

Mathematical and Computational Models for Simulating Transient Nuclear Criticality Excursions within Wetted Fissile Powder Systems

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Abstract

This paper describes a novel methodology for the analysis of nuclear criticality excursions in fissile powder beds under wetting conditions. These potentially hazardous powder, slurry and sludge systems may be found in nuclear fuel manufacturing and fabrication facilities. A point kinetics model was coupled with water infiltration, thermal-hydraulics and radiolysis models through the use of reactivity feedbacks. Good agreement in the water infiltration rate was found when comparing the water infiltration model used in this paper to experiments conducted by the French Commissariat à l'Énergie Atomique et aux Énergies Alternatives (CEA). A case study was proposed whereby a sheet of fine water droplets from a sprinkler system came into contact with an open-topped bed of low enriched UO_2 powder. Simulations indicate that the mean powder particle size had a strong effect on the time required for the water to percolate through the powder bed. Powder particle size was also predicted to have a moderate effect on the initial fission power spike. The fission energy released over the first 300 s of the nuclear criticality transient ranged from 65.28 MJ to 97.98 MJ depending on mean powder particle size. This is similar in magnitude to other simulated nuclear criticality excursions in powder beds. The model predicts that the initial fission power spike would be limited by the production of radiolytic gas and to a lesser extent the effects of Doppler broadening and thermal expansion. As expected, boiling and the associated steam production, was found to be an important phenomenon in the reduction of the fission rate through the negative void reactivity effect of the steam.

Keywords: nuclear criticality excursion and accident analysis, wetted fissile powder, water

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Nomenclature

Latin Symbols			
$A_{i,j}$	Interfacial area between material, i and material, j [m ²]	H_i	Height of region, i [m]
a_I	Interfacial area concentration [m ⁻¹]	$h_{l,v}$	Latent heat of vapourisation [J kg ⁻¹]
c	Gas advection speed empirical parameter [-]	$h_{p,v}$	Powder boiling heat transfer coefficient [W m ⁻² K ⁻¹]
C_i	Delayed neutron precursor concentration for group, i [precursors (cm ⁻³ · UO ₂)]	$h_{p,w}$	Powder water convective heat transfer coefficient [W m ⁻² K ⁻¹]
c_i	Specific heat capacity of material, i [J kg ⁻¹ K ⁻¹]	k	Permeability of powder bed [m ²]
D	Neutron diffusion coefficient [cm]	$K_{b,0}$	Gas advection speed empirical parameter [-]
d_i	Spherical diameter of part of material, i [m]	k_b	Boltzmann constant [eV K ⁻¹]
$D_{t,i}$	Turbulent mixing coefficient of region, i [m ² s ⁻¹]	k_{eff}	Effective multiplication factor [-]
E_{fast}	Thermal neutron energy [eV]	$K_{\text{sat},e}$	Saturated hydraulic conductivity [m s ⁻¹]
E_f	Energy generated per fission [J fission ⁻¹]	$M_{i,j}$	Mass of material, i, in region, j [kg]
E_i	Heat transfer rate for thermal process, i [J s ⁻¹]	Mo	Morton Number [-]
E_{therm}	Fast neutron energy [eV]	n	Neutron density [neutrons (cm ⁻³ · UO ₂)]
f_{imm}	Powder bed immersion fraction [-]	n_c	Gas advection speed empirical parameter [-]
$G(H_2)$	Volumetric yield of hydrogen from radiolysis [m ³ J ⁻¹]	$N_{p,i}$	Number of powder particles present in region, i [-]
h_ψ	Matric head [m]	P	Energy generation rate (fission power) [J s ⁻¹]
		Pr	Prandtl number [-]
		R	Powder drum radius [m]
		R_{fiss}	Range of fission fragments [m]

R_i	Reactivity of component, i [\$]	β_{eff}	Total delayed neutron fraction [-]
Re	Reynolds number [-]	β_i	Delayed neutron fraction for precursor group, i [-]
RR_f	Fission rate [fissions $\text{cm}^{-3} \text{ s}^{-1}$]		
S_A	Specific surface area [m^{-1}]	χ_p	Prompt fission energy spectrum [-]
s_n	Intrinsic neutron source density [neutrons $\text{s}^{-1} (\text{cm}^{-3} \cdot \text{UO}_2)$]	ΔS_w	Change in saturation across wetting front [-]
t	Time [s]	ϵ	Powder bed porosity [-]
$T_{i,j}$	Temperature of material, i, in region, j [K]	η	Thermal expansion coefficient of water [K^{-1}]
T_{in}	Water inlet temperature [K]	Γ_2	Diven factor [-]
T_I	Powder-water interface temperature [K]	$\hat{\phi}$	Normalised scalar neutron flux distribution [cm^{-1}]
T_{sat}	Water boiling temperature [K]		
U_{ade}	Advection-dispersion equation infiltration rate [m s^{-1}]	κ_i	Thermal conductivity of material, i [$\text{W m}^{-1} \text{ K}^{-1}$]
U_i	Superficial velocity of material, i [m s^{-1}]	Λ	Mean generation time [s]
U_{slump}	Slumping superficial velocity [m s^{-1}]	λ_i	Decay constant for precursor group, i [s^{-1}]
$U_{w,\text{in}}$	Boundary water flow speed [m s^{-1}]		
V	Prompt neutron speed, with energy, E [cm s^{-1}]	μ_i	Dynamic viscosity of material i [Pa s]
		$\rho_{i,\text{bulk}}$	Bulk density of material, i [kg m^{-3}]
$v_{f,i,j}$	Volume fraction of material, i, in region, j [-]	$\rho_{i,\text{th}}$	Theoretical density of material, i [kg m^{-3}]
$V_{i,j}$	Volume of gas of material, i, in region, j [m^3]	σ	Water surface tension [N m^{-1}]
		Σ_a	Macroscopic absorption cross-section [cm^{-1}]
Greek Symbols			
α_L	Longitudinal dispersivity [m]	Σ_f	Macroscopic fission cross-section [cm^{-1}]
$\bar{\nu}$	Average number of neutrons produced per fission [neutrons fission $^{-1}$]	Σ_s	Macroscopic scattering cross-section [cm^{-1}]

τ	Mean neutron lifetime near criticality [s]	rg	Radiolytic gas
Subscripts		th	Thermal reactivity component
boil	Boiling	T	Total reactivity component
conv	Inter-phase convection	void	Void reactivity component
dp	Dry powder region	v	Water vapour
eff	Effective region property	wp	Wet powder region
gen	Thermal energy generation	w	Water
g	Total gas	Superscripts	
imm	Water infiltration reactivity component	'	Spatial derivative (variable per unit height) [m^{-1}]
pooled	Pooled water region	-	Spatial average [-]
p	Fissile powder		

1. Introduction

Fissile powders, slurries and sludges are often processed in nuclear fuel facilities [1, 2]. A slurry refers to a fluid-solid mixture where solid particles are dispersed in a continuous fluid phase [3]. Slurries typically have a higher fluidity than sludges or cakes which consist of a more concentrated dispersed phase surrounded by liquid [3]. Colloids refer to powders that are suspended in a liquid phase without requiring motion of the liquid to remain suspended [3]. There are several occasions throughout the fuel life-cycle where a nuclear criticality excursion involving fissile powders may occur. The degree of the nuclear criticality, or reactivity, of a fissile system is influenced by a wide range of physical parameters. In the field of nuclear criticality safety assessment there are two useful mnemonics, or acronyms, that are useful in remembering the physical parameters that affect the criticality, or reactivity, of a system and these are given by: MAGIC MERV (for Mass, Absorption, Geometry, Interaction, Concentration, Moderation, Enrichment, Reflection, and Volume) [4] or alternatively MERMAIDS (for Mass, Enrichment, Reflection, Moderation, Absorption, Interaction, Density, and Shape) [5]. Nuclear criticality safety experts aim to prevent nuclear and radiation accidents that result from inadvertent, self-sustaining and divergent, nuclear chain reactions by controlling these parameters [6]. The nature of nuclear criticality excursions in fissile powder beds

may be captured using the models proposed in this paper, enabling the radiological impact to those persons in the vicinity to be quantified.

Uranium powders are the most commonly handled fissile material in the production of nuclear fuels [1, 2]. A specific type of uranium concentrate powder, referred to as Yellowcake, is obtained from leached solutions as an intermediate step in the processing of uranium ores. It is produced through the milling and chemical processing of uranium ore to form a coarse powder that contains approximately 80% uranium dioxide (UO_2). The facilities used to produce this powder often store powders, cakes, sludges and slurries in large cylindrical vessels [6]. One of the main concerns associated with the nuclear criticality safety assessment of such systems is the ingress of water into the vessels containing the powders, due to the activation of a fire sprinkler system. This may lead to a nuclear criticality excursion due to the extra moderation effect of the liquid entering the vessel. The POWDER code [7, 8], developed by the United Kingdom Atomic Energy Authority (UKAEA) in conjunction with the French Commissariat à l'Énergie Atomique et aux Énergies Alternatives (CEA) was developed to simulate these types of accidents whereby a drum of fissile powder becomes over-moderated due to the presence of water, leading to a criticality excursion. Code development was aided by three sub-critical experiments that were carried out by the CEA at the SILENE reactor [9]. Other sub-critical experiments carried out by Lichtenwalter [10], at the Y-12 National Security Complex located in Oak Ridge, Tennessee, USA, investigated the incursion of water from a sprinkler system into a container filled with depleted tri-uranium octoxide (U_3O_8) [11]. The retention of water in the powder bed was recorded as well as the change in bulk density as a result of wetting. Additional experiments by Dunn et al. [12] investigated the saturation of UO_2 powders and pellets when wetted, showing that water does not penetrate all of the pores in the powder bed. Finally, the Institut de Protection et Sécurité Nucléaire (IPSN), which is now the Institut de radioprotection et de sûreté nucléaire (IRSN), conducted the MARACAS approach-to-critical experiments from 1982 to 1986 at the Valduc research centre in France. The MARACAS experiment was a sub-critical facility of “split-table” type (a system for bringing two elements progressively closer to each other), able to contain up to six tonnes of nuclear material on two tables, one of which is stationary, while the other is mobile. IRSN performed approach-to-critical experiments with 5 % low-enriched UO_2 powder that was moistened to reach variable moderation ratios ($\text{H}/\text{U} = 2.0$ to 3.0) [13].

Fuel fabrication plants are also a focus of the nuclear criticality safety experts' attention due to

the large volumes of fuel present [14]. These facilities, commonly used to fabricate Mixed OXide (MOX) fuels, use paddles to stir a mixture of plutonium-uranium oxide fuel with uranium dioxide powder and a lubricant (zinc stearate) until it becomes homogeneous [15]. At different stages in the fabrication of the MOX fuel, low volumes of hydrogenated materials are used, posing a nuclear criticality safety risk [16]. Similarly, the fabrication of UO_2 fuels involves mixing a dry powder with other materials in a blender. Accidental leakage of the moderator (water) into the fissile system may lead to a nuclear criticality accident. Incomplete drying of fissile powders during manufacture or condensation during cooling of powders may also lead to nuclear criticality excursions [17]. These potential accidents highlight that one of the most important mechanisms for avoiding accidental, or unintended, nuclear criticality excursions in fissile powders is by controlling the amount of moderator (typically water) within the fissile system [14].

An experimental study by the European Union's European Atomic Energy Community (EURATOM) in association with the Keuring Van Electrotechnische Materialen (KEMA) labs investigated the possibilities and limitations of a homogeneous suspension power reactor [18]. A series of sub-critical experiments carried out on UO_2 suspensions investigated many aspects of the proposed suspension reactor. Their research focused on operational considerations such as the minimum volumetric flow rate through the system required for the suspension to remain homogeneous. Erosion, caking, fission fragment range and flocculation properties of the suspension in the power reactor were also examined through experiments outlined in the cited literature [19, 20, 21, 22].

A significant number of codes have been developed to model some of these nuclear criticality safety accident scenarios. The earliest of which, SKINATH-WP, designed by Basoglu [23], and also reported in Basoglu et al. [24], combines point kinetics with spatially independent nuclear thermal hydraulics to model criticality accidents initiated by the ingress of water into low enriched UO_2 powder contained within a conical stirrer. Another code developed by DiPeso and Peterson [25] also combined neutron kinetics with spatially independent nuclear thermal hydraulics and hydrodynamics to model criticalities in fissile slurry type systems. The FETCH (Finite Element Transient Criticality) code has also been applied to granular fuel contained within conical vessels housed in fuel fabrication facilities [26, 27, 28]. This code was used to model a nuclear criticality excursion in a layered Mixed OXide (MOX) powder system containing a zinc stearate lubricant. FETCH solves the time and spatially dependent neutron kinetics equations coupled with a multi-phase compressible flow model. The complex nature of this type of multi-physics model makes it

challenging to evaluate the importance of particular parameters on the model as well as to verify and
80 validate (V&V) the model. The model is very computationally demanding and usually, only very
short timescales are considered when applying the model to simulating transient nuclear criticality
excursions.

A suite of numerical codes produced by the Japan Atomic Energy Research Institute (JAERI)
for use on fissile powder systems, named AGNES-P, DOCTRINE and FEDEX-P have been designed
85 with varying levels of complexity with respect to the modelling of neutronics and nuclear thermal-
hydraulics phenomena. AGNES-P is a point model that has similar characteristics to SKINATH-
WP. DOCTRINE has been developed to consider more complicated geometry than AGNES-P whilst
still using point kinetics. FEDEX-P solves the time and spatially dependent neutron diffusion
equation alongside the Navier-Stokes equations using 3D Finite Element Modelling (FEM) [29].
90 Sakai et al. [30] used the discrete element method (DEM) to model the dynamics of MOX powder
particles during the agitation process. This was coupled with a continuous-energy Monte Carlo
neutron transport code to study how particle dynamics affect the criticality of the MOX powder as
it is stirred in a agitation tank [15].

Nuclear criticality safety experts are also concerned with the transport of fuel and scrap to
95 and from re-processing facilities. An investigation carried out by Artigas [31] modelled the nuclear
criticality safety of a shipping container of low-enriched UO_2 pellets, in UO_2 powder and water.
Other containers capable of transporting 540 kg of 5 % enriched UO_2 powder are manufactured
by Global Nuclear Fuels (GNF). Nuclear criticality safety analysis of these containers focused on
the addition of water to undamaged and damaged containers. They showed that for their specific
100 system, a nuclear criticality excursion could be mitigated for an unlimited amount of water entering
the shipping container [32]. This highlights the importance of modelling transient nuclear criticality
excursions in wetted powders to ensure that the potential for nuclear criticality excursions is fully
understood.

This paper proposes a novel phenomenological approach to modelling transient nuclear criticality
105 excursions in wetted fissile powders. The proposed model incorporates a multi-region approach,
of a cylindrical vessel, discretised in the axial direction using one-dimensional (1D) coordinates
and assuming cylindrical symmetry. A point kinetics model has been coupled with 1D nuclear
thermal hydraulics and phenomenologically based radiolysis models. This code directly models the
progression of a wetting front through the dry powder. Furthermore, the reactivity feedbacks are

110 specified as functions of both their primary independent variable (e.g. temperature, voidage) and also the bed immersion fraction. The model considers the dynamic motion of water and gas; albeit the powder particles remain stationary. This code is coupled with a steady-state deterministic neutron diffusion code to determine the axial scalar neutron flux distribution at any point in time.

The model described in this paper is applied to the specific case of water from a fire sprinkler 115 system infiltrating a bed of initially dry low-enriched UO_2 powder. This type of nuclear criticality accident has been described by both the International Atomic Energy Agency (IAEA) [17] and by the Japan Atomic Energy Research Institute (JAERI) [3] as a criticality safety case to consider. The neutron kinetics of the system are analysed and the fission power and energy released over a period of time after the initial nuclear criticality excursion are reported.

120 2. Model Phenomenology

2.1. Point Kinetics

The derivation of the point kinetics equations is well-known and may be found in Duderstadt and Hamilton [33] and Stacey [34]. The following equations define the change in neutron density and delayed neutron precursor concentration over time in the fissile powder system:

$$\begin{aligned} \frac{dn(t)}{dt} = & \frac{\beta_{\text{eff}} n(t)}{\Lambda} [R_{\text{imm}}(f_{\text{imm}}) + \Delta R_{\text{th,p,dp}}(T_{\text{p,dp}}, f_{\text{imm}}) + \Delta R_{\text{th,p,wp}}(T_{\text{p,wp}}, f_{\text{imm}}) \\ & + \Delta R_{\text{th,w,wp}}(T_{\text{w,wp}}, f_{\text{imm}}) + \Delta R_{\text{void,wp}}(v_{\text{f,g,wp}}, f_{\text{imm}}) + \Delta R_{\text{void,pooled}}(v_{\text{f,g,pooled}}, f_{\text{imm}}) - 1] \\ & + \sum_{i=1}^6 \lambda_i C_i(t) + s_n \quad (1) \end{aligned}$$

$$\frac{dC_i}{dt} = -\lambda_i C_i + \frac{\beta_i}{\Lambda} n(t) \quad (2)$$

where n [neutrons ($\text{cm}^{-3} \cdot \text{UO}_2$)] is the neutron density in the system, C_i is the amount of delayed neutron precursors per cm^3 of UO_2 for delayed neutron precursor group i , Λ [s] is the mean prompt neutron generation time, β_{eff} [-] is the total delayed neutron fraction, β_i [-] is the delayed neutron fraction for precursor group i and λ_i [s^{-1}] is the decay constant for neutrons in precursor group i . The variable s_n [neutrons s^{-1} ($\text{cm}^{-3} \cdot \text{UO}_2$)] is the intrinsic source density of neutrons, discussed further in Section 2.2. The reactivity of the system, measured in dollars (\$), is defined

in components where R_{imm} is the wetting-induced reactivity (usually positive reactivity effect), $\Delta R_{\text{th,p,dp}}$, $\Delta R_{\text{th,p,wp}}$ and $\Delta R_{\text{th,w,wp}}$ represent thermal reactivity feedbacks due to temperature changes in the dry powder, wet powder and pore water respectively. Void reactivity feedback is modelled in the pores and pooled water by the variables $\Delta R_{\text{void,wp}}$ and $\Delta R_{\text{void,pooled}}$ respectively. The bed immersion fraction, f_{imm} , is defined in Equation 3 as the ratio of the height of the wetted powder region to the total height of the powder bed.

$$f_{\text{imm}}(t) = \frac{H_{\text{wp}}(t)}{H_{\text{dp}}(t) + H_{\text{wp}}(t)} \quad (3)$$

125 where H_{dp} [m], H_{wp} [m] are the heights of the dry powder and wetted powder regions respectively.

2.2. Neutron Source for the Point Kinetics Equations

In point kinetics calculations, for nuclear reactor physics and nuclear criticality safety applications, there are usually either distributed intrinsic (or inherent) neutrons sources, localised extrinsic (or installed) neutron sources or combinations of both of these sources [35]. There are a number of physical processes that lead to distributed intrinsic neutron source such as spontaneous fission
130 [36, 37], (α, n) nuclear reactions [38], cosmic ray induced spallation neutron sources [39] and also (γ, n) nuclear reactions [40]. In general, spallation neutron sources and (γ, n) nuclear reactions are not important for nuclear criticality excursion analysis. However, spallation neutrons sources are important for accelerator driven systems (ADS) [41, 42]. Photo-neutron or (γ, n) nuclear reactions
135 are important for the analysis of heavy water (D_2O) moderated reactors such as the CANDU (Canadian Deuterium-Uranium) reactor [43]. In this case the point kinetics equations are modified by the inclusion of additional delayed neutron groups to represent the effect of the photo-neutron sources [44]. Moreover, it is important to account for photo-neutrons as not accounting for these neutron sources may lead to inaccuracies in the determination of the reactivity of the system [44].

140 In general, the most important neutron sources in nuclear criticality excursion problems are usually spontaneous fission and (α, n) nuclear reactions and these have been measured for fissile liquids such as uranyl nitrate and uranyl fluoride [45, 46]. The (α, n) neutron source occurs through interaction of the α particles, emitted in natural radioactivity of uranium, with light nuclei [38]. The relative strengths and importance of spontaneous fission and (α, n) nuclear reactions depends upon
145 the uranium enrichment of the fuel [35]. For sub-critical systems with weak neutron sources it may be important to model the stochastic fluctuations of the neutron field and this can be achieved using

either Monte Carlo methods [47] or the Pál-Bell equations [48, 49]. Hansen developed a criterion for determining whether systems were in the weak neutron source regime where stochastic effects dominate [50]. This weak neutron source regime is important in the start-up of nuclear reactors with highly enriched nuclear fuels [51]. This is the reason why primary and secondary extrinsic
150 neutron sources are installed within nuclear reactors to avoid potential blind nuclear reactor start-up scenarios [52]. Such blind nuclear reactor start-up scenarios can lead to overheating and damage of the nuclear fuel within the nuclear reactor core [53].

In nuclear criticality safety analysis the presence of weak neutron sources can lead to issues in measuring neutron fluxes as well as the degree of sub-criticality [54]. Moreover, it may lead to significant variations in burst wait times should a nuclear criticality excursion occur [55, 56]. Therefore, in general, for accurately modelling nuclear criticality excursions, it is useful to know whether the system is in the weak neutron source regime [50] and the magnitudes of the distributed intrinsic (or inherent) and any localised extrinsic (or installed) neutron sources to ensure that appropriate initial conditions are prescribed for the problem being analysed. These prescribed initial conditions for the nuclear criticality excursions analysed in this paper are derived by assuming the initially sub-critical systems have achieved some steady-state prior to the nuclear criticality excursion [53, 57]. This assumption leads to the following prescribed initial neutron and delayed neutron precursor densities [53, 57]:

$$n(0) = -\frac{s_n \Lambda}{R_T(0) \beta_{\text{eff}}} \quad (4)$$

and

$$C_i(0) = -\frac{\beta_i s_n}{\lambda_i R_T(0) \beta_{\text{eff}}} \quad (5)$$

where $n(0)$ [neutrons $\text{s}^{-1} \text{ cm}^{-3}$] is the initial sub-critical neutron density and $R_T(0)$ [\$\beta\$] is the initial
155 total reactivity of the system. These equations assume that the intrinsic and extrinsic neutron sources have been appropriately averaged over the volume of the system domain.

However, the averaging of the neutron sources over the system domain is itself a non-trivial task in order to develop a rigorous averaging procedure in order to account for any spatial, energy, angular and temporal variation of the neutron sources. In general, for point kinetics calculations,
160 an appropriately adjoint weighted fundamental mode neutron source must be derived to account for the relative spatial, angular, energy and temporal variation of the neutron sources [58]. The magnitude of the intrinsic neutron source can be determined using either the deterministic code

SOURCES-4C [59, 60] or an appropriate Monte Carlo code such as MC21 [61]. SOURCES-4C and MC21 can evaluate both the spontaneous fission and (α, n) neutron sources. Recent research
165 has indicated that the intrinsic neutron source values predicted by SOURCES-4C and MC21 are broadly in agreement for systems such as 2.4 % enriched uranium dioxide (UO_2) [62].

The nuclear criticality excursions that are presented in this paper involve a 500 kg powder bed made of 5 % enriched uranium dioxide (UO_2) powder. Furthermore, these simulations have assumed that there are no extrinsic neutron source present within the system. Therefore, it is assumed
170 that the only neutron sources are the intrinsic (or inherent) neutron sources from spontaneous fission and (α, n) nuclear reactions. In general, either SOURCES-4C or an ancillary Monte Carlo simulation would have to be performed in order to determine an appropriate magnitude for the intrinsic neutron sources. However, for the purposes of this paper and for all the nuclear criticality excursions simulations performed the spontaneous fission and (α, n) neutron source determined by
175 Griesheimer et al. [62] will be utilised. The reason for using the 2.4 % enriched UO_2 intrinsic neutron source value from the Griesheimer et al. [62] paper is because the magnitude, or strength, of this neutron source will not differ substantially to the value for the 5 % enriched UO_2 case. Sensitivity analysis of the intrinsic neutron source strength, outlined in Section 6.3, has shown that nuclear criticality transients in the present system are not sensitive to the prescribed variations of
180 the intrinsic neutron source strength. Griesheimer et al. [62] determined a value for 2.4 % enriched uranium dioxide (UO_2) of 6.248×10^{-4} (α, n) neutrons s^{-1} ($\text{g}^{-1} \cdot \text{UO}_2$) and 1.172×10^{-2} spontaneous fission (SF) neutrons s^{-1} ($\text{g}^{-1} \cdot \text{UO}_2$) using SOURCES-4C. For the MC21 Monte Carlo simulations Griesheimer et al. [62] determined a value of 6.248×10^{-4} (α, n) neutrons s^{-1} ($\text{g}^{-1} \cdot \text{UO}_2$) and 1.168×10^{-2} neutrons s^{-1} ($\text{g}^{-1} \cdot \text{UO}_2$). The agreement between the SOURCES-4C values and the
185 MC21 values for the neutron sources is approximately 2 %. In addition the spontaneous fission (SF) process is by far the most dominate process involved in producing the intrinsic neutron source as expected. Given that the theoretical density of UO_2 powder is 10.95 g cm^{-3} , the corresponding total neutron source due to (α, n) interactions and spontaneous fission is $s_n = 0.135 \text{ neutrons s}^{-1} (\text{cm}^{-3} \cdot \text{UO}_2)$.

In order to determine whether the 5 % enriched uranium dioxide (UO_2) powder systems studied in this paper is in the stochastic regime it is important to apply the Hansen criterion [50]. If the system obeys Hansen's criterion then the system is in a regime where stochastic effects in the neutron kinetics become important. Hansen [50] stated that weak neutron limit criterion was given

by:

$$\frac{2S_n\tau}{\bar{\nu}\Gamma_2} \ll 1 \quad (6)$$

where $\bar{\nu}$ is the mean number of neutrons per fission, S_n [neutrons s^{-1}] is the total distributed neutron source and Γ_2 [-] is the Diven factor, which can be expressed as:

$$\Gamma_2 = \frac{\overline{\nu(\nu - 1)}}{\bar{\nu}^2} \quad (7)$$

Basoglu [23] noted that generally for 5 % enriched UO_2 powder systems the value of $2/\bar{\nu}\Gamma_2 \approx 1$, given that the Diven factor, $\Gamma_2 \approx 0.8$ [50, 63, 64]. This yields Hansen's criterion as:

$$S_n\tau \ll 1 \quad (8)$$

The intrinsic neutron source density determined by Griesheimer et al. [62] is used as an approximation for the 5 % enriched UO_2 powder system analysed in this paper. Given the powder bed mass of 500 kg and approximate density of UO_2 of $0.01095 \text{ kg cm}^{-3}$, the total distributed intrinsic neutron source is calculated to be $S_n = 6172 \text{ neutrons s}^{-1}$. The mean neutron lifetime near criticality has been described in Section 4.7 as $\tau = 5.33 \times 10^{-5} \text{ s}$. Using the Hansen [50] criterion:

$$S_n\tau = 0.329 \quad (9)$$

190 This value only marginally satisfies the condition specified in Equation 8 [58]. However, in reality the system will largely be deterministic in its behaviour even with this marginal value [58]. Therefore, for the system analysed in this paper stochastic neutron kinetics effects do not need to be considered any further.

2.3. Water Infiltration

195 Once water has entered the system, it fills the interstitial voids between the powder particles, increasing the total system bulk density. Therefore, the infiltration of the water into the powder bed will yield a system with an overall increased macroscopic neutron scattering cross-section. Fast neutrons born through fission events are now more likely to be thermalised or moderated, through the scattering of neutrons with the hydrogen atoms of the water molecules within the interstitial
200 voids. This neutron thermalisation, or moderation effect, leads to an increased probability of subsequent thermal neutron fission occurring within the fissile material in the powder bed. The resulting increased fission rate leads to an overall positive reactivity effect on the system.

An accurate model of unsteady infiltration into a bed of initially dry, or partially saturated fissile powder is required to model the effect water has on the system reactivity. Water infiltration may be considered as a reactivity insertion and is the predominant driving force behind any subsequent nuclear criticality excursion. Prior research on the modelling of fissile powder systems assumed wetting caused a constant ramp reactivity insertion [9, 23, 24, 65]. However, without modelling the infiltration phenomenon, the variation in the reactivity over time as a direct result of wetting is unknown. Knowledge of the spatial water content during a nuclear criticality excursion is essential when analysing mechanisms that may quench the transient, such as thermal expansion, Doppler broadening, radiolysis and boiling. These factors affect the nuclear criticality assessment and safety case by limiting the fission power and determining the magnitude of negative reactivity feedback.

Infiltration phenomena have been extensively studied within the fields of geology and soil mechanics. This has led to the development of various numerical modelling techniques [66, 67, 68]. One such method defined by Bear [67] uses constitutive closure relations to solve two-phase porous media flow equations for phase saturations and pressures. Experiments to empirically determine necessary values for these closure correlations have not been carried out for fissile powder systems, posing a challenge when implementing this model.

An alternative approach proposed by Richards [69] assumes that the non-wetting phase is passive, thus reducing the complexity of the system to a single-phase [70]. Therefore, this approach assures that the gaseous phase remains at atmospheric pressure and does not influence the flow of water. This assumption is valid when the air can move freely to the atmosphere. Richards' equation assumes that the wetting phase fluid is incompressible. A constitutive relation is also required when implementing this method, which typically takes a non-linear form [71].

Another approach, first described by Green and Ampt [72] (GA), that does not require the use of empirical correlations, has been used in the proposed model. This model considers there to be a sharp interface between the two immiscible fluids, creating a fully saturated region, and a dry region. The GA equation also assumes that the air phase is passive and free to escape from the domain. Figure 1 shows how the domain may be separated using the GA infiltration model approach. The location of the planes separating individual regions changes as water percolates through the domain.

In an experimental study looking at the infiltration of water into earthen liners, Albrecht and Cartwright [73] found that the GA method was able to successfully model the propagation of the

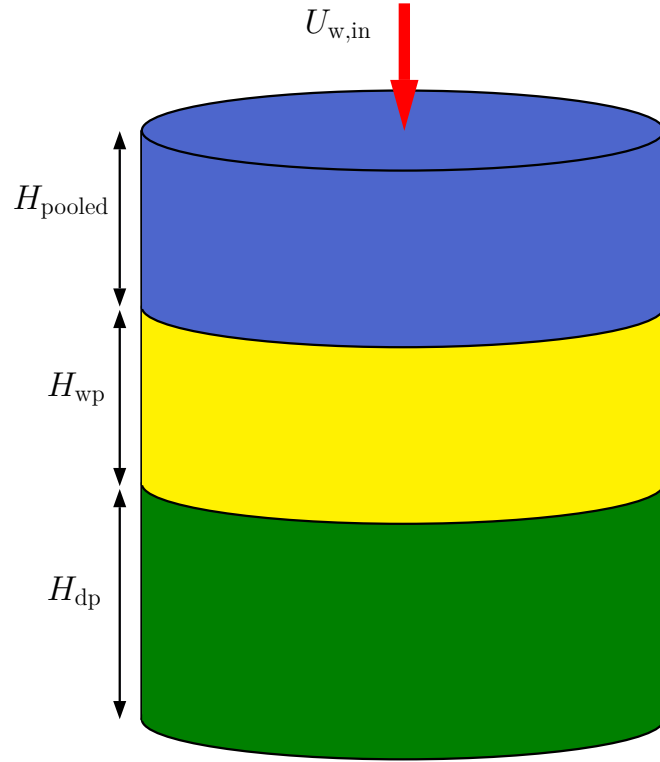


Figure 1: The GA representation of the three region powder drum. The top blue region represents the pooled water on the surface of the powder. The middle yellow region corresponds to the homogeneous mixture of water and UO_2 powder. The bottom green region represents the dry UO_2 powder. The change in height of each region as a function of time is represented through a set of ODEs that may be solved numerically.

235 wetting front. A study comparing the wetting front propagation using the GA model and Richards' equation by Govindaraju et al. [74] supports this finding.

Preliminary results have shown that when water infiltrates a fissile powder bed, a significant amount of radiolytic gas and water vapour may be produced. Therefore, the downward flow of water through the domain must be retarded to model the resistance to flow induced by the generation and advection of gas bubbles within the wetted fissile powder. The original GA equation may be found within the research literature [72, 75]. The proposed adjustment which accounts for the impedance to flow due to the presence of gas is:

$$\frac{dH_{wp}}{dt} = \frac{K_{sat,e}(1 - \bar{v}_{f,g,wp}(t))}{\epsilon \Delta S_w} \left(\frac{H_{pooled}(t) + h_\psi + H_{wp}(t)}{H_{wp}(t)} \right) \quad (10)$$

where:

$$\Delta S_w = S_{w,sat} - S_{w,0} \quad (11)$$

240 where H_{pooled} [m] is the height of the pooled water region, h_ψ [m] is the hydraulic head due to matric suction. To prevent a singularity arising in Equation 10, the wetted region height, H_{wp} [m], starts at a very small positive value. The variable $K_{sat,e}$ [m s⁻¹] represents the effective saturated hydraulic conductivity, ϵ [-] is the porous bed porosity, $v_{f,g}$ [-] is the fraction of gas present in the pore interstices and $S_{w,sat}$ [-] is the saturation level in the saturated case which may be less than one if there is residual air within the interstices of the pores, and $S_{w,0}$ [-] is the initial uniform saturation which may be greater than zero if there is any initial natural saturation of the porous medium due to hygroscopic effects. The present model assumes that all pores are completely filled with water.

The fraction of gas present in the pores may be defined as:

$$v_{f,g,wp}(t) = \frac{V_{rg,wp}(t) + V_{v,wp}(t)}{V_{rg,wp}(t) + V_{v,wp}(t) + V_{w,wp}(t) + V_{p,wp}(t)} \quad (12)$$

245 where $V_{rg,wp}$, $V_{v,wp}$, $V_{w,wp}$ and $V_{p,wp}$ are the total volumes present in the wet powder region of radiolytic gas, water vapour, water and UO₂ powder all in m³, respectively.

The porosity of the powder bed may be defined as:

$$\epsilon = 1 - \frac{\rho_{p,bulk}}{\rho_{p,th}} \quad (13)$$

where $\rho_{p,bulk}$ [kg m⁻³] is the bulk powder density and $\rho_{p,th}$ [kg m⁻³] is the density of solid UO₂. The bulk powder density may also be defined as:

$$\rho_{p,bulk} = \frac{V_p}{V_T} \rho_{p,th} \quad (14)$$

where V_p [m³] is the volume of powder within the volume element (V_T [m³]) under consideration.

The GA approach models the system as a three region problem, where the increase in height of the wet powder region is expressed by Equation 10. The change in the height of the pooled water and dry powder regions may be described respectively as:

$$\frac{dH_{\text{pooled}}}{dt} = U_{w,\text{in}}(t) - \frac{dH_{\text{wp}}}{dt} \frac{\epsilon \Delta S_w}{(1 - \bar{v}_{f,g,\text{wp}}(t))} \quad (15)$$

$$\frac{dH_{\text{dp}}}{dt} = -\frac{dH_{\text{wp}}}{dt} \quad (16)$$

where $U_{w,\text{in}}(t)$ [m s⁻¹] is the volumetric flow-rate per unit area through the top of the drum.

The hydraulic conductivity, which defines the ease at which water percolates through the system, is considered on the saturated side of the sharp wetting front. For saturated flows the hydraulic conductivity may be characterised using the following equation [66]:

$$K_{\text{sat}} = \frac{k \rho_w g}{\mu_w} \quad (17)$$

where k [m²] is the permeability at full saturation, g [m s⁻²] is the acceleration due to gravity, ρ_w [kg m⁻³] is the density of water and μ_w [Pa s] is the water dynamic viscosity.

Experiments investigating the measurement of hydraulic conductivity were performed by Bouwer [76]. These experiments suggested an adjustment to the saturated hydraulic conductivity of water was required in the unsaturated case, to account for impedance to the flow of water in porous media due to the entrapped air. This adjustment to the hydraulic conductivity led to an improved agreement between models and experiments of water ingress into unsaturated porous media [76]. Using this approach, the effective saturated hydraulic conductivity ($K_{\text{sat,e}}$ [m s⁻¹]) used in Equation 10 is:

$$K_{\text{sat,e}} = \frac{1}{2} K_{\text{sat}} \quad (18)$$

A comprehensive comparison regarding the various empirical and semi-empirical measures of saturated permeability carried out by Odong [77], indicated that the Koseny-Carman formula produced reliable results for particles sizes up to 300 μm . The anticipated particle size range for UO₂ powder produced using various manufacturing methods is from 1 μm to 100 μm distributed log-normally [78, 79]. Therefore, the Koseny-Carman relation has been considered appropriate for determining the saturated permeability. The equation to represent this is [66, 80]:

$$k = \frac{\epsilon^3}{5S_A^2(1 - \epsilon)^2} \quad (19)$$

where S_A [m^{-1}] is the specific surface area which is evaluated as the the surface area exposed to the fluid per unit volume of the solid.

UO₂ powders have different powder particle size distributions and levels of sphericity depending on the manufacturing method utilised [79]. Changes to the powder drum specific surface area arise because of differences in the orientation and packing structure of powders. For this investigation, the fissile porous medium is assumed to contain spherical, mono-dispersed powder particles with surfaces that are fully exposed to the wetting phase fluid. As a result, the specific surface area may be described as:

$$S_A = \frac{6}{d_p} \quad (20)$$

where d_p [m] is the mean powder particle diameter.

2.4. Neutronics Effects and Comparison of Diffusive and Sharp Wetting Fronts

Equation 10 assumes that the infiltrating water may be modelled by a sharp wetting front, neglecting the effect of dispersivity due to tortuosity and path length differences in the powder bed pores. Although it has been shown that the sharp wetting front model predicts infiltration well [73, 74], an understanding of how dispersivity around the wetting front affects the wetting-induced reactivity of the system is not known. A comparison between a sharp and diffusive front model has been carried out to investigate these differences. The Advection-Dispersion equation (ADE) may be used to model infiltration phenomena with dispersion [67, 75, 81]:

$$\frac{\partial S_w(t, z)}{\partial t} = \frac{\partial}{\partial z} \left(D(t) \frac{\partial S_w(t, z)}{\partial z} \right) - U_{\text{ade}}(t) \frac{\partial S_w(t, z)}{\partial z} \quad (21)$$

255 where the dispersivity $D(t) = \alpha_L U_{\text{ade}}(t)$ [$\text{m}^2 \text{s}^{-1}$], α_L [m] is the longitudinal dispersivity and U_{ade} [m s^{-1}] is the advection velocity of the moving plane wetting front.

Bear [67] estimates the longitudinal dispersivity as the ratio of the length of mixing cells, $\Delta l'$ [m], to the distance between the centre of cells, Δl [m]. An assumption is made that both the length of mixing cells and the distance between the centre of cells is on the order of a grain size, giving:

$$\alpha_L = \frac{(\Delta l')^2}{2\Delta l} \approx \frac{d_p}{2} \quad (22)$$

The velocity, $U_{\text{ade}}(t)$, is described in a similar way to Equation 10, accounting for the variable saturation around the wetting front:

$$U_{\text{ade}}(t) = \frac{K_{\text{sat,e}}(1 - \bar{v}_{\text{f,g,wp}}(t))}{\int_0^{H_{\text{p+wp}}} \epsilon(S_w(t, z) - S_{w,0}) \partial z} \left(\frac{H_{\text{pooled}}(t) + h_\psi + H_{\text{wp}}(t)}{H_{\text{wp}}(t)} \right) \quad (23)$$

A series of steady-state Monte Carlo neutron transport simulations were carried out using MCNP to compare the two methods for a range of bed immersion fractions. The sharp wetting front geometry was modelled as a cylinder containing three regions. The dispersed wetting front was modelled by sub-dividing the dispersed front into 100 smaller regions in the MCNP input file. A dry and wet powder region was specified on either side of the dispersed regions. The size of each region and the thermophysical properties of the system were identical to those used in the case study in Section 4, these are summarised in Table 1.

Table 1: System properties.

Property	Symbol	Value	Source
Drum radius	R	0.4 m	[9]
Initial Water height	$H_{\text{pooled},0}$	1.0 m	[9]
Dynamic viscosity of water	μ	0.001 Pa s	[82]
Density of water	ρ_w	1000 kg m ⁻³	[82]
Particle Mass	M_p	500 kg	-
Water Mass	M_w	502.65 kg	-
Enrichment	γ	5 %	[83]
Theoretical Bulk Density of UO ₂	$\rho_{p,\text{th}}$	10950 kg m ⁻³	[12]

The average change in the effective multiplication factor between the two methods is shown in Figure 2. Simulations indicate that there is an increasing difference between the sharp and dispersed wetting front models as both the porosity decreases and mean powder particle size increases. This is expected as larger powder particles at a greater number density create more tortuous paths for the water to percolate through the system. The increasing presence and size of powder particles also led to larger path length differences. Large differences between the two methods are predicted for tightly packed powder beds. For the expected maximum powder particle size ($d_p = 100 \mu\text{m}$) and porosity ($\epsilon = 0.8540$) of the system described in Section 4 the error is approximately $\Delta k_{\text{eff}} = 150$ pcm. This error is small relative to the large wetting-induced reactivities that are predicted as the powder bed becomes wet (see Section 4.8), supporting the use of the sharp wetting front approach. Furthermore, the sharp wetting front approximation also eliminates the necessity for very fine spatial discretisation around the wetting front which is required when small amounts of physical

dispersion are predicted, such as for the powder beds modelled here. Therefore, the sharp wetting front model shall be used for the case study outlined in Section 4.

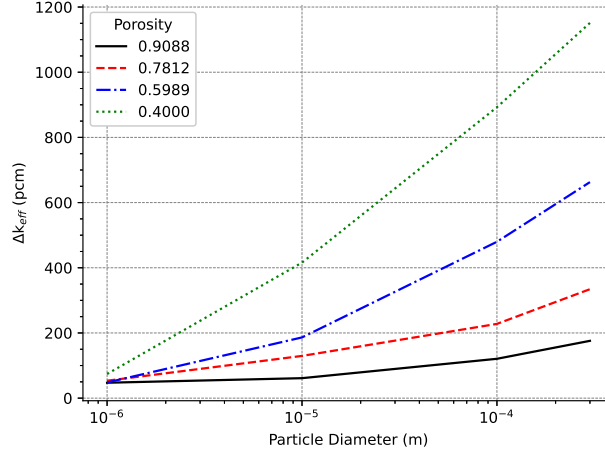


Figure 2: The average difference in the effective multiplication factor, k_{eff} , is shown as the mean powder size increases for the four powder bed porosities that were investigated.

2.5. Thermal Hydraulics and Boiling

The proposed thermal energy balance equations for the powder and water phase are based on several models that can be found in the cited literature [84, 85, 86, 87]. The thermal energy equation in the dry powder region that is valid over the height of the dry powder ($0 < z < H_p(t)$) is defined as:

$$M'_p c_p \frac{\partial T_{p,dp}(t, z)}{\partial t} = E'_{\text{gen},p}(t, z) + \frac{\partial}{\partial z} \left\{ \kappa_p(T_{p,dp})(1 - \epsilon) \pi R^2 \frac{\partial T_{p,dp}(t, z)}{\partial z} \right\} \quad (24)$$

where $T_{p,dp}$ [K] is the temperature of the dry fissile powder, c_p [$\text{J kg}^{-1} \text{K}^{-1}$] is the specific heat capacity of the powder, κ_p [$\text{W m}^{-1} \text{K}^{-1}$] is the thermal conductivity of the powder, R [m] is the radius of the cylindrical vessel and $E'_{\text{gen},p}$ [W m^{-1}] represents the generation of thermal energy in the fissile powder due to fission per unit height (see Equation 41). The mass per unit height of the powder, M'_p [kg m^{-1}], may be defined as:

$$M'_p = \rho_{p,th} \pi R^2 (1 - \epsilon) \quad (25)$$

The limits of the wet powder region relative to the base of the cylindrical vessel are:

$$H_{dp}(t) < z < H_{dp+wp}(t) \quad (26)$$

where $H_{dp+wp}(t) = H_{dp}(t) + H_{wp}(t)$. The thermal energy equation that describes the temperature variation of the wet powder bounded by the region in Equation 26 may be written as:

$$M'_p c_p \frac{\partial T_{p,wp}(t, z)}{\partial t} = \frac{\partial}{\partial z} \left\{ \kappa_p(T_{p,wp})(1 - \epsilon) \pi R^2 \frac{\partial T_{p,wp}(t, z)}{\partial z} \right\} + E'_{gen,p}(t, z) - E'_{convec}(t, z) - E'_{boil}(t, z) \quad (27)$$

where $T_{p,wp}$ [K] is the wet powder temperature. The transfer of energy per unit height between the powder and water via convection and boiling are respectively defined as E'_{convec} [W m⁻¹] (see Equation 47) and E'_{boil} [W m⁻¹] (see Equation 52).

The pore water temperature, bounded by Equation 26, may be modelled using the following:

$$c_w \left\{ \frac{\partial}{\partial t} [M'_w(t, z) T_{w,wp}(t, z)] + \frac{\partial}{\partial z} [M'_w(t, z) U_w(t, z) T_{w,wp}(t, z)] \right\} = E'_{gen,w}(t, z) + E'_{convec}(t, z) + \frac{\partial}{\partial z} \left\{ c_w D_{t,wp} \frac{\partial}{\partial z} [M'_w(t, z) T_{w,wp}(t, z)] \right\} \quad (28)$$

where $T_{w,wp}$ [K] is the temperature of the pore water, c_w [J kg⁻¹ K⁻¹] is the specific heat capacity of water and $D_{t,wp}$ [m² s⁻¹] represents the turbulent mixing in the pooled water region. The variable U_w [m s⁻¹] represents the advection of water (see Equation 38), and $E'_{gen,w}$ [W m⁻¹] represents the generation of heat in the water due to fission (see Equation 42). The mass per unit height of the water, M'_w [kg m⁻¹], may be defined as:

$$M'_w(t, z) = \rho_{w,th} \pi R^2 \epsilon S(t, z) \quad (29)$$

where $\rho_{w,th}$ [kg m⁻³] is the density of water at standard atmospheric conditions. Variations in the density of water as a function of temperature are not considered in the thermal hydraulic or infiltration equations. The effect of thermal expansion is considered in the thermal reactivity feedback.

The bounds of the pooled water region relative to the base of the cylindrical vessel are:

$$H_{dp+wp}(t) < z < H_{dp+wp+pooled}(t) \quad (30)$$

where $H_{\text{dp+wp+pooled}}(t) = H_{\text{dp}}(t) + H_{\text{wp}}(t) + H_{\text{pooled}}(t)$. The temperature variation of the pooled water may be described using the following equation:

$$c_w \left\{ \frac{\partial}{\partial t} [M'_w(t, z) T_{w, \text{pooled}}(t, z)] + \frac{\partial}{\partial z} [M'_w(t, z) U_w(t, z) T_{w, \text{pooled}}(t, z)] \right\} = \frac{\partial}{\partial z} \left\{ c_w D_{t, \text{pooled}} \frac{\partial}{\partial z} [M'_w(t, z) T_{w, \text{pooled}}(t, z)] \right\} \quad (31)$$

where $T_{w, \text{pooled}}$ [K] is the pooled water temperature, and $D_{t, \text{pooled}}$ [$\text{m}^2 \text{s}^{-1}$] represents the turbulent mixing in the pooled water region.

Equations 24, 27, 28 and 31 fully describe the axial temperature variation of the powder and water within the cylindrical vessel. These four equations have been solved numerically using prescribed boundary and interface conditions. The interface conditions are prescribed such that at the boundary between regions, there is a continuity of thermal energy between the phases. For the condition when $z = H_{\text{dp}}$, this may be described as:

$$k(T_{p, \text{dp}}) \frac{dT_{p, \text{dp}}(t, H_{\text{dp}})}{dz} = k(T_{p, \text{wp}}) \frac{dT_{p, \text{wp}}(t, H_{\text{dp}})}{dz} \quad (32)$$

When $z = H_{\text{dp+wp}}(t)$ the interface condition describing the thermal advection between the pooled and pore water may be simplified to:

$$T_{w, \text{wp}}(t, H_{\text{dp+wp}}) = T_{w, \text{pooled}}(t, H_{\text{dp+wp}}) \quad (33)$$

Thermal energy transfer to the container is not considered in the current model. A thermal gradient may be introduced at the open top of the vessel to model the effects of water ingress at a given temperature. The resulting boundary conditions at the bottom of the vessel ($z = 0$) and the top of the vessel ($z = H_{\text{dp+wp+pooled}}(t)$) may be respectively described as:

$$\frac{\partial T_{p, \text{dp}}(t, 0)}{\partial z} = 0 \quad (34)$$

$$D_{t, \text{pooled}} \frac{\partial T_{w, \text{pooled}}(t, H_{\text{dp+wp+pooled}})}{\partial z} = -U_{w, \text{in}} (T_{w, \text{pooled}}(t, H_{\text{dp+wp+pooled}}) - T_{\text{in}}) \quad (35)$$

where T_{in} [K] represents the temperature of the water entering the domain.

Two additional conditions have been applied to the wet powder region in the case where the powder bed is fully saturated, i.e. $H_{\text{dp}} = 0$:

$$\frac{\partial T_{p, \text{wp}}(t, 0)}{\partial z} = 0 \quad (36)$$

$$\frac{\partial T_{w,wp}(t, 0)}{\partial z} = 0 \quad (37)$$

290 The temperature of the fissile powder and surrounding water modelled by Equations 24, 27, 28 and 31 have been solved numerically by implementing the boundary and interface conditions outlined by Equations 32 to 37. Each term shown in the governing equations will now be described in more detail.

Fluid thermal advection is represented by the second term in Equations 28 and 31. A number of driving forces contribute to the advective motion of thermal energy throughout the wet powder and pooled water regions. These are distinguished by their forced ($U_{\text{forced,GA}}$ [m s⁻¹]) and free (U_{free} [m s⁻¹]) components:

$$U_w(t, z) = U_{\text{forced,GA}}(t) + U_{\text{free}}(t, z) \quad (38)$$

Forced convection due to the advection of water through the domain usually dominates the thermal convection present in the system. In the proposed model this is governed by the GA equation:

$$U_{\text{forced,GA}}(t) = \frac{K_{\text{sat,e}}}{\epsilon \Delta S_w} \left(\frac{H_{\text{pooled}}(t) + h_{\psi} + H_{\text{wp}}(t)}{H_{\text{wp}}(t)} \right) \quad (39)$$

Density differences between layers of fluid within the porous medium may lead to the onset of convection currents. Wooding [88] noted that the fluid will tend towards an equilibrium temperature due to this effect so long as the density difference persists. Typically these convection currents form vertical laminar flow paths throughout the porous medium. Bear [67] describes a model of free convection in porous media which can be directly derived from the Navier-Stokes equations for incompressible flow. If convective acceleration is also neglected, which is assumed when deriving Equation 10 from the Navier-Stokes equation, then the free convective velocity is:

$$U_{\text{free}}(t, z) = - \frac{kg(\rho_w(z, T_w) - \rho_w(z, T_{w,0}))}{\epsilon \Delta S_w \mu_w(T_w)} \quad (40)$$

295 where $T_{w,0}$ [K] is the initial reference temperature. Variations in the density and viscosity of water are considered only in the buoyancy-induced advection equation and are not present in the GA model. This is consistent with the derivation found in Bear [67].

A turbulent mixing parameter, $D_{t,wp}$, is introduced to model the effect of mixing in the pores between particles due to large volumes of gas that are expected to be present. A similar parameter,

$D_{t,pooled}$, has been defined to model the effects of turbulent mixing within the pooled water region. This approach was predicted to appropriately model the fissile solution system described by Cooling et al. [89].

Thermal diffusion through the powder via conduction is modelled by the final term in Equations 24 and 27.

The generation of energy due to the deposition of fission products within the powder and water has been defined as [87]:

$$E'_{gen,p}(t, z) = (g_p + (1 - g_p)v_{f,g,wp}(t, z))P'(t, z) \quad (41)$$

$$E'_{gen,w}(t, z) = (1 - g_p)(1 - v_{f,g,wp}(t, z))P'(t, z) \quad (42)$$

where g_p [-] is the fraction of kinetic energy held by a fission fragment that is deposited in the particle, and P [W] is the energy generation rate within the system.

The fraction of kinetic energy held by a fission fragment that is deposited in the particle, g_p , depends on both the relative magnitudes of the range of fission products and the size of the fissile powder particle in water. Theoretical considerations of this parameter by Gardner [90] indicated that for homogeneous slurry systems, the kinetic energy of fission fragments may be deposited within both the solid phase and the surrounding aqueous phase. For particle sizes over $\approx 20 \mu\text{m}$, g_p does not depend on powder particle concentration and may be defined as [90, 91]:

$$g_p = \frac{3d_p}{8R_{fiss}} ; d_p < R_{fiss} \quad (43)$$

$$g_p = 1 - \frac{3R_{fiss}}{4d_p} + \frac{R_{fiss}^3}{8d_p^3} ; d_p > R_{fiss} \quad (44)$$

where R_{fiss} [m] is the range of fission fragments in solid UO_2 .

The energy generation rate or fission power, P [W] at an instant in time, t , and at a position z within a cylindrically shaped differential volume element, $\Delta V = \pi R^2 \Delta z$, where Δz is the differential height of the volume element which has radius R . The energy generation rate, or fission power, is the product of the average energy released from a fission reaction, E_f [J fission $^{-1}$], multiplied by the fission rate, RR_f [fissions $\text{cm}^{-3} \text{s}^{-1}$] and the volume of the system. Mathematically, this may be represented as:

$$P(t, z) = E_f RR_f(t, z) \quad (45)$$

The fission rate in the cylindrically shaped volume element, ΔV , of height Δz and radius R is given by:

$$RR_f(t, z) = \int_0^\infty \int_z^{z+\Delta z} \Sigma_f(t, z, E) N(t) \hat{\phi}(z, E) v(E) dE dz \quad (46)$$

where E [J] represents the energy of the neutron, N is the total number of neutrons present in the system, Σ_f [cm^{-1}] is the macroscopic neutron fission cross-section at time, t and $\hat{\phi}$ [cm^{-1}] is the normalised, energy dependent, radially averaged, scalar neutron flux shape function that defines the distribution of neutrons within the system at position z with energy E . The velocity of the
310 neutrons, v [cm s^{-1}], is approximately $2.2 \times 10^5 \text{ cm s}^{-1}$ and $1.4 \times 10^9 \text{ cm s}^{-1}$ for thermal and fast neutrons respectively. The variable E_f represents the average energy released from fission, which for ^{235}U is $E_f = 3.2 \times 10^{-11} \text{ J}$. The normalised, energy dependent, radially averaged, scalar neutron flux, $\hat{\phi}$ at position z with energy E , is modelled using the steady-state neutron diffusion equation, this is discussed further in Section 2.7. The macroscopic fission cross-section is determined using
315 the Monte Carlo code Serpent, this is discussed further in Section 4.11.1.

The main mechanism of thermal energy transfer between the UO_2 powder and surrounding water is via convection. Heat transfer by thermal radiation between the fissile powder and surrounding water is negligible in comparison. Conduction is considered only through the process of boiling. The following equation defines the specific rate of transfer of thermal energy from the powder to the surrounding water:

$$E'_{\text{convec}}(t, z) = h_{p,w}(t, z) v_{f,w,wp}(t, z) A_{p,w}(t, z) (T_{p,wp}(t, z) - T_{w,wp}(t, z)) \quad (47)$$

where $A_{p,w}$ [m] is the interfacial area between the powder and water per unit height, $v_{f,w,wp} = 1 - (v_{f,g,wp} + v_{f,p,wp})$ is the volume fraction of water and $h_{p,w}$ [$\text{W m}^{-2} \text{ K}^{-1}$] is the convective heat transfer coefficient between the powder and water.

When assuming spherical, mono-disperse particles, the interfacial area per unit height may be readily defined as:

$$A_{p,w} = \frac{6\pi R^2(1 - \epsilon)}{d_p} \quad (48)$$

A correlation by Wakao and Kaguei [92] may be used to define the convective heat transfer in packed beds by matching to experimental data. This empirical correlation accurately predicted the interfacial convective heat transfer coefficient in packed beds with wide-ranging properties. An investigation by Kuwahara et al. [93] used microscopic pore-scale analysis to develop a correlation

that corroborates the results of the correlation in [92]. The correlation in [92], shown in Equation 49 has been considered for the proposed model.

$$h_{p,w}(t, z) = \frac{\kappa_w(T_{w,wp})}{d_p} \left[2 + 1.1 Pr^{1/3} Re_{d_p}^{0.6}(t, z) \right] \quad (49)$$

where κ_w [W m⁻¹ K⁻¹] is the fluid thermal conductivity, Re_{d_p} [-] represents the Reynolds number where the characteristic length is the powder diameter, and Pr [-] is the fluid Prandtl number. The Reynolds number may be defined as:

$$Re_{d_p}(t, z) = \frac{\rho_w U_w(t, z) d_p}{\mu_w} \quad (50)$$

The Prandtl number may be defined as:

$$Pr = \frac{c_w \mu_w}{\kappa_w} \quad (51)$$

Small variations in the convective heat transfer coefficient, through the selection of different correlations, are not expected to affect the rate of heat transfer between the powder and water. This is because the interfacial area between the powder and water is so great for small particles (see Equation 48), that the overall rate of heat transfer is relatively high.

When the system becomes supercritical, the increased rate of energy generation causes a corresponding increase in the rate of thermal energy deposited in the system. If the thermal reactivity feedback due to Doppler broadening and thermal expansion does not sufficiently decrease the energy generation rate of the system, the temperature of the water may increase up to the boiling point. At the boiling point, any further heat transfer into the water will lead to evaporation. The increased voidage due to the presence of vapour in the system will act as negative reactivity feedback. In this case, the production of vapour will strongly affect the fission rate, which is directly proportional to the energy generation rate. This is expected to be the main mechanism for heat loss from the powder and water. Therefore, this is an important phenomenon to consider when modelling such systems.

Once the edge of the powder particle reaches the boiling temperature of the surrounding water, any additional heat transferred to the water phase provides energy to vaporise the water. The proposed film boiling model, described within Faghri and Zhang [82], is shown in Equation 52. Using this model, energy is transferred from the powder, into the film and bulk liquid. This correlation is valid when the full surface area of an immersed sphere is in contact with water. Although this may

not be directly comparable to the geometry being considered, the high porosities simulated ensure that most of a single UO_2 powder particle is surrounded by water.

$$E'_{\text{boil}}(t, z) = h_{\text{p,v}}(t, z) a_{\text{p,w}}(t, z) v_{\text{f,w,wp}}(t, z) \max[0, (T_{\text{I}}(t, z) - T_{\text{sat}})] \quad (52)$$

where $h_{\text{p,v}}$ [$\text{W m}^{-2} \text{K}^{-1}$] is the boiling heat transfer coefficient, T_{sat} [K] is the boiling point of water at atmospheric pressure and T_{I} [K] is the interface temperature between the powder particle wall and surrounding liquid. The interface temperature is defined as the average between the powder and water temperature:

$$T_{\text{I}}(t, z) = \frac{1}{2} (T_{\text{p,wp}}(t, z) + T_{\text{w,wp}}(t, z)) \quad (53)$$

The proposed model does not consider condensation phenomena. The high rate of heat transfer between the powder and water means that both phases are expected to reach the boiling point of water at the same time. The corresponding generation of gas due to this latent heat transfer may be represented as follows:

$$\frac{\partial V'_{\text{v}}(t, z)}{\partial t} = \frac{E'_{\text{boil}}(t, z)}{\rho_{\text{v}} h_{\text{l,v}}} \quad (54)$$

where ρ_{v} [kg m^{-3}] and $h_{\text{l,v}}$ [J kg^{-1}] are the vapour density and latent heat of vaporisation, respectively. The variable V'_{v} [m^2] represents the volume of gas per unit height present in the domain. These values are specified at atmospheric pressure and the boiling point of water.

Research by Leinhard and Dhir [94] found a correlation, shown in Equation 55, for the film boiling heat transfer coefficient on the surface of an immersed sphere. The full surface area of the spherical particle is assumed to be in contact with the surrounding liquid, making the heat transfer coefficient similar to the case of a fully immersed sphere.

$$h_{\text{p,v}}(t, z) = 0.67 \left[\frac{\lambda_{\text{v}}^3 \rho_{\text{v}} g (\rho_{\text{w}} - \rho_{\text{v}}) h'_{\text{l,v}}}{\kappa_{\text{v}} d_{\text{p}} (T_{\text{I}}(t, z) - T_{\text{sat}})} \right]^{0.25} \quad (55)$$

where κ_{v} [$\text{W m}^{-1} \text{K}^{-1}$] is the vapour thermal conductivity and μ_{v} [Pa s] is the dynamic viscosity of the vapour. The adjusted latent heat of vaporisation, $h'_{\text{l,v}}$ [J kg^{-1}], may be defined as:

$$h'_{\text{l,v}}(t, z) = h_{\text{l,v}} + 0.4 c_{\text{v}} (T_{\text{I}}(t, z) - T_{\text{sat}}) \quad (56)$$

where c_{v} [$\text{J kg}^{-1} \text{K}^{-1}$] is the specific heat capacity of the vapour.

2.6. Radiolysis and Gas Advection

2.6.1. Radiolytic Gas Production

Ionising radiation causes molecules within the water to break into smaller molecules through a process known as radiolysis. Fission fragments are known to be the main cause of radiolysis in AHRs [95]. The problem of water decomposition within slurry reactors has been proposed to be similar to that of AHRs [96]. During primary radiolysis, these ionised particles dissociate into free radicals $\text{H}\cdot$ and $\cdot\text{OH}$. Many free radicals combine before they diffuse apart, forming more stable molecules. The yields of the primary species depend strongly on the linear energy transfer (LET) parameter. The LET is the energy that is transferred to the water per unit length along the fission track. Yields of H_2 and H_2O_2 are highest for radiations with high LET, whilst yields of free radicals such as H and OH are largest for radiations with low LET. Furthermore, free radicals re-combine in dense fission tracks very quickly and are therefore neglected in the proposed model [95]. The effect of hydrogen peroxide (H_2O_2) which decomposes in water is not considered in the present model.

Small pockets of gas get trapped in the interstices of the powder particles throughout water infiltration. These small pockets of gas are in dynamic equilibrium with the dissolved gasses in the pore water. The decomposition of water due to radiolysis leads to the production of small bubbles along the fission tracks. These small bubbles will collapse if the dissolved gas concentration is below their dynamic equilibrium concentration ($C_{\text{H}_2,\text{equil}}$). If the dissolved gas concentration rises above the equilibrium concentration, the direction of mass transfer reverses, now going from the dissolved to the gaseous phase. The theory of gas bubble formation is described in more detail by Spiegler et al. [97]. The following condition expresses the equilibrium concentration of gas within the pores of the porous medium:

$$C_{\text{H}_2,\text{equil}} = \left(P_w + \frac{2\sigma}{r_{\text{bubble}}} \right) H_{\text{H}_2,\text{sol}} y_{\text{H}_2} \quad (57)$$

where P_w [Pa] is the water pressure, $H_{\text{H}_2,\text{sol}}$ [$\text{mol m}^{-3} \text{ Pa}^{-1}$] is Henry's constant for hydrogen, σ [N m^{-1}] is the surface tension of water and y_{H_2} [-] is the mole fraction of hydrogen gas in the trapped bubble. The radius of the gas bubble, r_{bubble} [m], can play a significant role in the determination of the equilibrium concentration. For fast transients in fissile solution systems bubble nucleation typically occurs on fission track bubbles where the bubble size is on the order of 10^{-8} m [98]. However, bubble nucleation within porous media is expected to be different due to the presence of a large number of solid particles in the water. Direct numerical simulation (DNS) using microscopic pore-network models by Li and Yortsos [99] indicated that the radius of the

porous cavity where the trapped air is located will govern the equilibrium concentration. Mean
particle sizes of UO_2 powders indicate that gas trapped within a porous cavity may have a radius
360 on the order of 10^{-7} m to 10^{-5} m. Therefore, the gas held within porous cavities provides better
nucleation sites at a lower concentration than on fission track bubbles. Consequently, mass transfer
of radiolysis products to the gaseous phase is modelled at the porous cavity as opposed to across
the surface of the fission track bubbles.

The present model assumes that hydrogen produced via radiolysis is instantaneously present
365 within the gaseous phase. The low hydrogen content of atmospheric air means that only small
amounts of dissolved hydrogen would be required for mass transfer into the gaseous phase. For
this reason, the assumption has been considered appropriate. Oxygen is produced via secondary
reactions and is not a product of the initial break-up of water. The fission yield, typically governed
by the magnitude of the initial fission rate, is not affected by the presence of oxygen since it forms
370 much later in the transient. Therefore, the production of oxygen as a by-product of radiolysis has
not been considered in the proposed model.

The number of molecules produced of a given species X per 100 eV of fission energy absorbed
by the water is denoted $G(X)$. In the proposed model the hydrogen yield must be converted to a
volumetric hydrogen yield through the use of the ideal gas law:

$$V_{rg} = \frac{N_{rg}RT_{rg}}{P_{rg}} \quad (58)$$

where $R = 8.31 \text{ J mol}^{-1} \text{ K}^{-1}$ is the universal gas constant, N_{rg} is the number of moles of gas,
 T_{rg} [K] and P_{rg} [Pa] is the gas temperature and pressure respectively.

As noted by Steele et al. [96], a fraction of the energy (defined as g_p in Equations 43 and 44) from
fission recoil fragments are deposited in the water, the remaining energy is deposited in the powder.
An additional multiplier that accounts for the fractional occupation of the pores surrounding the
particle by water must also be included. This leads to the following definition for the volumetric
generation of H_2 per unit height, $v_{rg}(t, z)$ [m^2], within the water:

$$\frac{\partial V'_{rg}(t, z)}{\partial t} = (1 - g_p)(1 - v_{f,g}(t, z))G_V(\text{H}_2)P'(t, z) \quad (59)$$

where $G_V(\text{H}_2)$ [$\text{m}^3 \text{ J}^{-1}$] is the volumetric yield of hydrogen from radiolysis due to fission recoil
375 fragments.

2.6.2. Bubble Size and Advection Velocity

The advective velocity of the bubbles through the domain will ultimately govern the rate of loss of gas from the system and consequently the variation in reactivity feedback due to voidage. The advection of radiolytic gas and water vapour through the wet powder and pooled water region may be respectively defined as:

$$\frac{\partial V'_{\text{rg}}(t, z)}{\partial t} = -U_g(t, z) \frac{\partial V'_{\text{rg}}(t, z)}{\partial z} \quad (60)$$

$$\frac{\partial V'_v(t, z)}{\partial t} = -U_g(t, z) \frac{\partial V'_v(t, z)}{\partial z} \quad (61)$$

where U_g [m s^{-1}] is the gas advection speed through the domain and V'_v [m^2] represents the volume of gas per unit height present in the domain. In order to estimate this value, an understanding of bubble size and flow regimes is required.

380 Gas bubble sizes in the wetted fissile powder may be compared to those observed in three-phase fluidised bed systems due to their geometric similarities. For systems with small solid particles $\approx 88 \mu\text{m}$ in size, the Sauter mean diameter of bubbles were found to be similar to that of a two-phase system with no solid phase [100]. This is supported by experiments conducted by Quicker et al. [101] where bubble characteristics for low solid volume was similar to that of a two-phase
385 system. The proposed model expresses the three-phase powder bed as a two-phase system by using an effective medium viscosity approach [102].

The interfacial area concentration may be used to determine the Sauter mean gas bubble diameter. Comprehensive reviews of existing correlations may be found in Beenackers and Van Swaaij [103], and Jo [104]. Due to disparities in three-phase fluidised bed systems, large variations in the interfacial area concentration correlation exists in the cited literature. A number of correlations, based on experimental data, are only valid for the system which they were developed for [101, 105]. However, a correlation that has been shown to be valid for a wider range of solid volume fractions and particle sizes has been defined in Beenackers and Van Swaaij [103] as:

$$a_I = a_{I,0} \left(\frac{\mu_w}{\mu_{\text{eff}}} \right)^{0.21} \left(\frac{\rho_{\text{eff}}}{\rho_w} \right)^{0.1} \quad (62)$$

where a_I [m^{-1}] is the interfacial area concentration of the gas in the wetted powder, $a_{I,0}$ [m^{-1}] is the interfacial area concentration in pure water, and ρ_{eff} [kg m^{-3}], μ_{eff} [Pa s] are the effective medium densities and viscosities which are defined as follows:

$$\rho_{\text{eff}} = \rho_{\text{p,th}}(1 - \epsilon) + \rho_{\text{w,th}}\epsilon \quad (63)$$

$$\mu_{\text{eff}} = \mu_w \epsilon^{-2.8} \quad (64)$$

Research carried out by Zeitoun et al. [106] investigating interfacial area concentration in the bubbly flow regime for two-phase systems is shown in Equation 65.

$$a_{I,0} = 556.4 v_{f,g}^{0.74} \quad (65)$$

The Sauter mean diameter of a bubble (d_b [m]) may be defined as:

$$d_b = \frac{6 v_{f,g}}{a_I} \quad (66)$$

Substitution of Equations 62 and 65 into Equation 66 yields the following relation for the bubble diameter as a function of the void fraction:

$$d_b = 1.0784 \times 10^{-2} v_{f,g}^{0.26} \left(\frac{\mu_w}{\mu_{\text{eff}}} \right)^{-0.21} \left(\frac{\rho_{\text{eff}}}{\rho_w} \right)^{-0.1} \quad (67)$$

where the constant 1.0784×10^{-2} has units of metres, yielding d_b in units of metre.

A correlation may now be found to calculate the velocity of gas bubbles in the pore and pooled water. Experimental correlations developed by Zuber and Findlay [107] concerning vertical gas flow in pipes may be used for a porous medium model if considering the system as a bundle of tubules. Later work by Roosevelt and Corapcioglu [108] found that although gas transport in porous media may on occasion be modelled using a bundle of capillary tubes, actual gas transport in porous media deviates significantly from this idealised flow model. This could be due to the tortuosity of the porous medium causing significant lateral movement of bubbles.

For high slurry flows (< 5% powder by volume) Yang et al. [109] found that the gas advection velocity may be correlated to solid volume fraction within the porous medium. A review of gas advection velocities for slurries with higher volume fractions may be found in the cited literature [110]. Work by Cheng et al. [110] highlighted the importance of using a correlation that is valid for the expected mean particle size. It was found that smaller sized powders were easier to displace, meaning that less force was required for bubbles to push to the surface. At larger powder diameters, bubbles were more likely to become trapped [111]. For some porous media considered by Roosevelt and Corapcioglu [108], it was observed that bubbles below a critical size would never reach the surface and they would either become entrapped or coalesce into larger bubbles. A model for bubble

breakup and coalescence within the bubbly flow regime has been described by Jo and Revankar [112] but is out of the scope of this investigation.

The proposed model assumes that gas flows are predicted to be solely within the bubbly flow regime. Although the actual gas flow will vary between different flow regimes, this approximation enables the implementation of the correlation developed by Tsuchiya et al. [113], which applies to fluidised beds. This correlation models the fluidised bed using effective medium properties. Experimental analysis by Li and Prakash [114] found that this correlation correctly predicted the bubble rise velocity for slurry concentrations from 15 % to 40 % by volume. At higher solid particle concentrations this correlation fails because the granular flow of the powder becomes more dominant than fluid dynamic flow [115]. Using this model, the dimensionless bubble velocity, U_b , may be represented as:

$$U'_b = \left[\left(\frac{\text{Mo}^{-0.25}}{K_b} d'_b{}^2 \right)^{-n} + \left(\frac{2c}{d'_b} + \frac{d'_b}{2} \right)^{-0.5n} \right]^{-1/n} \quad (68)$$

where n , c and K_b are empirically determined parameters, Mo is the Morton number and d'_b is the effective bubble diameter. c ranges from 1.2 to 1.4 for mono-component and multi-component liquids respectively. n ranges from 1.6 to 0.8 for purified to contaminated liquids respectively. K_b may be found from:

$$K_b = \max(K_{b,0} \text{Mo}^{-0.038}, 12) \quad (69)$$

where $K_{b,0}$ varies from 14.7 to 10.2 for aqueous solutions (including water) to organic solvents (mixtures) respectively. The Morton number of the may be defined as:

$$\text{Mo} = \frac{g \mu_{\text{eff}}^4}{\rho_{\text{eff}} \sigma^3} \quad (70)$$

The effective bubble diameter may be found using:

$$d'_b = d_b \left(\frac{\rho_{\text{eff}} g}{\sigma} \right)^{0.5} \quad (71)$$

where the bubble diameter, d_b [m], may be found using the correlation defined by Equation 67.

The actual gas velocity used in the governing Equations 73 to 76 may then be defined as:

$$U_g = U'_b \left(\frac{\sigma g}{\rho_{\text{eff}}} \right)^{0.25} \quad (72)$$

2.6.3. Full Equations for Gas Production & Advection

The spatial and temporal fluctuation of vapour and radiolytic gas may now be summarised. The following equations are solved numerically using prescribed boundary and interface conditions.

Radiolysis has been modelled using the generation term specified by Equation 59 and the advection term specified by Equation 60. For the wet powder region, the volume of radiolytic gas per unit height, $V'_{\text{rg,wp}}$, is:

$$\frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial t} = (1 - g_p)(1 - \bar{v}_{\text{f,g,wp}}(t, z))G_V(\text{H}_2)P'(t, z) - U_g(t, z)\frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,wp}} \frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial z} \right) \quad (73)$$

For the pooled water region, the volume of radiolytic gas per unit height, $V'_{\text{rg,pooled}}$, is:

$$\frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial t} = -U_g(t, z)\frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,pooled}} \frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial z} \right) \quad (74)$$

Water vapour may be modelled using the generation term specified by Equation 54 and the advection term specified by Equation 61. For the wet powder region, the volume of water vapour per unit height, $V'_{\text{v,wp}}$, may be represented as:

$$\frac{\partial V'_{\text{v,wp}}(t, z)}{\partial t} = \frac{E'_{\text{boil}}(t, z)}{\rho_v h_{\text{l,v}}} - U_g(t, z)\frac{\partial V'_{\text{v,wp}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,wp}} \frac{\partial V'_{\text{v,wp}}(t, z)}{\partial z} \right) \quad (75)$$

For the pooled water region, the volume of water vapour per unit height, $V'_{\text{v,wp}}$, may be represented as:

$$\frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial t} = -U_g(t, z)\frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,pooled}} \frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial z} \right) \quad (76)$$

The interface conditions between the pore water and pooled water is defined such that mass between the regions is conserved. For the radiolytic gas this may be represented as:

$$\frac{\partial V'_{\text{rg,wp}}(t, H_{\text{dp+wp}})}{\partial z} = \frac{\partial V'_{\text{rg,pooled}}(t, H_{\text{dp+wp}})}{\partial z} \quad (77)$$

Similarly, for the vapour at the interface:

$$\frac{\partial V'_{\text{v,wp}}(t, H_{\text{dp+wp}})}{\partial z} = \frac{\partial V'_{\text{v,pooled}}(t, H_{\text{dp+wp}})}{\partial z} \quad (78)$$

The boundary condition may be found by assuming that gas transfer across the dry powder to wet powder region interface is not permitted. Gas exchange through the base of the vessel in the saturated case is also not permitted. The boundary condition for radiolytic gas and vapour may then be respectively defined as:

$$\frac{\partial V'_{\text{v,wp}}(t, H_{\text{dp}})}{\partial z} = 0 \quad (79)$$

$$\frac{\partial V'_{\text{rg,wp}}(t, H_{\text{dp}})}{\partial z} = 0 \quad (80)$$

Upwind differencing in the numerical discretisation scheme for the gas phase means that an upper boundary condition is not required. Using the interface and boundary conditions prescribed by Equations 77 to 80, the governing Equations 73 to 76 may be solved to determine the distribution of voids within the fissile powder system.

2.7. The Neutron Diffusion Equation and the Scalar Neutron Flux Distribution

An approximation of the axial distribution of neutrons within the system is required to predict how the energy generation rate is distributed throughout the system. The steady-state neutron diffusion equation may be used to capture the axial scalar neutron flux distribution at a prescribed bed immersion fraction. A two-group neutron diffusion model must be used to capture the effect of water on the effective multiplication factor (k_{eff}). The neutron diffusion equation has found widespread use within the field of nuclear reactor physics [33]. The energy-dependent neutron diffusion equation (NDE) is an elliptic partial-integral-differential equation (PIDE) defined as:

$$-\nabla \cdot D(\vec{r}, E) \nabla \phi(\vec{r}, E) + \Sigma_t(\vec{r}, E) \phi(\vec{r}, E) = \int_0^\infty dE' \Sigma_s(\vec{r}, E' \rightarrow E) \phi(\vec{r}, E') + \frac{\chi_p(E)}{k_{\text{eff}}} \int_0^\infty dE' \bar{\nu}(E') \Sigma_f(\vec{r}, E') \phi(\vec{r}, E') \quad (81)$$

where ϕ [neutrons $\text{cm}^{-2} \text{s}^{-1} \text{eV}^{-1}$] is the scalar neutron flux, k_{eff} [-] is the effective multiplication factor, D [cm] is the neutron diffusion coefficient, $\bar{\nu}$ [-] is the average number of neutrons created from a fission event and χ_p [-] is the prompt neutron energy spectrum. The parameters Σ_t and Σ_f are the macroscopic total and fission neutron cross-sections respectively with units cm^{-1} . The term $\Sigma_s(\vec{r}, E' \rightarrow E)$ is the macroscopic single differential scattering neutron cross-section for a neutron scattering from energy E' to a final energy E in the range E to $E + dE$ with units $\text{cm}^{-1} \text{eV}^{-1}$.

Since the porous medium is contained within a cylindrical vessel, spatial discretisation of the domain has been implemented using a two-dimensional (2D) r-z mesh. Vacuum boundary conditions have been prescribed at each boundary, which can be defined as [34]:

$$0 = \frac{1}{4} \phi(r, z) - \frac{1}{2} D(r, z) \frac{\partial}{\partial \vec{n}} \phi(r, z) \quad (82)$$

Energy condensed (or group-collapsed) and resonance self-shielded macroscopic neutron cross-sections have been determined using the results of neutron transport simulations performed using

the Monte Carlo code Serpent at different bed immersion fractions. These energy condensed and resonance self-shielded macroscopic neutron cross-sections are specified using a two-group structure. The thermal neutron energy group was prescribed using the following lower and upper energy range boundaries:

$$0 \text{ eV} < E_{\text{therm}} < 0.0625 \text{ eV} \quad (83)$$

where E_{therm} is the thermal neutron energy group. The fast neutron energy group was prescribed using the following lower and upper energy range boundaries:

$$0.0625 \text{ eV} < E_{\text{fast}} \quad (84)$$

where E_{fast} is the fast neutron energy group.

420 All prompt neutrons born through fission are assumed to be fast such that $\chi_p(E_{\text{fast}}) = 1$ and $\chi_p(E_{\text{therm}}) = 0$.

The conjugate gradient (CG) Krylov subspace method has been implemented in conjunction with fission source iteration to evaluate the fundamental eigenvalue (or k_{eff}) and corresponding multi-group scalar neutron fluxes. The resulting multi-group scalar neutron flux distribution is radially averaged and normalised such that the area under the energy and axially dependent multi-group scalar neutron flux distribution is one. Radial averaging and normalisation of the scalar neutron flux is required since the nuclear thermal hydraulics and gas creation equations are one-dimensional, meaning that the rate of fission energy deposition, described by Equation 45, must depend solely on the axial direction. Mathematically, the radially averaged and normalised scalar neutron flux may be calculated using the following:

$$\hat{\phi}(z, E) = \frac{\int_0^R r dr \phi(r, z, E)}{\int_0^H dz \int_0^R r dr \int_0^\infty dE \phi(r, z, E)} \quad (85)$$

where here, r represents the radial direction.

The steady-state neutron diffusion equation is solved at regular intervals of increasing wetting-induced reactivity. The solution to the steady-state neutron diffusion equation was recomputed 425 for every 0.5 \$ of wetting-induced reactivity insertion. Equation 85 defines the energy dependent, radially averaged, scalar neutron flux shape function, at position, z and with energy, E .

2.8. Spatial Discretisation, Time-dependent Algorithm and Software Workflow

The proposed model has been developed using modern Fortran [116]. The equations modelling thermal hydraulics, void generation and void migration were discretised using the finite volume

method in one-dimension. Using this methodology, the domain was separated into cylindrical slices of equal volume using a static Eulerian mesh. Crank [117] describes a methodology whereby interfaces between regions may be tracked. This enables a slice that contains an interface to be split into two smaller slices. Through the use of region interface conditions, the discretised governing equations may be solved for the two smaller slices as well as throughout the rest of the domain. The location of region interfaces varies as a function of time. The solution in time is found using an ODE algorithm and Fortran code developed by Shampine and Gordon [118].

The software workflow is shown in Figure 3. At each time step a data structure is populated with the most recent solution in time for the neutronics, voidage and phase temperature data. Each reactivity component may then be recalculated using this new data. The steady-state neutron diffusion equation is then solved, if the prescribed criterion is satisfied. Once this is completed, temporal gradients for the neutronics, voidage and phase temperatures are calculated on the discretised one-dimensional mesh. Using these gradients, the solution is then iterated in time using the ODE algorithm developed by Shampine and Gordon [118]. Each process step is repeated until the output time is reached.

3. Infiltration Model Comparison to Experiment

The infiltration ODE's defined by Equations 10, 15 and 16 may be solved in isolation to examine the accuracy of modelling infiltration into UO_2 powder using the Green and Ampt model. Data to perform this comparison is obtained from a paper by Rozain et al. [9] which describes an infiltration experiment carried out by the CEA. In this experiment, a jet of fine droplets was directed to impinge upon and wet the surface of a UO_2 powder contained within a cylindrical drum. Humidity sensors placed at equidistant points throughout the vessel were used to monitor the rate of infiltration into the powder bed. The experimental data required to investigate the accuracy of the infiltration model is documented in Table 2.

Data from the CEA experiment indicated that the powder density increased from 1000 kg m^{-3} to 2200 kg m^{-3} [9]. Since the proposed model does not consider the variable powder densities at this stage, an average density of 1600 kg m^{-3} has been used. Furthermore, the size distribution of the powder used in the CEA experiment was not reported. Therefore, to compare the results of the simulation to the experiment, the infiltration characteristics were obtained for several mean

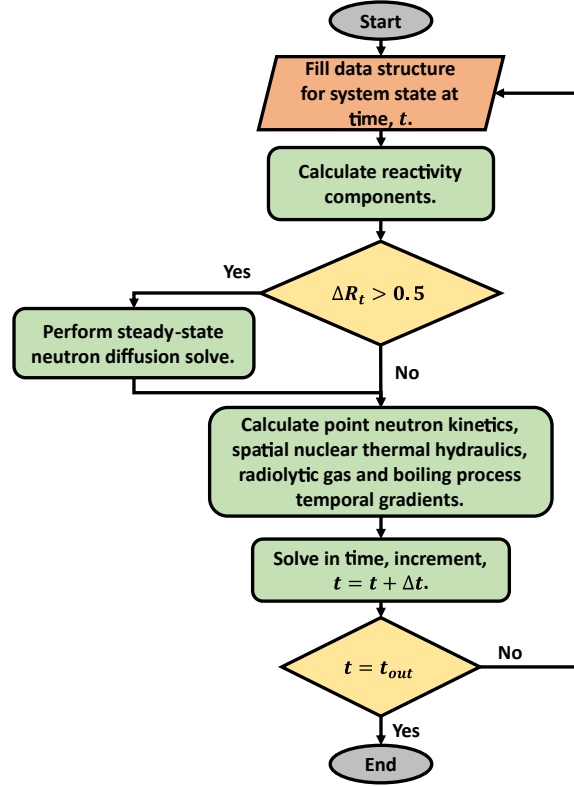


Figure 3: A flowchart diagram showing the workflow of the proposed numerical algorithm and model.

powder particle sizes. This was then compared to mean UO_2 powder particle sizes found during the manufacture of UO_2 powders.

Experimental results reported by Rozain et al. [9] indicated that water was detected at the lowest humidity sensors at a time of approximately 1850 s. The constant rate of penetration into the powder described by Rozain et al. [9] has been derived from experimental results as $1/3 \text{ mm s}^{-1}$. The geometry of the system at time $t = 0 \text{ s}$ in Figure 4 refers to the initial conditions specified in Table 2. The infiltration time into the powder bed may also be calculated using this constant infiltration rate and the estimated mean powder bulk density defined in Table 2. This value, calculated to be 1865 s, indicates that the mean powder bulk density chosen reflects the infiltration time estimated by Rozain et al. [9] to within approximately 1 %. The mean powder bulk density specified in the model (1600 kg m^{-3}) has therefore been considered appropriate.

The simulated size of the wetted powder region is shown as a function of time in Figure 4, for

Table 2: Infiltration simulation parameters.

Property	Symbol	Value
Drum height	H	1.0 m
Initial dry powder height	H_{dp}	0.6217 m
Initial wet powder height	H_{wp}	0 m
Initial water height	H_{pooled}	0 m
Powder mass	M_p	500 kg
Surface flow rate	$U_{w,in}$	$9.947 \times 10^{-4} \text{ m s}^{-1}$
Drum radius	R	0.4 m
UO ₂ powder bulk density	ρ_p	1600 kg m ⁻³
Solid UO ₂ powder density	$\rho_{p,th}$	10950 kg m ⁻³

several mean powder particle sizes. Also documented is the constant speed of penetration defined by Rozain et al. [9], determined from experimental data. The GA infiltration model found that for all mean powder particle sizes considered, the speed of penetration into the powder was slightly non-linear. As the mean powder particle size decreased, the permeability of the powder also decreased, leading to lower infiltration rates. The simulated speed of penetration satisfies experimental results when the mean powder particle size is between 9 μm and 10 μm . These mean powder particle sizes are similar to the range of UO₂ powder particle sizes observed in the cited literature [19, 78, 79].

4. Case Study Parameters

4.1. System Geometry & Bed Characteristics

The initial geometry of the drum was based on the data from the experimental/numerical investigation carried out by the CEA [9]. In this investigation, the total height of the system was 1.0 m and the cylindrical drum radius was 0.4 m. For a mean porosity of 0.8540, and fissile powder bed mass of 500 kg, the corresponding powder bed height was 0.6217 m. This porosity corresponds to a powder bulk density of 1600 kg m⁻³, which is within the range commonly observed for UO₂ powders produced from UF₆ using a conversion kiln [3, 12]. The remaining free space in the drum contained no water. Enrichment levels of the UO₂ powder used in this investigation are $\gamma = 5 \%$, reflecting the enrichment commonly seen in powders during shipping, fabrication and re-processing

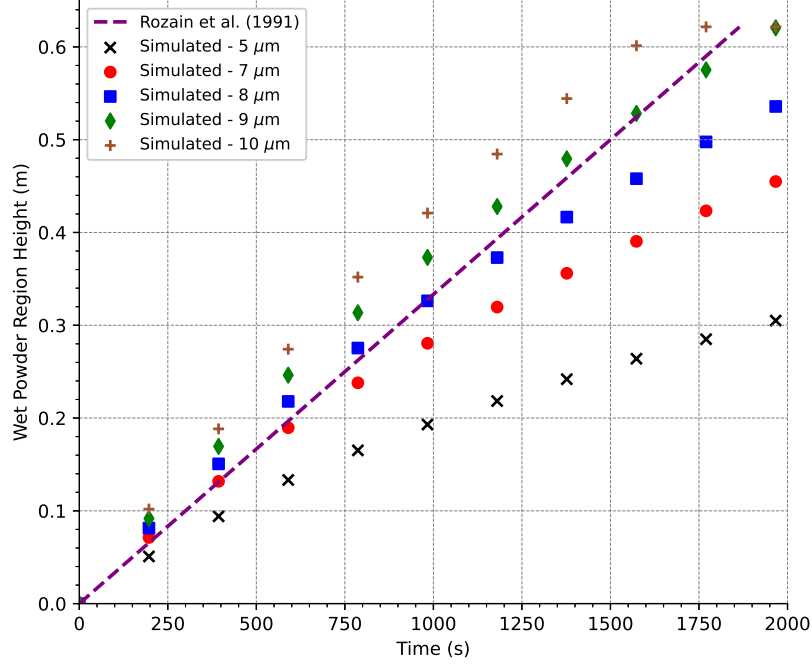


Figure 4: The propagation of the wetted powder region is shown for a number of different mean powder diameters. The dashed line represents the constant rate of infiltration that was assumed by Rozain et al. [9] having conducted an infiltration experiment on a UO_2 powder drum.

[12, 32]. This level of enrichment also reflects the UO_2 powder enrichment used in the sub-critical experiments by Rozain et al. [9]. At this level of enrichment, the theoretical density of UO_2 powder is different from that of natural UO_2 and is calculated to be $\rho_{p,\text{th}} = 10950 \text{ kg m}^{-3}$.

An activated sprinkler system, producing a uniform sheet of water, entered the domain through the top of the vessel at a speed of $U_{w,\text{in}} = 9.947 \times 10^{-4} \text{ m s}^{-1}$ ($= 0.5 \text{ L s}^{-1}$) and temperature of $T_{\text{in}} = 293.15 \text{ K}$. The speed of the water hitting the surface of the powder is assumed to be low enough such that the powder is not disturbed. If at any point in the transient, the height of the pooled water went above the maximum system height, the appropriate volume of water/gas was removed from the top of the domain.

4.2. Mean Powder Particle Sizes

Experimental work by Andreev et al. [78], and Clayton and Aronson [79], indicate that log-normal powder particle size distributions are commonly found for UO_2 powders manufactured using various techniques. This corresponds well with many other powder systems whose mean powder particle size are distributed log-normally [67]. The present model does not consider the poly-dispersion of powder particle sizes. To see the effect of the mean powder particle size on the nuclear criticality transient, a large and a small powder particle size of $100\ \mu\text{m}$ and $10\ \mu\text{m}$ were simulated.

4.3. Thermophysical Properties

4.3.1. UO_2 Powder

Excellent resources outlining the thermophysical properties of uranium dioxide can be found in Belle [119] and Haase et al. [120]. Further information regarding the theoretical density of UO_2 may be found within the cited literature [121]. Correlations for the specific heat capacity of UO_2 as a function of temperature were found by Fink et al. [122]. The variation of the specific heat capacity as a function of temperature for 5 % enriched uranium is shown in Figure 5. A constant value for the specific heat capacity was calculated by averaging the value over the expected temperature range of the UO_2 powder. The expected range of the UO_2 powder temperature was predicted to be between 293.15 K to 610 K, yielding an average UO_2 specific heat capacity of $c_p = 274\ \text{J kg}^{-1}\text{K}^{-1}$. Analysis shown in Figure 23b indicate that local UO_2 powder temperatures do not exceed 610 K.

The change in UO_2 powder density as a function of temperature is accounted for by the thermal feedback equations, but is neglected in the multiphysics part of the model. Figure 5 shows that by using a correlation defined by Ronchi et al. [123], the density does not change significantly over the expected temperature range.

The thermal conductivity of solid UO_2 powder (κ_p) may be modelled as a function of temperature using a correlation presented in Fink et al. [122]. Figure 5 highlights that over a temperature range of approximately 600 K, the thermal conductivity doubles. The correlation defined by Fink et al. [122] incorporated into the proposed model may be seen in Equation 86.

$$\kappa_p = \left[0.068337 + 1.6693 \times 10^{-4} T_{p/wp} + 3.1886 \times 10^{-8} T_{p/wp}^2 \right]^{-1} + 0.12783 T_{p/wp} e^{-1.1608/kT_{p/wp}} \quad (86)$$

where $T_{p/wp}$ is either the powder or wet powder region temperature depending on the region of interest, and $k = 8.6144 \times 10^{-5} \text{ eV K}^{-1}$ is the Boltzmann constant.

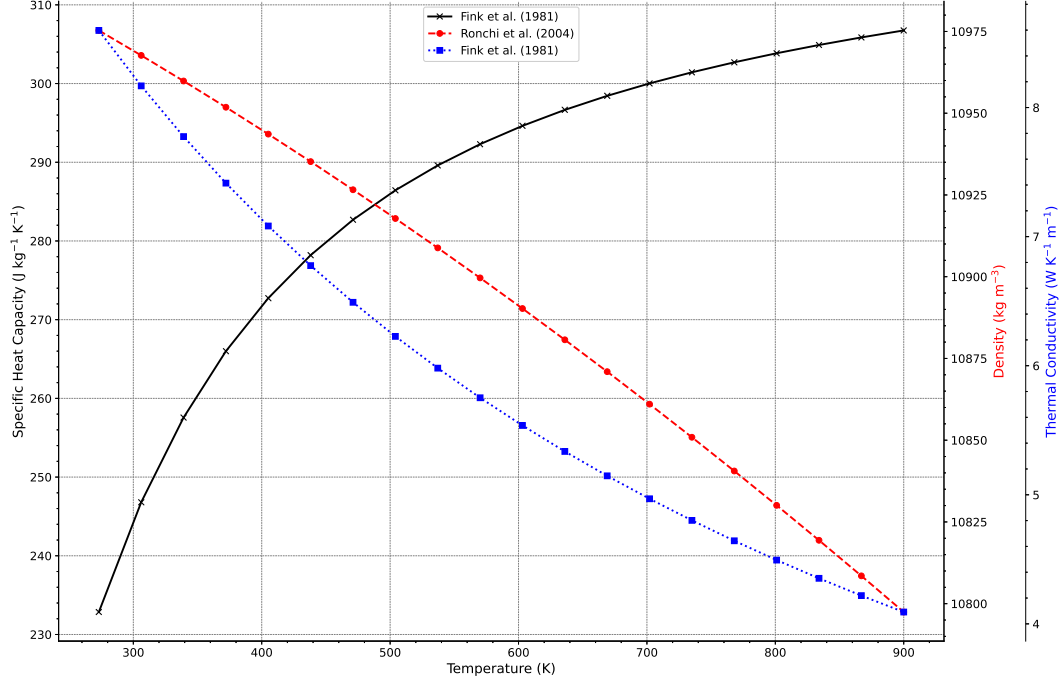


Figure 5: The thermophysical data for UO_2 powder at 5-wt% enrichment for the range of expected powder temperatures.

A summary of the thermophysical data for the UO_2 powder particles can be seen in Table 3.

Table 3: Thermophysical properties of UO_2 powder.

Property	Symbol	Value
Specific heat capacity	c_p	$274 \text{ J kg}^{-1} \text{ K}^{-1}$
Thermal conductivity	κ_p	see Equation 86
Bulk density	ρ_p	1600 kg m^{-3}
Theoretical density	$\rho_{p,th}$	10950 kg m^{-3}

4.3.2. Water

525 The specific heat capacity of water does not depend strongly on temperature, therefore a value of $4.2 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$ was used for the case study. By applying the Boussinesq approximation to the free convection equation, the effect of water temperature was modelled using the water thermal expansion coefficient. The density of water is specified at the initial system temperature of 293.15 K. The dynamic viscosity was determined using the correlation described by Pátek et al.
530 [124]. A summary of the thermophysical properties of water is shown in Table 4.

Table 4: Thermophysical properties of water.

Property	Symbol	Value
Specific heat capacity	c_w	$4.2 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$
Boiling point	T_{sat}	373.15 K
Thermal expansion coefficient	η	$2.14 \times 10^{-4} \text{ K}^{-1}$
Dynamic viscosity	μ_w	see [124]
Density	ρ_w	998.1 kg m^{-3}

4.3.3. Steam

The steam temperature was treated to be in equilibrium with the surrounding liquid. Therefore the thermophysical properties may be interpreted from steam tables contained in Faghri and Zhang [82] at a temperature of approximately 373 K. Table 5 outlines the constant thermophysical
535 properties of steam used in this case study.

Table 5: Thermophysical properties of water vapour at boiling.

Property	Symbol	Value
Specific heat capacity	c_v	$2.060 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$
Latent heat of vaporisation	$h_{l,v}$	$2.2512 \times 10^6 \text{ J kg}^{-1}$
Thermal conductivity	κ_v	$2.46 \times 10^{-2} \text{ W m}^{-1} \text{ K}^{-1}$
Dynamic viscosity	μ_v	$1.271 \times 10^{-5} \text{ Pa s}$
Density	ρ_v	$5.863 \times 10^{-1} \text{ kg m}^{-3}$

4.4. Turbulent Mixing Coefficient

Turbulence occurs when chaotic changes in flow velocity occur around a mean value [125]. This process manifests itself through the generation of turbulent eddies throughout the liquid medium. The varying shape of turbulent eddies may be understood to form via an energy cascade [126]. The phenomena of an energy cascade refers to the energy transfer between eddies as they go from the largest possible size, to the smallest. The varying eddy length and velocity scales observed during this energy cascade may be characterised by a mean length and velocity scale [127].

Full turbulence models are complex, computationally demanding and beyond the scope of the proposed model. Cooling et al. [89] models turbulent diffusion in fissile solutions as a Fickian diffusive process driven by the turbulence caused by gas bubble advection. The following equation described the turbulent mixing coefficient used in the proposed model based on [89]:

$$D = \frac{v_{\text{eddy}} r_{\text{eddy}}}{\pi} \quad (87)$$

where v_{eddy} [m s^{-1}] and r_{eddy} [m] are the velocity and radius of the largest eddies that are predicted to be in the system.

Turbulent mixing is expected to occur throughout all regions where advecting gas bubbles are present. The size and velocity of turbulent eddies are expected to vary depending on whether they are present in the wet powder region of pooled water. The proposed model estimates the turbulent mixing coefficient using a constant value for each region, where $D_{\text{t,wp}} = 0.0001 \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{t,pooled}} = 0.001 \text{ m}^2 \text{ s}^{-1}$ respectively. Sensitivity analysis has been performed on the values to examine their importance in determining the peak power, energy released and time to boiling for the nuclear criticality transient.

4.5. Radiolytic Gas Generation Coefficient

Experiments to evaluate $G(X)$ for uranium-thorium slurry systems due to fission recoil particles have been carried out by Steele et al. [96]. The fraction of the total fission energy deposited in the surrounding water was found to depend on particle size and concentration. The number of hydrogen molecules produced through the break-up of water per 100 eV of fission energy was found to be 2.1 ± 0.2 molecules per 100 eV, near the maximum value of 2.35 molecules per 100 eV expected for water. This is contrary to the work in Lane et al. [95] which states that the maximum value of $G(\text{H}_2)$ for pure water is 1.8 molecules per 100 eV. In the proposed model the more conservative value that minimises radiolytic gas production has been chosen, $G(\text{H}_2) = 1.8$ molecules per

100 eV. The proposed model considers constant vapour properties of 1 atm and 373.15 K, leading to a volumetric generation coefficient of $G_V(\text{H}_2) = 5.37 \times 10^{-9} \text{ m}^3 \text{ J}^{-1}$. For the range of radiolytic gas temperatures expected (273.15 K to 373.15 K), $G_V(\text{H}_2)$ varies from $4.2 \times 10^{-9} \text{ m}^3 \text{ J}^{-1}$ to $5.37 \times 10^{-9} \text{ m}^3 \text{ J}^{-1}$.

565 4.6. Range of UO_2 Fission Fragments

The average range of fission fragments in UO_2 varies depending on the mass of the fission fragment considered and the initial kinetic energy of fission fragments. Experimental data produced by Eichenberg et al. [128] indicates that there is a linear relationship between the atomic mass of the fission fragment and its range in UO_2 . The fission fragment range varied from approximately 8 μm for lighter fragments, down to approximately 5 μm for heavier fragments. Based on this data, 570 the mean fission fragment range for the case study was set as 7 μm .

4.7. Delayed Neutron Data and Generation Time

The fraction of neutrons born via delayed neutron precursors and the decay constant of delayed neutron precursors were determined through a series of neutron transport simulations using the 575 Monte Carlo code MCNP [129], ranging in bed immersion fraction from 0 to 1. The results were then averaged and the errors calculated through the use of the central limit theorem. The results for each of the six delayed neutron precursor groups are shown in Table 6. The fraction of neutrons born via delayed neutron precursors and decay constant of delayed neutron precursors did not vary widely for the range of bed immersion fractions considered. The mean prompt neutron generation 580 time was found to be $\Lambda = (5.32959 \pm 0.0094) \times 10^{-5} \text{ s}$. The total fraction of neutrons born via delayed neutron precursors was found to be $\beta_{\text{eff}} = (733 \times \pm 8) \times 10^{-5}$.

4.8. Wetting-Induced Reactivity Insertion

The ingress of water into the interstices between the UO_2 powder particles has been modelled in dollars (\$) as a reactivity insertion, denoted R_{imm} . MCNP was used to determine the effective multiplication factor (k_{eff}) at a range of bed immersion fractions. The reactivity was calculated using:

$$R_{\text{imm}} = \frac{k - 1}{k\beta_{\text{eff}}} \quad (88)$$

Table 6: Delayed neutron precursor group data.

Group, i	$\beta_i (\times 10^{-5})$	$\lambda_i (\text{s}^{-1})$
1	25 ± 1	0.01335 ± 0.00000
2	129 ± 4	0.03264 ± 0.00000
3	122 ± 3	0.12099 ± 0.00000
4	279 ± 5	0.30482 ± 0.00001
5	125 ± 4	0.85704 ± 0.00003
6	53 ± 2	2.87888 ± 0.00015

The polynomial best-fit line described by Equation 89 and shown in Figure 6 has been used to describe the reactivity insertion due to wetting between known values found using MCNP.

$$R_{\text{imm}}(f_{\text{imm}}) = \sum_{i=0}^n C_i f_{\text{imm}}^i \quad (89)$$

where $f_{\text{imm}} = H_{\text{wp}}/H_{\text{dp+wp}}$ is the bed immersion fraction, $n = 4$ is the polynomial order, the coefficients of the polynomial C_i are defined in Table 7.

Table 7: Bed immersion feedback coefficients.

i	0	1	2	3	4
C_i	-90.8692	531.7354	-973.8299	832.3316	-270.8543

585 At low bed immersion fractions the system is deeply sub-critical. The case study simulation began from a point near criticality. Table 8 shows the starting height of each region used for the simulations. This system configuration corresponds to a bed saturation of $f_{\text{imm}} = 0.29$ and initial reactivity of $-0.179 \$$.

Table 8: Initial region heights, neutron density and delayed neutron precursor concentrations.

Property	Symbol	Value
Initial dry powder height	H_{dp}	0.4414 m
Initial water height	H_{pooled}	0.3783 m
Initial wet powder height	H_{wp}	0.1803 m

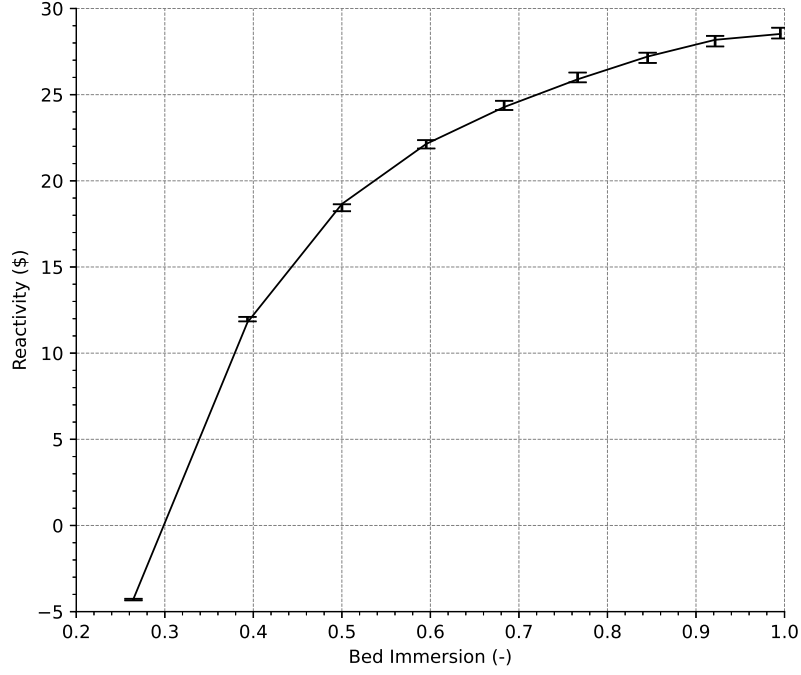


Figure 6: The reactivity inserted due to the infiltration of water into the dry powder calculated using $\beta_{\text{eff}} = (733 \pm 8) \times 10^{-5}$. The line represents the correlation specified by Equation 89.

4.9. Thermal Reactivity Feedback

590 Powder and water thermal expansion, and the associated density changes of the powder bed, occurring as a result of increasing system temperatures, leads to a decreased probability of interactions between neutrons and target nuclei. This usually reduces the reactivity of the system. Increasing temperatures also leads to Doppler broadening, which broadens microscopic nuclear cross-sections, increasing the probability of interaction between neutrons and ^{238}U nuclei. For UO_2 powder systems moderated by water, this decreases the system reactivity. A thermal reactivity feedback term
595 incorporated into the point kinetics equation (see Equation 1) has been used to model these effects. The proposed model has separated thermal reactivity feedbacks by phase and region. The change in reactivity due to temperature variations, denoted $\Delta R_{\text{th}, X}(T_X, f_{\text{imm}})$, is a function of both temperature and the bed immersion fraction. Here, the subscript X refers to the region/phase of interest

600 and may refer to the dry powder, wet powder or pore water.

The effects of Doppler broadening and thermal expansion on the system reactivity were determined using the Monte Carlo code MCNP. For each bed immersion fraction considered, the effective multiplication factor (k_{eff}) was calculated for all expected region/phase specific temperatures. Room temperature (293.15 K) was the lower temperature limit for the MCNP simulations. Preliminary analysis indicated that the powder temperature would not rise above 800 K, therefore this was set as the upper limit. The upper temperature limit for the water was set just above the boiling point (383.15 K), given atmospheric conditions. These values represent the range of validity for the reactivity feedback correlations created using the results of MCNP simulation data. The ENDF/B-VII nuclear data library was used to obtain temperature adjusted nuclear cross-sections for MCNP simulations. The method of stochastic mixing described in Donnelly [130] was used to evaluate thermal feedback effects at temperatures not explicitly tabulated in the ENDF/B-VII nuclear data library.

The results of the MCNP simulations are shown in Figures 7, 8 and 9. A line of best-fit that is a function of both temperature and bed immersion has also been shown.

For water in the pore space between fissile powder particles, the reactivity was found to be a linear function of temperature and a second order polynomial function of bed immersion. In the wet powder, the thermal reactivity feedback was found to be a linear function of both temperature and bed immersion. Doppler broadening in the dry powder region decreased the reactivity. Thermal expansion of the dry powder due to increased temperatures caused a fraction of the dry powder to penetrate the wetting front and become surrounded by water. Although the wetting front location did not change, the greater mass of fissile powder in the wet powder region due to this expansion introduced a positive reactivity feedback. The effect of this can be seen in Figure 9 which shows that the reactivity due to a combination of Doppler broadening and thermal expansion resulted in a net positive reactivity feedback. The following equation defines the general thermal reactivity feedback for a given phase/region:

$$\Delta R_{\text{th, X}}(T_X, f_{\text{imm}}) = R_{\text{th, X}}(T_X, f_{\text{imm}}) - R_{\text{th, X}}(T_0, f_{\text{imm}}) \quad (90)$$

where T_0 is the reference temperature at the start of the simulation. The correlations are of the form:

$$R_{\text{th, X}}(T_X, f_{\text{imm}}) = (C_0 f_{\text{imm}}^2 + C_1 f_{\text{imm}} + C_2) T_X(t) \quad (91)$$

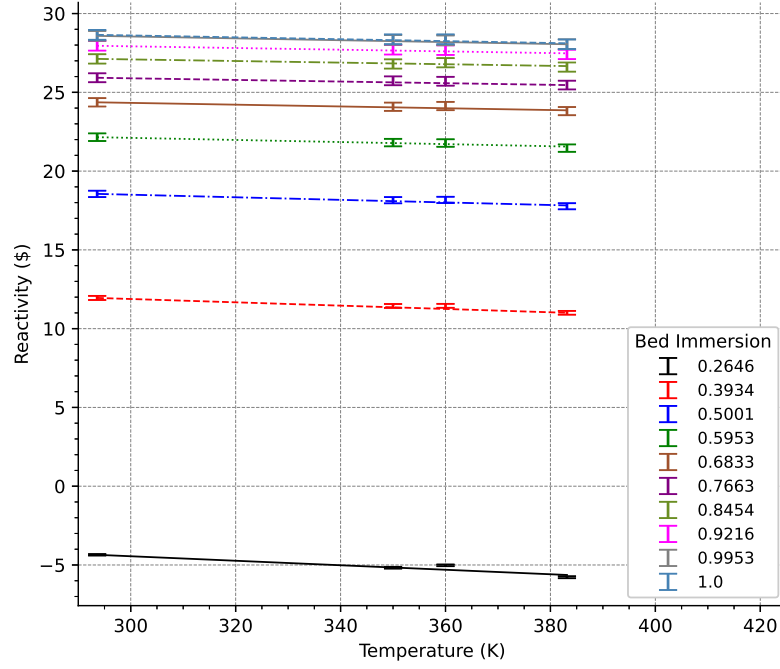


Figure 7: The points represent the reactivity calculated using MCNP due to variations in temperature from Doppler broadening and expansion of pore water. The lines represents the correlation described by Equation 91.

where $T_X(t)$ is the region and phase averaged temperature. Substituting Equation 91 into Equation 90, the following may be found:

$$\Delta R_{th, X}(T_X, f_{imm}) = (C_0 + C_1 f_{imm} + C_2 f_{imm}^2) (T_X(t) - T_{0,X}) \quad (92)$$

615 Table 9 shows the coefficients for each of the regions and phases used to describe the thermal reactivity feedback correlation, along with the R-squared value.

4.10. Void Reactivity Feedback

Void reactivity feedback has been separated into pore water and pooled water components. This is because voids in the interstices between fissile powder particles are expected to have a
620 much larger effect on system reactivity than voids located in the pooled water, which does not contain fissile material. Voids present in the system provide pathways for neutrons to travel with a

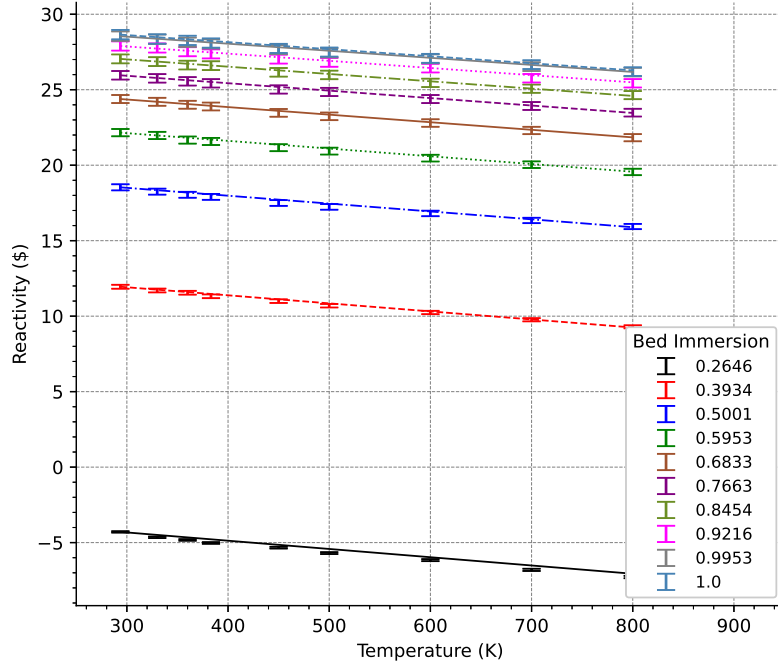


Figure 8: MCNP data points are shown alongside the correlation used for temperature variations in the wet powder.

Table 9: Thermal feedback coefficients and R-squared for correlation.

Region	$C_0 (\times 10^{-2})$	$C_1 (\times 10^{-2})$	$C_2 (\times 10^{-2})$	R^2
dry powder	0.4219	-0.8590	0.4700	0.99994
wet powder	-0.5759	0.1086	0.0000	0.99986
pore water	-2.5051	4.8433	-2.9357	0.99992

very low probability of collision, increasing neutron leakage and usually decreasing the macroscopic absorption neutron cross-section.

Using MCNP, a series of simulations were carried out to investigate the reactivity of the system at various void fractions and bed immersion fractions. Voids in MCNP were modelled by decreasing the density of water in the appropriate region, any excess volume of water was moved upwards. The maximum system height was 1 m, meaning that in some cases water left the system.

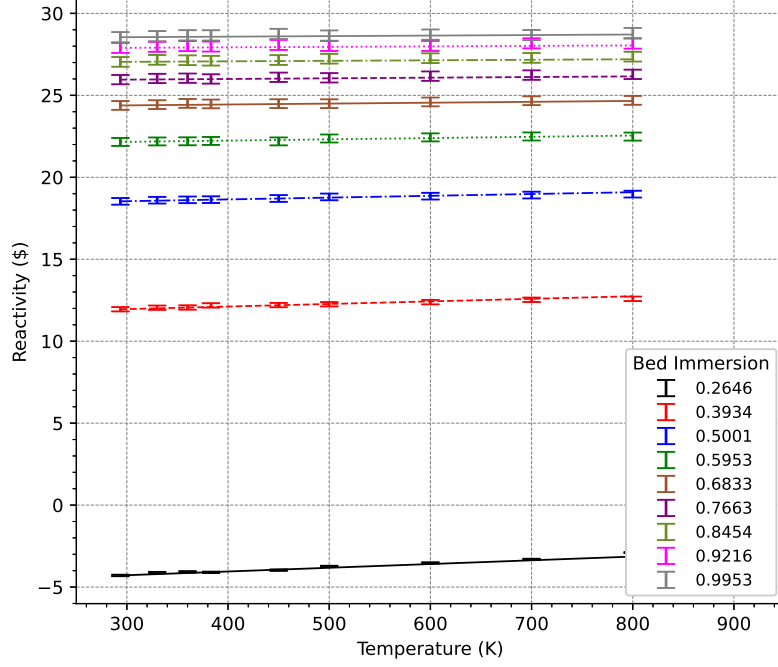


Figure 9: MCNP data points are shown alongside the correlation used for temperature variations in the dry powder.

Figure 10 highlights that within the wet powder region there is a non-linear relationship between the reactivity, voidage and bed immersion fraction. This differs from data for the pooled water region, shown in Figure 11, which shows a linear relationship between reactivity and voidage, and a non-linear relationship between reactivity and bed immersion fraction. These graphs confirm that voids present in the pores have a greater effect on the reactivity of the system than voids present in the pooled water.

The reactivity feedback due to the presence of voids is:

$$\Delta R_{\text{void, X}}(v_{\text{f,g,X}}, f_{\text{imm}}) = R_{\text{void, X}}(v_{\text{f,g,X}}, f_{\text{imm}}) - R_{\text{void, X}}(v_{0,\text{f,g,X}}, f_{\text{imm}}) \quad (93)$$

where $v_{0,\text{f,g,X}}$ is the reference or initial voidage in the region being considered.

The correlation for the reactivity as a function of bed immersion fraction and voidage within the pores takes the following form:

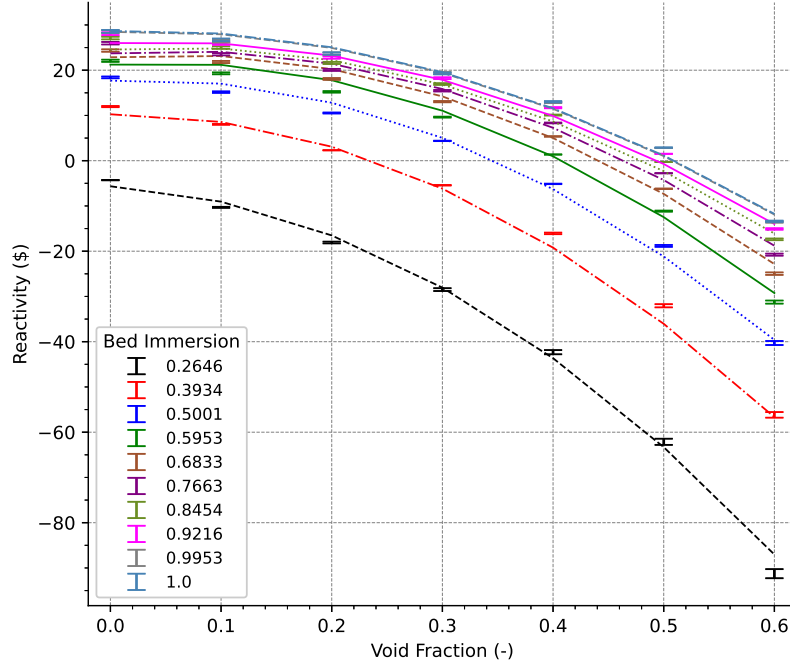


Figure 10: The reactivity due to the presence of voids located in the pores around the wet powder. The lines represent the correlation described by Equation 94.

$$\begin{aligned}
 R_{\text{void,wp}}(v_{\text{f,g,wp}}, f_{\text{imm}}) = & C_0 + C_1 f_{\text{imm}} + C_2 v_{\text{f,g,wp}} \\
 & + C_3 f_{\text{imm}}^2 + C_4 v_{\text{f,g,wp}} f_{\text{imm}} + C_5 v_{\text{f,g,wp}}^2 + C_6 f_{\text{imm}}^3 \\
 & + C_7 f_{\text{imm}}^2 v_{\text{f,g,wp}} + C_8 f_{\text{imm}} v_{\text{f,g,wp}}^2 \quad (94)
 \end{aligned}$$

The coefficients for Equation 94, which are described in Table 10, give a close fit to MCNP simulated data ($R^2 = 0.99556$).

Reactivity as a result of voids present in the pooled water region has been defined by the following correlation:

$$R_{\text{void,pooled}}(v_{\text{f,g,pooled}}, f_{\text{imm}}) = (C_0 f_{\text{imm}}^2 + C_1 f_{\text{imm}} + C_2) v_{\text{f,g,pooled}} \quad (95)$$

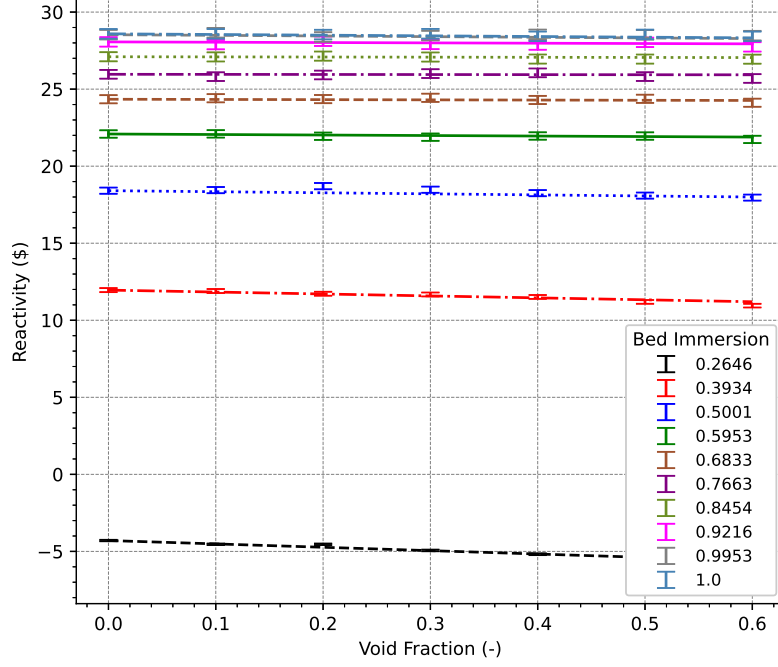


Figure 11: The reactivity due to the presence of voids within the pooled water. The lines represent the correlation described by Equation 95.

Table 10: Void feedback coefficients for voids present within pores.

i	0	1	2	3	4	5	6	7	8
C_i	-72.72	362.1	-62.1	-464.4	224.1	-232.0	203.7	-155.4	108.7

The coefficients for Equation 95, which are described in Table 11, give a close fit to MCNP simulated data ($R^2 = 0.99972$).

Table 11: Void feedback coefficients for voids present within pooled water.

i	0	1	2
C_i	-4.929841	12.476281	-7.983281

4.11. Axial Neutron Distribution

A three region neutron diffusion code has been used to evaluate the shape function describing the distribution of neutrons in the system. The regions in the diffusion code are a dry UO_2 powder region, a homogeneous UO_2 powder and water region, and a pooled water region.

4.11.1. Energy Condensed and Resonance Self-Shielded Macroscopic Neutron Cross-Section and Associated Correlations as a Function of Immersion Fraction

Group condensed and resonance self-shielded macroscopic cross-sections were determined using the Monte Carlo neutron transport code Serpent [131] for the two-group macroscopic neutron cross-sections required by the neutron diffusion code. The bed immersion fraction was varied to investigate the functional dependence of the macroscopic cross-sections on the height of the wet powder region.

A homogenised three region approach was used as an approximate geometry for the Monte Carlo neutron transport simulations. This neglects the effect of double heterogeneity spatial self-shielding which causes nuclear cross-sections to vary radially through an individual powder particle as well as in space overall [132]. Research on ^{239}Pu systems by Shmakov et al. [133] showed that typically the homogenisation of dispersed particulates in a continuous moderator produces appropriate values for nuclear cross-sections when the mean free path of thermal neutrons is greater than the size of the particulates [134]. Similar conclusions were made by Yamamoto et al. [135] investigating Mixed OXide (MOX) fuel pellets. In this case, double heterogeneity spatial self-shielding effects do not significantly affect the nuclear cross-sections. Furthermore, increasing the number density of UO_2 powder particles in the continuous water phase decreases the impact of the double heterogeneity on the macroscopic nuclear cross-sections [136]. However, there is still spatial self-shielding effects associated with the natural spatial variation of the energy-dependent scalar neutron flux within the homogeneous material regions. Using MCNP, Laramore et al. [137] showed that a homogeneous approach was appropriate for the range of UO_2 powder sizes used in this paper. This was further supported by the Working Group on Nuclear Criticality Safety Data at JAERI [3] who showed that the change in the infinite multiplication factor for particles of size $d_p = 200 \mu\text{m}$ was less than 0.3 % for 5 % enriched UO_2 powder at a bulk density of 2.0 g cm^{-3} . Using a homogenised region approximation significantly reduces the computing complexity of the Monte Carlo simulations of the solution domain. If one was to use Monte Carlo methods to model the problem as a heterogeneous

dispersed particulate multiphase material [138], where all the individual UO_2 particles or grains are modelled, would be computational very challenging [139].

675 Another factor to consider is the agglomeration, or clumping, of dispersed particulates which would lead to particles that are effectively larger than their constituents. Large particles may be formed by a process known as soft agglomeration, where smaller primary particles are loosely bound by Van der Waals forces [140, 141]. There may be a case where the size of the agglomerate becomes large enough that the double heterogeneity spatial self-shielding effects become significant. In
680 this case, the heterogeneity of the system must be taken into account. The present model does not consider the agglomeration of powder particles. Statistical methods for modelling the agglomeration of the powder particles in a continuous medium may be found within the cited literature [136, 142].

Figure 12 shows the variation of the macroscopic neutron cross-sections as a function of bed immersion. Thermal group fission and absorption neutron cross-sections in the dry powder region
685 vary non-linearly as a function bed immersion. All other group and region cross-sections are considered to be independent of the bed immersion fraction. Figure 13 represents the two-group neutron scattering cross-sections for the dry powder, wet powder and pooled water regions respectively.

Near saturation, large standard deviations were observed for the macroscopic neutron scattering cross-sections in the dry powder region. At this point, the dry powder region is of the size where
690 these standard deviations are not expected to affect the neutron distribution.

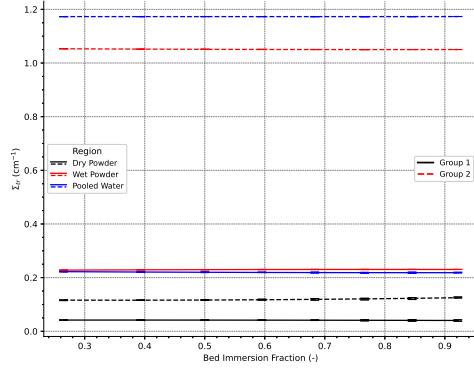
The correlation, as a function of immersion fraction, defining the thermal fission and absorption macroscopic neutron cross-sections within the dry powder region are of the following form:

$$\Sigma_X = C_0 + C_1 f_{\text{imm}} + C_2 f_{\text{imm}}^2 \quad (96)$$

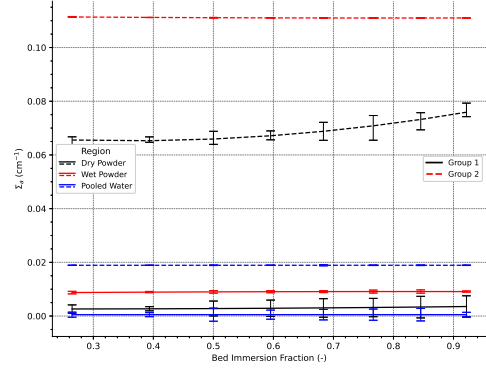
where Σ_X may be the macroscopic absorption cross-section (Σ_a), the macroscopic fission cross-section (Σ_f) or the average number of neutrons born through fission multiplied by the macroscopic fission cross-section ($\bar{\nu}\Sigma_f$). Table 12 highlights the coefficients used in the correlation.

Table 12: Dry powder region thermal fission and absorption macroscopic neutron cross-section correlation coefficients.

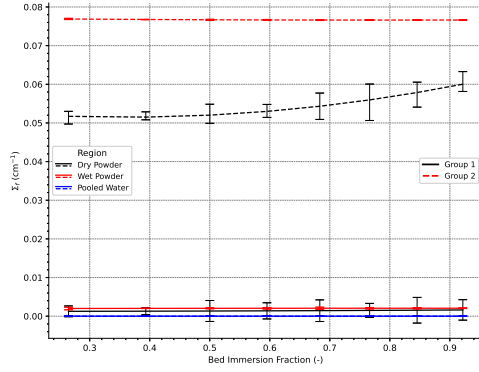
Cross-Section (cm^{-1})	C_0	C_1	C_2
$\bar{\nu}\Sigma_f$	0.14448371	-0.09262251	0.10812776
Σ_a	0.07504803	-0.04757275	0.0554874
Σ_f	0.05929682	-0.03801429	0.0443752



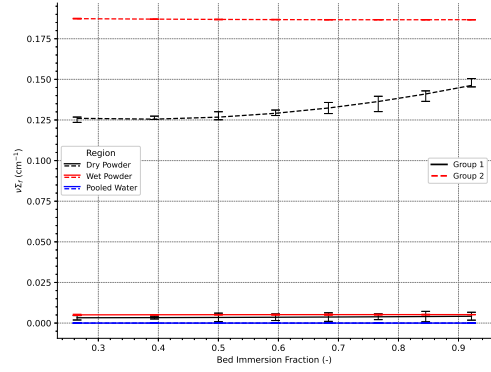
(a) Σ_{tr} .



(b) Σ_a .

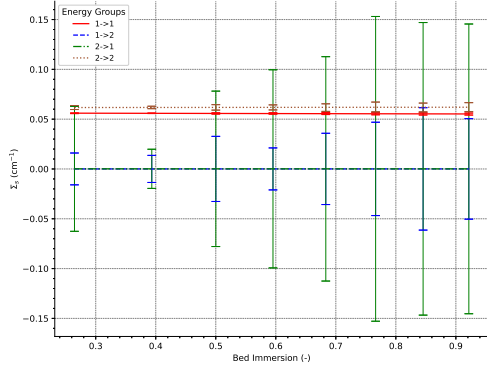


(c) Σ_f .

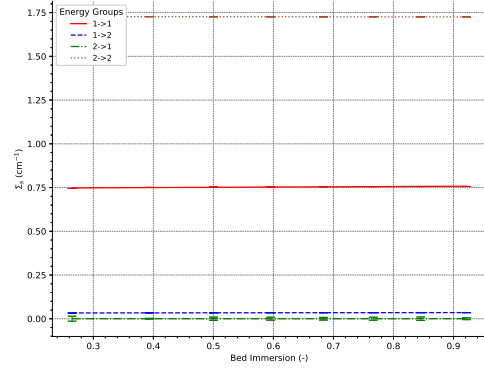


(d) $\bar{\nu}\Sigma_f$.

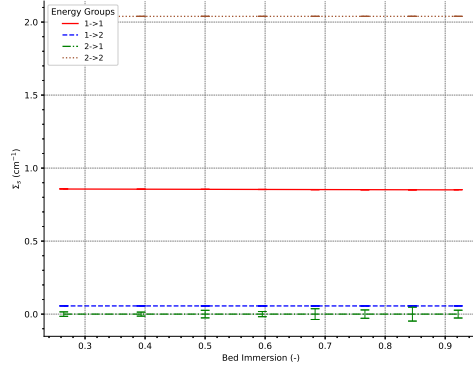
Figure 12: The fast and thermal macroscopic neutron cross-sections for each region is shown. The points represent data calculated using Serpent, the lines represent the values/correlations used in the proposed model.



(a) Dry powder.



(b) Wet powder.



(c) Pooled water.

Figure 13: The macroscopic neutron scattering cross-sections for each region. The lines represent the values defined to model these cross-sections in the neutron diffusion code.

The remaining data for the macroscopic neutron cross-sections have been considered independent of bed immersion, and are shown in Table 13.

Table 13: Region specific two-group macroscopic neutron cross-sections. Subscripts 1 and 2 correspond to fast and thermal groups respectively.

Region g	Dry Powder		Wet Powder		Pore Water	
	1	2	1	2	1	2
$\bar{\nu}\Sigma_{f,g}$	0.00343	see eqn 96	0.00490	0.18813	0.00000	0.00000
$\Sigma_{a,g}$	0.00363	see eqn 96	0.00835	0.11183	0.00049	0.01890
$\Sigma_{f,g}$	0.00132	see eqn 96	0.00193	0.07717	0.00000	0.00000
$\Sigma_{s,1 \rightarrow g}$	0.05598	0.00003	0.74489	0.03248	0.85863	0.05621
$\Sigma_{s,2 \rightarrow g}$	0.00018	0.06079	0.00032	1.72766	0.00007	2.03920
$\Sigma_{tr,g}$	0.04073	0.12236	0.22794	1.05325	0.22295	1.17310

All neutrons born through fission are assumed to be fast neutrons, i.e. $\chi_p(E_{fast}) = 1$ and $\chi_p(E_{therm}) = 0$.

4.11.2. Correlation Example Results

The macroscopic neutron cross-section correlations may now be used within the two-group, three region neutron diffusion code to evaluate the scalar neutron flux distribution and the effective multiplication factor (k_{eff}). To verify the neutron diffusion code and correlations, the results have been compared to values calculated using Serpent. In the case of the effective multiplication factor (k_{eff}) results from MCNP were also used. Figure 14 highlights how the effective multiplication factor (k_{eff}) varies as a function of bed immersion fraction. As expected, MCNP and Serpent predict similar effective multiplication factors. The neutron diffusion approach under-predicts the effective multiplication factor. Differences between the Monte Carlo neutron transport codes and the diffusion code are expected for several reasons. The diffusion approximation is not well suited to evaluate the scalar neutron flux at interfaces well. Given that there are three interfaces present within the model, this may be the main source of error. Furthermore, the diffusion model implemented reduces the continuous energy group structure in the Monte Carlo code to a two-group approximation. Using a coarsely discretised energy group structure will inherently introduce some

error when compared to the continuous energy approach implemented by Serpent and MCNP.

Discrepancies in the effective multiplication factor, k_{eff} , between the neutron diffusion code and Monte Carlo codes will not affect the nuclear criticality transient as this data is not directly used by the proposed model. Instead, the reactivity components described in the previous sections by the derived correlations are used to determine the total reactivity of the system at a given time, t . This approach eliminates computational complexity by having simple polynomial approximations of the reactivity rather than having to solve the time-dependent neutron diffusion equation at every time step. Solving the neutron diffusion equation solely for the axial scalar neutron flux distribution enables the use of a quasi-static approach for the neutron diffusion equation, which has much less computational requirements than the time-dependent neutron diffusion equation. Furthermore, employing the time-dependent neutron diffusion equation to model the system's reactivity would require correlations for the macroscopic nuclear cross-sections as functions of both voidage and temperature. This would produce a single reactivity value, making it hard to evaluate individual effects of each reactivity component, whereas the point kinetics approach can directly model the effects of each reactivity component on the fission power of the system.

The normalised scalar neutron flux distribution calculated using Serpent and the neutron diffusion model are shown in Figure 15. The peak of the flux distribution can always be seen in the wet powder region. As the bed becomes more saturated, the peak moves down the drum and spreads out. The dry powder region appears to show a non-linear increase in neutron flux from the base to the interface with the wet powder region. A secondary bump can be seen in the pooled water region due to the effect of thermal neutrons in the neutron diffusion model. Good agreement between the scalar neutron flux profiles for the two models indicate that the correlations used for the macroscopic neutron cross-sections are appropriate.

For the case studies considered, the neutron flux distribution was re-calculated every 0.5 \$ of wetting-induced reactivity insertion.

5. Model Summary

5.1. Summary of Equations

Below are a summary of the ODE's and PDE's solved in the proposed model using a finite volume method (FVM) along with the time discretisation algorithm described by Shampine and Gordon [118].

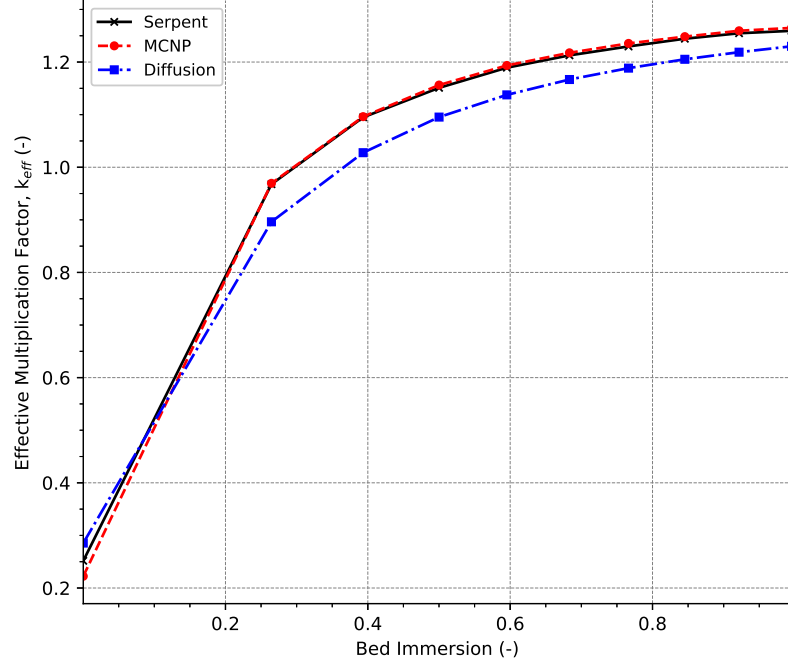


Figure 14: The effective multiplication factor (k_{eff}) as a function of bed immersion calculated with the three codes used throughout this study. The two transport codes produce very similar results, whereas the neutron diffusion code differs.

5.1.1. Point Kinetic Equations

$$\begin{aligned} \frac{dn(t)}{dt} = \frac{\beta_{\text{eff}} n(t)}{\Lambda} [R_{\text{imm}}(f_{\text{imm}}) + \Delta R_{\text{th,p,dp}}(T_{\text{p,dp}}, f_{\text{imm}}) + \Delta R_{\text{th,p,wp}}(T_{\text{p,wp}}, f_{\text{imm}}) \\ + \Delta R_{\text{th,w,wp}}(T_{\text{w,wp}}, f_{\text{imm}}) + \Delta R_{\text{void,wp}}(v_{\text{f,g,wp}}, f_{\text{imm}}) + \Delta R_{\text{void,wp}}(v_{\text{f,g,wp}}, f_{\text{imm}}) - 1] \\ + \sum_{i=1}^6 \lambda_i C_i(t) + s_n \end{aligned} \quad (97)$$

$$\frac{dC_i}{dt} = -\lambda_i C_i + \frac{\beta_i}{\Lambda} n(t) \quad (98)$$

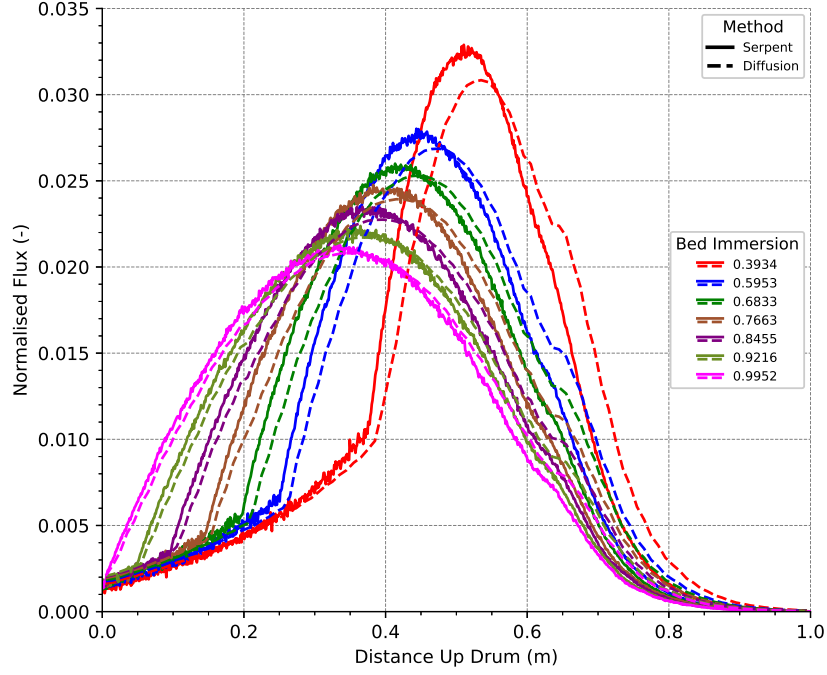


Figure 15: The normalised flux distribution computed at various bed immersion fractions using the Serpent Monte Carlo code and the developed deterministic neutron diffusion code.

5.1.2. Neutron Diffusion Equation (NDE)

The two-group neutron diffusion equation is outlined as follows:

$$-\nabla \cdot D_1(\vec{r})\nabla\phi_1(\vec{r}) + \Sigma_{t,1}(\vec{r})\phi_1(\vec{r}) = \Sigma_{s,1\rightarrow 1}(\vec{r})\phi_1(\vec{r}) + \Sigma_{s,2\rightarrow 1}(\vec{r})\phi_2(\vec{r}) + \frac{\chi_{p,1}(\vec{r})}{k_{\text{eff}}} [\bar{\nu}\Sigma_{f,1}(\vec{r})\phi_1(\vec{r}) + \bar{\nu}\Sigma_{f,2}(\vec{r})\phi_2(\vec{r})] \quad (99)$$

$$-\nabla \cdot D_2(\vec{r})\nabla\phi_2(\vec{r}) + \Sigma_{t,2}(\vec{r})\phi_2(\vec{r}) = \Sigma_{s,1\rightarrow 2}(\vec{r})\phi_1(\vec{r}) + \Sigma_{s,2\rightarrow 2}(\vec{r})\phi_2(\vec{r}) \quad (100)$$

Note that the prompt fission spectrum probability for group 1, $\chi_{p,1} = 1$ and prompt fission spectrum probability for group 2, $\chi_{p,2} = 0$.

745

5.1.3. Infiltration Equations

$$\frac{dH_{\text{wp}}}{dt} = \frac{K_{\text{sat,e}}(1 - \bar{v}_{\text{f,g}}(t))}{\epsilon \Delta S_{\text{w}}} \left(\frac{H_{\text{pooled}}(t) + h_{\psi} + H_{\text{wp}}(t)}{H_{\text{wp}}(t)} \right) \quad (101)$$

$$\frac{dH_{\text{pooled}}}{dt} = U_{\text{w,in}}(t) - \frac{dH_{\text{wp}}}{dt} \frac{\epsilon \Delta S_{\text{w}}}{(1 - \bar{v}_{\text{f,g}}(t))} \quad (102)$$

$$\frac{dH_{\text{dp}}}{dt} = - \frac{dH_{\text{wp}}}{dt} \quad (103)$$

$$f_{\text{imm}} = \frac{H_{\text{wp}}(t)}{H_{\text{dp}}(t) + H_{\text{wp}}(t)} \quad (104)$$

5.1.4. Thermal Energy Equations

$$M'_{\text{p}} c_{\text{p}} \frac{\partial T_{\text{p,dp}}(t, z)}{\partial t} = E'_{\text{gen,p}}(t, z) + \frac{\partial}{\partial z} \left\{ \kappa_{\text{p}}(T_{\text{p,dp}})(1 - \epsilon) \frac{\partial T_{\text{p,dp}}(t, z)}{\partial z} \right\} \quad (105)$$

$$M'_{\text{p}} c_{\text{p}} \frac{\partial T_{\text{p,wp}}(t, z)}{\partial t} = \frac{\partial}{\partial z} \left\{ \kappa_{\text{p}}(T_{\text{p,wp}})(1 - \epsilon) \frac{\partial T_{\text{p,wp}}(t, z)}{\partial z} \right\} + E'_{\text{gen,p}}(t, z) - E'_{\text{convect}}(t, z) - E'_{\text{boil}}(t, z) \quad (106)$$

$$c_{\text{w}} \left\{ \frac{\partial}{\partial t} [M'_{\text{w}}(t, z) T_{\text{w,wp}}(t, z)] + \frac{\partial}{\partial z} [M'_{\text{w}}(t, z) U_{\text{w}}(t, z) T_{\text{w,wp}}(t, z)] \right\} = E'_{\text{gen,w}}(t, z) + E'_{\text{convect}}(t, z) + \frac{\partial}{\partial z} \left\{ c_{\text{w}} D_{\text{t,wp}} \frac{\partial}{\partial z} [M'_{\text{w}}(t, z) T_{\text{w,wp}}(t, z)] \right\} \quad (107)$$

$$c_{\text{w}} \left\{ \frac{\partial}{\partial t} [M'_{\text{w}}(t, z) T_{\text{w,pooled}}(t, z)] + \frac{\partial}{\partial z} [M'_{\text{w}}(t, z) U_{\text{w}}(t, z) T_{\text{w,pooled}}(t, z)] \right\} = \frac{\partial}{\partial z} \left\{ c_{\text{w}} D_{\text{t,pooled}} \frac{\partial}{\partial z} [M'_{\text{w}}(t, z) T_{\text{w,pooled}}(t, z)] \right\} \quad (108)$$

5.1.5. Gas Equations

$$\frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial t} = \frac{2}{3}(1 - g_p)(1 - v_{\text{f,g}}(t, z))G_V(\text{H}_2)P'(t, z) - U_g(t, z)\frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,wp}} \frac{\partial V'_{\text{rg,wp}}(t, z)}{\partial z} \right) \quad (109)$$

$$\frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial t} = -U_g(t, z)\frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,pooled}} \frac{\partial V'_{\text{rg,pooled}}(t, z)}{\partial z} \right) \quad (110)$$

$$\frac{\partial V'_{\text{v,wp}}(t, z)}{\partial t} = \frac{E'_{\text{boil}}(t, z)}{\rho_v h_{\text{l,v}}} - U_g(t, z)\frac{\partial V'_{\text{v,wp}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,wp}} \frac{\partial V'_{\text{v,wp}}(t, z)}{\partial z} \right) \quad (111)$$

$$\frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial t} = -U_g(t, z)\frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial z} + \frac{\partial}{\partial z} \left(D_{\text{t,pooled}} \frac{\partial V'_{\text{v,pooled}}(t, z)}{\partial z} \right) \quad (112)$$

5.2. Summary of Interface and Boundary Conditions

Below are a summary of the boundary conditions used in the proposed model. A boundary condition describing the advection of gas from the top boundary is not explicitly required due to the use of a first-order upwind discretisation scheme on all advection terms.

5.2.1. Thermal Interface and Boundary Conditions

$$k(T_{\text{p,dp}})\frac{dT_{\text{p,dp}}(t, H_{\text{dp}})}{dz} = k(T_{\text{p,wp}})\frac{dT_{\text{p,wp}}(t, H_{\text{dp}})}{dz} \quad (113)$$

$$T_{\text{w,wp}}(t, H_{\text{dp+wp}}) = T_{\text{w,pooled}}(t, H_{\text{dp+wp}}) \quad (114)$$

$$D_{\text{t,pooled}}\frac{\partial T_{\text{w,pooled}}(t, H_{\text{dp+wp+pooled}})}{\partial z} = -U_{\text{w,in}}(T_{\text{w,pooled}}(t, H_{\text{dp+wp+pooled}}) - T_{\text{in}}) \quad (115)$$

If $H_{\text{dp}} > 0$:

$$\frac{\partial T_{\text{p,dp}}(t, 0)}{\partial z} = 0 \quad (116)$$

otherwise:

$$\frac{\partial T_{\text{p,wp}}(t, 0)}{\partial z} = 0 \quad (117)$$

$$\frac{\partial T_{\text{w,wp}}(t, 0)}{\partial z} = 0 \quad (118)$$

5.2.2. Gas Void Interface and Boundary Conditions

$$\frac{\partial V'_{\text{rg,wp}}(t, H_{\text{dp+wp}})}{\partial z} = \frac{\partial V'_{\text{rg,pooled}}(t, H_{\text{dp+wp}})}{\partial z} \quad (119)$$

$$\frac{\partial V'_{\text{v,wp}}(t, H_{\text{dp+wp}})}{\partial z} = \frac{\partial V'_{\text{v,pooled}}(t, H_{\text{dp+wp}})}{\partial z} \quad (120)$$

$$\frac{\partial V'_{\text{v,wp}}(t, H_{\text{dp}})}{\partial z} = 0 \quad (121)$$

$$\frac{\partial V'_{\text{rg,wp}}(t, H_{\text{dp}})}{\partial z} = 0 \quad (122)$$

755 5.2.3. Neutron Diffusion Boundary Conditions

$$0 = \frac{1}{4}\phi(r, z) - \frac{1}{2}D(r, z)\frac{\partial}{\partial n}\phi(r, z) \quad (123)$$

5.3. Summary of Inputs Parameters

A summary of the input parameters for simulating the nuclear criticality in the wetted UO₂ powder bed are shown in Table 14.

5.4. Summary of Initial Conditions

760 The sub-critical system is assumed to be in thermodynamic equilibrium at a temperature of 293.15 K. The reference temperature used to evaluate the thermal reactivity feedback is therefore set at 293.15 K. No gas is present in the system at the start of the simulation. The initial neutron and delayed neutron precursor densities are calculated from Equations 4 and 5 using an initial reactivity of, $R_t = -0.179$ \$, initial source density of $s_n = 0.135$ neutrons s⁻¹ (cm⁻³ · UO₂) and delayed
765 neutron data and generation time described in Section 4.7. The initial reactivity, $R_t = -0.179$ \$, has been found using the near-critical geometry of the powder bed, outlined in Section 4.8. These calculated initial conditions are specified alongside the other prescribed conditions in Tables 15 and 16.

Table 14: Summary of input parameters for sub-critical partially wetted UO₂ powder.

Property	Symbol	Value	Discussed
UO ₂ specific heat capacity	c_p	$274 \text{ J kg}^{-1} \text{ K}^{-1}$	sec 4.3.1
Vapour specific heat capacity	c_v	$2.060 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$	sec 4.3.3
Water specific heat capacity	c_w	$4.2 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$	sec 4.3.2
UO ₂ mean powder particle size	$d_{p,1}, d_{p,2}$	$10 \text{ } \mu\text{m}, 100 \text{ } \mu\text{m}$	sec 4.2
Pooled water turbulent mixing coefficient	$D_{t,\text{pooled}}$	$10^{-3} \text{ m}^2 \text{ s}^{-1}$	sec 4.4
Pore water turbulent mixing coefficient	$D_{t,\text{wp}}$	$10^{-4} \text{ m}^2 \text{ s}^{-1}$	sec 4.4
Radiolytic gas generation coefficient	$G_V(\text{H}_2)$	$5.36777 \times 10^{-9} \text{ m}^3 \text{ J}^{-1}$	sec 4.5
Drum height	H	1.0 m	sec 4.1
Latent heat of vaporisation	$h_{l,v}$	$2.2512 \times 10^6 \text{ J kg}^{-1}$	sec 4.3.3
Surface water flow rate	$U_{w,\text{in}}$	$9.947 \times 10^{-4} \text{ m s}^{-1}$	sec 4.1
Drum radius	R	0.4 m	sec 4.1
UO ₂ fission fragment range	R_{fiss}	$7 \text{ } \mu\text{m}$	sec 4.6
Water boiling point	T_{sat}	373.15 K	sec 4.3.2
Water input temperature	T_{in}	293.15 K	sec 4.1
UO ₂ powder enrichment	γ	$5 \text{ } \%$	sec 4.1
UO ₂ powder porosity	ϵ	0.8539	sec 4.1
Water thermal expansion coefficient	η	$2.14 \times 10^{-4} \text{ K}^{-1}$	sec 4.3.2
UO ₂ thermal conductivity	κ_p	see Equation 86	sec 4.3.1
Vapour thermal conductivity	κ_v	$2.46 \times 10^{-2} \text{ W m}^{-1} \text{ K}^{-1}$	sec 4.3.3
Solid UO ₂ powder particle density	$\rho_{p,\text{th}}$	$10.95 \times 10^3 \text{ kg m}^{-3}$	sec 4.1
Vapour dynamic viscosity	μ_v	$1.271 \times 10^{-5} \text{ Pa s}$	sec 4.3.3
Water dynamic viscosity	μ_w	$1.0 \times 10^{-3} \text{ Pa s}$	sec 4.3.2
Vapour density	ρ_v	$5.863 \times 10^{-1} \text{ kg m}^{-3}$	sec 4.3.3
Water density	ρ_w	998.1 kg m^{-3}	sec 4.3.2

Table 15: Group-wise initial delayed neutron precursor concentrations.

Group, i	1	2	3	4	5	6
C_i (precursor number per cm^3 of UO ₂)	1.93	4.07	1.04	0.94	0.15	0.02

Table 16: Initial conditions for sub-critical partially wetted UO_2 powder bed.

Property	Symbol	Value	Discussed
Dry powder height	H_{dp}	0.4414 m	sec 4.8
Wet powder height	H_{wp}	0.1803 m	sec 4.8
Water height	H_{pooled}	0.3783 m	sec 4.8
Neutron density	n	0.0055 neutrons ($\text{cm}^{-3} \cdot \text{UO}_2$)	sec 2.1
Intrinsic source	s_{n}	0.135 neutrons s^{-1} ($\text{cm}^{-3} \cdot \text{UO}_2$)	sec 2.1
Initial dry powder region saturation	$S_{\text{w},0}$	0	-
Reference temperature	T_0	293.15 K	-
All-region water temperature	T_{w}	293.15 K	-
All-region void fraction	$V_{\text{f,g}}$	0	-

6. Case Study Results

6.1. Large Particle Size

Results are displayed as functions of time from the start of the simulation as opposed to the start of water infiltration. For the 100 μm mean powder particle size considered here, criticality occurs approximately 150 seconds after water infiltration begins. The simulation has begun at a point just before criticality, this is reflected in the results by stating $t = T - 150$ seconds where t and T represent the the time since wetting began and the time since the simulation began respectively. Data is shown logarithmically in time due to the short timescales over which the nuclear criticality transient occurs for the 100 μm mean powder particle size case. This is contrary to the 10 μm powder size case which is not shown logarithmically due to the fission power fluctuations occurring on the order of tens of seconds (see Figure 20a).

The variation in the fission power and total fission energy generated 300 s after the initial nuclear criticality excursion is shown in Figure 16a. The initial super-criticality causes the fission power to reach a magnitude of 2.7169×10^8 W (271.69 MW). The system briefly reaches prompt super-critical, with the total reactivity going above unity at approximately $t = 0.3$ s after the first wetting induced criticality is reached. Figures 16b and 18a show that the initial fission power spike leads to the production of radiolytic gas which subsequently causes the system to go sub-critical and then plateau. The plateau in system reactivity and fission power is predicted until boiling occurs as the wetting-induced reactivity matches the reactivity due to the presence of radiolytic gas voids. The reactivity effect of temperature at such high positive reactivity insertions has been shown to not affect the system reactivity as much as voidage in the wet powder. The same may be predicted for voids in the pooled water which do not develop a large negative reactivity feedback, even at high voidage.

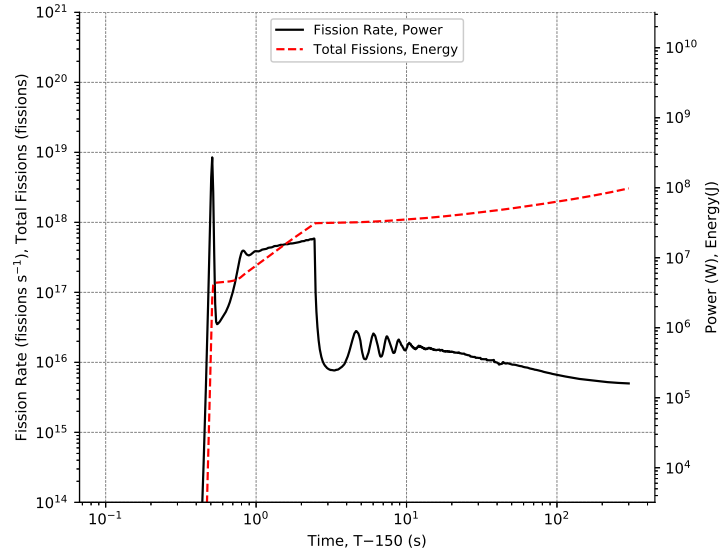
Figure 16b shows that the infiltration of water adds approximately 15 \$ over 5 seconds of time since the initial criticality. Over this time-frame the majority of fissions occur, producing 3.1×10^7 J (31 MJ) out of the total 9.79×10^7 J (97.98 MJ) of fission energy over the duration of the simulation. Figure 16a also shows that the nuclear criticality transient does not terminate once the simulation ends at $t = 300$ s. Instead, the system reaches a new equilibrium at a fission power of 1.8×10^5 W (180 kW). For a direct comparison of the energy released by systems of different particle sizes, the smaller particle size simulation was also limited to 300 s.

The high rate of reactivity insertion causes a sharp increase in the UO_2 powder and pore water temperature, this can be seen in Figure 19a. The increase in temperature leads to boiling in the water filled interstices between powder particles at approximately 2.6 s after the wetting-induced criticality is first reached (see Figure 18a). Due to the large surface area between the powder and water, large volumes of steam were created over a short timescale. This resulted in lower interaction cross-sections in the regions with high voidage, causing the system to go deeply sub-critical. Due to the presence of delayed neutron precursors, boiling did not cause the fission power of the system to decrease below 2.5×10^5 W (250 kW).

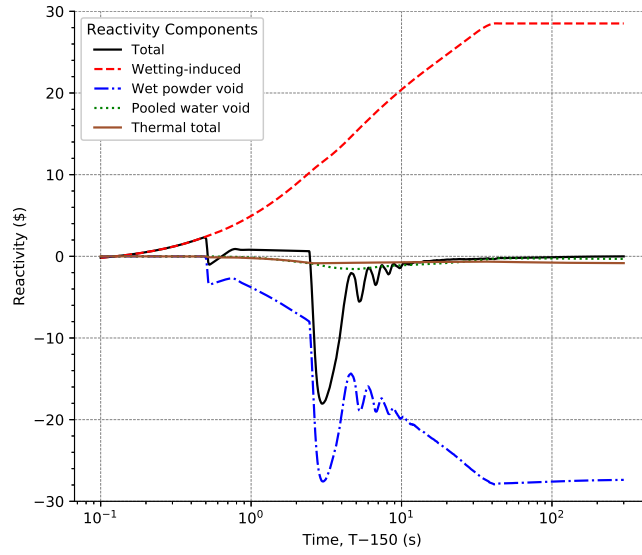
The region heights and species' mass, shown in Figure 17, indicate that at the onset of boiling there is a decrease in the mass of water in the system. This is predominantly due to the overflowing of the containment vessel as large volumes of gas are generated in the wet powder region. Over longer timescales the water varies a small amount due to a balance between the incoming water from the sprinkler system and the vaporisation of water on the particle surface as well as radiolysis.

As gas passes into the pooled water region and out of the system, the void reactivity feedback decreases, leading to an increased total system reactivity. The spatial variation in the void fraction at $t = 300$ s is shown in Figure .

The axial temperature distribution for the saturated UO_2 powder is shown in Figure 19b. The temperature of the powder bed tends to decrease further up the drum. Pooled water remained under the boiling point for the duration of the simulation. As predicted, the local powder and water temperatures in the wet powder region were almost the same, as a result of the high interfacial area between the phases.

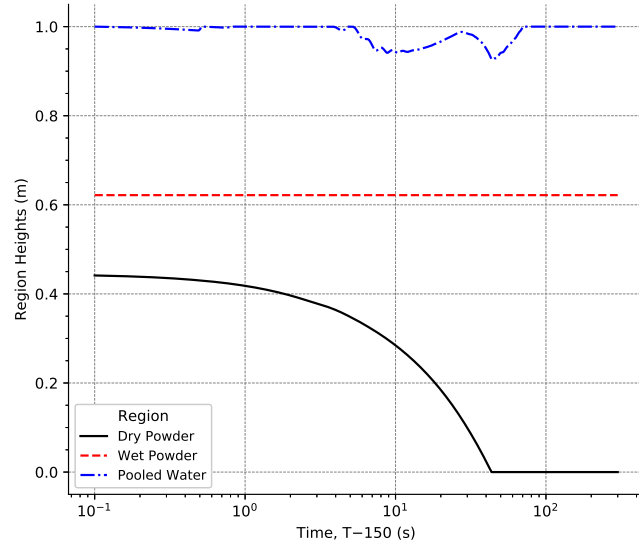


(a)

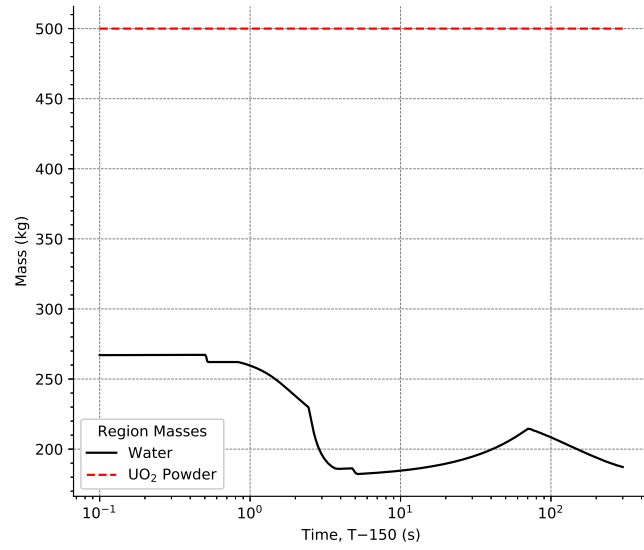


(b)

Figure 16: (a) The variation in fission power and fission energy over time. (b) Variation of each reactivity component as well as total system reactivity. Both for the case where $d_p = 100 \mu\text{m}$.

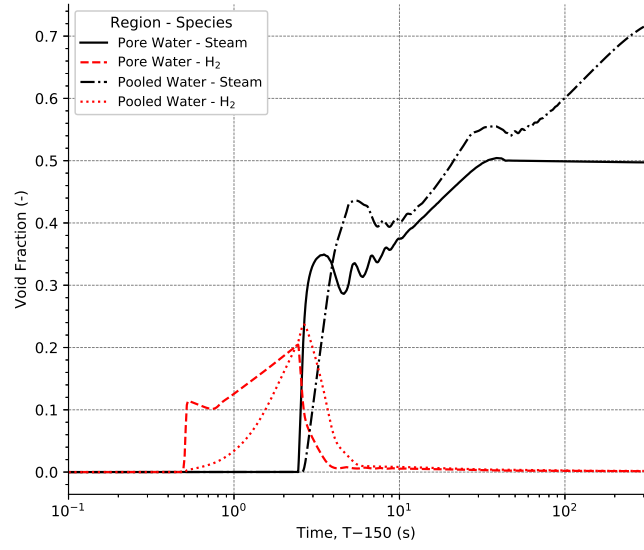


(a)

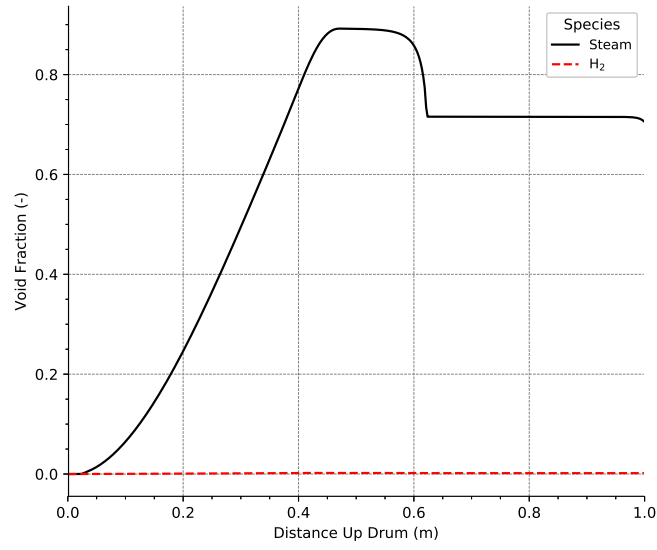


(b)

Figure 17: (a) The variation in region height shows the infiltration of water into the domain. (b) Variation in the total UO_2 powder and water mass over time. Both for the case where $d_p = 100 \mu\text{m}$.

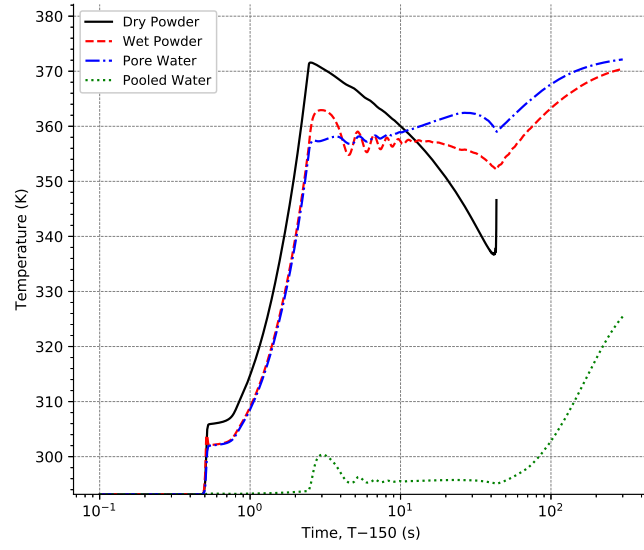


(a)

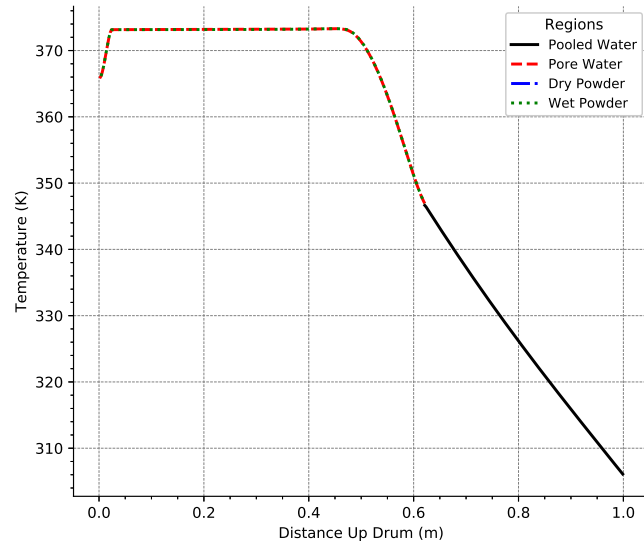


(b)

Figure 18: Voidage data for the case where $d_p = 100 \mu\text{m}$. (a) The variation in void fraction over time. (b) The spatial variation in void fraction throughout the domain at $t = 300 \text{ s}$.



(a)



(b)

Figure 19: Temperature data for the case where $d_p = 100 \mu\text{m}$. (a) The variation in region and species temperatures over time. (b) The spatial variation in temperature throughout the domain at $t = 300 \text{ s}$.

When $d_p = 10 \mu\text{m}$, criticality occurred approximately 410 seconds after infiltration began. The simulation was started at a point just before criticality, this is reflected in the results by stating $t = T - 410$ seconds. The presence of gas within the system impeded the progression of the wetting front.

825 The variation in various reactivity components is shown in Figure 20b. The rate of increase in the wetting-induced reactivity is less for the $10 \mu\text{m}$ than the $100 \mu\text{m}$ case. Figure 20a shows that the initial criticality excursion causes the fission power to spike at $3.1 \times 10^6 \text{ W}$ (3.1 MW). The subsequent steady power rise increases the power to $3.6 \times 10^6 \text{ W}$ (3.5 MW) before boiling occurs. Subsequent fission power oscillations that occur after the onset of boiling do not exceed the
830 initial fission power spike. The total energy released over the 300 s of simulated time is $6.5 \times 10^7 \text{ J}$ (65 MJ).

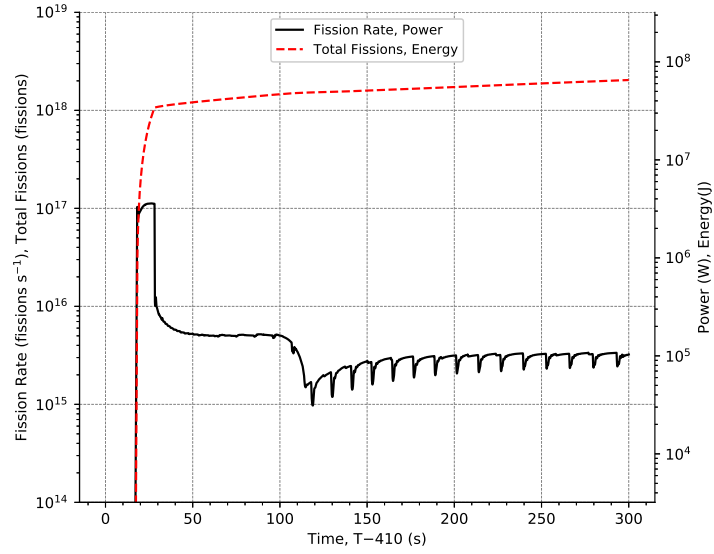
The system reaches prompt critical at a later time than for the larger particle size (approximately $t = 12 \text{ s}$) because of the lower rate of water infiltration caused by a lower powder bed permeability. During the initial delayed super-critical stage of the transient, the concentration of delayed neutron
835 precursors begins to build and the fission power rises at a lower rate than in the prompt super-critical stage. Once the system reaches prompt super-critical the fission power is limited by the production of radiolytic gas and to a lesser extent, Doppler broadening. A plateau can be seen with steadily increasing fission power until boiling occurs.

Figure 22 shows a sharp increase in the volume of radiolytic gas in the pores around the powder
840 particles at the time of prompt super-criticality. This gas advects into the pooled water region and eventually leaves the system. Figures 21a and 21b show that the production of radiolytic gas pushes water out of the top of the container. Significant volumes of radiolytic gas cease to be produced when boiling occurs due to the large volumes of steam that take up space between the powder particles.

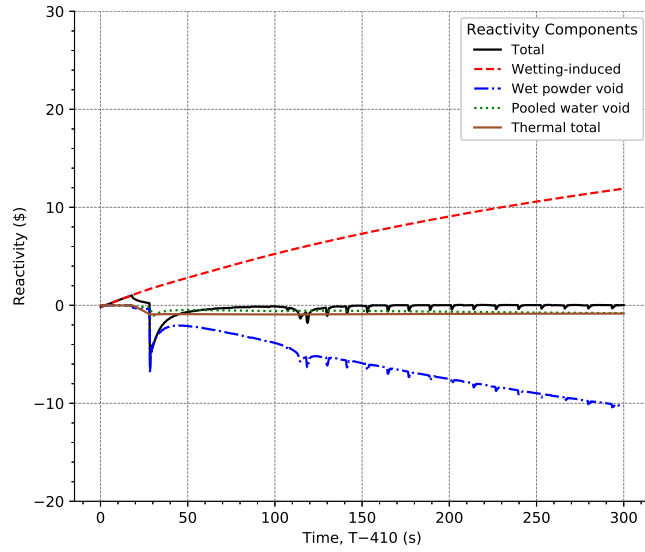
845 A significant reduction in the fission power is predicted at $t = 28 \text{ s}$ as steam generated in the wetted powder adds approximately 7 \$ of negative reactivity into the system. After this time, the power decreases to an order of magnitude of 10^5 W . Advection of the steam from the domain (see Figure 22) causes the system reactivity to slowly approach critical again. A cyclical fission power response with small amplitude is predicted as water hits highly superheated dry powder at the
850 wetting front. This causes violent boiling, adding negative reactivity, preventing further boiling

until the gas advects from the region around the wetting front and the process repeats.

The steep gradient between the dry and wet powder shown in Figure 23b may be attributed to a number of reasons. A combination of efficient convection between the wet powder and pore water, as well as the high heat capacity of the pore water, means that significantly more energy is required to raise the wet powder temperature than the dry powder. Also, the production of steam adds a cooling effect in the wet powder region, this is not present in the dry powder. The result is a dry powder region that heats up quickly when compared to the wetted powder. The low thermal conductivity of UO_2 combined with the high porosity prevents a significant exchange of thermal energy between the dry and wet powder.

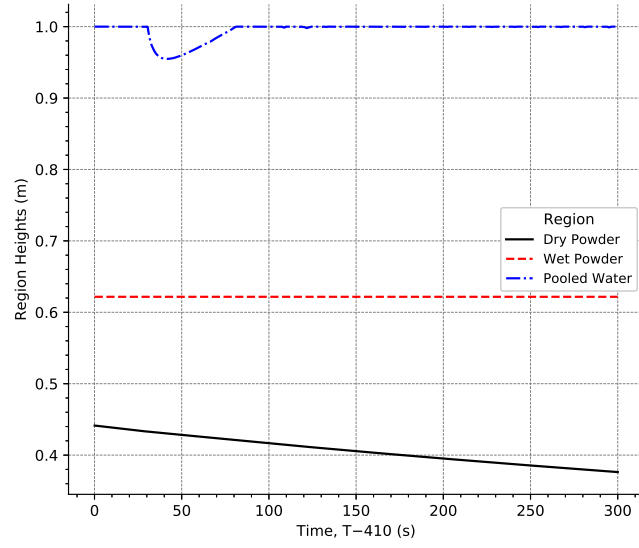


(a)

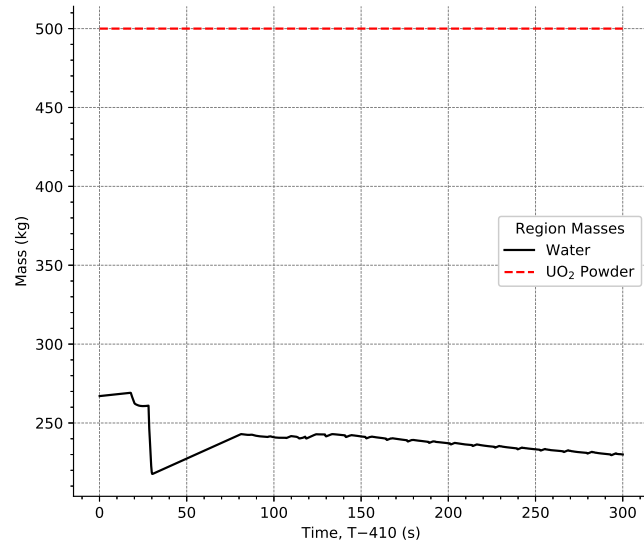


(b)

Figure 20: (a) The variation in fission power and fission energy over time. (b) Variation of each reactivity component as well as total system reactivity. Both for the case where $d_p = 10 \mu\text{m}$.



(a)



(b)

Figure 21: (a) The variation in region height shows the infiltration of water into the domain. (b) Variation in the total UO_2 powder and water mass over time. Both for the case where $d_p = 10 \mu\text{m}$.

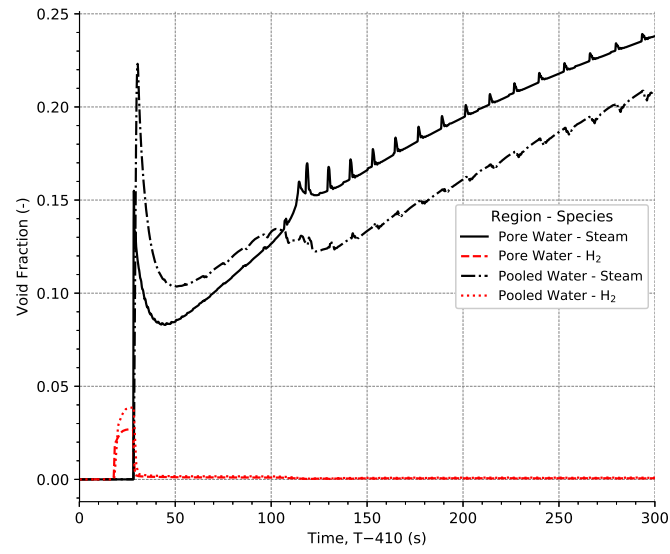
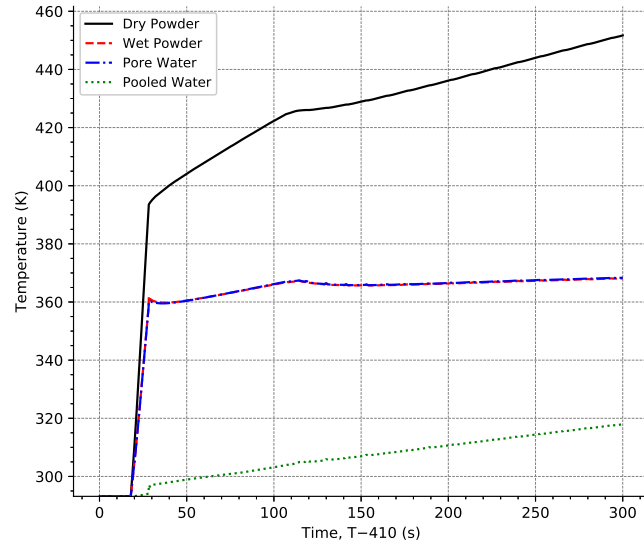
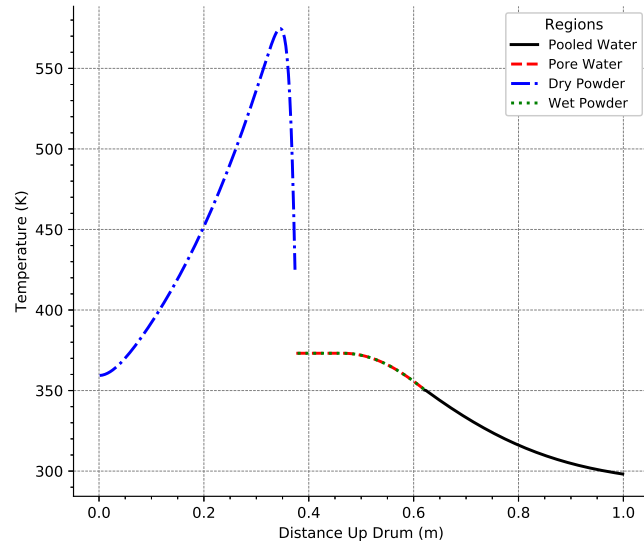


Figure 22: The region and species specific void fraction for the case where $d_p = 10 \mu\text{m}$.



(a)



(b)

Figure 23: Temperature data for the case where $d_p = 10 \mu\text{m}$. (a) The variation in region and species temperatures over time. (b) The spatial variation in temperature throughout the domain at $t = 300 \text{ s}$.

860 6.3. Sensitivity of Turbulent Mixing Coefficient and Intrinsic Neutron Source

The turbulent mixing coefficient is a complex parameter to correctly determine due to the chaotic nature of turbulent flows caused by the generation and advection of gas bubbles in the pore and pooled water. By altering the turbulent mixing coefficient for each region by an order of magnitude, the effect that this parameter has on the initial fission power spike, total energy released and time to boiling may be evaluated. Table 17 shows that over the 300 s of simulated time, the primary fission power spike that occurs after the initial criticality excursion has a strong dependence on powder particle size and not the turbulent mixing coefficient. Similarly, the time to boiling did not vary widely by changing the turbulent mixing coefficient by an order of magnitude in each region. Small variations in the fission energy released 300 s after the initial criticality excursion are predicted as the turbulent mixing coefficient varies. The particle size is predicted to strongly affect the fission energy released over the first 300 s of the nuclear criticality excursion. Decreasing the turbulent mixing coefficient in the wet powder region prevented energy and voids from diffusing through the system. For the small powder size case this suppressed the subsequent fission power spikes. The effect on the energy released for the larger powder size was less pronounced. Fission energy release for the 100 μm powder particles were similar in magnitude regardless of the turbulent mixing coefficient.

The intrinsic neutron source was varied by 50 % above and below the nominal value of $s_n = 0.135 \text{ neutrons s}^{-1} (\text{cm}^{-3} \cdot \text{UO}_2)$ and the effect on the initial peak fission power and energy released 300 s after the initial nuclear criticality excursion for the case where $d_p = 100 \mu\text{m}$ was studied. Corresponding new neutron density and delayed neutron precursor initial conditions were calculated. Results shown in Figure 24 indicate that there is a small dependence on the initial peak fission power due to the intrinsic neutron source density. This amounts to initial peak fission power variations no greater than 2.85 %. A larger intrinsic source density led to a decreased peak fission power because reactivity feedback's were able to be greater in magnitude before the prompt super-critical state was reached. The total energy released from fission 300 s after the initial nuclear criticality excursion did not differ by more than 0.005 % relative to the energy produced from fission for the nominal neutron source density case. This is expected since the energy produced from fission of neutrons from intrinsic sources is very low compared to the energy produced from neutrons born through fission during the transient.

Table 17: Sensitivity of initial peak fission power, energy and boiling time to mean powder particle size and turbulent mixing coefficients.

Case	d_p (μm)	$D_{t,wp}$ ($\text{m}^2 \text{s}^{-1}$)	$D_{t,pooled}$ ($\text{m}^2 \text{s}^{-1}$)	Primary fission power Spike (MW)	Fission Energy (MJ)	Time to Boiling (s)
1	10	10^{-4}	10^{-3}	3.60	65.30	28.12
2	100	10^{-4}	10^{-3}	271.69	97.98	2.46
3	10	10^{-5}	10^{-3}	3.59	65.09	28.13
4	100	10^{-5}	10^{-3}	271.66	97.94	2.46
5	10	10^{-4}	10^{-4}	3.60	65.28	28.12
6	100	10^{-4}	10^{-4}	271.69	97.98	2.46
7	10	10^{-3}	10^{-3}	3.56	65.14	28.22
8	100	10^{-3}	10^{-3}	272.06	97.99	2.47
9	10	10^{-4}	10^{-2}	3.60	65.28	28.12
10	100	10^{-4}	10^{-2}	271.69	97.98	2.46

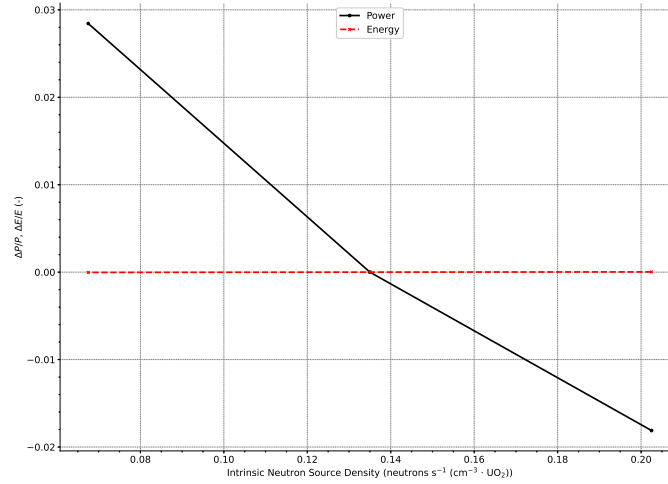


Figure 24: The relative variation of fission power and energy as a function of the intrinsic neutron source density.

890 7. Conclusions

This paper proposed a novel model coupling point nuclear reactor kinetics with phenomenologically based models of powder bed wetting and nuclear thermal hydraulics, including models for the generation and advection of steam and radiolytic gas, for the analysis of nuclear criticality excursions in fissile powder bed systems under wetting conditions. The point nuclear reactor kinetics
895 model used non-linear multi-dimensional reactivity components that are dependent on the powder bed immersion fraction, temperature and system voidage. These types of systems can be found in nuclear fuel production and fabrication facilities around the world. Understanding potential nuclear criticality excursions, under processing and storage conditions, is an important aspect of nuclear criticality safety assessment. Using this model, the initial fission power spike and fission yield, over
900 the 300 s of simulated time, could be quantified.

A comparative study found good agreement between the proposed Green and Ampt infiltration model and experimental data by Rozain et al. [9], for UO_2 mean powder particle sizes observed in the literature.

A hypothetical nuclear criticality excursion scenario was considered which consisted of a container of low-enriched UO_2 powder which becomes open to the outside environment at the top surface. A sprinkler system, at a storage or fabrication facility holding these containers then becomes
905 activated, causing water to impinge upon the surface of the powder, leading to water infiltration. Two mean powder particle sized beds were considered, the larger being $100\text{ }\mu\text{m}$, the smaller being $10\text{ }\mu\text{m}$.

The simulations predicted that the magnitude of the first fission power spike was similar for
910 both UO_2 powder particle sizes modelled. The first fission power spike was predominantly limited by the production of radiolytic gas. Due to the presence of water in the system, the powder temperature could not increase enough to counter the significant positive reactivity insertion due to water infiltration. Consequently, the system remained super-critical until enough energy had
915 been deposited in the powder-water mixture for boiling to occur and for steam to be produced. The decreased rate of wetting due to the variation in the powder bed size caused the geometry of the system to change less rapidly around the point of criticality. This may be the reason for the varying neutron kinetic response predicted by the simulations of the powder beds.

For both powder beds considered, boiling caused the system to become significantly sub-critical
920 due to the negative void reactivity effect caused by the presence of large volumes of steam in

the pore interstices. The larger mean powder particle sized bed reached a new critical state at a lower fission rate. An oscillatory response in the fission power was predicted for the smaller mean powder particle sized bed as water percolated into super-heated dry powder. Although powder motion has not been modelled, the large volumes of gas generated over these short timescales are likely to provide enough force to overcome the weight of gravity of the powder, resulting in powder motion. This corresponds well with the study performed by Rozain et al. [9] who also noted that the production of gas was likely to result in significant motion of the powder bed.

The fission power response and peak fission power was found to vary depending on the powder particle size. A maximum fission power of 272 MW was predicted for the larger powder particles compared to 3.60 MW for the smaller sized powder particles, indicating that the initial release of neutrons is far greater for the UO_2 powder bed consisting of larger particles. The fission energy released during the first 300 s of the transient nuclear criticality excursion was predicted to be a strong function of the mean powder particle size at 65 MJ and 98 MJ for the small and larger powder particle sized cases, respectively. In the absence of experimental data, one may note that the total yield was similar in order of magnitude to previous simulations of powder bed systems by Rozain et al. [9] and Basoglu et al. [24].

Although the peak fission power and time to boiling was found to be almost independent of the range of turbulent mixing coefficient, they depended strongly on the mean powder particle size used. Similarly, the fission energy released over the duration of the simulation was a strong function of mean powder particle size and to a lesser extent, the turbulent mixing coefficient. The energy was found to be independent of the intrinsic neutron source density used in the investigation. A weak dependence was predicted for the initial peak fission power reached after the initial nuclear criticality excursion and the intrinsic neutron source density.

8. Data Statement

In accordance with EPSRC funding requirements, all supporting data used to create figures in this paper may be accessed at the following URL: <https://doi.org/10.5281/zenodo.5636825>.

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