.
- [216] L. Meng, M. Yang, X. Chen, Y. Hu, and M. Feng, “Physically short fatigue crack growth from notch described by plasticity-corrected stress intensity factor,” *Int J Mech Sci*, vol. 176, no. February, 2020, doi: 10.1016/j.ijmecsci.2020.105544.
- [217] P. Lazzarin, F. Berto, and M. Zappalorto, “Rapid calculations of notch stress intensity factors based on averaged strain energy density from coarse meshes: Theoretical bases and applications,” *Int J Fatigue*, vol. 32, no. 10, pp. 1559–1567, 2010, doi: 10.1016/j.ijfatigue.2010.02.017.
- [218] G. J. Pataky, M. D. Sangid, H. Sehitoglu, R. F. Hamilton, H. J. Maier, and P. Sofronis, “Full field measurements of anisotropic stress intensity factor ranges in fatigue,” *Eng Fract Mech*, vol. 94, pp. 13–28, 2012, doi: 10.1016/j.engfracmech.2012.06.002.
- [219] V. Karamitros, D. W. MacLachlan, and F. P. E. Dunne, “Mechanistic fatigue in Ni-based superalloy single crystals: A study of crack paths and growth rates,” *J Mech Phys Solids*, vol. 158, no. June 2021, p. 104663, 2022, doi: 10.1016/j.jmps.2021.104663.
- [220] R. Bandyopadhyay, V. Prithivirajan, A. D. Peralta, and M. D. Sangid, “Microstructure-sensitive critical plastic strain energy density criterion for fatigue life prediction across various loading regimes,” *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 476, no. 2236, 2020, doi: 10.1098/rspa.2019.0766.
- [221] H. Jahed, A. Varvani-Farahani, M. Noban, and I. Khalaji, “An energy-based fatigue life assessment model for various metallic materials under proportional and non-proportional loading conditions,” *Int J Fatigue*, vol. 29, no. 4, pp. 647–655, 2007, doi: 10.1016/j.ijfatigue.2006.07.017.

- [222] T. M. Ahn, “Delayed-hydride cracking (DHC) of cladding materials: An analysis for storage and disposal of spent nuclear fuel,” *Nuclear Engineering and Design*, vol. 350, no. April, pp. 128–136, 2019, doi: 10.1016/j.nucengdes.2019.04.010.
- [223] J. Hong, M. Park, A. A. Holston, and J. Stjärnsäter, “Threshold stress intensity factor of delayed hydride cracking in irradiated and unirradiated zircaloy-4 cladding,” *Journal of Nuclear Materials*, vol. 543, p. 152596, 2021, doi: 10.1016/j.jnucmat.2020.152596.
- [224] J. Miao, T. M. Pollock, and J. Wayne Jones, “Microstructural extremes and the transition from fatigue crack initiation to small crack growth in a polycrystalline nickel-base superalloy,” *Acta Mater*, vol. 60, no. 6–7, pp. 2840–2854, Apr. 2012, doi: 10.1016/j.actamat.2012.01.049.
- [225] D. Texier *et al.*, “Short crack propagation from cracked non-metallic inclusions in a Ni-based polycrystalline superalloy,” *Acta Mater*, vol. 165, pp. 241–258, Feb. 2019, doi: 10.1016/j.actamat.2018.11.051.
- [226] M. Waddell *et al.*, “Small fatigue crack growth behavior of Ti-6Al-4V produced via selective laser melting: In situ characterization of a 3D crack tip interactions with defects,” *Int J Fatigue*, vol. 137, Aug. 2020, doi: 10.1016/j.ijfatigue.2020.105638.
- [227] J. R. Rice and R. Thomson, “Ductile versus brittle behaviour of crystals,” *Philosophical Magazine*, vol. 29, no. 1, pp. 73–97, 1974, doi: 10.1080/14786437408213555.
- [228] P. Neumann, “The geometry of slip processes at a propagating fatigue crack II,” *Acta Metallurgica*, vol. 22, pp. 1167–1178, 1974.
- [229] P. Neumann, “Coarse slip model of fatigue,” *Acta Metallurgica*, vol. 17, pp. 1219–1225, 1969.
- [230] L. Zhang, L. G. Zhao, A. Roy, V. V. Silberschmidt, and G. McColvin, “In-situ SEM study of slip-controlled short-crack growth in single-crystal nickel superalloy,” *Materials Science and Engineering A*, vol. 742, pp. 564–572, Jan. 2019, doi: 10.1016/j.msea.2018.11.040.
- [231] J. D. Carroll, W. Abuzaid, J. Lambros, and H. Sehitoglu, “High resolution digital image correlation measurements of strain accumulation in fatigue crack growth,” *Int J Fatigue*, vol. 57, pp. 140–150, 2013, doi: 10.1016/j.ijfatigue.2012.06.010.

- [232] A. Rovinelli, M. D. Sangid, H. Proudhon, Y. Guilhem, R. A. Lebensohn, and W. Ludwig, “Predicting the 3D fatigue crack growth rate of small cracks using multimodal data via Bayesian networks: In-situ experiments and crystal plasticity simulations,” *J Mech Phys Solids*, vol. 115, pp. 208–229, 2018, doi: 10.1016/j.jmps.2018.03.007.
- [233] K. D. Pierson, J. D. Hochhalter, and A. D. Spear, “Data-Driven Correlation Analysis Between Observed 3D Fatigue-Crack Path and Computed Fields from High-Fidelity, Crystal-Plasticity, Finite-Element Simulations,” *JOM*, vol. 70, no. 7, pp. 1159–1167, Jul. 2018, doi: 10.1007/s11837-018-2884-2.
- [234] B. Lin, L. G. Zhao, and J. Tong, “A crystal plasticity study of cyclic constitutive behaviour, crack-tip deformation and crack-growth path for a polycrystalline nickel-based superalloy,” *Eng Fract Mech*, vol. 78, no. 10, pp. 2174–2192, Jul. 2011, doi: 10.1016/j.engfracmech.2011.04.006.
- [235] V. Karamitros, D. W. MacLachlan, and F. P. E. Dunne, “Modelling of short crack growth in single crystal Ni $\gamma - \gamma'$ microstructure,” *Acta Mater*, p. 118305, Nov. 2022, doi: 10.1016/j.actamat.2022.118305.
- [236] H. han Guo *et al.*, “Microscopic fatigue crack propagation model for polycrystalline alloys,” *Int J Fatigue*, vol. 170, May 2023, doi: 10.1016/j.ijfatigue.2023.107526.
- [237] F. Farukh, L. G. Zhao, R. Jiang, P. Reed, D. Proppentner, and B. A. Shollock, “Realistic microstructure-based modelling of cyclic deformation and crack growth using crystal plasticity,” *Comput Mater Sci*, vol. 111, pp. 395–405, Jan. 2016, doi: 10.1016/j.commatsci.2015.09.054.
- [238] S. Srivastava, M. Yaghoobi, and V. Sundararaghavan, “A graph-theoretic approach for multiscale modeling and prediction of crack propagation in polycrystalline materials,” *Eng Fract Mech*, vol. 241, no. November 2020, p. 107406, 2021, doi: 10.1016/j.engfracmech.2020.107406.
- [239] J. Chang, J. Q. Xu, and Y. Mutoh, “A general mixed-mode brittle fracture criterion for cracked materials,” *Eng Fract Mech*, vol. 73, no. 9, pp. 1249–1263, Jun. 2006, doi: 10.1016/j.engfracmech.2005.12.011.

- [240] T. C. Wang, “Fracture criteria for combined cleavage and dislocation emission,” *Philosophical Magazine A: Physics of Condensed Matter, Structure, Defects and Mechanical Properties*, vol. 74, no. 4, pp. 983–1001, 1996, doi: 10.1080/01418619608242172.
- [241] C. T. Sun and Z.-H. Jin, “Chapter 5 - Mixed Mode Fracture,” in *Fracture Mechanics*, C. T. Sun and Z.-H. Jin, Eds., Boston: Academic Press, 2012, pp. 105–121. doi: <https://doi.org/10.1016/B978-0-12-385001-0.00005-5>.
- [242] P. Ravi, D. Naragani, P. Kenesei, J. S. Park, and M. D. Sangid, “Direct observations and characterization of crack closure during microstructurally small fatigue crack growth via in-situ high-energy X-ray characterization,” *Acta Mater*, vol. 205, p. 116564, 2021, doi: 10.1016/j.actamat.2020.116564.
- [243] Y. Otsuka, M. Sakaguchi, Y. Kurokawa, and H. Inoue, “Strain field measurement around the crack tip in Ni-base superalloy using Digital Image Correlation,” *Transactions of the JSME (in Japanese)*, vol. 84, no. 866, pp. 18-00246-18-00246, 2018, doi: 10.1299/transjsme.18-00246.
- [244] F. Li *et al.*, “Slip band formation in plastic deformation zone at crack tip in fatigue stage II of 2xxx aluminum alloys,” *Int J Fatigue*, vol. 91, pp. 68–78, Oct. 2016, doi: 10.1016/j.ijfatigue.2016.05.014.
- [245] G. Lu, K. Miyazaki, H. Kitahara, and S. Ando, “Effects of Cerium on Crystal Orientation Dependence of Fatigue Fracture Behavior of Magnesium Single Crystals,” *Mater Trans*, vol. 63, no. 1, pp. 27–32, 2022, doi: 10.2320/matertrans.MT-L2021013.
- [246] S. Hémery *et al.*, “Strain localization and fatigue crack formation at (0001) twist boundaries in titanium alloys,” *Acta Mater*, vol. 219, Oct. 2021, doi: 10.1016/j.actamat.2021.117227.
- [247] D. L. McDowell, “Basic issues in the mechanics of high cycle metal fatigue,” *Int J Fract*, vol. 80, pp. 103–145, 1996.
- [248] M. Liu, Y. Gan, D. A. H. Hanaor, B. Liu, and C. Chen, “An improved semi-analytical solution for stress at round-tip notches,” *Eng Fract Mech*, vol. 149, pp. 134–143, 2015, doi: 10.1016/j.engfracmech.2015.10.004.

- [249] T. L. Anderson, *Fracture Mechanics - Fundamentals and Applications*. CRC Press, Taylor & Francis Group, 1996.
- [250] P. A. S. Reed, X. D. Wu, and I. Sinclair, “Fatigue crack path prediction in UDIMET 720 nickel-based alloy single crystals,” *Metall Mater Trans A Phys Metall Mater Sci*, vol. 31, no. 1, pp. 109–123, 2000, doi: 10.1007/s11661-000-0058-6.
- [251] F. Briffod, T. Shiraiwa, M. Enoki, and S. Emura, “Effect of macrozones on fatigue crack initiation and propagation mechanisms in a forged ti-6Al-4V alloy under fully-reversed condition,” *Materialia (Oxf)*, vol. 22, no. March, p. 101401, 2022, doi: 10.1016/j.mtla.2022.101401.
- [252] W. Wan and F. P. E. Dunne, “Microstructure-interacting short crack growth in blocky alpha Zircaloy-4,” *Int J Plast*, vol. 130, no. February, p. 102711, 2020, doi: 10.1016/j.ijplas.2020.102711.
- [253] M. Su, L. Xu, C. Peng, Y. Han, and L. Zhao, “Fatigue short crack growth, model and EBSD characterization of marine steel welding joint,” *Int J Fatigue*, vol. 156, no. September 2021, p. 106689, 2022, doi: 10.1016/j.ijfatigue.2021.106689.
- [254] V. Doquet and G. Bertolino, “Local approach to fatigue cracks bifurcation,” *Int J Fatigue*, vol. 30, no. 5, pp. 942–950, May 2008, doi: 10.1016/j.ijfatigue.2007.06.001.
- [255] P. A. Sabnis, S. Forest, and J. Cormier, “Microdamage modelling of crack initiation and propagation in FCC single crystals under complex loading conditions,” *Comput Methods Appl Mech Eng*, vol. 312, pp. 468–491, Dec. 2016, doi: 10.1016/j.cma.2016.04.018.
- [256] S. Suzuki, M. Sakaguchi, M. Domen, T. Karato, and K. Suzuki, “Temperature and ΔK dependence of grain boundary effect on fatigue crack propagation in a two-dimensional polycrystalline Ni-base superalloy,” *Acta Mater*, vol. 240, Nov. 2022, doi: 10.1016/j.actamat.2022.118288.
- [257] K. Nikolakopoulos, J. P. Crete, and P. Longere, “Volume averaging based integration method in the context of XFEM-cohesive zone model coupling,” *Mech Res Commun*, vol. 104, Mar. 2020, doi: 10.1016/j.mechrescom.2020.103485.
- [258] H. L. Ewalds and R. J. H. Wanhill, *Fracture mechanics*, 4th ed. London: Edward Arnold, 1989.

- [259] B. V. Cockeram and B. F. Kammenzind, “The increase in fatigue crack growth rates observed for Zircaloy-4 in a PWR environment,” *Journal of Nuclear Materials*, vol. 499, pp. 111–125, 2018, doi: 10.1016/j.jnucmat.2017.11.015.
- [260] A. Cattivelli, M. J. Roy, M. G. Burke, J. Dhers, T. L. Lee, and J. A. Francis, “Internal stresses in a clad pressure vessel steel during post weld heat treatment and their relevance to underclad cracking,” *International Journal of Pressure Vessels and Piping*, vol. 193, p. 104448, 2021, doi: <https://doi.org/10.1016/j.ijpvp.2021.104448>.
- [261] Z. Zheng, P. Zhao, M. Zhan, S. Shen, Y. Wang, and M. W. Fu, “The roles of rise and fall time in load shedding and strain partitioning under the dwell fatigue of titanium alloys with different microstructures,” *Int J Plast*, vol. 149, Feb. 2022, doi: 10.1016/j.ijplas.2021.103161.
- [262] C. T. Poon, R. A. Barrett, and S. B. Leen, “Global and local modeling for inter-wire fretting in multi-wire copper conductors,” *Fatigue Fract Eng Mater Struct*, vol. 45, no. 6, pp. 1618–1634, Jun. 2022, doi: 10.1111/ffe.13683.
- [263] M. T. Milan, D. Spinelli, W. W. Bose Filho, M. F. V Montezuma, and V. Tita, “Failure analysis of a SAE 4340 steel locking bolt,” *Eng Fail Anal*, vol. 11, no. 6, pp. 915–924, 2004, doi: <https://doi.org/10.1016/j.engfailanal.2003.12.003>.
- [264] M. D. Sangid, T. Ezaz, H. Sehitoglu, and I. M. Robertson, “Energy of slip transmission and nucleation at grain boundaries,” *Acta Mater*, vol. 59, no. 1, pp. 283–296, Jan. 2011, doi: 10.1016/j.actamat.2010.09.032.
- [265] P. B. Chowdhury, H. Sehitoglu, R. G. Rateick, and H. J. Maier, “Modeling fatigue crack growth resistance of nanocrystalline alloys,” *Acta Mater*, vol. 61, no. 7, pp. 2531–2547, Apr. 2013, doi: 10.1016/j.actamat.2013.01.030.
- [266] P. B. Chowdhury, H. Sehitoglu, and R. G. Rateick, “Predicting fatigue resistance of nano-twinned materials: Part i - Role of cyclic slip irreversibility and Peierls stress,” *Int J Fatigue*, vol. 68, pp. 277–291, 2014, doi: 10.1016/j.ijfatigue.2014.05.014.
- [267] P. B. Chowdhury, H. Sehitoglu, and R. G. Rateick, “Predicting fatigue resistance of nano-twinned materials: Part II - Effective threshold stress intensity factor range,” *Int J Fatigue*, vol. 68, pp. 292–301, 2014, doi: 10.1016/j.ijfatigue.2014.06.006.

- [268] P. R. Dawson, “Computational crystal plasticity,” *Int J Solids Struct*, vol. 37, pp. 115–130, 2000.
- [269] J. Shen, S. Kotha, R. Noraas, V. Venkatesh, and S. Ghosh, “Developing parametrically upscaled constitutive and crack nucleation models for the α/β Ti64 alloy,” *Int J Plast*, vol. 151, Apr. 2022, doi: 10.1016/j.ijplas.2021.103182.
- [270] N. Grilli, E. Tarleton, and A. C. F. Cocks, “Coupling a discrete twin model with cohesive elements to understand twin-induced fracture,” *Int J Fract*, vol. 227, no. 2, pp. 173–192, 2021, doi: 10.1007/s10704-020-00504-9.
- [271] J. J. C. Remmers, R. de Borst, and A. Needleman, “The simulation of dynamic crack propagation using the cohesive segments method,” *J Mech Phys Solids*, vol. 56, no. 1, pp. 70–92, 2008, doi: 10.1016/j.jmps.2007.08.003.
- [272] Q. Zhao, M. Abdel Wahab, Y. Ling, and Z. Liu, “Fatigue crack propagation across grain boundary of Al-Cu-Mg bicrystal based on crystal plasticity XFEM and cohesive zone model,” *J Mater Sci Technol*, vol. 126, pp. 275–287, Nov. 2022, doi: 10.1016/j.jmst.2022.03.020.
- [273] J. Dolbow, N. Moës, and T. Belytschko, “Discontinuous enrichment in finite elements with a partition of unity method,” *Finite elements in analysis and design*, vol. 36, no. 3, pp. 235–260, 2000, doi: 10.1016/S0168-874X(00)00035-4.
- [274] S. A. Silling, “Reformulation of elasticity theory for discontinuities and long-range forces,” *J Mech Phys Solids*, vol. 48, no. 1, pp. 175–209, 2000, doi: [https://doi.org/10.1016/S0022-5096\(99\)00029-0](https://doi.org/10.1016/S0022-5096(99)00029-0).
- [275] A. Bamgboye, T. A. Haynes, and M. R. Wenman, “Predicting crack patterns in SiC-based cladding for LWR applications using peridynamics,” in *Procedia Structural Integrity*, Elsevier B.V., 2020, pp. 1520–1535. doi: 10.1016/j.prostr.2020.10.125.
- [276] O. Karpenko, S. Oterkus, and E. Oterkus, “Titanium alloy corrosion fatigue crack growth rates prediction: Peridynamics based numerical approach,” *Int J Fatigue*, vol. 162, p. 107023, 2022, doi: <https://doi.org/10.1016/j.ijfatigue.2022.107023>.

- [277] K. Hong, S. Oterkus, and E. Oterkus, “Peridynamic analysis of fatigue crack growth in fillet welded joints,” *Ocean Engineering*, vol. 235, p. 109348, Sep. 2021, doi: 10.1016/J.OCEANENG.2021.109348.
- [278] D. Wang, L. Zhao, L. Xu, Y. Han, and K. Hao, “A microstructure-based study of irradiation hardening in stainless steel: Experiment and phase field modeling,” *Journal of Nuclear Materials*, vol. 569, p. 153940, 2022, doi: <https://doi.org/10.1016/j.jnucmat.2022.153940>.
- [279] W. Ai, B. Wu, and E. Martínez-Pañeda, “A coupled phase field formulation for modelling fatigue cracking in lithium-ion battery electrode particles,” *J Power Sources*, vol. 544, p. 231805, 2022, doi: <https://doi.org/10.1016/j.jpowsour.2022.231805>.
- [280] D. Montes de Oca Zapiain, J. A. Stewart, and R. Dingreville, “Accelerating phase-field-based microstructure evolution predictions via surrogate models trained by machine learning methods,” *NPJ Comput Mater*, vol. 7, no. 1, p. 3, 2021, doi: 10.1038/s41524-020-00471-8.
- [281] H. Zhou, Y. Suzuki, M. Kinefuchi, S. Schmauder, K. Dogahe, and K. Shibamura, “Bridging strategy between microscopic and macroscopic crack growth simulations to predict fatigue strength of steels,” *Int J Fatigue*, vol. 168, Mar. 2023, doi: 10.1016/j.ijfatigue.2022.107386.
- [282] H. Zhou, Z. Liu, M. Kinefuchi, and K. Shibamura, “Multiscale modelling strategy for predicting fatigue lives and limits of steels based on a generalised evaluation method of grain boundaries effects,” *Int J Fatigue*, vol. 158, May 2022, doi: 10.1016/j.ijfatigue.2022.106749.
- [283] H. Zhou, Z. Liu, S. Kikuchi, and K. Shibamura, “Analysis of fatigue performance of austenitic stainless steels with bimodal harmonic structures based on multiscale model simulations,” *Mater Des*, p. 111657, Feb. 2023, doi: 10.1016/j.matdes.2023.111657.
- [284] H. Ito, Y. Suzuki, H. Nishikawa, M. Kinefuchi, M. Enoki, and K. Shibamura, “Multiscale model prediction of ferritic steel fatigue strength based on microstructural information, tensile properties, and loading conditions (no adjustable material constants),” *Int J Mech Sci*, vol. 170, Mar. 2020, doi: 10.1016/j.ijmecsci.2019.105339.

- [285] N. Recho, *Fracture Mechanics and Crack Growth*, 1. Aufl. London Hoboken, N.J: Wiley-ISTE, 2012.
- [286] E. E. Gdoutos, *Fracture mechanics criteria and applications* . in Engineering application of fracture mechanics ; v.10. Dordrecht: Kluwer Academic Publishers, 1990.
- [287] J. Pan and S. H. Lin, *Fracture Mechanics and Fatigue Crack Propagation*, 1st ed. Elsevier Inc., 2005. doi: 10.1016/B978-075067719-6/50007-5.
- [288] J. E. Allison, “The measurement of crack tip stresses by X-ray diffraction,” Ohio, 1978.
- [289] A. J. Allen, M. T. Hutchings, C. G. Windsor, and C. Andreani, “Neutron diffraction methods for the study of residual stress fields,” *Adv Phys*, vol. 34, no. 4, pp. 445–473, Jan. 1985, doi: 10.1080/00018738500101791.
- [290] S. Liang, Y. Zhu, M. Huang, and Z. Li, “Studying dislocation-induced shielding effect on the crack-tip in polycrystal by discrete dislocation dynamics,” *Int J Solids Struct*, vol. 156–157, pp. 148–162, 2019, doi: 10.1016/j.ijsolstr.2018.08.012.
- [291] C. S. Pande and R. Goswami, “Dislocation emission and crack dislocation interactions,” *Metals (Basel)*, vol. 10, no. 4, pp. 1–14, 2020, doi: 10.3390/met10040473.
- [292] C. G. Park, C. S. Lee, and Y. W. Chang, “In Situ TEM Observations of Crack Tip Dislocation Behaviors Under a Mixed Mode Loading,” in *Mechanical Behaviour of Materials VI*, Kyoto, Japan, 1991, pp. 3–9. doi: <https://doi.org/10.1016/B978-0-08-037890-9.50395-1>.
- [293] R. Goswami, “Dislocation Crack Interactions and Lattice Rotation Under Uniaxial and Cyclic Loading,” Naval Research Laboratory, Washington, DC, 2018.
- [294] B. A. Auld, *Acoustic Fields And Waves In Solids Volume I*, 1st ed. New York: Wiley, 1973.
- [295] S. Joseph, I. Bantounas, T. C. Lindley, and D. Dye, “Slip transfer and deformation structures resulting from the low cycle fatigue of near-alpha titanium alloy Ti-6242Si,” *Int J Plast*, vol. 100, pp. 90–103, 2018, doi: 10.1016/j.ijplas.2017.09.012.

- [296] M. Connolly *et al.*, “In situ high energy X-ray diffraction measurement of strain and dislocation density ahead of crack tips grown in hydrogen,” *Acta Mater*, vol. 180, pp. 272–286, Nov. 2019, doi: 10.1016/j.actamat.2019.09.020.
- [297] L. A. James, “Environmentally aggravated fatigue cracking of Zircaloy-2,” *Nuclear Applications and Technology*, vol. 9, no. 2, pp. 260–267, 1970, doi: 10.13182/nt70-a28815.
- [298] S. B. Wisner, M. B. Reynolds, and R. B. Adamson, “Fatigue behaviour of irradiated and unirradiated Zircaloy and Zirconium,” in *Tenth International Symposium on Zirconium in the Nuclear Industry*, A. M. Garde and R. E. Bradley, Eds., Baltimore: ASTM, Jun. 1993, pp. 499–520.
- [299] W. A. Logsdon and P. K. Liaw, “Tensile, Fracture Toughness, and Fatigue Crack Growth Rate Properties of Zirconium Copper,” Springer-Verlag New York Inc, 1987.
- [300] J. J. Kearns, “Thermal expansion and preferred orientation in Zircaloy (LWBR Development Program),” United States, 1965. [Online]. Available: <https://www.osti.gov/biblio/6736579>
- [301] Y. Murakami, “4 - Effect of size and geometry of small defects on the fatigue limit,” in *Metal Fatigue (Second Edition)*, Y. Murakami, Ed., Academic Press, 2019, pp. 39–59. doi: <https://doi.org/10.1016/B978-0-12-813876-2.00004-2>.
- [302] A. R. Warwick *et al.*, “Dislocation density transients and saturation in irradiated zirconium,” *Int J Plast*, p. 103590, May 2023, doi: 10.1016/j.ijplas.2023.103590.
- [303] T. Seymour *et al.*, “Evolution of dislocation structure in neutron irradiated Zircaloy-2 studied by synchrotron x-ray diffraction peak profile analysis,” *Acta Mater*, vol. 126, pp. 102–113, Mar. 2017, doi: 10.1016/j.actamat.2016.12.031.
- [304] M. Topping *et al.*, “Investigating the thermal stability of irradiation-induced damage in a zirconium alloy with novel in situ techniques,” *Acta Mater*, vol. 145, pp. 255–263, Feb. 2018, doi: 10.1016/j.actamat.2017.11.051.
- [305] T. Torimaru, T. Yasuda, and M. Nakatsuka, “Changes in mechanical properties of irradiated Zircaloy-2 fuel cladding due to short term annealing,” 1996.

- [306] L. M. Howe and W. R. Thomas, “The effect of neutron irradiation on the properties of Zircaloy-2,” *Journal of Nuclear Materials*, pp. 248–260, 1960.
- [307] T. Yasuda, M. Nakatsuka, and K. Yamashita, “Deformation and Fracture Properties of Neutron-irradiated Recrystallized Zircaloy-2 Cladding under Uniaxial Tension,” in *Zirconium in the Nuclear Industry*, West Conshohocken, PA: ASTM International, 1987, pp. 734–747. doi: 10.1520/STP28156S.
- [308] F. Onimus, L. Dupuy, and F. Mompiau, “In situ TEM observation of interactions between gliding dislocations and prismatic loops in Zr-ion irradiated zirconium alloys,” in *Progress in Nuclear Energy*, May 2012, pp. 77–85. doi: 10.1016/j.pnucene.2011.10.005.
- [309] M. S. Weschler, “The Inhomogeneity of Plastic Deformation,” *ASM, Metals Park, Ohio*, pp. 19–52, 1973.
- [310] J. V. Sharp, “Deformation of neutron-irradiated copper single crystals,” *Philosophical Magazine*, vol. 16, no. 139, pp. 77–96, 1967, doi: 10.1080/14786436708229258.
- [311] J. Drouet, L. Dupuy, F. Onimus, and F. Mompiau, “A direct comparison between in-situ transmission electron microscopy observations and Dislocation Dynamics simulations of interaction between dislocation and irradiation induced loop in a zirconium alloy,” *Scr Mater*, vol. 119, pp. 71–75, Jul. 2016, doi: 10.1016/j.scriptamat.2016.03.029.
- [312] M. Sauzay and M. O. Moussa, “Prediction of grain boundary stress fields and microcrack initiation induced by slip band impingement,” in *International Journal of Fracture*, Nov. 2013, pp. 215–240. doi: 10.1007/s10704-013-9878-4.
- [313] W. Liu, L. Chen, L. Yu, J. Fu, and H. Duan, “Continuum modeling of dislocation channels in irradiated metals based on stochastic crystal plasticity,” *Int J Plast*, vol. 151, Apr. 2022, doi: 10.1016/j.ijplas.2021.103211.
- [314] P. Lin, J. Nie, and M. Liu, “Multiscale crystal plasticity finite element model for investigating the irradiation hardening and defect evolution mechanism of A508-3 steel,” *Nuclear Materials and Energy*, vol. 32, Sep. 2022, doi: 10.1016/j.nme.2022.101214.

- [315] X. Xiao, S. Li, and L. Yu, “Effect of irradiation damage and indenter radius on pop-in and indentation stress-strain relations: Crystal plasticity finite element simulation,” *Int J Mech Sci*, vol. 199, Jun. 2021, doi: 10.1016/j.ijmecsci.2021.106430.
- [316] A. Xu, T. Wei, and D. Bhattacharyya, “The effect of strain rate and orientation on He ion irradiated Ni single crystals - An in situ micro-tensile study,” *Int J Plast*, vol. 126, Mar. 2020, doi: 10.1016/j.ijplas.2019.11.006.
- [317] N. H. Packan, K. Farrell, and J. O. Stiegler, “Correlation of neutron and heavy ion damage: I. The influence of dose rate and injected helium on swelling in pure nickel,” *Journal of Nuclear Materials*, vol. 78, pp. 143–155, 1978.
- [318] Y. Liu, S. El Chamaa, M. R. Wenman, C. M. Davies, and F. P. E. Dunne, “Hydrogen concentration and hydrides in Zircaloy-4 during cyclic thermomechanical loading,” *Acta Mater*, vol. 221, Dec. 2021, doi: 10.1016/j.actamat.2021.117368.
- [319] Y. Xu, “A non-local methodology for geometrically necessary dislocations and application to crack tips,” *Int J Plast*, vol. 140, no. March, p. 102970, 2021, doi: 10.1016/j.ijplas.2021.102970.
- [320] R. A. Holt, A. R. Causey, N. Christodoulou, M. Griffiths, E. T. C. Ho, and C. H. Woo, “Non-linear irradiation growth of cold-worked Zircaloy-2,” *ASTM Special Technical Publication*, vol. 1295, pp. 623–637, 1996, doi: 10.1520/stp16193s.
- [321] C. Dai, L. Balogh, Z. Yao, and M. R. Daymond, “Atomistic simulations of the formation of <c>-type dislocation loops in α -zirconium,” *Journal of Nuclear Materials*, vol. 478, pp. 125–134, Sep. 2016, doi: 10.1016/j.jnucmat.2016.06.002.
- [322] C. Zheng, S. Maloy, and D. Kaoumi, “Effect of dose on irradiation-induced loop density and Burgers vector in ion-irradiated ferritic/martensitic steel HT9,” *Philosophical Magazine*, vol. 98, no. 26, pp. 2440–2456, Sep. 2018, doi: 10.1080/14786435.2018.1490825.
- [323] K. Nordlund *et al.*, “Improving atomic displacement and replacement calculations with physically realistic damage models,” *Nat Commun*, vol. 9, no. 1, p. 1084, Mar. 2018, doi: 10.1038/s41467-018-03415-5.

- [324] L. K. Béland *et al.*, “Features of primary damage by high energy displacement cascades in concentrated Ni-based alloys,” *J Appl Phys*, vol. 119, no. 8, Feb. 2016, doi: 10.1063/1.4942533.
- [325] K. Nordlund *et al.*, “Primary radiation damage in materials,” 2015.
- [326] R. Kathiria, D. E. Wolf, R. Raj, and M. Jongmanns, “Frenkel pairs cause elastic softening in zirconia: Theory and experiments,” *New J Phys*, vol. 23, no. 5, May 2021, doi: 10.1088/1367-2630/abf704.
- [327] P. T. Heald and K. M. Miller, “Point defect sink strengths in irradiated materials,” *Journal of Nuclear Materials*, vol. 66, no. 1–2, pp. 107–111, Apr. 1977, doi: 10.1016/0022-3115(77)90139-8.
- [328] P. M. Derlet and S. L. Dudarev, “Microscopic structure of a heavily irradiated material,” *Phys Rev Mater*, vol. 4, no. 2, p. 023605, Feb. 2020, doi: 10.1103/PhysRevMaterials.4.023605.
- [329] R. E. Stoller and L. K. Mansur, “The influence of displacement rate on damage accumulation during the point defect transient in irradiated materials,” Oak Ridge National Laboratory, 1990.
- [330] E. Aydogan, O. El-Atwani, M. Li, and S. A. Maloy, “In-situ observation of nano-oxide and defect evolution in 14YWT alloys,” *Mater Charact*, vol. 170, Dec. 2020, doi: 10.1016/j.matchar.2020.110686.
- [331] G. J. C. Carpenter, R. H. Zee, and A. Rogerson, “Irradiation growth of zirconium single crystals: a review,” *Journal of Nuclear Materials*, vol. 159, pp. 86–100, 1988.
- [332] B. L. Eyre and D. M. Maher, “Neutron irradiation damage in molybdenum part V. Mechanisms of vacancy and interstitial loop growth during post-irradiation annealing,” *Philosophical Magazine*, vol. 24, no. 190, pp. 767–797, 1971, doi: 10.1080/14786437108217049.
- [333] A. C. P. Jain, P. A. Burr, and D. R. Trinkle, “First-principles calculations of solute transport in zirconium: Vacancy-mediated diffusion with metastable states and interstitial diffusion,” *Phys Rev Mater*, vol. 3, no. 3, p. 033402, Mar. 2019, doi: 10.1103/PhysRevMaterials.3.033402.

- [334] F. Onimus and J. L. Béchade, “Radiation Effects in Zirconium Alloys,” in *Comprehensive Nuclear Materials*, Elsevier, 2012, pp. 1–31. doi: 10.1016/B978-0-08-056033-5.00064-1.
- [335] R. Thetford, “Theory of defect interactions in metals,” PhD, University of Oxford, Oxford, 1989.
- [336] J. P. Hirth and J. Lothe, *Theory of dislocations*. in McGraw-Hill series in materials science and engineering. New York: McGraw-Hill, 1967.
- [337] J. Ribis *et al.*, “Experimental study and numerical modelling of the irradiation damage recovery in zirconium alloys,” *Journal of Nuclear Materials*, vol. 403, no. 1–3, pp. 135–146, Aug. 2010, doi: 10.1016/j.jnucmat.2010.06.012.
- [338] R. B. Adamson and W. L. Bell, “Microstructure and mechanical behaviour of materials,” in *International Symposium*, Xian, China, 1985, p. 237.
- [339] E. Eidelpes, L. F. Ibarra, and R. A. Medina, “Probabilistic Assessment of Peak Cladding Hoop Stress and Hydrogen Content of PWR SNF Rod Cladding,” *Nucl Technol*, vol. 205, no. 8, pp. 1095–1118, Aug. 2019, doi: 10.1080/00295450.2019.1575127.
- [340] R. Zino, R. Chosson, M. Ollivier, E. Serris, and L. Favergeon, “Parallel mechanism of growth of the oxide and α -Zr(O) layers on Zircaloy-4 oxidized in steam at high temperatures,” *Corros Sci*, vol. 179, Feb. 2021, doi: 10.1016/j.corsci.2020.109178.
- [341] F. Long, D. Kerr, G. Domizzi, Q. Wang, and M. R. Daymond, “Microstructure characterization of a hydride blister in Zircaloy-4 by EBSD and TEM,” *Acta Mater*, vol. 129, pp. 450–461, May 2017, doi: 10.1016/j.actamat.2017.03.016.
- [342] National Academies of Sciences Engineering and Medicine, *Reducing the Use of Highly Enriched Uranium in Civilian Research Reactors*. Washington, DC: The National Academies Press, 2016. doi: 10.17226/21818.
- [343] M. Griffiths, “Effect of Neutron Irradiation on the Mechanical Properties, Swelling and Creep of Austenitic Stainless Steels,” *Materials*, vol. 14, no. 10, p. 2622, May 2021, doi: 10.3390/ma14102622.

- [344] T. R. Allen, J. I. Cole, H. Tsai, S. Ukai, S. Mizuta, and T. Yoshitake, “The effect of low dose rate irradiation on the swelling of 12% cold-worked 316 stainless steel,” Newport Beach, Aug. 1999.
- [345] Q. Yu, S. Chatterjee, K. J. Roche, G. Po, and J. Marian, “Coupling crystal plasticity and stochastic cluster dynamics models of irradiation damage in tungsten,” *Model Simul Mat Sci Eng*, vol. 29, no. 5, Jul. 2021, doi: 10.1088/1361-651X/ac01ba.
- [346] Y. Li, G. Po, and N. Ghoniem, “Coupled cluster-dislocation dynamics of irradiation-induced defects,” *Materialia (Oxf)*, vol. 14, p. 100891, Dec. 2020, doi: 10.1016/j.mtla.2020.100891.
- [347] R. J. H. Wanhill, D. A. Ryder, and T. J. Davies, “Metallographic studies of fatigue in 20% cold-worked zircaloy-2 containing zirconium hydride,” *Journal of Nuclear Materials*, vol. 43, no. 2, pp. 75–85, May 1972, doi: 10.1016/0022-3115(72)90141-9.
- [348] M. Kerr, M. R. Daymond, R. A. Holt, and J. D. Almer, “Strain evolution of zirconium hydride embedded in a Zircaloy-2 matrix,” *Journal of Nuclear Materials*, vol. 380, no. 1–3, pp. 70–75, Oct. 2008, doi: 10.1016/j.jnucmat.2008.07.004.
- [349] E. Garlea *et al.*, “Hydride-Phase Formation and its Influence on Fatigue Crack Propagation Behavior in a Zircaloy-4 Alloy,” *Metallurgical and Materials Transactions A*, vol. 41, no. 11, pp. 2816–2828, Nov. 2010, doi: 10.1007/s11661-010-0342-z.
- [350] J. Li *et al.*, “Effect of hydride precipitation on the fatigue cracking behavior in a zirconium alloy cladding tube,” *Int J Fatigue*, vol. 129, p. 105230, Dec. 2019, doi: 10.1016/j.ijfatigue.2019.105230.
- [351] M. Taherijam, S. Marashi, A. Tondro, and H. Abdolvand, “On the effects of transformation strain induced by hydride precipitation,” *Acta Mater*, vol. 261, p. 119356, Dec. 2023, doi: 10.1016/j.actamat.2023.119356.
- [352] R. M. Birch, J. O. Douglas, and T. Ben Britton, “Characterization of local deformation around hydrides in Zircaloy-4 using conventional and high angular resolution electron backscatter diffraction,” *Mater Charact*, vol. 202, p. 112988, Aug. 2023, doi: 10.1016/j.matchar.2023.112988.

- [353] Y. Zhang, L. You, X. Li, J. Zhou, and X. Song, “In-situ investigation of the fatigue crack initiation and propagation behavior of Zircaloy-4 with different hydrogen contents at RT and 300 °C,” *Journal of Nuclear Materials*, vol. 532, p. 152065, Apr. 2020, doi: 10.1016/j.jnucmat.2020.152065.
- [354] B. V. Cockeram and B. F. Kammenzind, “The increase in fatigue crack growth rates observed for Zircaloy-4 in a PWR environment,” *Journal of Nuclear Materials*, vol. 499, pp. 111–125, Feb. 2018, doi: 10.1016/j.jnucmat.2017.11.015.
- [355] R. L. Eadie, D. R. Metzger, and M. Léger, “The thermal ratchetting of hydrogen in zirconium-niobium — An illustration using finite element modelling,” *Scripta Metallurgica et Materialia*, vol. 29, no. 3, pp. 335–340, Aug. 1993, doi: 10.1016/0956-716X(93)90509-Q.
- [356] H. Li, M. Wen, G. Chen, W. Yu, and X. Chen, “Constitutive modeling for the anisotropic uniaxial ratcheting behavior of Zircaloy-4 alloy at room temperature,” *Journal of Nuclear Materials*, vol. 443, no. 1–3, pp. 152–160, Nov. 2013, doi: 10.1016/j.jnucmat.2013.06.052.
- [357] G. Chen, C. Xu, H. Qu, and X. Chen, “Ratcheting behavior of zirconium alloy tubes under combined cyclic axial load and internal pressure at 350 °C,” *Journal of Nuclear Materials*, vol. 491, pp. 138–148, Aug. 2017, doi: 10.1016/j.jnucmat.2017.05.006.
- [358] P. D. Honniball, L. Cogez, and C. F. Gee, “Influence of Hydrides upon the Fatigue Initiation Behavior of Irradiated Zircaloy-2,” in *Zirconium in the Nuclear Industry: 19th International Symposium*, ASTM International, 2021, pp. 365–384. doi: 10.1520/stp162220190026.
- [359] C. Lee and Y. Lee, “Simulation of hydrogen diffusion along the axial direction in zirconium cladding tube during dry storage,” *Journal of Nuclear Materials*, vol. 579, Jun. 2023, doi: 10.1016/j.jnucmat.2023.154352.
- [360] B. F. Kammenzind, D. G. Franklin, W. J. Duffin, and H. R. Peters, “Hydrogen pickup and redistribution in alpha-annealed Zircaloy-4,” West Mifflin, PA, Jun. 1996. doi: 10.2172/244661.

- [361] S. Alekseeva *et al.*, “Grain boundary mediated hydriding phase transformations in individual polycrystalline metal nanoparticles,” *Nat Commun*, vol. 8, no. 1, p. 1084, Oct. 2017, doi: 10.1038/s41467-017-00879-9.
- [362] X. Zeng, C. Zhang, W. Zhu, M. Zhu, Y. Wang, and X. Zhang, “Quantitative characterization of short fatigue crack and grain boundary interaction behavior in zirconium,” *Int J Fatigue*, vol. 161, Aug. 2022, doi: 10.1016/j.ijfatigue.2022.106894.
- [363] H. Bhadeshia, *Geometry of Crystals, Polycrystals, and Phase Transformations*, 1st ed. CRC Press, Taylor & Francis, 2018.
- [364] G. J. C. Carpenter, “The dilatational misfit of zirconium hydrides precipitated in zirconium,” *Journal of Nuclear Materials*, vol. 48, no. 3, pp. 264–266, Oct. 1973, doi: 10.1016/0022-3115(73)90022-6.
- [365] P. A. T. Olsson, A. R. Massih, J. Blomqvist, A.-M. Alvarez Holston, and C. Bjerkén, “Ab initio thermodynamics of zirconium hydrides and deuterides,” *Comput Mater Sci*, vol. 86, pp. 211–222, Apr. 2014, doi: 10.1016/j.commatsci.2014.01.043.
- [366] I. A. Khan, A. C. F. Cocks, and J. Chattopadhyay, “A study of hydride precipitation in zirconium,” *Mechanics of Materials*, vol. 155, no. January, p. 103773, 2021, doi: 10.1016/j.mechmat.2021.103773.
- [367] P. Saha *et al.*, “Issues and future direction of thermal-hydraulics research and development in nuclear power reactors,” *Nuclear Engineering and Design*, vol. 264, pp. 3–23, 2013, doi: 10.1016/j.nucengdes.2012.07.023.
- [368] M. Nakatsuka, T. Kubo, Y. I. Hayashi, and M. T. Kubo, “Fatigue Behavior of Neutron Irradiated Zircaloy-2 Fuel Cladding Tubes,” American Society for Testing and Materials, 1991. [Online]. Available: www.aslm.org
- [369] A. Soniak, S. Lansart, J. Royer, J.-P. Mardon, and N. Waeckel, “Irradiation Effect on Fatigue Behavior of Zircaloy-4 Cladding Tubes,” American Society for Testing and Materials, 1994. [Online]. Available: www.astm.org
- [370] K. Zhang, X. Wu, and C. H. J. Davies, “Effect of microtexture on short crack propagation in two-phase titanium alloys,” *Int J Fatigue*, vol. 104, pp. 206–220, 2017, doi: 10.1016/j.ijfatigue.2017.07.022.

- [371] L. Li, Z. Zhang, and G. Shen, “Influence of grain size on fatigue crack propagation and acoustic emission features in commercial-purity zirconium,” *Materials Science and Engineering A*, vol. 636, pp. 35–42, 2015, doi: 10.1016/j.msea.2015.03.046.
- [372] W. Liu, L. Chen, L. Yu, J. Fu, and H. Duan, “Continuum modeling of dislocation channels in irradiated metals based on stochastic crystal plasticity,” *Int J Plast*, vol. 151, Apr. 2022, doi: 10.1016/j.ijplas.2021.103211.
- [373] J. Shi *et al.*, “Experiments and/or crystal plasticity finite element modeling of the mechanical properties of pristine and irradiated tungsten single crystal,” *Int J Plast*, vol. 154, Jul. 2022, doi: 10.1016/j.ijplas.2022.103293.
- [374] A. Patra and D. L. McDowell, “Crystal plasticity-based constitutive modelling of irradiated bcc structures,” *Philosophical Magazine*, vol. 92, no. 7, pp. 861–887, Mar. 2012, doi: 10.1080/14786435.2011.634855.
- [375] N. M. Jose, M. K. Samal, P. V Durgaprasad, A. Alankar, and B. K. Dutta, “Crystal Plasticity Modelling of Neutron Irradiation Effects on the Flow and Damage Behaviour of Zircaloy-4,” in *Advances in Structural Integrity*, K. Jonnalagadda, A. Alankar, N. J. Balila, and T. Bhandakkar, Eds., Singapore: Springer Singapore, 2022, pp. 255–265.