Lattice Strain Evolution During Creep in Single Crystal Superalloys

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Abstract

\textit{In-situ} neutron diffraction studies are carried out to characterise the micro-mechanical deformation occurring during tensile creep of a typical single crystal nickel-based superalloy, CMSX-4. The loading responses of the matrix $\gamma$ phase and the precipitate $\gamma'$ are distinct. Moreover, the behaviour in the tertiary creep regime (in which the $\gamma'$ phase remains intact) is qualitatively different from that in the primary creep regime (when $\gamma'$ is sheared). In tertiary creep, initial deformation of the matrix leads to a release of misfit between the phases in the (100), resulting in elastic compression of the $\gamma$ in the loading direction. The load state then remains fairly constant during creep. During the initial stages of primary creep, elastic compression of the $\gamma$ phase is observed until at around 2-4\% creep strain this compression stabilises as the (100) misfit is released. This is the point at which $\gamma'$ shearing is thought to begin. Subsequently, the load in the $\gamma$ increases by around 200 MPa until a maximum is reached at around 8\% creep strain. This load is then suddenly released, which may be due to the release of back stress.

\textit{Key words:} neutron diffraction, nickel alloys, creep, dislocations, creep

1. Introduction

Certain questions remain unanswered about the micromechanics of deformation of the single crystal superalloys. For example, to what extent is deformation on the microscale restricted to the face-centered cubic $\gamma$ matrix which resides between the strengthening, ordered $\gamma'$ precipitates? Or alternatively, under what conditions are the $\gamma'$ particles sheared by dislocation activity? Moreover, how do the different modes of deformation on the microscale depend upon the temperature $T$ and the applied stress level $\sigma$? And from an engineering perspective, how does the underlying micromechanical mode of deformation influence the macroscopic response? Answers to these questions would allow a greater understanding of the deformation characteristics of these materials, and would allow more physically-faithful constitutive models to be constructed \cite{1, 2}. In turn, such understanding would enable more accurate estimates to be made of the deformation induced under engine operating conditions.

To answer these questions unequivocally, novel and high resolution characterisation techniques are needed. Unfortunately, the micromechanics of deformation are most usually studied by post-mortem characterisation of the microstructure and dislocation activity, often under conditions very different from those at which deformation was induced in the first place. For example, in the case of creep in nickel-based superalloys, the regimes of temperature and stress relevant to practical applications are 700 – 1100°C and 100 – 850 MPa, as experienced in gas turbine applications. Yet it is probably true to state that transmission electron microscopy (TEM) has been, until now, the most viable and widely used tool for studying the modes of dislocation activity \cite{3, 4}. However, a weakness of this approach is that one is unable to probe the kinematics of deformation. Moreover, it is necessary to make sometimes rather subjective interpretations based upon the dislocation configurations, which are often relaxed because of the necessity to examine thin foils at room temperature. Finally, it is necessary to be sure that one samples a volume of material whose deformation characteristics are representative of the bulk behaviour, which might not always be the case.

In superalloys, conventionally creep is studied in three regimes \cite{2–4}. At low temperatures and high stresses, dislocation shear of the $\gamma'$ precipitates is observed and the creep rate initially decreases in a conventional manner. This is followed by a so-called steady state regime and finally, acceleration of the creep rate associated with void formation. Low temperature and high stress creep is termed the primary creep regime due to the large primary creep strains observed. At intermediate temperatures and stresses shear of the precipitates does not occur and little initial primary creep is observed. Instead the creep rate increases continuously with time; this is termed the tertiary regime since only the final accelerating creep rate is observed. Morphological changes of the $\gamma'$ occur at the highest temperatures. This change is termed rafting and therefore this regime is called the rafting creep regime.

In this paper, neutron diffractometry is used to directly study the micromechanisms occurring during the creep de-
formation of a typical single crystal superalloy. Whilst neutron diffractometry is now a widely used method for characterising engineering alloys [5–18], it is still rare for the measurements to be made in-situ at temperatures and loading conditions beyond simple tensile or compression testing, as is attempted here. Moreover, the nature of specimens which are monocristalline in form means that the experimental setup is more complicated than that used conventionally for polycrystalline materials [7, 9–11, 19, 20]. Here, it is demonstrated that neutron diffractometry can be used to elucidate the micromechanisms of creep deformation in these materials; moreover some of the first in-situ measurements made on this class of material are presented. This is done by making phase-specific determinations of the load-strain response during creep deformation.

2. Experimental Details

Tensile-creep bars of the single crystal nickel superalloy CMSX-4 were machined from homogenized, heat treated and aged castings provided by Rolls-Royce Plc., Derby, UK. The orientation of the three bars were with the loading axis within $\theta < 5.8^\circ$ of the [100] direction. The mosaic spread of the crystals was $\sim 1^\circ$ in the casting [100] direction and around $\sim 7^\circ$ in the secondary [010] and [001] orientations, as is typical for industrial Bridgman castings.

In time-of-flight diffraction, the measurement is radial in reciprocal space. For this reason the sample mosaic contributions to the peak $d$-spacing width are observed only indirectly. The samples were tested on the Engin-X time-of-flight (TOF) neutron diffractometer at ISIS, Didcot, UK [21, 22]. The samples were mounted such that the stress was applied along the nominal [100] direction each time. The loading axis was horizontal and the rig was positioned to give the longitudinal lattice displacement in one detector and the transverse lattice displacement in the other. The detectors were fixed at $90^\circ$ to the incident beam (Figure 1), and data obtained from the full $\pm 15^\circ$ radial detector bank.

In general, if more than one Bragg peak is required, a TOF source outperforms a constant-wavelength (reactor) source, as a TOF pulse contains a wide range of wavelengths and thus can obtain a complete diffraction pattern. The variable in TOF is the wavelength, and the measurements are radial in reciprocal space in a single detector within a bank. The set-up of a single crystal sample for neutron diffraction experimentation is more complicated than that for a polycrystal. For a single crystal to be correctly aligned, measurements need to be taken and the sample subsequently rotated until the desired lattice planes are found in each detector. An example of the diffraction spectra obtained is shown in Figure 2.

Three creep tests were performed; a tertiary creep test at 900°C 460 MPa (sample c), and two primary creep tests at 650°C 825 MPa (samples a, b). Following sample alignment, the temperature $T$ was increased to the test temperature. The stress was stepped in order to measure the elastic constants, and then ramped to the creep stress. The elastic data generated serves to improve confidence in the experimental methodology and diffraction-spectrum fitting routine performed, by providing a reference case for comparison with measurements made using other techniques in other nickel alloys [23–29]. This provides some confidence in the measurements, as the elastic properties are relatively well known, and the $d$-spacing must be linear with stress. In addition, the thermal expansion coefficients and constrained misfit of the phases were found. The results from the elastic analysis have been presented elsewhere [22].

The first primary creep test (sample b) was commenced at 700°C 825 MPa. However, the initial creep rate was found to be rather high and therefore after $\sim 0.2$ h the temperature was dropped to 650°C. To remove any ambiguity, the test was repeated at 650°C on a subsequent visit to ISIS, sample (a). Unfortunately, the sample provided in this test (sample a) had not been subjected to the final ageing heat treatment, while samples (b) and (c)
had the final ageing heat treatment. This was noticed after the experiments had been performed, and should be a consideration when comparing the experimental results.

The macroscopic creep curves are shown in Figure 3, with an inset to show the early stages of creep and the temperature drop in sample (b). It can be observed that the creep rate accelerated continuously in (c) with no primary creep, and that there was a small incubation period after the experiments had been performed, and should be a consideration when comparing the experimental results.

The macroscopic creep curves are shown in Figure 3, with an inset to show the early stages of creep and the temperature drop in sample (b). It can be observed that the creep rate accelerated continuously in (c) with no primary creep, and that there was a small incubation period in samples (a-b), as noted by [2].

The intensity ratio was found by testing the fit of diffracted peaks. Often, TOF neutron diffraction peaks can be asymmetric, leading to the use of a leading or trailing exponential convoluted with the pseudo-Voigt, and the asymmetry can be wavelength-dependent. In the present case, both the (100) and (300) single phase peaks could be adequately described without resorting to such measures, and so were not used. The (200) composite peak, Figure 4, was fitted in a similar manner to that performed by Stone et al. [10], by assuming i) the instrumental peak widths of both phases are equal, ii) the Voigt shape function of both peaks are equal and iii) that the (200) γ’ position is fixed from the $d'_{(100)}$, $d'_{(200)} = 0.5 \times d'_{(100)}$.

The intensity ratio was found by testing the fit of different ratios to measurements which contained widely separated γ' and γ (200) peaks. It was found that $I'_{(200)} = 0.25 \times I_{(200)}$. The final fitting parameters for the (200) doublet were therefore the width, shape function, overall intensity and the location of the γ peak. The (100) and (300) peak widths were found to be the same, ruling out a wavelength dependence of peak width. The primary contributions to the peak widths were felt to be (i) strain, i.e. dislocations, (ii) the initial sample mosaic spread and (iii) instrumental broadening. Given that the γ’ precipitated from the γ matrix, the assumption that the peak widths were similar therefore seems reasonable, and was consistent with the data observed (Figure 4).

The individual detectors used in ENGIN-X are 196 mm high and 3 mm wide, providing a horizontal angular resolution of around 0.12°. Hence the (x00) peaks covered around 10 detector elements horizontally but was contained within a single element vertically; the (0x0) peaks covered more elements owing to their higher mosaic. Each element was calibrated by reference to a standard powder sample to provide a composite detector response. During straining, the peaks rotate as the crystal rotates, but since the d-spacing measurement is in time-of-flight (wavelength), such movements are not of concern. The comparability of this approach to that obtained at a constant wavelength source has previously been examined in [22].

In the case of the (200) peak, at times the d-spacing of the two phases was very similar, giving rise to difficulty in accurately fitting the doublet. Several different peaks that arose in this experimentation are shown in Figure 5. When the γ and γ’ components of the (200) were widely spread apart (case (a) and (c)), the fitting routine could readily determine the position of the smaller γ peak. Similarly for cases (b) and (d), with a small separation between the peaks, the methodology employed was satisfactory, with the exception of some early measurements in the tertiary creep test. Where the two overlap, the solution for the γ’ position becomes increasingly indeterminate (as measured by stability of the fitting routine solution to the initial trial location of the γ peak). Therefore care must be taken when interpreting the γ d-spacing output when both phases have similar d-spacing values (case (c) of Figure 5).

3. Results

The macroscopic creep behaviour observed during creep in the primary regime, at 650 °C and 825 MPa, is quite different to that observed during tertiary creep at 900 °C and 460 MPa, Figure 3. The macroscopic strain is defined by the standard formula $\varepsilon = (l - l_0)/l_0$, where l is the instantaneous length of the sample and $l_0$ is the original sample length at 0MPa. The macroscopic creep strain is defined as $\varepsilon_{creep} = (l - l_x)/l_x$ where $l_x$ is the sample length following initial elastic loading when the applied creep stress is applied. Thus any subsequent strain is, to first order, plastic deformation due to creep. Of course, elastic softening due to rotation of the crystal may occur and therefore this experimental decomposition is not quite the same.
as that classically used in micromechanical models where the total strain is decomposed into its elastic and inelastic (creep) components. The tertiary creep test was run to failure, while the first primary creep test finished at approximately 4% macroscopic strain due to failure of the grips. The second primary creep test was run for 24 h.

The evolution of measured \(d\)-spacing during the test, after sample heating is shown in Figure 6. The error bars have been removed from the \(d\)-spacing graphs (Figure 6), corrected lattice strain graphs (Figure 7), and misfit graphs (Figure 8). Their inclusion obscures the data, making it difficult to distinguish the \(\gamma\) from the \(\gamma'^\prime\) data points. Error bars have been included on two \(\gamma\) and two \(\gamma'^\prime\) data points in the (200), Figure 6 (a), to give a representation of the small calculated error from the fitting routine, ranging from \(\sim \pm 2 \times 10^{-5}\) to \(3 \times 10^{-4}\) Å. Of course, the actual uncertainty in peak location has additional contributions; typically an uncertainty of \(\pm 5 \times 10^{-5}\) Å is achievable on polycrystal samples at this instrument [21] when performing strain measurement. One would expect a similar accuracy to be achievable for CMSX-4 measurements, given the similarity in experimental procedure and the high intensity of the peaks measured, Figure 2. There exists an additional error associated from the furnace control (\(\pm 1^\circ\)C) [30]. A fluctuation of \(\pm 1^\circ\)C will give a corresponding change in \(d\)-spacing of \(\pm 2.8 \times 10^{-5}\) Å in the \(\gamma\) and \(\pm 5.4 \times 10^{-5}\) Å in the \(\gamma'^\prime\), from the calculation of thermal expansion coefficients in the longitudinal direction [22]. Thus the associated error in \(d\)-spacing from furnace control is of a similar value to the error from the peak fitting routine, and both will contribute to the overall total error in \(d\)-spacing measurement. Observing the \(d\)-spacing results, there is very little observable scatter of the \(\gamma'^\prime\) in any of the graphs, due to the clean data and suitable single-peak fitting routine. Thus it is fair to assume that the \(\gamma'^\prime\) \(d\)-spacing error is of the order of \(10^{-4}\) Å when

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{The different (200) peaks observed from experimentation. From left (a) to right (e), different situations with the \(\gamma\) peak moving from the left of the \(\gamma'^\prime\) to the right of the \(\gamma'^\prime\). The example in (c) is the most challenging for peak fitting analysis, and illustrates the necessity of the fitting assumptions made. (a) \(T = 650^\circ\)C \(\sigma = 825\) MPa \(\varepsilon = 11.74\%\) (200) (b) \(T = 650^\circ\)C \(\sigma = 825\) MPa \(\varepsilon = 10.14\%\) (200) (c) \(T = 650^\circ\)C \(\sigma = 825\) MPa \(\varepsilon = 9.34\%\) (200) (d) \(T = 650^\circ\)C \(\sigma = 825\) MPa \(\varepsilon = 0.29\%\) (020) (e) \(T = 650^\circ\)C \(\sigma = 825\) MPa \(\varepsilon = 11.74\%\) (020).}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{The \(d\)-spacing response during elastic loading and creep under two different operating conditions; top row, longitudinal (200) data, bottom row, transverse (020) data. \(\gamma\) \(d\)-spacing data from peaks which proved difficult to fit accurately are highlighted in grey in (a) and (c).}
\end{figure}
the contributions of peak location, peak fit, and thermal instability are accounted for. The use of a doublet peak fit in order to deconvolute the $\gamma$ from the (200) and (020) doublet peaks introduces additional uncertainty for this phase, particularly when the $\gamma$ and $\gamma'$ d-spacing are not widely spread. This is clearly seen in the range of scatter in $\gamma$ in Figure 6. The (020) transverse measurements have more widely spread $\gamma$ and $\gamma'$ peaks compared with the longitudinal (200), giving less scatter for the transverse $\gamma$ d-spacing from the fitting routine. The scatter in the transverse $\gamma$ d-spacing results is estimated to be $\sim 14 \times 10^{-4}$ Å, from Figure 6b, which to an approximation is $\sim \pm 7 \times 10^{-4}$ Å about an averaged free-hand fit to the data. The $\gamma$ d-spacing scatter is worse in the longitudinal direction, as the (200) $\gamma$ and $\gamma'$ contributions are not as widely spread making the deconvolution from the doublet more difficult. The scatter in this data in the worst case approximately $\sim \pm 3 \times 10^{-3}$ Å about a free-hand graphical fit to the $\gamma$ data in Figure 6(b). This is much greater than the error calculated from the fitting routine and a more realistic estimate for the error in achieving the $\gamma$ d-spacing results.

The $\gamma$ d-spacing measurements are an average of contributions from both the horizontal and vertical channels. Considering the three dimensional cubic precipitate system dispersed amongst the $\gamma$ matrix, there are twice as many vertical channels than horizontal. Unfortunately it is not possible to separate the contributions from each channel type, further complicating analysis. This is discussed further in the data analysis.

Initially, the d-spacing of both phases in each test increases in the (200) loading direction as the stress is ramped through the elastic loading regime, Figure 6(a - c). This is followed by an evolution during creep at elevated stress, Figure 6(a - c). The d-spacing is temperature dependent, and at zero stress, the (200) and (020) measurements should be equal. However, this is not observed in Figure 6(a - c). The authors can only attribute this to a possible offset in experimental set-up and recognise that absolute d-spacing measurements in two detectors are inherently difficult.

Following the elastic loading regime, the (200) $\gamma'$ d-spacing increases quite linearly during tertiary creep, Figure 6(c). The $\gamma$ data points shaded grey between $\varepsilon \sim 0.5 \rightarrow 1.0\%$ are due to unsatisfactory fits of the (200) doublet peak. By minimisation of errors the $\gamma$ peak fits to the left of the fixed $\gamma'$ peak position in the (200) doublet. However by visual inspection of the data it is seen that the $\gamma$ is clearly to the right of the $\gamma'$, thus the outputted d-spacing incorrectly gives $d_{(200)}^{\gamma} < d_{(200)}^{\gamma'}$. The only good fit between $\varepsilon \sim 0.5 \rightarrow 1.0\%$ occurred at $\varepsilon = 0.6\%$, $d_{(200)}^{\gamma'} = 1.829$ Å. Following this region the fits were all good.

Neglecting the poor fits in the (200) tertiary creep d-spacing data, there is a sudden decrease in (200) $\gamma$ d-spacing following the elastic loading, Figure 6(c). After this sudden decrease, the $\gamma$ d-spacing increases almost linearly. This is in contrast to the (020) data for each creep test. The elastic loading leads to a decrease in d-spacing of both phases in the transverse direction (due to Poisson contraction). Once the load is stabilized in the tertiary regime, the d-spacing of the (020) $\gamma$ initially increases, Figure 6(c). The rate at which the $\gamma$ d-spacing is increasing declines during early creep, after which the d-spacing increases slowly at a constant rate. The $\gamma'$ lattice parameter continuously decreases during tertiary creep, almost the exact reverse of the behaviour observed in the (020) $\gamma$

In the primary creep test, Figure 6(b), the temperature drop from 700 → 650°C is seen in both phases as a decrease in d-spacing, at $\varepsilon \sim 1.4\%$. Following the elastic region, the (020) d-spacing is constant in the $\gamma$ phase and decreases in the $\gamma'$, Figure 6(b). The second primary creep test performed without the temperature drop, Figure 6(a), agrees with the first in the (020). In the (200), the $\gamma'$ d-spacing increases during creep after the first $\sim 1\%$ strain, Figure 6(a and b). $d_{(200)}^{\gamma'}$ is initially greater than $d_{(200)}^{\gamma}$; seen clearly in Figure 6(b). Unfortunately it was not possible to accurately fit the first three $\gamma$ creep measurements of the second primary creep test, shaded grey in Figure 6(a). On visual inspection of these three fits it was observed that the $\gamma$ fit was too far to the right, giving larger $\gamma$ d-spacing values.

Between 2 and 3% strain in Figure 6(b), there is a drop in (200) $\gamma$ lattice parameter. From Figure 6(b), this drop in $\gamma$ lattice parameter is equivalent to a lattice strain of approximately (1.830 – 1.824)/1.830 = $3 \times 10^{-3}$, or roughly 320 MPa, where the elastic constant for $\gamma$ at 650°C is 106 ± 7GPa, Table 1. The $\gamma$ d-spacing of both primary creep tests steadily increases from $\sim 3\%$ strain in the longitudinal direction. At a macroscopic strain of $\sim 9\%$ a maximum $\gamma$ d-spacing is reached in Figure 6(a). The first primary creep test ended due to grip failure before this maximum was observed, Figure 6(b). Following the maximum $\gamma$ d-spacing value in Figure 6(a), there is a quite sudden drop in the $\gamma$ (200) lattice parameter, and hence average elastic lattice strain, over the course of around 1.5% macroscopic creep strain. This is followed by a slightly increasing $\gamma$ d-spacing.

In order to relate the d-spacing measurements to strain, conventionally, we define the (elastic) lattice strain as follows

$$\varepsilon_{h00}^x = \frac{d}{d_0}$$

where $\varepsilon_{h00}^x$ is the lattice strain of phase $x$ in the (h00) lattice plane. $d_0$ is the d-spacing at the creep temperature and zero stress. The strain was then corrected to account for true stress and temperature changes

$$\varepsilon_{h00}^{x,corr} = \varepsilon_{h00}^x - \frac{\sigma}{E_{h00}^x} (1 + \varepsilon) - \alpha x \Delta T$$

where $\sigma$ is the applied macroscopic nominal stress, $E_{h00}^x$ is the apparent or effective stiffness of the (h00) plane, $\varepsilon$ is the macroscopic engineering strain, $x$ denotes the phase and $\alpha$ the thermal expansion coefficient. The strain $\varepsilon_{h00}^{x,corr}$
is therefore the measured elastic strain due to creep, after correcting for the strain due to elastic loading, changes in section area due to plasticity and thermal expansion. This allows for the test which began at 700°C to be corrected for the drop in temperature using the measured lattice thermal expansion coefficients, published in [22]. The modulus $E_{\text{hao}}$ is the diffraction elastic constant measured for each phase, given in Table 1. This should be distinguished from the $< h00 >$ moduli of the isolated phases. The diffraction elastic constants are the measured apparent moduli and differ from the isolated moduli by the constraint between the two phases.

The evolution of this corrected lattice strain during each creep test is shown in Figure 7. Pollock and Argon [3] discuss their TEM observations and finite element study of creep in CMSX-3 in terms of misfit. This suggests another way to view the diffraction measurements presented within this paper. Defining the apparent misfit $\delta_{\text{app}}$ as follows

$$\delta_{\text{app}} = 2 \times \frac{d_{\gamma'} - d_\gamma}{d_{\gamma'} + d_\gamma}$$

we obtain Figure 8. It should be emphasized that misfits measured always include a degree of stress accommodation between the phases at the temperature and stress conditions that pertain to the measurement made, whether this is in TEM or in a bulk diffraction measurement.

Examining the apparent lattice misfit of the (100) and (010) in the tertiary regime (900°C 460 MPa), Figure 8(c), there is some agreement with conclusions made from the finite element model for CMSX-3 of Pollock and Argon in the (100) direction, but not the (010) [3]. The finite element model of CMSX-3 suggests that at 850°C and an applied stress of 552 MPa (a tertiary creep regime), all misfit stresses are totally relieved after $t = 1170 \text{s}$, i.e. 19.5 minutes. Figure 8(c) shows that the (100) misfit is nearly totally released at a macroscopic strain of 0.62%. This data point corresponds to the earliest (100) $\gamma$ lattice data point in Figure 7(c) that came from a good peak fit (i.e. the first data point not shaded grey), at 0.14% macroscopic creep strain. From Figure 3(c), a macroscopic creep strain of 0.14% was reached at $t = 0.85 \text{~h}$, i.e. $\sim 50 \text{~minutes}$.

Unfortunately the earlier data points were poor fits so we cannot say for certain at what point in time before 50 minutes this release occurred.

The misfit derived from the tertiary creep transverse strain data also decreases slightly in magnitude from $-4 \times 10^{-3}$ to $-3 \times 10^{-3}$ on initial elastic loading, and then increases in magnitude continuously during creep to nearly $-5 \times 10^{-3}$, Figure 8(c). This shows that the (010) misfit is never fully released, in contradiction of Pollock and Argon [3].

The misfit analysis aids the explanation of the corrected strain evolution of both phases during tertiary creep, Figure 7(c). In the tertiary regime, the release of misfit stress in the (100) direction during the first 1% of creep causes the observed compressive strain in the (100) $\gamma$, Figures 8(c) and 7(c) respectively. The corrected strains of both phases then increase in tandem, Figure 7(c). This might be taken to imply that co-deformation of both phases was occurring, as both phases would strain harden resulting in an increase in observed elastic strains in the (100). However it is generally agreed that tertiary creep is confined to the $\gamma$ matrix [3, 31–33]. The effect of changing cross section during deformation, which increases the true stress, has been removed from the lattice strain data to obtain the corrected lattice strains. Thus the increase of corrected lattice strain in both phases cannot be attributed solely to an increasing true stress.

We suggest that this change, which is a strain of around $1.0 \times 10^{-3}$ in the $\gamma'$ (200) between creep strains of 1 and 8% is due to lattice rotation. A lattice rotation towards the less stiff (100) would give an increase in observed lattice strain. In order to measure the degree of lattice rotation, the post-crept sample was cut along the tensile axis and electron backscattered diffraction (EBSD) analysis was performed at the fracture region and at the threads of the sample (where no deformation occurs during testing). From analyses of the pole figures and by calculating the degree of relative rotation from the Euler angles [34], the crystal rotated $\sim 10.5^\circ$, with a rotation of (100) $\sim 6^\circ$ towards the tensile axis. The consequent change in modulus could be responsible for at least some of the increase in corrected lattice strain observed in Fig 7(c): approximate calculations yield a value of order $1 \times 10^{-3}$ for the magnitude of this effect.

It is unclear how the matrix (200) compresses to release misfit, with no apparent effect on the $\gamma'$. It may be possible that plastic flow of the matrix around the precipitate acts in such a way as to release the (200) misfit strain.

The longitudinal lattice strain measurements measured by Ma et al. [19] for CM247LC differ quite markedly to those presented here. This is not surprising as single crystal creep lifetimes are much greater than those of polycrystals, indicating differences in the creep mechanics. For example, the creep lifetime of a first generation single-crystal, MAR-M200, when tested at 982°C and 206 MPa increases from approximately 36 h with a failure strain of 4% when cast as a polycrystal, to 110 h with a failure strain of 28% when cast as a single crystal [35]. The final stress of a multiple stress creep test performed by Ma et al. was 425 MPa at a temperature of 900°C, i.e. a tertiary creep regime. The (100) $\gamma'$ lattice strain increased by approximately $1.3 \times 10^{-3}$ while the (100) $\gamma$ lattice strain decreased.

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### Table 1: Measured diffraction elastic constants in the loading and transverse directions of each phase [22].

<table>
<thead>
<tr>
<th>$T$ (°C)</th>
<th>$E_{[100]}^{\gamma'}$ (GPa)</th>
<th>$E_{[100]}^{\gamma}$ (GPa)</th>
<th>$E_{[010]}^{\gamma'}$ (GPa)</th>
<th>$E_{[010]}^{\gamma}$ (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>128 ± 3</td>
<td>109 ± 7</td>
<td>-192 ± 3</td>
<td>-208 ± 16</td>
</tr>
<tr>
<td>650</td>
<td>105 ± 4</td>
<td>106 ± 7</td>
<td>-198 ± 12</td>
<td>-211 ± 12</td>
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<tr>
<td>700</td>
<td>102 ± 2</td>
<td>108 ± 6</td>
<td>-227 ± 5</td>
<td>-196 ± 12</td>
</tr>
<tr>
<td>900</td>
<td>87 ± 2</td>
<td>89 ± 3</td>
<td>-217 ± 7</td>
<td>-171 ± 17</td>
</tr>
</tbody>
</table>
by $0.8 \times 10^{-3}$ over 8% macroscopic creep strain. The lattice strain response of (100) $\gamma'$ is similar in this work, however the [100] lattice strain of $\gamma$ increases in CMSX-4, Figure 2. Ma et al. believed that the observed tertiary stage behaviour of CM247LC is due to a build up of dislocations in the vertical $\gamma$ channel [19]. It is difficult to compare the lattice strain responses of a polycrystal and single crystal alloy without noting more than they differ markedly, as the lattice strain response of a grain within a polycrystal is dependent upon the deformation of surrounding grains of different orientations. That is to say the lattice strain response of a (100) grain will be dependent on the deformation of surrounding grains of (110), (111) etc. This is not a consideration when studying the response of a single crystal as there is only one grain. Ma et al. did not present the transverse data, so it is not possible to compare the (010) lattice strain response. Furthermore grain boundary sliding is not a consideration of single crystals, which in turn allows the single crystal to be alloyed very differently to polycrystals as grain boundary strengthening elements are not required.

In the tertiary creep transverse measurements, the (020) $\gamma$ lattice strain increases rapidly during the first 1% macroscopic creep strain after which the rate decreases, presumably due to strain hardening, Figure 7(c). Following an initial compression in the (020) $\gamma'$ during early tertiary creep, the $\gamma'$ strain slightly decreases. Focusing solely on the (020) data, this could be interpreted as a load transfer from the $\gamma$ to the $\gamma'$. Considering the (020) data in light of what was suggested as the interpretation of the (200) data, if the lattice is rotating towards (020) in the transverse direction, this would result in increasing compressive lattice strains of both phases. Pollock and Argon [3] showed that under a similar temperature and stress condition in CMSX-3 there is much greater stress in the matrix (020) than (200). Thus the authors hypothesize that in the (020) the creeping matrix continues to transfer compressive load into the stronger $\gamma'$ phase, with the change in strain being far greater in the $\gamma$ phase due to the effect of volume fraction. It is clear from this analysis that
the interpretation of the transverse data is extremely difficult and is sometimes omitted from papers for this reason, see for example Ma et al. [19].

In conclusion of the tertiary creep analysis, the measurements taken can be interpreted to be somewhat consistent with the tertiary creep analysis of Pollock and Argon [3]: The γ initially yields to relieve the misfit stresses in the (100), and then dislocation motion is confined to the γ channels without precipitate shear. The lattice is rotating and true stress is increasing, seen clearly in the (200). There appears to be a lot of stress transfer from γ to γ′ in the (020) direction which could be indicative of a lot of creep occurring in the (020) γ, as suggested by Pollock and Argon [3].

Analysing the macroscopic creep curves of the two primary creep tests, it is seen that the creep responses differ quite markedly, despite being performed on bars that have very similar orientations, Figure 3. The initial primary creep strain before a steady-state regime is exhibited is almost three times greater in test (a) than (b). A possible explanation for this ambiguity is the sample in test (b) had been subjected to the final aging heat treatment, the sample in (a) had not. The post-creep samples were tested for ovalisation; the ratio of the minor to major diameters was 0.99 for test (b), and 0.79 for test (a), i.e. test (b) showed no ellipticity. The degree of ellipticity has been shown to be a function of macroscopic primary creep strain [31]. The thinning along the <110> direction during primary creep has proved controversial. Pan et al. [36] proposed that slip occurs along the 6 cube (100)<110> and 12 normal {111}<110> slip systems, while others emphasize the importance of the {111}<112> slip system [37–43]. There is strong evidence that suggests the dominant slip mode is temperature and stress dependent. Matan et al. [31] showed that macroscopic shape deformation at 750°C and 750 MPa is due to {111}<112>, but at 950°C and 185 MPa slip is due to {111}<110>. The primary creep test conditions employed here are similar to 750°C 750 MPa, so one would expect the {111}<112> slip mode to dominate. The ovalisation that occurs during test (a) is a first indication that {111}<112> ribbon shear has occurred, however the lack of ovalisation in test (b) indicates that, if particle cutting has occurred, it has not occurred to a degree where it is evident by optical analysis, i.e. the primary creep strain was not large enough. The diffraction data allows us to probe further into the micro-mechanics of primary creep.

During both primary creep tests, the initial misfit is relaxed by elastic compression of the γ phase in the (200). This occurs within the first 2% of creep, seen clearly in Figure 7(b). The release of misfit corresponds to compression of the (200) γ in the horizontal channel, however a relaxation of misfit will also cause expansion of the (200) γ in the vertical channels. Although these vertical channels are twice as numerous, the net effect is still a compression of the (200) γ matrix, as the contribution from the direct compression in the (200) horizontal channels outweighs the expansion due to Poisson’s effect in the vertical channels. As the initial period of rapid (primary) creep begins to slow, the (200) γ corrected lattice strain gradually increases from a minimum of −4.5 × 10−3 to around −2 × 10−3 at a creep strain of ∼ 8% and a total strain of ∼ 9%, Figure 7(a). The γ′ phase gradually increases from 1 × 10−3 to 2 × 10−3 during the same period of creep. This would be interpreted, in the light of Rae et al. [4, 37, 38, 44], to correspond to the shearing of both phases. Plastic deformation of both phases causes an increase in hardness due to the increase in dislocation density, which would be seen as an increase in lattice strain in both phases. However one can’t ignore that lattice rotation towards (100) could also cause an increase in lattice strain in both phases due to the change in sample modulus as it rotates.

Then, quite suddenly, there is a large change in misfit and decrease in the longitudinal (200) strain in the γ phase in test (a) between 8 and 9% creep strain, followed by a plateau (test (b) did not run to a large enough strain for this to be observed), Figure 7a. There is a corresponding drop of ∼ 1 × 10−3 lattice strain in the γ′. The rapid decrease of γ lattice strain could be indicative of a release of accommodation stress (sometimes termed back stress) between the phases, however this would be accompanied by a corresponding increase in γ′ lattice strain. A decrease of 4.5 × 10−3 corresponds to a stress of around 450 MPa, Table 1, i.e. relaxation of approximately 55% of the applied stress of 825 MPa. Notably, there is no corresponding change in the macroscopic creep curve, suggesting that compression of the γ is balanced by a decrease in the γ′ shear rate during a period of stress release. A decrease in γ′ shear results in a lower γ′ lattice strain rate. One explanation might be that a recovery mechanism may be operating, allowing the (200) γ′ lattice strain to also decrease.

Similarly to the tertiary creep test, although the thermal misfit is relieved in the (200) during primary creep by compression of the γ matrix (Figures 6b and 8b), the misfit in the (020) is never relieved, even up to 12% macroscopic strain, Figure 8(a). The stress transfer from γ to γ′ observed in the transverse data during tertiary creep also occurs in the (020) for the duration of the first primary creep test, Figure 7(b), and up to 9% of the second creep test, Figure 7(a). As the lattice rotates towards the (100) in the longitudinal direction, one would expect the lattice strain of both phases to decrease in the (020), however the γ phase lattice strain is approximately constant in Figure 7(b) and increases to a maximum at 9% macroscopic strain in Figure 7(a), while the γ′ lattice strain decreases in both. This is indicative of a stress transfer from (020) γ to (020) γ′. At 9% macroscopic strain a change in the creep mechanics is observed in the (020). This is the same macroscopic strain at which the (100) creep mechanics change. Thus the data, although very difficult to interpret, is real. At this macroscopic strain the (020) γ lattice strain decreases while the γ′ increases, Figure 7(a).
This is consistent with the hypothesis put forward for the (100) primary creep data. A release of back stress from $\gamma'$ to $\gamma$ results in an increase in the (020) $\gamma'$ lattice strain and a reduction in the (020) $\gamma$ lattice strain. This results in an increase of the creep rate in the $\gamma$ phase, however there is no dramatic change in macroscopic creep rate at this strain (Figure 3). The authors propose that dislocation shear of the $\gamma'$ was occurring to this strain and then stops so there is no observed overall increase in the creep rate, as a possible interpretation. Certain works suggest that shearing of the $\gamma'$ ceases after the end of primary creep [45], while others have concluded that shearing of the $\gamma'$ continues through secondary creep [4, 46, 47].

In summary, then, primary creep proceeds in three phases. 1. An increase in misfit towards zero in the (100). There is stress transfer from $\gamma$ to $\gamma'$ in the (020). 2. The stress gradually increases in both phases in the (200) until 3, there is an increase of the creep rate in the $\gamma$ phase when a stress is released from the $\gamma'$. The macroscopic strain rate is fairly constant when this increase of creep rate in the $\gamma$ occurs, and the authors take this to be indicative that shearing of $\gamma'$ was occurring to this point, at which point it halts, effectively keeping the net macroscopic creep rate steady.

Finally, the samples were examined after testing, Figure 9. The $\gamma'$ of the primary creep samples still possessed a cubic morphology, while the tertiary creep samples possess a partially rafted structure in the (010) plane. Therefore, even at a temperature below that traditionally recognised as causing rafting in CMSX-4, some rafting can be observed. If tertiary creep is confined to the $\gamma$, presumably a point would eventually be reached where rafting would be so extensive as to require either glissile sliding of the $\gamma/\gamma'$ interfaces to accommodate creep of the $\gamma$, or shearing of both phases would be required. However, there is no indication in the diffraction data that such a point was reached in the present testing, which was performed to failure.

4. Summary and Conclusions

The elastic lattice strain evolution during the tertiary and primary creep of single crystals of the nickel-base superalloy CMSX-4 has been examined using in-situ neutron diffraction and analysed in light of our current understanding of the dislocation mechanisms operative during creep from TEM studies. The following conclusions can be drawn from this work.

1. In-situ neutron diffractometry can be used to examine the evolution of loading state in the two phases in a single crystal superalloy during creep deformation using the peak positions as an internal elastic strain gauge.

2. The lattice strain response observed in the two regimes is quite distinct, indicating a difference in the operative micromechanisms of deformation during primary and tertiary creep.

3. During the initial stages of tertiary creep deformation of the matrix phase leads to a release of misfit between the two phases in the (100), resulting in elastic compression of the $\gamma$ phase in the loading direction. The load state in the two phases then remains fairly constant during creep in the (100), with a gradual increase observed that is thought to be due to lattice rotation. There is no unequivocal evidence of shearing of the $\gamma'$ phase. There is load transfer from $\gamma$ to $\gamma'$ in the (020).

4. In primary creep, during the initial stages elastic compression of the $\gamma$ phase is also observed in the (100) until at a creep strain of around 2-4% this compression stabilises as the misfit is released. This is expected to be the point at which shearing of the $\gamma'$ begins. Subsequently, the load in the $\gamma$ increases by around 200 MPa until a maximum is reached at around 8% creep strain. A large fraction of the load in the $\gamma'$ is then suddenly released into the $\gamma$ between 8 and 9% creep strain. This is thought to be the point where shearing of the $\gamma'$ halts.

5. Limited rafting is observed to occur during tertiary creep testing at 900°C and 460 MPa.

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