

# Transient analysis of the 1970 Windscale nuclear criticality incident

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## ABSTRACT

This paper describes a novel methodology for the analysis of transient nuclear criticality in layered aqueous-emulsion-organic plutonium nitrate systems. The presented methodology includes point neutron kinetics equations coupled with phenomenological one-dimensional nuclear thermal hydraulics models, which describe the variation in mass, power, reactivity, temperature and voidage within the system. Equations to describe the mean neutron generation time and changes in reactivity due to changes in the system's temperature and void fraction are formulated from fits of the results predicted by the criticality transient multiphysics model. Using these developed models, the nuclear criticality incident that occurred at the Windscale Works in 1970 is analysed and presented. In this criticality transient, two immiscible fissile liquids (an aqueous solution and an organic solvent) were disturbed such that an emulsion layer formed between them, inducing a transient nuclear criticality excursion and yielding around  $10^{15}$  fissions. It was determined that parameters such as emulsion globule size and vessel outlet pipe radius had negligible impact on the total fission yield of the corresponding transient when compared to variations in organic solvent volume and emulsion band thickness. Several possible configurations were identified which could result in a transient similar to that of the Windscale Works in 1970. Notably, a system containing 39.75 L of organic solvent which underwent a transient with the formation of an emulsion band that was a maximum of 5.4 cm thick and 54% aqueous solution by volume yielded  $9 \times 10^{14}$  fissions, and thus is expected to closely resemble the 1970 Windscale nuclear criticality incident. However, the uncertainties associated with the original transient and the system parameters (e.g. thermophysical properties of the three layers) are such that the configuration is likely to vary from these exact quantities.

## 1. Introduction

Solvent extraction processes naturally involve fissile liquids, and are present throughout the nuclear fuel cycle, for example dissolving ores during in-situ leaching and reprocessing waste using the PUREX process (Crossland, 2012; Wilson, 1996; Zohuri, 2019). During the PUREX process, TBP-kerosene is frequently used as the solvent (or organic phase), with the solution being aqueous uranyl or plutonium nitrate (Irish and Reas, 1957). Additionally, fissile liquid systems can also be found in a range of experimental and research facilities (Crossland, 2012; McLaughlin et al., 2000; Wilson, 1996; Zohuri, 2019). Process criticality accidents are more likely to occur in fissile solutions or liquid media (such as plutonium-bearing aqueous and organic liquids) than in dry systems (McLaughlin, 2003).

Accidental criticality has associated hazards and dangers. Primarily, unexpected and unwanted radiation release during accidental criticality can expose facility workers, the general public and the wider environment to ionising radiation (Crossland, 2012; Kimpland, 1993; Knief, 1985; Srinivasan et al., 1992), which can prove harmful to health, or

even be fatal (Crossland, 2012; Knief, 1985). Additionally, the need for cleanup, on-site repair, increased healthcare and mitigation actions can result in significant economic costs (Bizet and Lévêque, 2017).

Thus, the thorough understanding of the behaviour of nuclear systems is paramount for the prevention of nuclear criticality excursions (International Atomic Energy Agency, 2014); learning from past accidents helps to prevent and prepare for future accidents of a similar nature. As such, past accidents and excursions influence present-day criticality control measures (Pruvost and Paxton, 1996; Thomas, 1978). Further, emergency planning and preparedness can be aided by well-developed phenomenological codes, so that risks and costs associated with nuclear criticality accidents can be minimised (Kimpland, 1993; McLaughlin, 2003; Srinivasan et al., 1992; Whitesides et al., 2010).

In 1970, a nuclear criticality excursion occurred at the Windscale Works, in a transfer vessel of a plutonium recovery facility. Unknown to workers, an organic tributyl phosphate (TBP)-kerosene solvent, rich in plutonium (IV) nitrate, was trapped in the vessel. It was estimated that criticality was maintained for less than 10 s, and around  $10^{15}$  fissions

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**Nomenclature****Acronyms**

CP Collision probability

**Latin Symbols**

|            |                                                                                |
|------------|--------------------------------------------------------------------------------|
| $A$        | Area [m <sup>2</sup> ]                                                         |
| $C$        | Delayed neutron precursor population [–]                                       |
| $C_p$      | Specific heat capacity [J kg <sup>−1</sup> K <sup>−1</sup> ]                   |
| $D_{FS}$   | Mixing coefficient [m <sup>2</sup> s <sup>−1</sup> ]                           |
| $E$        | Energy [J]                                                                     |
| $E_n$      | Neutron energy [MeV]                                                           |
| $F$        | Total number of fissions [–]                                                   |
| $g$        | Acceleration due to gravity [m s <sup>−2</sup> ]                               |
| $G_{rg}$   | Radiolytic gas generation coefficient [m <sup>3</sup> J <sup>−1</sup> ]        |
| $H$        | Height [m]                                                                     |
| $h$        | Heat transfer coefficient [J m <sup>−2</sup> K <sup>−1</sup> s <sup>−1</sup> ] |
| $k_{eff}$  | Effective neutron multiplication factor [–]                                    |
| $M$        | Mass [kg]                                                                      |
| $n$        | Neutron population [–]                                                         |
| $P$        | Power [J s <sup>−1</sup> ]                                                     |
| $p$        | Pressure [MPa]                                                                 |
| $R$        | Vessel radius [m]                                                              |
| $r_{glob}$ | Emulsion globule radius [m]                                                    |
| $r_{out}$  | Vessel outlet radius [m]                                                       |
| $s_n$      | Intrinsic neutron source [neutrons s <sup>−1</sup> ]                           |
| $T$        | Temperature [K]                                                                |
| $t$        | Time [s]                                                                       |
| $U$        | Velocity [m s <sup>−1</sup> ]                                                  |
| $V$        | Volume [m <sup>3</sup> ]                                                       |
| $z$        | Axial location [m]                                                             |

**Greek Symbols**

|              |                                                                           |
|--------------|---------------------------------------------------------------------------|
| $\alpha$     | Volume fraction of an emulsion that is the aqueous phase [–]              |
| $\beta$      | Delayed neutron fraction [–]                                              |
| $\epsilon$   | Energy released per fission [J fission <sup>−1</sup> ]                    |
| $\hat{\phi}$ | Normalised scalar neutron flux distribution [cm <sup>−1</sup> ]           |
| $\kappa$     | Thermal conductivity [J m <sup>−1</sup> K <sup>−1</sup> s <sup>−1</sup> ] |
| $\Lambda$    | Mean neutron generation time [s]                                          |
| $\lambda$    | Delayed neutron precursor decay constant [s <sup>−1</sup> ]               |
| $\mu$        | Dynamic viscosity [kg m <sup>−1</sup> s <sup>−1</sup> ]                   |
| $\omega$     | Mass fraction of an emulsion that is the aqueous phase [–]                |

|            |                                                                                    |
|------------|------------------------------------------------------------------------------------|
| $\phi$     | Radially averaged scalar neutron flux [neutrons cm <sup>−2</sup> s <sup>−1</sup> ] |
| $\rho$     | Density [kg m <sup>−3</sup> ]                                                      |
| $\sigma$   | Surface tension [J m <sup>−2</sup> ]                                               |
| $\Sigma_f$ | Macroscopic neutron fission cross section [cm <sup>−1</sup> ]                      |
| $\theta$   | Void fraction [–]                                                                  |
| $\xi$      | Reactivity [–]                                                                     |
| $v$        | Neutron speed [cm s <sup>−1</sup> ]                                                |
| $\zeta$    | Effective thickness [cm]                                                           |

**Subscripts**

|      |                                          |
|------|------------------------------------------|
| aq   | Aqueous                                  |
| eff  | Effective                                |
| em   | Emulsion                                 |
| or   | Organic                                  |
| rg   | Radiolytic gas                           |
| th   | Thermal                                  |
| vess | Vessel                                   |
| $i$  | Index of delayed neutron precursor group |

Section 2 describes the nuclear criticality incident that occurred at the Windscale Works in 1970. Sections 3–6 discuss the phenomenology of criticality transients in multi-layered fissile liquid systems, and the parameters required to employ such phenomenology in transient analysis, with application to the 1970 Windscale nuclear criticality incident. The use of the developed model and parameters is presented in Sections 7 and 8: Section 7 details the overall behaviour of a range of multi-layered fissile liquid transients, forming an investigation into the effects of selected parameters on fission yield. The detailed description of the behaviour of an example transient is presented in Section 8.

The motivation for the presented research includes a more thorough understanding of criticality transients involving layered fissile liquid systems, for risk assessment and safety analysis, using the 1970 Windscale nuclear criticality incident as an example.

## 2. The 1970 Windscale criticality transient

The criticality incident at the Windscale Works in 1970 occurred in a transfer vessel which was part of the Works' plutonium residue recovery plant. In this plant, plutonium was recovered from solid and liquid residues by TBP/OK solvent extraction (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000). Figs. 1 and 2 show isometric and side elevation schematics of the facility. A batch of the plutonium recovery process involved vacuum lifting an aqueous solution of plutonium nitrate from a conditioner into a transfer vessel. On completion of the vacuum lift, the vessel began to drain, feeding solution to the remainder of the recovery process (British Nuclear Fuels Limited, no date; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000).

At the time of the incident, it was thought that the transfer vessel was empty between batches. However, there was in fact a liquid organic phase continually present in the vessel (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000). The exact reason the organic phase came to exist in the vessel is unknown. It was postulated that its origin was due to the entrainment of the organic phase within the aqueous phase, either earlier on in the process line or during the recycling of the aqueous material as shown in Fig. 2 (Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972).

occurred. Due to the low fission yield, it is thought that geometric shutdown mechanisms (i.e. emulsion dissipation and aqueous drainage) brought the system back to a subcritical level (British Nuclear Fuels Limited, no date; Chalmers, 1971; Evans, 1984a,b; Daniels et al., 1971; Hughes, 1972; McLaughlin et al., 2000).

This paper presents the development of a mathematical and computational model to calculate transient behaviour of an aqueous-emulsion-organic layered fissile liquid system. Using this model, an investigation into the 1970 Windscale nuclear criticality incident has been performed.

**Table 1**

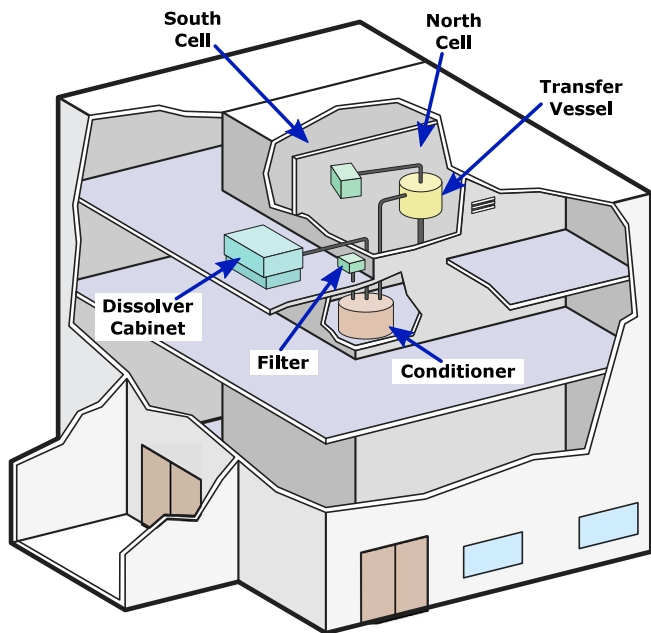
Atomic compositions of the aqueous and organic phases used in the presented research (Chalmers, 1971).

| Nuclide           | Atomic weight | Aqueous                                                     |          |          | Organic                                                     |          |          |
|-------------------|---------------|-------------------------------------------------------------|----------|----------|-------------------------------------------------------------|----------|----------|
|                   |               | Number density (atoms barn <sup>-1</sup> cm <sup>-1</sup> ) | Weight % | Atomic % | Number density (atoms barn <sup>-1</sup> cm <sup>-1</sup> ) | Weight % | Atomic % |
| Hydrogen          | 1.0080        | $5.90890 \times 10^{-2}$                                    | 7.750    | 56.986   | $6.08950 \times 10^{-2}$                                    | 10.671   | 61.744   |
| Carbon            | 12.0112       | –                                                           | –        | –        | $3.18698 \times 10^{-2}$                                    | 66.550   | 32.314   |
| Nitrogen          | 14.0067       | $4.28602 \times 10^{-3}$                                    | 7.812    | 4.134    | $7.70981 \times 10^{-4}$                                    | 1.877    | 0.782    |
| Oxygen            | 15.9994       | $4.02906 \times 10^{-2}$                                    | 83.889   | 38.862   | $4.42273 \times 10^{-3}$                                    | 12.302   | 4.484    |
| Phosphorus        | 30.9738       | –                                                           | –        | –        | $5.27448 \times 10^{-4}$                                    | 2.840    | 0.535    |
| <sup>239</sup> Pu | 239.0522      | $1.65321 \times 10^{-5}$                                    | 0.514    | 0.016    | $1.29913 \times 10^{-4}$                                    | 5.399    | 0.132    |
| <sup>240</sup> Pu | 240.0540      | $1.09754 \times 10^{-6}$                                    | 0.034    | 0.001    | $8.62475 \times 10^{-6}$                                    | 0.360    | 0.009    |

**Table 2**

Molecular compositions of the two phases, calculated using Table 1.

| Molecule          | Chemical formula                                              | Weight (g mol <sup>-1</sup> ) | Aqueous    |          | Organic    |          |
|-------------------|---------------------------------------------------------------|-------------------------------|------------|----------|------------|----------|
|                   |                                                               |                               | Molecular% | Weight % | Molecular% | Weight % |
| Water             | H <sub>2</sub> O                                              | 18.015                        | 86.632     | 64.315   | –          | –        |
| Nitric acid       | HNO <sub>3</sub>                                              | 63.013                        | 13.312     | 34.568   | 6.766      | 2.375    |
| OK (approximated) | C <sub>11</sub> H <sub>20</sub>                               | 152.282                       | –          | –        | 72.452     | 61.471   |
| TBP               | (C <sub>4</sub> H <sub>9</sub> ) <sub>3</sub> PO <sub>4</sub> | 266.320                       | –          | –        | 16.459     | 24.421   |
| Plutonium nitrate | Pu(NO <sub>3</sub> ) <sub>4</sub>                             | 487.134                       | 0.056      | 1.118    | 4.323      | 11.733   |

**Fig. 1.** An isometric elevation schematic of the facility.

Source: Adapted from British Nuclear Fuels Limited (no date) and originally published by Daniels et al. (2024b).

As aqueous solution entered the vessel, it fell as a jet through the less dense organic phase. This generated a cone-like plug of aqueous solution within the organic phase, and the disruption to the organic phase induced the formation of an emulsion layer between the aqueous and organic phases, as presented in Fig. 3 (Chalmers, 1971; Hughes, 1972; McLaughlin et al., 2000). This emulsion layer consisted of organic globules dispersed within the continuous aqueous phase (Chalmers, 1971). Due to the density difference of the aqueous (dense) and organic (less dense) phases, the aqueous solution accumulated at the bottom of the vessel, as shown in Fig. 3. As such, when the vessel drained, the aqueous solution drained first, and the organic phase remained hydraulically trapped in the vessel after each batch process, balanced by the aqueous solution within the lute between the transfer vessel and constant volume feeder (as shown in Fig. 2). Thus, it is likely that the organic phase built in volume and plutonium concentration

over time, until it contained enough fissile mass to induce a nuclear criticality excursion during the processing of an aqueous batch (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000). The chemical examination of the organic phase, once it had been recovered from the vessel, indicated high concentrations of degradation products, which were produced by alpha irradiation originating from the plutonium in the system. This is an indication that the organic phase could have been trapped in the vessel from several months to two years (Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972).

Evans (1984a) reported that there were four distinct stages to the transient:

1. “Filling”: a jet of the aqueous solution entered the vessel, creating a coarse emulsion whilst sinking through the solvent. The jet’s impact onto the solvent was seen to move from the centre of the vessel towards the vessel wall (i.e. moving from Figs. 4(a) to 4(b)). This stage lasted around 4 min.
2. “In-rush”: for around 2 s, the aqueous jet impacted the vessel wall (as in Fig. 4(b)), not the solvent. However, the emulsion layer was maintained during this time, with no significant changes.
3. “Separation”: the emulsion layer dispersed into the separate phases, which took between 5 and 10 s.
4. “Draining”: the aqueous solution drained from the system, starting simultaneously to separation. The drainage rate was initially 1 litre per second, slowing to 0.1 litres per second after 10 s.

These stages are illustrated diagrammatically in Fig. 4.

As the aqueous solution entered the vessel, the system’s reactivity increased — as the volume of the aqueous solution in the vessel increased, the emulsion band (containing the plutonium-rich organic solvent) became thicker, and more geometrically central. Additionally, the high hydrogen content of the aqueous phase aided neutron moderation, and the reflection of neutrons into the plutonium-rich organic phase. After an entire batch of aqueous solution had been lifted into the vessel, there was no process disturbing the organic phase, or the aqueous–organic interface. As such, the emulsion layer dissipated over time. The emulsion dissipation, and aqueous draining, were seen as the most significant criticality shut-down mechanisms (Daniels et al., 1971; Evans, 1984a; Hughes, 1972; McLaughlin et al., 2000).

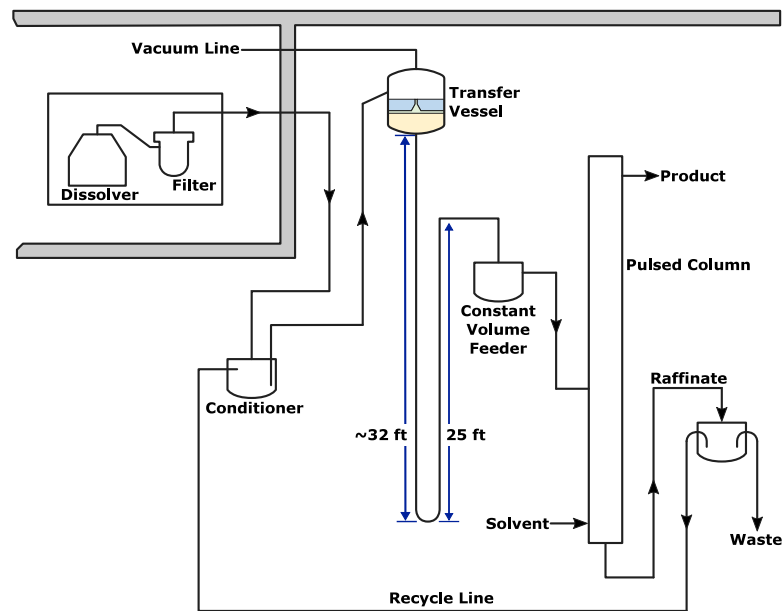


Fig. 2. A side elevation schematic of the relevant section of the processing facility. Source: Adapted from McLaughlin et al. (2000) and originally published by Daniels et al. (2024b).

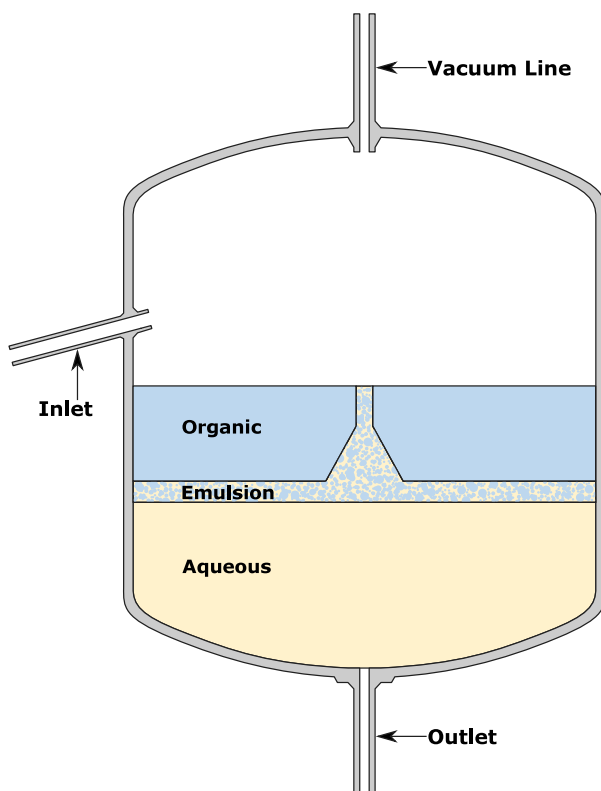


Fig. 3. A side elevation schematic of the 1970 Windscale transfer vessel. Source: Adapted from Chalmers (1971) and originally published by Daniels et al. (2024b).

### 2.1. Properties of the aqueous and organic phases

The atomic compositions of the aqueous and organic phases are presented in Table 1. Using this data, the molecular composition of the two phases can be calculated, and are given in Table 2. In Table 2,

the kerosene of the organic phase has been approximated as having chemical formula  $C_{11}H_{20}$ , though in reality it is a mixture of different hydrocarbons with varied molecular formulae.

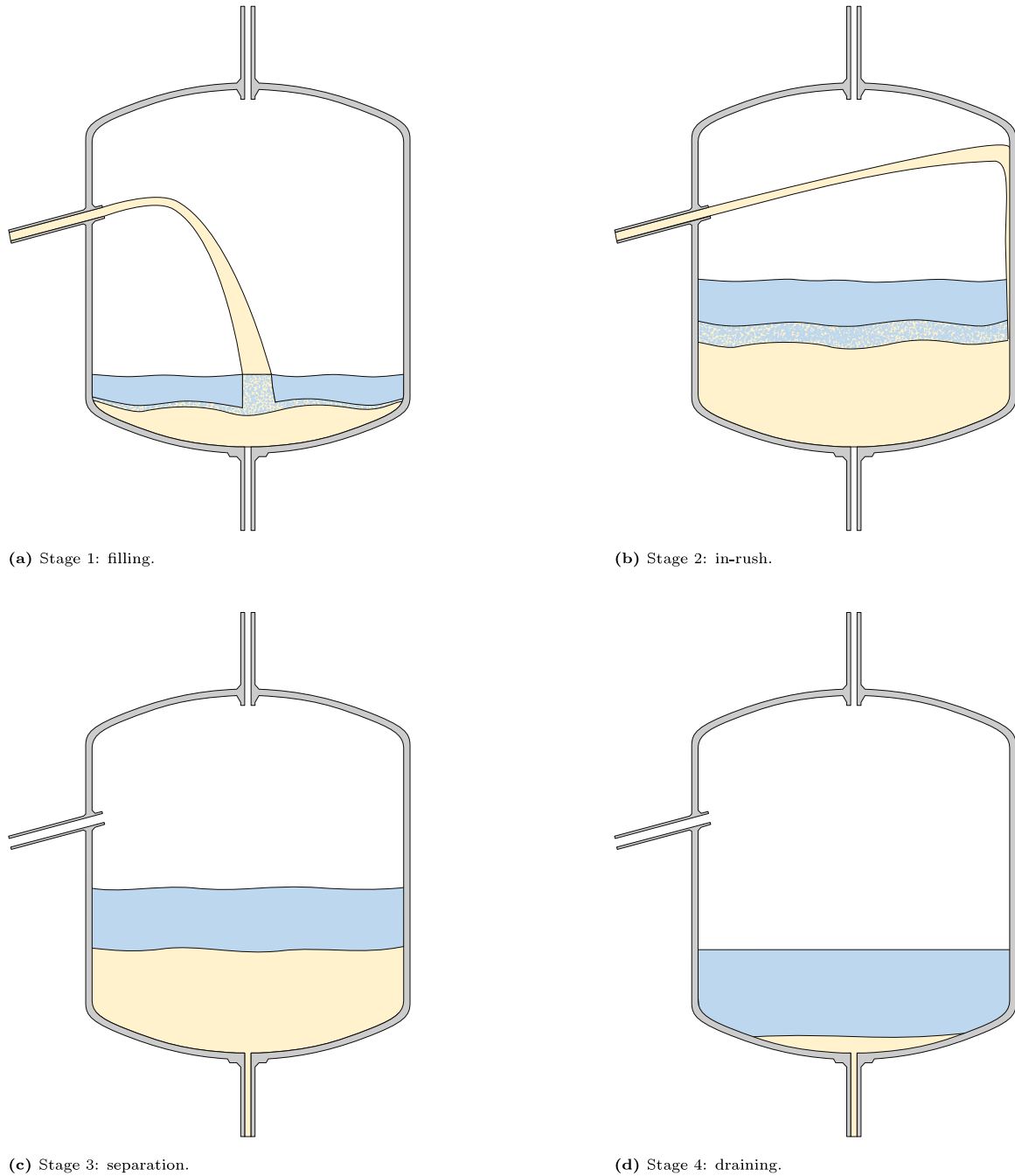
As reported by Chalmers (1971), the plutonium isotopic composition of both phases was 93.750%  $^{239}\text{Pu}$  and 6.250%  $^{240}\text{Pu}$  by weight. The densities of the aqueous and organic phases were  $1.276 \text{ g cm}^{-3}$  and  $0.955 \text{ g cm}^{-3}$ , respectively, and the plutonium concentrations were  $7 \text{ g L}^{-1}$  and  $55 \text{ g L}^{-1}$ , for the aqueous and organic phases, respectively (Chalmers, 1971; Daniels et al., 1971; Evans, 1984a; Hughes, 1972; McLaughlin et al., 2000). Finally, the excess nitric acid molarities were 7.0 M and 0.36 M for the aqueous and organic solutions, respectively (Chalmers, 1971). In the presented research, to remain consistent with the MCNP evaluations of later sections, it will be assumed that these material properties were determined at 293.6 K.

### 2.2. Parameters and uncertainties associated with the 1970 Windscale nuclear criticality incident

In addition to the material data given in Section 2.1, the height, radius and volume of the transfer vessel are known to be 68.6 cm, 30.5 cm and 176 L respectively, and the size of a batch of aqueous solution was 50 L (British Nuclear Fuels Limited, no date; Chalmers, 1971; Hughes, 1972; McLaughlin et al., 2000). The limited amount of data and literature available results in numerous unknowns and uncertainties being associated with various details of the 1970 Windscale nuclear criticality incident.

Firstly, the components of the vessel geometry that are unknown are: the vacuum line, inlet pipe and outlet pipe radii, and the exact shape of the curved top and base of the vessel. In the presented research, the vessel schematic reported by Chalmers (1971) (adapted and presented in Fig. 3) will be assumed to be a to-scale model of the transfer vessel.

The uncertainties regarding the different phases present in the system include the maximum thickness of the emulsion band being reported to have values in the range 5 to 7.6 cm (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a; Hughes, 1972; McLaughlin et al., 2000). Similarly, conflicting organic phase volumes are recorded in the literature, with reported volumes in the range of 39 to 41.5 L (British Nuclear Fuels Limited,



**Fig. 4.** Side elevation schematics of the four main stages of the 1970 Windscale nuclear criticality incident.  
 Source: Adapted from [Evans \(1984a\)](#) and originally published in [Daniels et al. \(2024b\)](#).

no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000). Additionally, the exact properties of the emulsion layer are unknown. Previous work (Chalmers, 1971; Evans, 1984a) uses the estimate that the aqueous and organic phases contributed to the emulsion layer in the same proportions as they would have existed if fully separate. Unsurprisingly, information regarding the size distribution of the globules in the emulsion is not detailed in the available literature.

The length of time the organic phase remained trapped in the vessel is unknown, with estimates of as little as a few months, to over two years, being suggested (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes,

1972; McLaughlin et al., 2000). Although degradation of the organic phase due to alpha irradiation had occurred (British Nuclear Fuels Limited, no date; Evans, 1984a,b; Hughes, 1972), the quantity and structure of degradation products has not been detailed within the available literature. Though fission products could also be present in the system, their quantity and structure are also not detailed in the available literature.

The exact timings of each of the transient's four distinct stages are unknown. During the post-excision testing, the filling stage took an "average of 4 minutes", and the separation stage took between 5 and 10 s (British Nuclear Fuels Limited, no date; Chalmers, 1971; Daniels et al., 1971; Evans, 1984a,b; Hughes, 1972; McLaughlin et al., 2000).

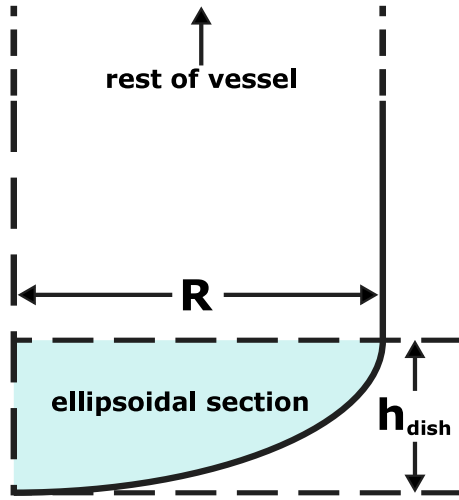


Fig. 5. A side elevation schematic, half-plane view of the lower half of transfer vessel geometry used in the presented research.

### 2.3. Vessel geometry employed in neutron transport codes

As described previously, the vessel schematic reported by Chalmers (1971) and presented in Fig. 3 is assumed to be a to-scale model of the transfer vessel. Previous work by Daniels et al. (2024b) investigated the effect of vessel geometry on results of neutron transport research code EVENT. Two curvilinear geometries were studied: one in which the to-scale geometry of Chalmers (1971) was recreated using splines, and one in which ellipsoids were employed to approximate the top and bottom ‘dished ends’ of the vessel. It was concluded that there was negligible difference between the two geometries. Thus, the ellipsoidal model will be used in the presented research.

Fig. 5 depicts a diagram of the geometry used in the presented research. The dished ends of the vessel are modelled by two hemi-ellipsoids. The full geometry is modelled using the following equations for the inside surface of the vessel wall:

$$1 = \begin{cases} \frac{x^2+y^2}{R^2} + \frac{(z-h_{\text{dish}}-t_{\text{steel}})^2}{h_{\text{dish}}^2} & \text{if } z < (h_{\text{dish}} + t_{\text{steel}}), \\ \frac{x^2+y^2}{R^2} & \text{if } (h_{\text{dish}} + t_{\text{steel}}) \leq z \leq (H_{\text{vess}} - h_{\text{dish}} - t_{\text{steel}}) \\ \frac{x^2+y^2}{R^2} + \frac{(z-H_{\text{vess}}+h_{\text{dish}}+t_{\text{steel}})^2}{h_{\text{dish}}^2} & \text{if } z > (H_{\text{vess}} - h_{\text{dish}} - t_{\text{steel}}). \end{cases} \quad (1)$$

and the following equations for the outside surface of the vessel wall:

$$1 = \begin{cases} \frac{x^2+y^2}{(R+t_{\text{steel}})^2} + \frac{(z-h_{\text{dish}}-t_{\text{steel}})^2}{(h_{\text{dish}}+t_{\text{steel}})^2} & \text{if } z < (h_{\text{dish}} + t_{\text{steel}}), \\ \frac{x^2+y^2}{(R+t_{\text{steel}})^2} & \text{if } (h_{\text{dish}} + t_{\text{steel}}) \leq z \leq (H_{\text{vess}} - h_{\text{dish}} - t_{\text{steel}}) \\ \frac{x^2+y^2}{(R+t_{\text{steel}})^2} + \frac{(z-H_{\text{vess}}+h_{\text{dish}}+t_{\text{steel}})^2}{(h_{\text{dish}}+t_{\text{steel}})^2} & \text{if } z > (H_{\text{vess}} - h_{\text{dish}} - t_{\text{steel}}), \end{cases} \quad (2)$$

where  $t_{\text{steel}}$  is the thickness of the vessel wall,  $R$  is the vessel’s inner radius,  $H_{\text{vess}}$  is the total (outer) height of the vessel, and  $h_{\text{dish}}$  is the width of a dished end (i.e the semi minor axis of the spheroid).

To conserve the 176 L volume of the vessel, and to ensure the height of the vessel (from inner wall to inner wall) is 68.6 cm,  $h_{\text{dish}}$  is calculated to be 12.56535 cm, with  $H_{\text{vess}} = 70.505$  cm,  $R = 30.5$  cm, and  $t_{\text{steel}} = 0.9525$  cm.

### 2.4. Vessel outflow pipe radius

As the organic phase remains trapped in the vessel between vacuum lifts, it will be assumed that the total volume of the outflow pipe connecting the transfer vessel to the constant volume feeder will have a maximum value,  $V_{\text{out,max}}$ , of 50 L, and the pipe was empty at the start of the transient. As depicted in Fig. 2, the vessel outflow pipe has an initial vertical section of length 32 ft ( $h_1$ ), after which it U-bends to another vertical section of 25 ft ( $h_2$ ), before bending at 90° to become horizontal. As the lengths of the U-bend and horizontal section of the pipe are unknown, it will be assumed that their volumes will be negligible.

Using this information, the equality

$$r_{\text{out,max}} \leq \sqrt{\frac{V_{\text{out,max}}}{\pi(h_1 + h_2)}} \quad (3)$$

can be formed, resulting in an upper limit,  $r_{\text{out,max}}$ , to the outflow radius of 3.03 cm.

However, assuming the vessel schematic of Chalmers (1971) is to-scale, it is more likely the outflow pipe radius,  $r_{\text{out}}$  was between 0.5 and 1.0 cm.

## 3. Transient model phenomenology

### 3.1. Point kinetics

Point neutron kinetics equations describe the overall transient behaviour of a fissile system (Lewins, 1978). The derivation of the point neutron kinetics equations is well-documented, for example in Duderstadt and Hamilton (1976) and Stacey (2018). Eqs. (4) and (5) describe the change in neutron population,  $n$ , and delayed neutron precursor population,  $C_i$ , in a three-layered fissile liquid system over time,  $t$ .

$$\begin{aligned} \frac{dn(t)}{dt} = & \frac{\beta_{\text{eff}} n(t)}{\Lambda(t)} \left( \xi_{\text{geom}}(t) + \Delta\xi_{\text{th,aq}}(\bar{T}_{\text{aq}}, t) + \Delta\xi_{\text{th,em}}(\bar{T}_{\text{em}}, t) \right. \\ & + \Delta\xi_{\text{th,or}}(\bar{T}_{\text{or}}, t) \\ & + \Delta\xi_{\text{rg,aq}}(\bar{\theta}_{\text{aq}}, t) + \Delta\xi_{\text{rg,em}}(\bar{\theta}_{\text{em}}, t) + \Delta\xi_{\text{rg,or}}(\bar{\theta}_{\text{or}}, t) - 1 \Big) \\ & + \sum_{i=1}^6 \lambda_i C_i(t) + s_n(t) \end{aligned} \quad (4)$$

$$\frac{dC_i(t)}{dt} = -\lambda_i C_i(t) + \frac{\beta_i}{\Lambda(t)} n(t) \quad (5)$$

In the above equations,  $\beta_{\text{eff}}$ , is the total delayed neutron fraction,  $\beta_i$ , is the delayed neutron fraction for precursor group  $i$ ,  $\lambda_i$  is the decay constant for neutrons in precursor group  $i$  and  $\Lambda$  is the mean prompt neutron generation time.  $s_n$  is the total intrinsic neutron source in the system. The reactivity of the system,  $\xi$ , measured in dollars (\$), has been defined in components. Firstly,  $\xi_{\text{geom}}$  is the geometry-induced reactivity (e.g. from aqueous ingress or emulsion formation).  $\Delta\xi_{\text{th,aq}}$ ,  $\Delta\xi_{\text{th,em}}$ , and  $\Delta\xi_{\text{th,or}}$  represent the thermal reactivity feedbacks due to temperature change in the aqueous, emulsion, and organic layers, respectively:  $\bar{T}_{\text{yy}}$  represents the average temperature in layer yy. Similarly,  $\Delta\xi_{\text{rg,aq}}$ ,  $\Delta\xi_{\text{rg,em}}$ , and  $\Delta\xi_{\text{rg,or}}$  represent the reactivity feedbacks due to the presence of voids in the aqueous, emulsion, and organic layer, respectively:  $\bar{\theta}_{\text{yy}}$  represents the average void fraction in layer yy.

Expressions for the power,  $P$ , and energy,  $E$ , and total number of fissions,  $F$ , are defined in Eq. (6), where  $\epsilon$  is the average energy released per fission event ( $3.318 \times 10^{-11}$  J fission<sup>-1</sup> for <sup>239</sup>Pu Sher and Beck, 1981).  $E_n$  and  $z$  are the neutron energy and axial location, respectively.  $H$  is the total height of the fissile liquid system,  $\Sigma_f$  is the macroscopic neutron fission cross section, and  $v$  is the neutron speed. Finally,  $\hat{\phi}$  is a shape function defining the radially averaged, energy and height dependent neutron distribution.

$$\begin{aligned} \frac{1}{\epsilon} \frac{dE(t)}{dt} = & \frac{1}{\epsilon} P(t) = \frac{dF(t)}{dt} \\ = & n(t) \int_0^\infty dE_n \int_0^H dz \Sigma_f(t, z, E_n) \hat{\phi}(t, z, E_n) v(t, z, E_n) \end{aligned} \quad (6)$$

### 3.1.1. Intrinsic neutron source

Distributed intrinsic and/or localised extrinsic neutron source(s) are often present in fissile systems (Radkowsky, 1964). Intrinsic neutron sources can include spontaneous fissions (SFs),  $(\alpha, n)$  reactions,  $(\gamma, n)$  reactions and cosmic ray spallation (Rossi, 1933), although SF and  $(\alpha, n)$  reactions are often dominant in transient nuclear criticality excursion problems, such that the  $(\gamma, n)$  reactions and cosmic ray spallation can be neglected.

For systems containing plutonium, the relative neutron source density generated from SF and  $(\alpha, n)$  reactions depends upon the relative concentration of the system's plutonium isotopes (Dolan et al., 2014). For example,  $^{240}\text{Pu}$  has a high SF rate compared to other isotopes of plutonium (Marcath et al., 2016). Additionally, for fissile liquid systems, the intrinsic neutron source strength will depend on the concentration and acidity of the solution (Kudryavtsev et al., 2020; Swinhoe, 2011).

In the literature surrounding the 1970 Windscale nuclear criticality incident, there is no mention of any extrinsic neutron sources being present. However, SF and  $(\alpha, n)$  intrinsic neutron sources would have been present. In this and previous (Daniels et al., 2024b) research, the isotopic composition of the plutonium is assumed to be 93.750%  $^{239}\text{Pu}$  and 6.250%  $^{240}\text{Pu}$  by weight, for both the aqueous and organic phase. In actuality, the materials were likely to contain some  $^{238}\text{Pu}$ , a strong  $(\alpha, n)$  source.

In general, to determine the total distributed neutron source across a system domain, spatial, energy, angular and temporal variations of the neutron source must be adequately accounted for, in addition to accounting for neutron importance via adjoint weighting (Spriggs et al., 1999). Currently, the deterministic code SOURCES-4C is the standard when calculating SF and  $(\alpha, n)$  neutron sources (Griesheimer et al., 2017; Wilson et al., 2002). It can calculate  $(\alpha, n)$  and SF source rates and spectra in homogeneous media, two-region interface problems, three-region interface problems, and  $(\alpha, n)$  reactions induced by a monoenergetic beam of  $\alpha$ -particles incident on a slab of target material (Wilson et al., 2002).

### 3.1.2. Initial conditions

Prescribed initial conditions are often derived by assuming the initially subcritical systems are at a steady state prior to the transient nuclear criticality excursion (Kerlin and Upadhyaya, 2019; Lewis, 1977), as in Eqs. (7) and (8):

$$n(0) = \frac{s_n(0)A}{\xi(0)\beta_{\text{eff}}}, \quad (7)$$

$$C_i(0) = \frac{s_n(0)\beta_i}{\lambda_i\xi(0)\beta_{\text{eff}}}. \quad (8)$$

### 3.1.3. Geometry-induced reactivity and the scalar neutron flux distribution

As demonstrated in Daniels et al. (2024a,b), the CP method can be used to perform steady state analysis of systems such as configurations expected to occur during the 1970 Windscale nuclear criticality incident. As such, the CP code developed and verified in Daniels et al. (2024a) will be used to calculate the effective neutron multiplication factor,  $k_{\text{eff}}$ , with the geometry-induced reactivity of the transient 1970 Windscale nuclear criticality incident subsequently being calculated using

$$\xi_{\text{geom}} = \frac{k_{\text{eff}} - 1}{k_{\text{eff}}\beta_{\text{eff}}}. \quad (9)$$

In addition, the CP code calculates the radially averaged scalar neutron flux,  $\phi(z, E_n)$ , such that the shape function  $\hat{\phi}(z, E_n)$  can be calculated using

$$\hat{\phi}(z, E_n) = \frac{\phi(z, E_n)}{\int_0^H dz \int_0^\infty dE_n \phi(z, E_n)}. \quad (10)$$

## 3.2. Fissile liquid dynamics

### 3.2.1. The emulsion

As aqueous solution was poured into the vessel, it penetrated the organic layer, undoubtedly disrupting the surface of the organic layer. Subsequently, the surface of the aqueous and emulsion layers would also have been disrupted. The disruption could have involved ripples on the surface, or splashes or displacement up the vessel's sides. There is no mention of this behaviour in the available literature, and thus it will be assumed that these phenomena can be neglected.

Additionally, as the aqueous solution poured through the organic layer, organic globules would have been entrained in the flow. Thus, it is almost certain that some organic globules would have existed, distributed, in the aqueous layer. Again, there is no mention of this behaviour in the available literature, and thus it will be assumed that this phenomenon can also be neglected. It will also be assumed that the rate of formation of the emulsion layer will be constant.

When the aqueous solution ceased entering the vessel, the emulsion immediately began to separate, due to the density difference of the organic globules and the continuous aqueous phase (British Nuclear Fuels Limited, no date). An investigation into emulsion separation dynamics is presented in Appendix A: in the presented research, emulsion dissipation will be characterised using the Richardson–Zaki equation (Richardson and Zaki, 1954), as shown in Eq. (11):

$$U_{\text{sep}} = U_0 \alpha^n \quad (11)$$

where,  $U_{\text{sep}}$  is the bulk emulsion separation velocity,  $U_0$  is the rise velocity of an isolated (free flowing) emulsion globule, and  $n$  is a suitably chosen constant (Richardson and Zaki, 1954, 1997). The fraction of the emulsion layer that is occupied by the continuous aqueous phase,  $\alpha$ , is defined in Eq. (12).

$$\alpha = \frac{V_{\text{aq}}}{V_{\text{aq}} + V_{\text{or}}} \quad (12)$$

In the presented research,  $\alpha$  will be evaluated with both aqueous and organic phases at 293.6 K.

Rowe (1987) formed an empirical relation for determining  $n$ , as presented in Eq. (13):

$$n = 2.35 \frac{2 + 0.175 Re^{0.75}}{1 + 0.175 Re^{0.75}}, \quad (13)$$

where the Reynolds number,  $Re$ , used by Rowe (1987), is defined as:

$$Re = \frac{2\rho_{\text{aq}}U_0r_{\text{glob}}}{\mu_{\text{aq}}}, \quad (14)$$

where  $\rho$  and  $\mu$  are the density and dynamic viscosity, respectively, and  $r_{\text{glob}}$  is the radius of an emulsion globule. Subscripts 'aq' and 'or' refer to the aqueous and organic phases, respectively.

Using the assumptions and approximations described above, the following Equation can be formed:

$$\frac{dM_{\text{em}}(t)}{dt} = \begin{cases} \frac{M_{\text{em,max}}}{t_{\text{form}}} & \text{if } t \leq t_{\text{form}}, \\ 0 & \text{if } t_{\text{form}} < t \leq t_{\text{fill}}, \\ U_{\text{sep}}A_{\text{em}}(t, H_{\text{em}})\frac{1}{V_{\text{em}}(t)} \int_V dV \rho_{\text{em}}(t, z) & \text{if } t > t_{\text{fill}} \text{ and } M_{\text{em}}(t) > 0, \\ 0 & \text{otherwise,} \end{cases} \quad (15)$$

where  $t_{\text{form}}$  is the time taken for the emulsion to reach its maximum volume from its starting volume (0.0 L in this case),  $t_{\text{fill}}$  is the time taken for the aqueous material to be fully vacuum lifted into the vessel.  $M_{\text{em,max}}$  is the maximum mass of the emulsion layer, and  $A_{\text{em}}$  is the surface area of the emulsion's top surface.

### 3.2.2. The organic phase

The dynamics of the organic phase are relatively simple: as this phase remains hydraulically trapped within the vessel, its mass flow rate depends only on the rate of emulsion formation or dissipation:

$$\frac{dM_{or}(t)}{dt} = -(1 - \omega) \frac{dM_{em}(t)}{dt}. \quad (16)$$

Here,  $\omega$ , the fraction of the emulsion that is aqueous by mass, has been defined.

### 3.2.3. The aqueous phase

It will be assumed that the mass flow rate of aqueous solution into the vessel is constant:

$$\frac{dM_{aq,in}(t)}{dt} = \frac{M_{aq,max}}{t_{fill}} \quad \text{if } t \leq t_{fill} \quad (17)$$

where  $M_{aq,max}$  is the volume of the aqueous material that is vacuum lifted into the vessel. In addition, the aqueous phase contributes to the emulsion layer, a phenomenon which will be modelled using Eq. (18):

$$\frac{dM_{aq \rightarrow em}(t)}{dt} = -\omega \frac{dM_{em}(t)}{dt}. \quad (18)$$

To determine the drainage rate of aqueous liquid from the vessel, Bernoulli's equation (Eq. (20)), along with the continuity equation (Eq. (19)), can be used. The two reference points chosen are the height of the surface of the organic layer ( $z = H_{or}$ ), and the height of the drainage orifice ( $z = 0$ ). It is assumed that the pressure at both reference points are equal, and the flow in all liquids is incompressible.

$$U(t, 0)A(t, 0)\rho(t, 0) = U(t, H)A(t, H)\rho(t, H) \quad (19)$$

$$\frac{1}{2}\rho(t, H)U^2(t, H) + g \int_0^H dz \rho(t, z) = \frac{1}{2}\rho(t, 0)U^2(t, 0) \quad (20)$$

By substituting Eq. (19) into Eq. (20) and rearranging, the following equation for aqueous drainage rate is obtained:

$$\frac{dM_{aq,out}(t)}{dt} = - \left[ \frac{2g\rho(t, H_{or})\rho(t, 0)}{\rho(t, H_{or})A^{-2}(t, 0) - \rho(t, 0)A^{-2}(t, H_{or})} \int_0^{H_{or}} dz \rho(t, z) \right]^{0.5} \quad (21)$$

### 3.3. Thermal hydraulics

In the presented work, a simple thermal hydraulics model has been employed, in which thermal equilibrium between the emulsion globules and continuous phase is assumed. Eqs. (22) and (23) govern the time-dependent thermal energy distribution in the fissile liquid and the vessel.

$$\begin{aligned} \frac{\partial E_{th}'''(t, z)}{\partial t} = & \frac{\partial}{\partial V} \left\{ P(t)\hat{\phi}(t, z) - [T(t, z) - T_{vess}(t, z)]h_{int}(t, z)A_{int}(t, z) \right. \\ & \left. + \sum_x \frac{dM_x(t, z)}{dt} C_{p,x}(t, z)T_x(t, z) \right\} \\ & + \nabla \cdot [D_{FS}(t, z)\nabla E_{th}'''(t, z)] \end{aligned} \quad (22)$$

$$\begin{aligned} \frac{\partial E_{vess}(t, z)}{\partial t} = & [T(t, z) - T_{vess}(t, z)]h_{int}(t, z)A_{int}(t, z) \\ & - [T_{vess}(t, z) - T_{ext}]h_{ext}A_{ext}(t, z) \end{aligned} \quad (23)$$

In these equations, subscripts 'vess' and 'ext' refer to the vessel and its surroundings, respectively. As such, Eq. (22) is used to determine the thermal energy of the fissile liquids, and (23) describes the thermal energy of the vessel.  $E_{th}'''$  represents the thermal energy per unit volume of fissile liquid.

The first term on the right-hand side of Eq. (22) represents the rate of production of thermal energy due to fission. The second term represents heat convection between the liquids and the vessel:  $h_{int}$  and

$A_{int}$  represent the heat transfer coefficient and contact area between the liquid and the vessel, respectively. Similarly, the final term in Eq. (23) represents heat convection between the vessel and its surroundings:  $h_{ext}$  and  $A_{ext}$  represent the heat transfer coefficient and contact area between the vessel and the environment, respectively. It is assumed that the surroundings remain at constant temperature  $T_{ext}$ .

The summation term in Eq. (22) represents the rate of energy addition (or subtraction) due to the movement of mass around the vessel, where  $x$  is each component of mass movement. Mass moves around the vessel due to aqueous ingress and drainage, in addition to emulsion formation and separation, as discussed in Section 3.2. Here,  $C_{p,x}$  is the specific heat capacity of the moving mass.

Finally, the last term in Eq. (22) represents the rate of energy change due to the effect of turbulent mixing: here,  $D_{FS}$  is the mixing coefficient. The vacuum lifting process was likely to cause a large degree of turbulence due to the force of the aqueous solution falling into, and through, the organic layer — this is the mechanism which created the emulsion layer. However, once the vacuum lifting was complete, and the aqueous layer started to drain, it could be that there was a decrease in turbulent mixing. As such, the turbulent mixing coefficient is assumed to vary with both time and location.

Assuming that the three fissile liquid layers (aqueous, emulsion, organic) are immiscible and no turbulent mixing occurs between them, the following interface conditions can be defined in order to conserve thermal energy:

$$\begin{aligned} D_{FS,y}(t, H_x) \frac{\partial E_{th,y}'''(t, H_x)}{\partial z} - D_{FS,x}(t, H_x) \frac{\partial E_{th,x}'''(t, H_x)}{\partial z} \\ = [T_y(t, H_x) - T_x(t, H_x)]h_{sur}(t, H_x) \end{aligned} \quad (24)$$

where  $x$  is the index of the lower region, and  $y$  is the index of the upper region.  $h_{sur}$  and  $A_{sur}$  represent the heat transfer coefficient and contact area between the two regions, respectively.

Similarly, at the top of the fissile liquid system ( $z = H_{total} = H_{or}$ ), the following boundary condition, to account for loss of thermal energy to the plenum gas, is prescribed:

$$\frac{\partial E_{th}'''(t, H_{or})}{\partial z} = [T(t, H_{or}) - T_{ext}]h_{sur}(t, H_{or}). \quad (25)$$

Finally, at the base of the vessel:

$$\frac{\partial E_{th}'''(t, 0)}{\partial z} = 0. \quad (26)$$

### 3.4. Radiolytic gas production and motion

#### 3.4.1. Radiolysis

The fission of a  $^{239}\text{Pu}$  atom usually results in the emission of two charged fission fragments (Gönnenwein, 2004; Perfilov et al., 1963) and of an average of 2.9 neutrons (Badikov et al., 2007), alongside the release of gamma ray photons (Sher and Beck, 1981).

Radiolysis is the process by which ionising radiation dissociates molecules. As the ionising radiation travels through a material, tracks of radiolysis products are left in its wake, the quantities and proportions of which depend on the linear energy transfer (LET) of the ionising radiation (Lane et al., 1958). The LET is the average radiation energy deposited per unit length along the track of the ionising particle (Seltzer, 2011). In radiation chemistry, the  $G$  value refers to the number of molecules of product formed (or reactant consumed) per 100 eV of energy absorbed (Baird et al., 1990). The theory and processes of radiolysis and radiolytic gas formation are described in, for example, Baird et al. (1990), Henry and Banks (1803), Lane et al. (1958) and Spiegler et al. (1962).

A solution is capable of containing a certain amount of dissolved gas up to a critical concentration, which is dictated by Henry's law (Henry and Banks, 1803). The evaluation of Henry's law is considered outside the scope of the presented research. Thus, it will be assumed that, at the start of the transient, radiolytic gases exist in both aqueous and

organic liquids at the critical concentration, and there will be no delay to radiolytic gas production.

The radiolytic gas within bubbles (identified by subscript 'b') obeys the ideal gas law, as per Eq. (27):

$$\frac{V_b}{N_b} = \frac{RT_b}{p_b} \quad (27)$$

where  $V_b$ ,  $T_b$  and  $N_b$  are the volume, temperature, and number of moles of gas within the bubble, respectively;  $p_b$  is the pressure within the bubble; and  $R$  is the universal gas constant, equal to  $8.31 \text{ J mol}^{-1} \text{ K}^{-1}$ . As such, an expression for the radiolytic gas generation coefficient ( $G_{\text{rg}}$ , measured in  $\text{m}^3 \text{ J}^{-1}$ ) can be formed:

$$G_{\text{rg}} = \frac{1}{1.602 \times 10^{-17}} \frac{GRT_b}{N_A p_b} \quad (28)$$

where  $G$  is the total number molecules of radiolytic gas produced per 100 eV of deposited fission energy,  $N_A$  is the Avogadro constant, and other symbols are as previously defined.

Finally, the rate of radiolytic production can be calculated using Eq. (29):

$$\frac{\partial V_{\text{gen}}'''(t, z)}{\partial t} = \frac{\partial}{\partial V} \left[ G_{\text{rg}}(t, z) P(t) \hat{\phi}(t, z) \right] \quad (29)$$

where  $V_{\text{gen}}'''$  is the volume of radiolytic gas formed per volume of fissile liquid. The product  $P(t) \hat{\phi}(t, z)$  is the fission power per at time  $t$  and location  $z$ , with  $\hat{\phi}(t, z)$  defined in Section 3.1.3.

### 3.4.2. Radiolytic gas advection

Bubbles of radiolytic gas will be advected through a fissile liquid system due to the effects of buoyancy. The speed at which bubbles advect is dependent on their radius, which is governed by properties of the bulk liquid. The review and research by [Beenackers and Van Swaaij \(1993\)](#) resulted in the following correlation:

$$a = a_0 \left( \frac{\mu_0}{\mu_{\text{eff}}} \right)^{0.21} \left( \frac{\rho_{\text{eff}}}{\rho_0} \right)^{0.1} \quad (30)$$

which describes the interfacial area concentration,  $a$  (units  $\text{m}^{-1}$ ), of gas in a gas-liquid slurry reactor, with particle sizes in the range 100–1000  $\mu\text{m}$ . In Eq. (30), subscript '0' refers to properties of the liquid that would exist if no solids were present and subscript 'eff' refers to the effective properties of the fluidised system. Due to the similarities between suspensions of solid particles and suspensions of liquid particles (emulsions), Eq. (30) will be assumed valid when describing the interfacial area in a three-phase liquid-liquid-gas (emulsion) system.

[Kundu et al. \(2015\)](#) reports that the viscosity of an emulsion tends to increase with increased volume fraction of the dispersed phase, due to non-Newtonian behaviour being induced in the emulsion due to crowding of emulsion globules. The effective viscosity of an emulsion has been widely investigated: a comprehensive review is given by [Kundu et al. \(2015\)](#). In the presented research, the Krieger-Dougherty equation ([Krieger and Dougherty, 1959](#)), presented in Eq. (31), has been selected to determine  $\mu_{\text{eff}}$ :

$$\mu_{\text{eff}} = \mu_0 \left( 1 - \frac{\phi}{\phi_m} \right)^{-[\mu]\phi_m} \quad (31)$$

In this equation,  $\mu_0$  is the viscosity of the continuous phase,  $\phi$  is the volume fraction of the dispersed phase,  $\phi_m$  is the maximum packing fraction of particles in the dispersed phase, and  $[\mu]$  is the intrinsic viscosity.

[De La Rosa et al. \(2021\)](#) reports that the intrinsic viscosity depends on the ratio of the viscosities of the dispersed and continuous phase, in addition to the volume fraction of the dispersed phase. For polydispersed systems, a value of  $[\mu]$  equal to 2.5 is reported to be suitable ([De La Rosa et al., 2021](#); [Pal, 1998](#)). For polydispersed systems, it is common for  $\phi_m$  to exceed 0.7 ([De La Rosa et al., 2021](#)). In addition, for emulsions (as opposed to suspensions of solid particles), the globules in the dispersed phase can deform, leading to higher values for  $\phi_m$

([De La Rosa et al., 2021](#); [Frising et al., 2008](#); [Kundu et al., 2015](#); [Pal et al., 1986](#); [Pal, 1996, 1998](#)). [Pal \(1998\)](#) performed experiments in which both 'fine' and 'coarse' droplets were present in an oil-in-water emulsion. [Pal \(1998\)](#) reported that  $\phi_m$  for an emulsion of this kind will depend on the ratio of Sauter mean diameters of the fine and coarse emulsion components, as well as the volume ratio of fine to coarse droplets. In the research presented by [Pal \(1998\)](#),  $\phi_m$  values of up to 0.875 were observed (for a Sauter mean diameter ratio of 0.138 and a volume fraction of fine emulsion of around 0.1) ([Pal, 1998](#)). In the presented research,  $\phi_m$  equal to 0.8 has been assumed. As such, Eq. (32) describes the effective viscosity of the emulsion present in the 1970 Windscale nuclear criticality incident:

$$\mu_{\text{eff}} = 1.25 \mu_{\text{aq}} (\alpha - 0.2)^{-2}. \quad (32)$$

Eq. (33) is used to determine the emulsion's effective density,  $\rho_{\text{eff}}$ :

$$\rho_{\text{eff}} = \alpha \rho_{\text{aq}} + (1 - \alpha) \rho_{\text{or}} \quad (33)$$

In the aqueous and organic layers, any radiolytic gas production will induce two-phase (gas-liquid) systems to form. Assuming that the produced radiolytic gas bubbles travel in the bubbly flow regime, Eq. (34) ([Zeitoun et al., 1994](#)) can be used to determine the interfacial area concentration for two phase flow:

$$a_0 = 556.4 v_g^{0.74} \quad (34)$$

where  $v_g$  is the void fraction of bubbles. The Sauter mean diameter,  $d_b$ , of gas bubbles can be defined using Eq. (35) ([Beenackers and Van Swaaij, 1993](#)).

$$d_b = \frac{6 v_g}{a} \quad (35)$$

Substitution of Eqs. (30) and (34) into Eq. (35) yields an expression for bubble diameter as a function of void fraction, as in Eq. (36) ([Jones et al., 2022](#)).

$$d_b = 1.0784 \times 10^{-2} v_g^{0.26} \left( \frac{\mu_{\text{aq}}}{\mu_{\text{eff}}} \right)^{-0.21} \left( \frac{\rho_{\text{eff}}}{\rho_{\text{aq}}} \right)^{-0.1} \quad (36)$$

With the bubble diameter determined, an expression can be found to calculate the advection velocity of bubbles in two-phase flow. [Fan and Tsuchiya \(1990\)](#) reported that Eq. (37) adequately describes the dimensionless bubble velocity,  $U'_b$ , for a wide range of bubble sizes in two phase flow. Later research by [Tsuchiya et al. \(1997\)](#) determined that, under certain conditions and solid volume fractions ( $0.430 \leq (1 - \alpha) \leq 0.526$ ), Eq. (37) was also applicable for three-phase flow in fluidised beds. Eq. (37) was shown to be the most accurate when  $d_b > 30d_p$ , where  $d_p$  is the particle diameter. For  $d_b \leq 30d_p$ , Eq. (37) underestimated the bubble advection velocity by around 10% for 0.430  $\leq (1 - \alpha) \leq 0.451$ . Due to the similarities between emulsion layers and a three-phase fluidised bed, Eq. (37) will be deemed acceptable for all flow conditions present in the 1970 Windscale nuclear criticality incident.

$$U'_b = \left[ \left( \frac{\text{Mo}^{0.25} d_b'^2}{K_b} \right)^{-n} + \left( \frac{2c}{d_b'} + \frac{d_b'}{2} \right)^{-n/2} \right]^{-1/n} \quad (37)$$

In the above,  $n$  and  $c$  are empirically determined parameters, dependent on the bulk liquid and its properties ([Fan and Tsuchiya, 1990](#); [Jones et al., 2022](#)). [Fan and Tsuchiya \(1990\)](#) recommends setting  $c$  to 1.2 for a mono-component liquid, or to 1.4 for a multi-component liquid. Additionally,  $n$  will depend on the purity of the liquid, with  $n$  equal to 0.8 for a pure liquid, and  $n$  equal to 1.6 for a highly contaminated liquid ([Fan and Tsuchiya, 1990](#); [Jones et al., 2022](#)). The Morton number,  $\text{Mo}$ , is defined in Eq. (38), and the dimensionless bubble diameter,  $d_b'$  is defined in Eq. (39).

$$\text{Mo} = \frac{g \mu_{\text{eff}}^4}{\rho_{\text{eff}} \sigma^3} \quad (38)$$

$$d'_b = d_b \left( \frac{\rho_{\text{eff}} g}{\sigma} \right)^{0.5} \quad (39)$$

$K_b$ , another empirically determined parameter, can be calculated using Eq. (40), where  $K_{b,0}$  can be set to 14.7 for aqueous solutions, or to 10.2 for organic mixtures (Fan and Tsuchiya, 1990).

$$K_b = \max(K_{b,0} \text{Mo}^{-0.038}, 12) \quad (40)$$

Finally, the actual bubble velocity can be determined from its dimensionless parameter by using Eq. (41) (Fan and Tsuchiya, 1990; Jones et al., 2022):

$$U_b = U'_b \left( \frac{\sigma g}{\rho_{\text{eff}}} \right)^{0.25} \quad (41)$$

The advection of radiolytic gas through the system can now be described using Eq. (42):

$$\frac{\partial V_{\text{adv}}'''(t, z)}{\partial t} = -U_b(t, z) \frac{\partial V_{\text{rg}}'''(t, z)}{\partial z} \quad (42)$$

where  $V_{\text{adv}}'''$  is the volume of radiolytic gas advected per volume of fissile liquid.

#### 3.4.3. Complete radiolytic gas equation, boundary conditions and interface conditions

Using the previously-defined variables and expressions, Eq. (43) can be used to calculate the space- and time-dependent behaviour of radiolytic gas voids in a layered fissile liquid system:

$$\begin{aligned} \frac{\partial V_{\text{rg}}'''(t, z)}{\partial t} = & \frac{\partial}{\partial V} \left[ G_{\text{rg}}(t, z) P(t) \hat{\phi}(t, z) \right] - U_b(t, z) \frac{\partial V_{\text{rg}}'''(t, z)}{\partial z} \\ & + \nabla \cdot \left[ D_{\text{FS}}(t, z) \nabla V_{\text{rg}}'''(t, z) \right] \end{aligned} \quad (43)$$

where  $V_{\text{rg}}'''$  is the volume of radiolytic gas per volume of fissile liquid. The first term on the right-hand side governs radiolytic gas production due to radiolysis, the second term on the right-hand side determines the rate at which radiolytic gas advects upwards in the system, and the final term represents the rate of change of radiolytic gas distribution due to turbulent mixing.

With the previously-made assumption that the three fissile liquid layers (aqueous, emulsion, organic) are immiscible and no turbulent mixing occurs between them, the following interface conditions can be defined in order to account for the advection of radiolytic gas between layers:

$$\begin{aligned} U_{b,y}(t, H_x) V_{\text{rg},y}'''(t, H_x) - U_{b,x}(t, H_x) V_{\text{rg},x}'''(t, H_x) \\ = D_{\text{FS},y}(t, H_x) \frac{\partial V_{\text{rg},y}'''(t, H_x)}{\partial z} - D_{\text{FS},x}(t, H_x) \frac{\partial V_{\text{rg},x}'''(t, H_x)}{\partial z} \end{aligned} \quad (44)$$

where  $x$  is the index of the lower region, and  $y$  is the index of the upper region.

As upwind differencing is employed in the numerical discretisation scheme in the presented research, no boundary condition is needed on the top surface of the fissile system. In addition, no gas can travel between the fissile liquids and the vessel, resulting in the following boundary condition at the base of the vessel:

$$\frac{\partial V_{\text{rg}}'''(t, 0)}{\partial z} = 0. \quad (45)$$

#### 4. Summary of equations, interface conditions and boundary conditions

In this section, a summary of the ODEs and PDEs solved by the proposed model are presented. The solution uses a Lagrangian finite volume method to axially discretise the system. The time discretisation algorithm described by Shampine and Gordon (1975) is employed, which uses a modified divided difference form of the Adams PECE (predict-evaluate-correct-evaluate) formulas (Bose, 2019; Jeffreys and

Jeffreys, 2000; LeVeque, 2007; Shampine and Gordon, 1975). It employs local extrapolation to improve stability and accuracy, with the local error per unit step controlled by adjusting the step size (Shampine and Gordon, 1975).

##### 4.1. Point kinetics equations

$$\begin{aligned} \frac{dn(t)}{dt} = & \frac{\beta_{\text{eff}} n(t)}{\Lambda(t)} \left( \xi_{\text{geom}}(t) + \Delta \xi_{\text{th,aq}}(\bar{T}_{\text{aq}}, t) + \Delta \xi_{\text{th,em}}(\bar{T}_{\text{em}}, t) \right. \\ & + \Delta \xi_{\text{th,or}}(\bar{T}_{\text{or}}, t) \\ & + \Delta \xi_{\text{rg,aq}}(\bar{\theta}_{\text{aq}}, t) + \Delta \xi_{\text{rg,em}}(\bar{\theta}_{\text{em}}, t) + \Delta \xi_{\text{rg,or}}(\bar{\theta}_{\text{or}}, t) - 1 \Big) \\ & + \sum_{i=1}^6 \lambda_i C_i(t) + s_n(t) \end{aligned} \quad (46)$$

$$\frac{dC_i(t)}{dt} = -\lambda_i C_i(t) + \frac{\beta_i}{\Lambda(t)} n(t) \quad (47)$$

$$\begin{aligned} \frac{1}{\epsilon} \frac{dE(t)}{dt} = & \frac{1}{\epsilon} P(t) = \frac{dF(t)}{dt} \\ = & n(t) \int_0^\infty dE_n \int_0^{H(t)} dz \Sigma_f(t, z, E_n) \hat{\phi}(t, z, E_n) v(t, z, E_n) \end{aligned} \quad (48)$$

##### 4.2. Layer mass flow rates

$$\frac{dM_{\text{aq}}(t)}{dt} = \dot{M}_{\text{ingress}}(t) - \omega \frac{dM_{\text{em}}(t)}{dt}, \quad (49)$$

$$\frac{dM_{\text{em}}(t)}{dt} = \dot{M}_{\text{form}}(t), \quad (50)$$

$$\frac{dM_{\text{or}}(t)}{dt} = (\omega - 1) \frac{dM_{\text{em}}(t)}{dt}, \quad (51)$$

where

$$\dot{M}_{\text{form}}(t) = \begin{cases} \frac{M_{\text{em,max}}}{t_{\text{form}}} & \text{if } t \leq t_{\text{form}} \\ 0 & \text{if } t_{\text{form}} < t \leq t_{\text{fill}} \\ -U_{\text{sep}} A_{\text{em}}(t, H_{\text{em}}) \frac{1}{V_{\text{em}}(t)} \int_V dV \rho_{\text{em}}(t, z) & \text{if } t > t_{\text{fill}} \text{ and } M_{\text{em}}(t) > 0 \\ 0 & \text{otherwise} \end{cases} \quad (52)$$

and

$$\dot{M}_{\text{ingress}}(t) = \begin{cases} \frac{M_{\text{aq,max}}}{t_{\text{fill}}} & \text{if } t \leq t_{\text{fill}} \\ - \left[ \frac{2 g \rho(t, H_{\text{or}}) \rho(t, 0)}{\rho(t, H_{\text{or}}) A^{-2}(t, 0) - \rho(t, 0) A^{-2}(t, H_{\text{or}})} \int_0^{H_{\text{or}}} dz \rho(t, z) \right]^{0.5} & \text{if } t > t_{\text{fill}} \text{ and } M_{\text{aq}}(t) > 0 \\ 0 & \text{otherwise} \end{cases} \quad (53)$$

##### 4.3. Thermal hydraulics and voidage

$$\begin{aligned} \frac{\partial E_{\text{th}}'''(t, z)}{\partial t} = & \frac{\partial}{\partial V} \left\{ P(t) \hat{\phi}(t, z) - [T(t, z) - T_{\text{vess}}(t, z)] h_{\text{int}}(t, z) A_{\text{int}}(t, z) \right. \\ & + \sum_x \frac{dM_x(t, z)}{dt} C_{p,x}(t, z) T_x(t, z) \Big\} \\ & + \nabla \cdot \left[ D_{\text{FS}}(t, z) \nabla E_{\text{th}}'''(t, z) \right] \end{aligned} \quad (54)$$

$$\begin{aligned} \frac{\partial E_{\text{vess}}(t, z)}{\partial t} = & [T(t, z) - T_{\text{vess}}(t, z)] h_{\text{int}}(t, z) A_{\text{int}}(t, z) \\ & - [T_{\text{vess}}(t, z) - T_{\text{ext}}] h_{\text{ext}} A_{\text{ext}}(t, z) \end{aligned} \quad (55)$$

$$\begin{aligned} \frac{\partial V_{\text{rg}}'''(t, z)}{\partial t} = & \frac{\partial}{\partial V} \left[ G_{\text{rg}}(t, z) P(t) \hat{\phi}(t, z) \right] - U_b(t, z) \frac{\partial V_{\text{rg}}'''(t, z)}{\partial z} \\ & + \nabla \cdot \left[ D_{\text{FS}}(t, z) \nabla V_{\text{rg}}'''(t, z) \right] \end{aligned} \quad (56)$$

#### 4.4. Interface conditions

The following interface conditions ensure continuity of energy and account for heat and voidage transfer between two adjacent fissile liquid layers. In these Equations,  $x$  refers to the lower of the two layers, and  $y$  refers to the upper of the two layers.

$$D_{FS,y}(t, H_x) \frac{\partial E'''_{\text{therm},y}(t, H_x)}{\partial z} - D_{FS,x}(t, H_x) \frac{\partial E'''_{\text{therm},x}(t, H_x)}{\partial z} = [T_y(t, H_x) - T_x(t, H_x)] h_{\text{sur}}(t, H_x) \quad (57)$$

$$U_{b,y}(t, H_x) V'''_{\text{rg},y}(t, H_x) - U_{b,x}(t, H_x) V'''_{\text{rg},x}(t, H_x) = D_{FS,y}(t, H_x) \frac{\partial V'''_{\text{rg},y}(t, H_x)}{\partial z} - D_{FS,x}(t, H_x) \frac{\partial V'''_{\text{rg},x}(t, H_x)}{\partial z} \quad (58)$$

#### 4.5. Boundary conditions

The following equations account for heat loss from the surface of the fissile liquid system to the vessel's plenum gas, and ensure no energy or voidage is lost through the bottom of the fissile liquid system.

$$\frac{\partial E'''_{\text{th}}(t, H_{\text{or}})}{\partial z} = [T(t, H_{\text{or}}) - T_{\text{ext}}] h_{\text{sur}}(t, H_{\text{or}}). \quad (59)$$

$$\frac{\partial E'''_{\text{th}}(t, 0)}{\partial z} = 0. \quad (60)$$

$$\frac{\partial V'''_{\text{rg}}(t, 0)}{\partial z} = 0. \quad (61)$$

#### 4.6. Scalar neutron flux distribution and geometry-induced reactivity

The radially averaged scalar neutron flux is calculated using the CP code of Daniels et al. (2024a) is normalised using

$$\hat{\phi}(z, E_n) = \frac{\phi(z, E_n)}{\int_0^H dz \int_0^\infty dE_n \phi(z, E_n)}, \quad (62)$$

and the geometry-induced reactivity is calculated using

$$\xi_{\text{geom}} = \frac{k_{\text{eff}} - 1}{k_{\text{eff}} \beta_{\text{eff}}}. \quad (63)$$

In the presented research, a 30 group energy discretisation, presented in Table B.17, was used to solve the CP equations.

#### 4.7. Initial conditions

The following equations describe the initial conditions of the fissile liquid system. It has been assumed that the system begins in thermodynamic equilibrium: a constant temperature of  $T_{\text{ext}} = 293.6$  K is applied throughout the system. It is also assumed that no radiolytic gas is present at the start of the transient.

$$n(0) = \frac{s_n(0) \Lambda}{\xi(0) \beta_{\text{eff}}} \quad (64)$$

$$C_i(0) = \frac{s_n(0) \beta_i}{\lambda_i \xi(0) \beta_{\text{eff}}} \quad (65)$$

$$E(0) = F(0) = 0 \quad (66)$$

$$M_{\text{aq}}(0) = M_{\text{em}}(0) = 0 \quad (67)$$

$$M_{\text{or}}(0) = \rho_{\text{or}, 293.6 \text{ K}} V_{\text{or}}(0) \quad (68)$$

$$E'''_{\text{th}}(0, z) = \rho(0, z) C_p(0, z) T_{\text{ext}} \quad (69)$$

$$E'''_{\text{vess}}(0, z) = \rho_{\text{vess}} C_{p,\text{vess}} T_{\text{ext}} \quad (70)$$

$$V'''_{\text{rg}}(0, z) = 0 \quad (71)$$

#### 4.8. Spatial and temporal discretisation and software workflow

Fig. 6 describes the proposed workflow of the numerical algorithm and model. At the start of the simulation, the variable  $st$  is entered as an input. This variable represents the type of solve required: if  $st = \text{'CP'}$ , the algorithm will calculate  $k_{\text{eff}}$  (once per value of time index  $j$ ), and only solve the time- and space-dependent mass equations. If  $st = \text{'PK'}$ , the point neutron kinetics, spatial thermal hydraulics, and spatial radiolytic gas equations will be solved in time, in addition to the mass equations. Finally, if  $st = \text{'full'}$ , the algorithm will first solve the system as if  $st = \text{'CP'}$ , and then re-run by updating  $st$  to  $\text{'PK'}$ .

Other inputs included in Fig. 6 include  $t_{\text{end}}$ , the simulation end time, from which the array of times  $(t_1, \dots, t_j, \dots, t_I)$  are calculated using a user-specified time step. Different time steps can be specified for  $\text{'CP'}$ - or  $\text{'PK'}$ -specific solves.

### 5. Material properties of aqueous, emulsion and organic phases

Despite their use in the PUREX process, there is limited literature surveying the material properties of aqueous plutonium nitrate and plutonium-containing TBP in kerosene. As such, many approximations and assumptions have been made.

#### 5.1. Thermophysical properties

##### 5.1.1. Density

Chalmers (1971) gives the densities of the aqueous and organic phases ( $\rho_{\text{aq}}$  and  $\rho_{\text{or}}$ ) to be 1276 kg m<sup>-3</sup> and 955 kg m<sup>-3</sup> respectively. It will be assumed these densities were measured at a temperature of 293.6 K.

Sakurai and Tachimori (1996) determined a correlation to describe the density of an aqueous solution containing tetravalent plutonium, hexavalent uranium, and nitric acid over a range of temperatures. To use this correlation to describe the density of the plutonium nitrate present in the 1970 Windscale criticality incident (i.e., to ensure the 7 g L<sup>-1</sup> plutonium nitrate has a density of 1.276 g cm<sup>-3</sup> at 293.6 K), the result of this correlation needs to be modified by a factor of around 1.04. Accounting for this factor, and omitting terms containing uranyl nitrate, Eq. (72) can be used to describe the temperature dependence of  $\rho_{\text{aq}}$  (in kg m<sup>-3</sup>):

$$\begin{aligned} \rho_{\text{aq}} = & 1.0342 \times 10^3 + 1.7846 c_{\text{Pu}} + 4.0667 \times 10^1 c_{\text{HN}} - 1.0150 \times 10^{-4} c_{\text{Pu}}^2 \\ & - 7.6680 \times 10^{-1} c_{\text{HN}}^2 \\ & - 5.7794 \times 10^{-3} (T - 273.15)^2 - 5.0793 \times 10^{-2} c_{\text{Pu}} c_{\text{HN}} \\ & - 1.4865 \times 10^{-3} c_{\text{Pu}} (T - 273.15) \\ & - 8.8482 \times 10^{-2} c_{\text{HN}} (T - 273.15) \quad \text{for } 283.15 < T < 333.15 \end{aligned} \quad (72)$$

where  $T$  is the temperature in Kelvin,  $c_{\text{Pu}}$  is the plutonium concentration (in g L<sup>-1</sup>) and  $c_{\text{HN}}$  is the concentration of nitric acid (in mol L<sup>-1</sup>).

Zheng et al. (2010) investigated the temperature dependence of the density of TBP, kerosene and phosphoric acid mixtures. It was seen that, when the mixture contained only TBP and kerosene, the density of the mixture decreased linearly with temperature. Assuming the same relation is observed for a mixture of TBP, kerosene, plutonium nitrate and small quantities of nitric acid, Eq. (73), based on the results of Zheng et al. (2010), can be used to describe the density of the organic phase present in the 1970 Windscale nuclear criticality incident. Here,  $T$  is measured in Kelvin, and  $\rho_{\text{or}}$  measured in kg m<sup>-3</sup>.

$$\rho_{\text{or}} = -8.526667 \times 10^{-1} T + 1.205343 \times 10^3 \quad \text{for } 293.15 < T < 333.15 \quad (73)$$

The temperature dependent emulsion density,  $\rho_{\text{em}}$ , will be calculated using Eq. (33).

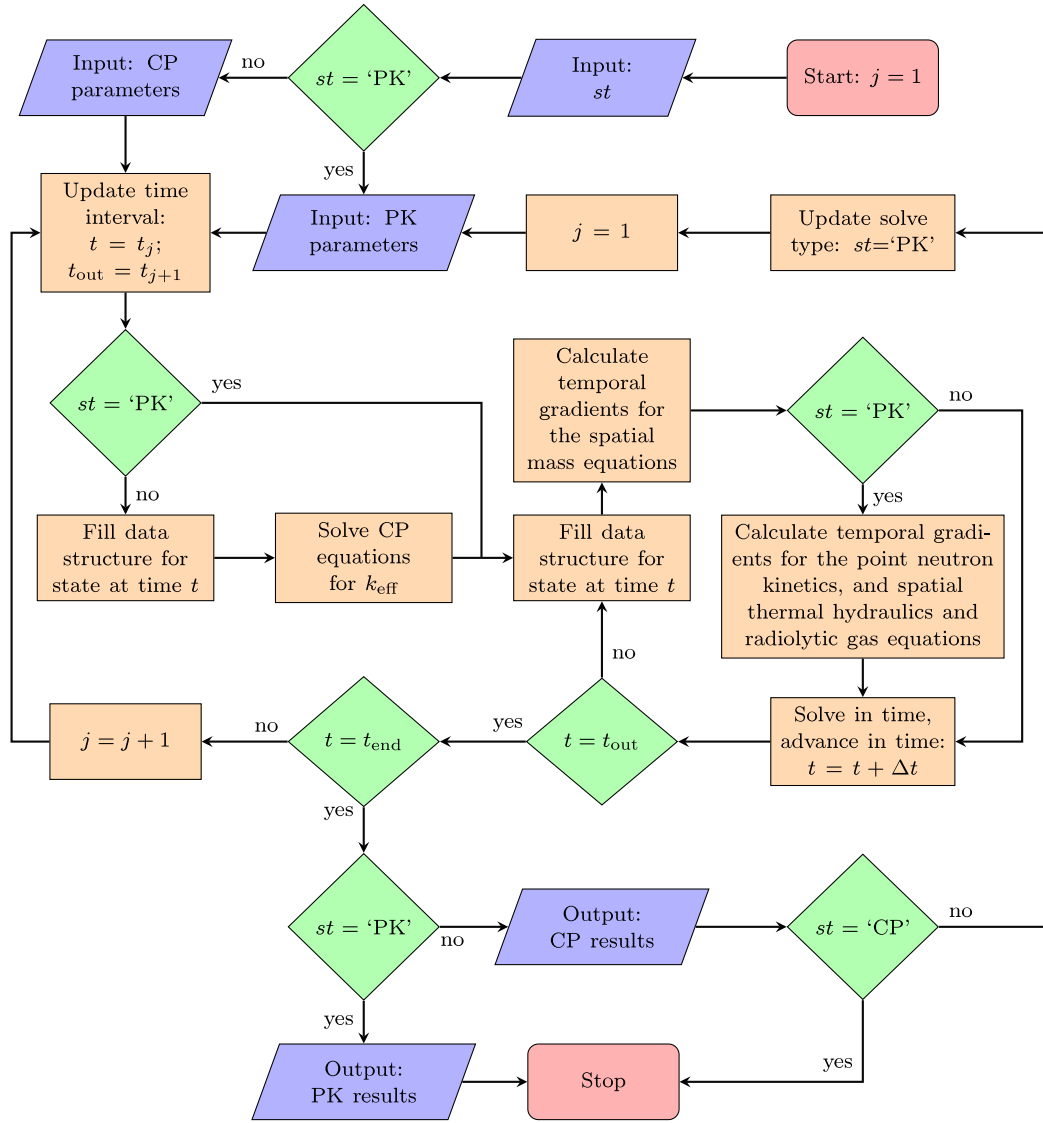


Fig. 6. Flowchart demonstrating the workflow of the proposed numerical algorithm and model.

### 5.1.2. Dynamic viscosity

Eqs. (74) and (75) describe the viscosities of water and nitric acid ( $\mu_{\text{H}_2\text{O}}$  and  $\mu_{\text{HNO}_3}$  respectively), where viscosity is given in mPa s, and temperature,  $T$ , in Kelvin.

$$\log_{10}(\mu_{\text{H}_2\text{O}}) = -1.16225 \times 10^1 + 1.94896 \times 10^3 T^{-1} + 2.16414 \times 10^{-2} T - 1.59901 \times 10^{-5} T^2 \quad \text{for } 273.15 < T < 646.15 \quad (74)$$

$$\log_{10}(\mu_{\text{HNO}_3}) = -3.42600 + 7.19635 \times 10^2 T^{-1} + 3.65191 \times 10^{-3} T - 1.90240 \times 10^{-6} T^2 \quad \text{for } 273.15 < T < 367.90 \quad (75)$$

The viscosity of aqueous solutions can be estimated using Eq. (76), where subscript 'w' represents water,  $m_n$  is the mass fraction of solute  $n$ , and  $N$  is the number of solutes in the solution (Laliberté, 2009).

$$\mu = \mu_w^{m_w} \prod_{n=1}^N \mu_n^{m_n} \quad (76)$$

Thus, assuming the aqueous plutonium has a negligible effect on dynamic viscosity, Eqs. (74) and (75) can be used to determine  $\mu_{\text{H}_2\text{O}}$

and  $\mu_{\text{HNO}_3}$  at a given temperature, such that Eq. (76) can be used to determine the viscosity of the aqueous phase,  $\mu_{\text{aq}}$ , in mPa s.

Experimental data from Wongsawa et al. (2018) gives values for the viscosity of TBP and kerosene mixtures at various TBP concentrations, at 303.15 K and 0.1 MPa. Data from Zheng et al. (2010) gives values for the viscosity of TBP and kerosene mixtures at various temperatures. Combining this data, and using Table 2, the following equation can be used to determine the temperature-dependent variation of the dynamic viscosity of the organic phase present in the 1970 Windscale nuclear criticality incident:

$$\mu_{\text{or}} = 1.10740 \exp(-2.24593 \times 10^{-2} T) \quad \text{for } 293.15 < T < 333.15 \quad (77)$$

where  $\mu_{\text{or}}$  is the dynamic viscosity of the organic phase (in Pa s),  $T$  is the temperature in Kelvin. It has been assumed that the presence of plutonium nitrate will have a negligible effect on the viscosity of the organic phase.

The dynamic viscosity in the emulsion region,  $\mu_{\text{em}}$ , will be calculated using Eq. (32).

### 5.1.3. Specific heat capacity

Regarding the aqueous phase, Laliberté (2009) used experimental data from Enea et al. (1977), Hovey et al. (1988), Hovey and Hepler

(1989) and Patterson and Woolley (2002) to determine correlations describing the specific heat capacity of aqueous solutions over a range of temperatures and compositions, however data for solutions of  $\text{HNO}_3$  were limited to low molalities.

Using data from the tables of the thermophysical properties of saturated water (Incropera et al., 2013), it can be determined that an increase in water temperature from 290.0 K to 335.0 K corresponds to a 0.05% increase in  $C_{p,\text{H}_2\text{O}}$ . Thus, it will be assumed that the specific heat capacity of the aqueous phase will remain constant with temperature.

With the further assumption that  $\text{Pu}(\text{NO}_3)_4$  molecules have a negligible effect on the aqueous phase's heat capacity, Eq. (78) (Yaws, 2014) can be used to describe  $C_{p,\text{aq}}$ , the specific heat capacity of the aqueous phase at 293.15 K:

$$C_{p,\text{aq}} = 4.1860 - 6.0121m + 9.1307m^2 - 5.4941m^3 \quad \text{for } 0.0 < m < 0.8 \quad (78)$$

where  $m$  is the mass fraction of  $\text{HNO}_3$  in the solution. Using the data from Table 2,  $C_{p,\text{aq}}$  is calculated to be  $2965 \text{ J kg}^{-1} \text{ K}^{-1}$ .

Regarding the organic phase, the contribution of kerosene, TBP,  $\text{Pu}(\text{NO}_3)_4$  and  $\text{HNO}_3$  to the mixture's specific heat capacity,  $C_{p,\text{or}}$ , need to be considered. If a mixture behaves ideally, its specific heat capacity,  $C_p$ , can be calculated using Eq. (79):

$$C_p = \sum_{n=1}^N m_n C_{p,n} \quad (79)$$

where  $N$  is the number of pure substances (components) in the mixture, and  $m_n$  and  $C_{p,n}$  are the mass fraction and specific heat capacity of component  $n$  (Günther and Steimle, 1997). However, if there are deviations in ideal behaviour, additional functions need to be included in Eq. (79) to account for the interaction between the mixture's components (Günther and Steimle, 1997). It is unlikely that the organic phase was an ideal mixture of its constituents. However, given the lack of data available relevant to the calculation of  $C_{p,\text{or}}$ , it will be assumed that Eq. (79) can be used to effectively determine  $C_{p,\text{or}}$ .

For kerosene at 298.15 K, Yaws (2014) gives  $C_{p,\text{kerosene}}$  to be  $2010 \text{ J kg}^{-1} \text{ K}^{-1}$ , whereas (Nord and Bolland, 2020) gives a value of  $1968 \text{ J kg}^{-1} \text{ K}^{-1}$ . Using the assumption that the average molecular composition of the kerosene present in the 1970 Windscale criticality incident was  $\text{C}_{11}\text{H}_{20}$ , for all organic compounds with the formula  $\text{C}_{11}\text{H}_{20}$  listed in Yaws (2014), the only one with a specific heat capacity between 1968 and  $2010 \text{ J kg}^{-1} \text{ K}^{-1}$  is 1-hexylcyclopentene. When the temperature of 1-hexylcyclopentene is raised from 293.15 to 348.15 K, a 4.78% increase in  $C_{p,\text{C}_{11}\text{H}_{20}}$  is observed (Yaws, 2014). Thus, it will be assumed that, in the range  $293.15 < T \text{ (K)} < 348.15$ ,  $C_{p,\text{kerosene}}$  is constant and equal to  $1989 \text{ J kg}^{-1} \text{ K}^{-1}$ , the average of the values cited by Yaws (2014) and Nord and Bolland (2020).

There is limited thermophysical data available for  $\text{Pu}(\text{NO}_3)_4$ . Therefore, the apparent heat capacities of the  $\text{Pu}^{4+}$  and  $\text{NO}_3^-$  ions will be used, as these values are available in the literature. Hepler and Hovey (1996) gives  $C_{p,\text{NO}_3^-} = -71 \text{ J K}^{-1} \text{ mol}^{-1}$  at 298.15 K, and Lemire and Tremaine (1980) gives an average value of  $C_{p,\text{Pu}^{4+}} = -63 \text{ J K}^{-1} \text{ mol}^{-1}$  over the temperature range  $298.15 < T \text{ (K)} < 473.15$ . Using these two values, it will be assumed that the presence of  $\text{Pu}(\text{NO}_3)_4$  will modify the specific heat capacity of the organic phase by  $-712 \text{ J kg}^{-1} \text{ K}^{-1}$ , and that the effect of temperature can be neglected. Thus,  $C_{p,\text{Pu}(\text{NO}_3)_4} = -712 \text{ J kg}^{-1} \text{ K}^{-1}$ .

Using data from Design Institute for Physical Properties, Sponsored by AIChE (2020), the specific heat capacity for TBP,  $C_{p,\text{C}_{12}\text{H}_{27}\text{O}_4\text{P}}$ , at 293.6 K was calculated to be  $1587 \text{ J kg}^{-1} \text{ K}^{-1}$ . Using data from Forsythe and Giauque (1942), the specific heat capacity for  $\text{HNO}_3$ ,  $C_{p,\text{HNO}_3}$ , at 293.6 K was calculated to be  $1748 \text{ J kg}^{-1} \text{ K}^{-1}$ .

Thus, using Eq. (79), organic phase composition data from Table 2, and the constituent specific heat capacities precisely defined,  $C_{p,\text{or}}$  was calculated to be  $1568 \text{ J kg}^{-1} \text{ K}^{-1}$ .

It will be assumed that the specific heat capacity of the emulsion,  $C_{p,\text{em}}$  can be calculated as the mass-weighted average of the aqueous and organic specific heat capacities:

$$C_{p,\text{em}} = \omega C_{p,\text{aq}} + (1 - \omega) C_{p,\text{or}} \quad (80)$$

**Table 3**

Parameters in Eqs. (82) and (83).

| Component                       | A (mN m <sup>-1</sup> ) | B      | C        | T <sub>min</sub> (K) | T <sub>max</sub> (K) | Reference    |
|---------------------------------|-------------------------|--------|----------|----------------------|----------------------|--------------|
| C <sub>11</sub> H <sub>20</sub> | 56.21150                | 650.99 | 1.22222  | 248.16               | 650.99               | <sup>a</sup> |
| TBP                             | 0.06717                 | 1.8629 | -0.54604 | 194.31               | 716.7                | <sup>b</sup> |

<sup>a</sup> Yaws (2014).

<sup>b</sup> Design Institute for Physical Properties, Sponsored by AIChE (2020).

#### 5.1.4. Surface tension

Using the proposed equation for the surface tension of water by Jones (1984) and Hosoma et al. (2000) used Eq. (81) to estimate surface tension of a solution of aqueous plutonium nitrate,  $\sigma_{\text{aq}}$ :

$$\begin{aligned} \sigma_{\text{aq}} = & 7.5675 \times 10^{-2} - 1.3762 \times 10^{-4}(T - 273.15) \\ & - 3.938 \times 10^{-7}(T - 273.15)^2 \\ & + 1.076 \times 10^{-9}(T - 273.15)^3 + 1.2 \times 10^{-5}c_{\text{Pu}} - 5.6 \times 10^{-4}c_{\text{HN}} \end{aligned} \quad (81)$$

where  $c_{\text{Pu}}$  is the plutonium concentration (in  $\text{g L}^{-1}$ ) of the solution,  $c_{\text{HN}}$  is the free nitric acid concentration (in  $\text{mol L}^{-1}$ ) in the solution and the temperature  $T$  is measured in K.

Several values of the surface tension of kerosene are reported in the literature. Knovel Corporation (2008) reports a value of  $26.4 \text{ mN m}^{-1}$  at 298.15 K, whereas Wypych (2015) gives a value of  $26 \text{ mN m}^{-1}$  for kerosene, and 26 and  $27.5 \text{ mN m}^{-1}$  for industrial chemicals Shellsol 16 and Shellsol 2046 AR. As no temperature is given for these values, 298.15 K will be assumed. As such, the average surface tension of kerosene from the literature is  $26.475 \text{ mN m}^{-1}$  at 298.15 K. Yaws (2014) reports that the temperature-dependent surface tension of organic products can be determined using the following relation:

$$\sigma = A \left(1 - \frac{T}{B}\right)^C \quad \text{for } T_{\min} < T < T_{\max} \quad (82)$$

where  $A$ ,  $B$ , and  $C$  are material-dependent constants, and  $T$  is the temperature in Kelvin. Using the assumption that the kerosene present in the organic phase has molecular formula  $\text{C}_{11}\text{H}_{20}$ , data from Yaws (2014) can be used to approximate the temperature dependent surface tension of the kerosene present in the 1970 Windscale nuclear criticality incident. Of all substances with formula  $\text{C}_{11}\text{H}_{20}$ , 1-undecyne has the closest surface tension ( $26.590 \text{ mN m}^{-1}$ ) at 298.15 K to the  $26.475 \text{ mN m}^{-1}$  average determined previously. The values  $A$ ,  $B$ , and  $C$  in Eq. (82) for 1-undecyne are presented in Table 3, along with the temperature limits  $T_{\min}$  and  $T_{\max}$ .

Similarly, Design Institute for Physical Properties, Sponsored by AIChE (2020) determined that Eq. (83) could be used to calculate the temperature-dependent surface tension of TBP:

$$\sigma_{\text{TBP}} = A \left(1 - \frac{T}{T_{\text{crit}}}\right)^{B+C \frac{T}{T_{\text{crit}}}} \quad \text{for } T_{\min} < T < T_{\max} \quad (83)$$

where  $T_{\text{crit}}$  is the critical temperature of TBP, equal to 716.7 K, and  $A$ ,  $B$  and  $C$  are constants, defined in Table (Design Institute for Physical Properties, Sponsored by AIChE, 2020), in addition to the temperature limits  $T_{\min}$  and  $T_{\max}$ .

The surface tension of a mixture is noted by many to be a complex function (Escobedo and Mansoori, 1998; Kahl et al., 2003). This is attributed to the fact that a mixture's composition at its surface is not necessarily the same as the bulk composition (Meissner and Michaels, 1949). In lieu of correlations or experimental data describing the surface tension of a TBP/OK mixture, temperature dependent or otherwise, evaluating the correlations described in references such as Escobedo and Mansoori (1998) are outside the scope of the presented research. Allen et al. (1999) validated the use of mass-weighted averaging to predict the surface tension of biodiesel fuels, though it was noted that this method tended to overestimate the surface tension of a mixture. Shu et al. (2008) built on the work of Allen et al. (1999) by using topological indices and attempting to account for the relative concentration of mixture components at the mixture's surface. The simpler

**Table 4**  
Parameters in Eq. (85) for the materials presented in this research.

| Liquid                                           | A (W m <sup>-1</sup> K <sup>-1</sup> ) | B (W m <sup>-1</sup> K <sup>-2</sup> ) | C (W m <sup>-1</sup> K <sup>-3</sup> ) | T <sub>min</sub> (K) | T <sub>max</sub> (K) | Reference    |
|--------------------------------------------------|----------------------------------------|----------------------------------------|----------------------------------------|----------------------|----------------------|--------------|
| H <sub>2</sub> O                                 | -2.98749 × 10 <sup>-1</sup>            | 4.70537 × 10 <sup>-3</sup>             | -5.62090 × 10 <sup>-6</sup>            | 273.16               | 633.15               | <sup>a</sup> |
| HNO <sub>3</sub>                                 | 1.21070 × 10 <sup>-1</sup>             | 5.38300 × 10 <sup>-4</sup>             | 0.0                                    | 233.15               | 433.15               | <sup>a</sup> |
| C <sub>12</sub> H <sub>27</sub> O <sub>4</sub> P | 1.30990 × 10 <sup>-1</sup>             | -1.32700 × 10 <sup>-4</sup>            | 0.0                                    | 194.31               | 562.15               | <sup>b</sup> |
| Kerosene                                         | 1.96945 × 10 <sup>-1</sup>             | -1.63636 × 10 <sup>-4</sup>            | 0.0                                    | 293                  | 348                  | <sup>c</sup> |

<sup>a</sup> Yaws (2014).

<sup>b</sup> Design Institute for Physical Properties, Sponsored by AIChE (2020).

<sup>c</sup> Chhabra and Shankar (2018).

approach taken by Allen et al. (1999) has been used in the presented research. It was be assumed that the surface tension of the organic phase,  $\sigma_{or}$ , of the 1970 Windscale nuclear criticality incident can be suitably approximated using the mass-weighted average of the surface tensions of kerosene and TBP, viz:

$$\sigma_{or} = \frac{m_{C_{11}H_{20}}\sigma_{C_{11}H_{20}} + m_{TBP}\sigma_{TBP}}{m_{C_{11}H_{20}} + m_{TBP}} \quad (84)$$

where  $m_{C_{11}H_{20}}$  and  $m_{TBP}$  are the masses of kerosene and TBP in the organic phase, respectively. This assumes that the plutonium nitrate and nitric acid in the organic phase have a negligible effect on the phase's surface tension. Eqs. (82) and (83) are used to determine the surface tensions of the constituents, where data for 1-undecyne is used to approximate the surface tension of kerosene.

#### 5.1.5. Thermal conductivity

For each of the liquid components of the aqueous and organic phases in the 1970 Windscale nuclear criticality incident, Eq. (85) can be used to determine the thermal conductivity,  $\kappa$ , of the liquid at temperature  $T$ . Table 4 gives the values of  $A$ ,  $B$ ,  $C$ ,  $T_{min}$  and  $T_{max}$  for each liquid.

$$\kappa = A + B T + C T^2 \quad \text{for } T_{min} < T < T_{max} \quad (85)$$

A number of correlations have been postulated to determine the heat capacity of liquid mixtures. Of these, that of Filippov and Novoselova (1955) will be used (as in Eq. (86)), due to the lack of experimental data limiting the usability of other correlations (Gaitonde et al., 1978).

$$\kappa_m = \kappa_1 m_1 + \kappa_2 m_2 - 0.72[\kappa_1 - \kappa_2]m_1 m_2 \quad (86)$$

In Eq. (86), subscript 'm' denotes 'mixture' (either aqueous or organic), and  $m$  refers to the mass fraction of component 1 or 2. For the aqueous phase, the components are H<sub>2</sub>O and HNO<sub>3</sub>, and for the organic, the components are C<sub>12</sub>H<sub>27</sub>O<sub>4</sub>P and kerosene. It will be assumed that the effects of other components in each of the mixtures are negligible.

#### 5.2. Mixing coefficient

Turbulence is the chaotic variation in flow velocity about a mean value (Coussot, 1997), where energy cascades from the largest to smallest turbulent eddies (Friedlander, 1957). The mean length and velocity scale characterise the varying length and velocity scales of the eddies (Csanady, 1973).

In the system being considered, turbulent mixing would have occurred in each of the three regions, due to the entry of aqueous material (including the disruption to the organic layer); non-uniform heating (from the  $r, z$  power profile) causing buoyancy-driven flow; and production and advection of radiolytic gas bubbles. The rate of mixing will therefore be a function of  $r$ ,  $z$ , and  $t$ .

A simplified approach describing turbulence as a Fickian diffusive process, originally developed by Cooling (2014), will be used in the proposed model. Cooling (2014) characterises the rate of mixing in a fuel solution through a mixing coefficient,  $D_{FS}$ , which is defined as

$$D_{FS}(t, z) = \frac{U_{eddy}(t)r_{eddy}(t, z)}{\pi} \quad (87)$$

where  $U_{eddy}$  and  $r_{eddy}$  are the velocity and radius, respectively, of the largest eddies predicted to be in the system. In the proposed model, it will be assumed that  $U_{eddy}$  is constant throughout the vessel, taking a value of 15 cm s<sup>-1</sup> during aqueous ingress and 5 cm s<sup>-1</sup> otherwise. The parameter  $r_{eddy}$  is assumed to be constant within each fissile liquid. Due to the aqueous solution ingress disrupting the liquids, it will be assumed that turbulent eddies cannot cross the radial centre of the vessel. As such, and to account for both radial and axial directions,  $r_{eddy}$  is taken to be the average of the half-thickness and half-radius of each layer:

$$r_{eddy}(t, z) = \frac{1}{4} \left( (H_x(t, z) - H_{x-1}(t, z)) + \bar{R}_x(t, z) \right) \quad (88)$$

where  $x$  is the layer index, and  $\bar{R}_x$  is the average radius in layer  $x$ .

#### 5.3. Heat transfer between phases

##### 5.3.1. Convective heat transfer between the fissile liquids and the vessel

Convective heat transfer coefficients (HTCs) between a fluid and solid depend on the speed of circulating currents within the fluid, which in turn will depend on fluid temperature profiles, rising gas bubbles, and any forced convection (e.g. due to mechanical agitation). During the 1970 Windscale nuclear criticality incident, the liquids were agitated, initially due to aqueous entry, and then due to aqueous drainage. The exact effects of this agitation are unknown.

Using CFD, Milian Pérez et al. (2015) determined the speed of circulating currents within aqueous homogeneous reactor ARGUS. Average fluid velocities of 0.016 m s<sup>-1</sup> and 0.104 m s<sup>-1</sup> were determined for a systems without and with gas voids, respectively. In the absence of any detailed description of the fluid agitation due to aqueous entry, the higher speed of 0.104 m s<sup>-1</sup> will be assumed as the average fluid velocity.

Chilton et al. (1944) proposed a correlation to determine the HTCs between the solution and wall of agitated vessels. This correlation is presented in Eq. (89), where  $Nu = hD/\kappa$ ,  $Ro = L^2 N \rho_s / \mu$  and  $Pr = c_p \mu / \kappa$  are the Nusselt, Roshko and Prandtl numbers, respectively.  $D$  and  $h$  are the characteristic length and HTC. Subscript  $s$  means the property is to be taken at the wall, though for this work, it will be assumed that properties are constant throughout the domain, at a temperature of 293.15 K and atmospheric pressure.  $L$  is the length of the impeller blade used in Chilton et al. (1944) to agitate the vessel, and  $N$  is the impeller shaft speed (in revolutions per second). Other variables are as previously defined.

$$Nu \left( \frac{\mu_s}{\mu} \right)^{0.14} = 0.36 Ro^{2/3} Pr^{1/3} \quad (89)$$

This correlation has been successfully been used by Winter (2019) to model aqueous homogeneous reactors, and as such will be used in the presented research.

Using a vessel radius,  $R_v$ , of 30.5 cm,  $D = 2R_v$ , and ignoring the effect of the vessel's dished end, Fig. 7 shows the convective HTC for both the aqueous and organic liquids present in the 1970 Windscale nuclear criticality incident. The HTC between the aqueous phase and vessel wall,  $h_{int,aq}$ , was chosen to be 440 W m<sup>-2</sup> K<sup>-1</sup>, whereas the HTC between the organic phase and the vessel wall,  $h_{int,or}$ , was chosen to be 110 W m<sup>-2</sup> K<sup>-1</sup>, as shown in Fig. 7. For both materials, the selected HTCs correspond to either a large impeller ( $L = 1.50R_v$ ) moving slowly

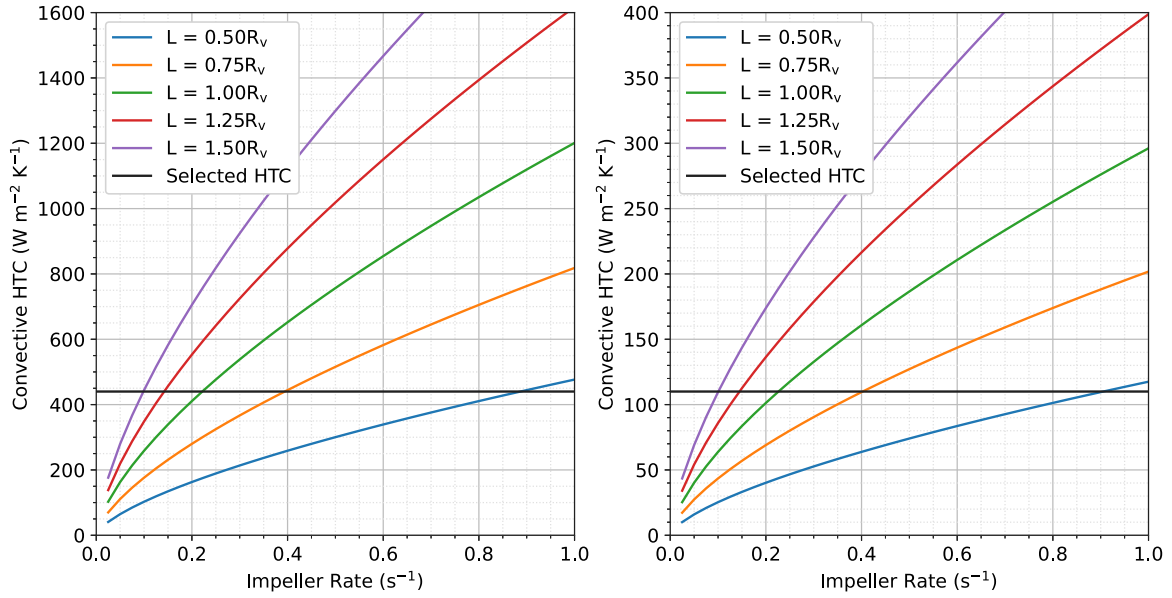


Fig. 7. Predicted HTCs for the aqueous (left) and organic (right) liquids, as a function of impeller speed, for a variety of impeller blade lengths.

( $\sim 0.1 \text{ s}^{-1}$ ), or a small impeller ( $L = 0.50R_v$ ) moving relatively quickly ( $\sim 0.9 \text{ s}^{-1}$ ). These rotational speeds correspond to fluid velocities of  $0.072 \text{ m s}^{-1}$  and  $0.216 \text{ m s}^{-1}$  respectively, which is of the same order of magnitude as the  $0.104 \text{ m s}^{-1}$  calculated by Milian Pérez et al. (2015).

It will be assumed that the HTC between the emulsion layer and the vessel,  $h_{\text{int,em}}$ , can be calculated as the volume-weighted average of the aqueous- and organic-vessel HTCs:

$$h_{\text{int,em}} = \alpha h_{\text{int,aq}} + (1 - \alpha) h_{\text{int,or}} \quad (90)$$

### 5.3.2. Heat transfer between the vessel and surroundings

Heat can be transferred from the reactor vessel to its surroundings by either radiation or convection. The rate of radiation released from a surface is described by Eq. (91) (Incropera et al., 2013), where  $\sigma_{\text{SB}} = 5.67 \times 10^{-8} \text{ W m}^{-2} \text{ K}^{-4}$  is the Stefan–Boltzmann constant,  $T_{\infty}$  is the temperature of the surroundings, and  $T_{\text{vess}}$  is the temperature of the vessel (emitting radiative energy). The emissivity,  $\epsilon$ , of a surface describes how efficiently the surface emits energy:  $\epsilon = 1$  indicates blackbody radiation, but real surfaces have emissivity in the range  $0 < \epsilon < 1$ . Coulson (1999) gives the emissivity of sheet steel to be 0.657, which will be used in the presented research as the emissivity of the vessel.

$$E = \epsilon \sigma_{\text{SB}} (T_{\text{vess}}^4 - T_{\infty}^4) \quad (91)$$

Assuming a constant ambient temperature of 293.6 K, and an estimate that the vessel reaches a maximum temperature of 348.6 K, Eq. (91) gives the radiative heat flux from the vessel's outer surface to be  $273 \text{ W m}^{-2}$ .

By assuming the vessel's walls can be modelled as a flat plate, an Equation of the form presented in Eq. (92) can be used to estimate the rate of heat loss due to natural convection from the reactor surface to its surroundings. In this equation,  $Nu = h_{\text{ext}} L / \kappa_{\text{air}}$ ,  $Gr = g \rho_{\text{air}} (T_{\text{vess}} - T_{\infty}) L^3 / \nu_{\text{air}}^2$  and  $Pr = c_{p,\text{air}} \mu_{\text{air}} / \kappa_{\text{air}}$  are the Nusselt, Grashof and Prandtl numbers, respectively. In these definitions,  $L$  is the characteristic length (i.e. the height of the vessel, equal to 0.686 m),  $g$  is the acceleration due to gravity and  $T_{\text{vess}}$  and  $T_{\infty}$  are as previously defined. Subscripts 'air' indicate the property is that of air; these properties are defined in Table 5.

$$Nu = f(Gr, Pr) \quad (92)$$

Using the properties of air presented in Table 5, the Grashof and Nusselt numbers are calculated to be  $2.559 \times 10^9$  and  $1.316 \times 10^2$  respectively. For  $Gr > 10^9$ , Eq. (93) has been shown to adequately describe

Table 5  
Properties of air at 293.6 K.

| Property                         | Symbol                | Value                  | Unit                              | Reference |
|----------------------------------|-----------------------|------------------------|-----------------------------------|-----------|
| Density                          | $\rho_{\text{air}}$   | 1.191                  | $\text{kg m}^{-3}$                | b         |
| Viscosity                        | $\mu_{\text{air}}$    | $1.814 \times 10^{-5}$ | $\text{Pa s}$                     | b         |
| Thermal conductivity             | $\kappa_{\text{air}}$ | $2.579 \times 10^{-2}$ | $\text{W m}^{-1} \text{ K}^{-1}$  | b         |
| Specific heat capacity           | $c_{p,\text{air}}$    | $1.007 \times 10^3$    | $\text{J kg}^{-1} \text{ K}^{-1}$ | b         |
| Coefficient of thermal expansion | $\beta_{\text{air}}$  | $3.406 \times 10^{-3}$ | $\text{K}^{-1}$                   | a         |
| Prandtl number                   | $Pr$                  | $7.083 \times 10^{-1}$ | –                                 | –         |

a Yaws (2014).

b Incropera et al. (2013).

the heat transfer from vertical cylinders (Coulson, 1999; Hiroharu et al., 1968). Using Eq. (93),  $h_{\text{ext}}$  is calculated to be  $4.949 \text{ W m}^{-2} \text{ K}^{-1}$ . For  $T_{\text{vess}} = 348.6$  and  $T_{\infty} = 293.6 \text{ K}$ , this HTC corresponds to a heat transfer rate of  $272 \text{ W m}^{-2}$ .

$$Nu = 0.138 Gr^{0.36} (Pr^{0.175} - 0.55) \quad (93)$$

Thus, the rates of heat loss due to radiation and natural convection are on a similar scale.

### 5.3.3. Heat transfer from the organic phase surface to the plenum gas

Heat transfer from the surface of the organic phase to the plenum gas could occur via forced or natural convection. As the aqueous solution was being vacuum-lifted into the vessel, the vacuum-lifting could have induced forced convection by disrupting the gas at the top of the vessel. When aqueous entry ceased, the convection would have returned to 'natural'. Correlations for natural and forced convection are presented in Eqs. (94) and (95) respectively (Incropera et al., 2013). In these equations, the surface of the organic phase is treated as a solid flat plate, and the characteristic length  $L$  is equal to the surface area of the organic phase divided by the surface's perimeter.  $Re = \rho_{\text{air}} u_{\text{air}} L / \mu_{\text{air}}$  is the Reynolds number of the gas, where  $u_{\text{air}}$  is the speed of the air.

$$Nu = 0.15 (GrPr)^{1/3} \quad \text{for } 10^7 < GrPr < 10^{11} \quad (94)$$

$$Nu = 0.66 Re^{1/2} Pr^{1/3} \quad \text{for } Re \leq 2500 \quad (95)$$

In this application,  $GrPr = 1.991 \times 10^7$ , resulting in a HTC due to natural convection,  $h_{\text{nat,or} \rightarrow \text{air}}$  of  $6.875 \text{ W m}^{-2} \text{ K}^{-1}$ . The HTCs due to forced convection are presented in Fig. 8 as a function of the velocity of the plenum gas. Fig. 8 shows that, even at gas velocities of  $24.8 \text{ cm}$

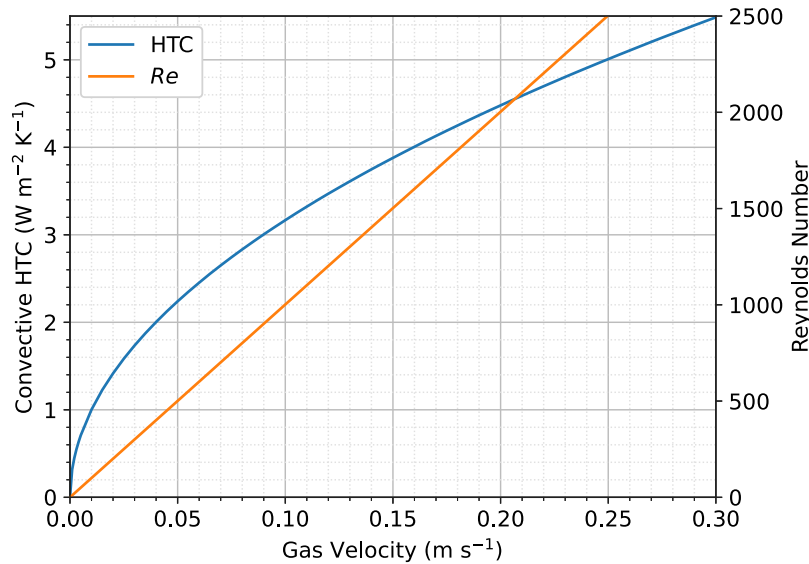


Fig. 8. HTC describing heat transfer from the surface of the organic phase to the plenum gas due to forced convection, and the corresponding Reynolds numbers.

$s^{-1}$  (and a corresponding  $Re = 2483.657$ ), the convective HTC is equal to  $4.988 \text{ W m}^{-2} \text{ K}^{-1}$ .

The overall HTC between the organic solution and the plenum gas will also depend on the resistance to heat transfer from the solution bulk and solution surface to the plenum gas. Taking this into account, the overall HTC between the fuel solution and the surface,  $h_{sur}$ , can be calculated using Eq. (96):

$$\frac{1}{h_{sur,or \rightarrow air}} = \frac{1}{h_{int,or}} + \frac{1}{h_{nat,or \rightarrow air}}. \quad (96)$$

The first term on the right-hand side represents the resistance to heat transfer between the bulk and surface of the organic phase, and has been approximated as  $h_{int,or}$ , calculated in Section 5.3.1 to be  $110 \text{ W m}^{-2} \text{ K}^{-1}$ . Regarding the heat transfer from the surface of the organic phase to the plenum gas, the HTC for natural convection ( $6.875 \text{ W m}^{-2} \text{ K}^{-1}$ ) will be used, as it is greater than the HTC for forced convection. Thus,  $h_{sur,or \rightarrow air}$  is calculated to be  $6.470 \text{ W m}^{-2} \text{ K}^{-1}$ .

#### 5.3.4. Heat transfer between fissile liquid layers

Using the assumption that the fissile liquid layers (aqueous, emulsion, organic) are completely immiscible, heat transfer between the layers will be largely due to convection.

Using the same methodology as Section 5.3.3, the HTCs between the emulsion layer and the aqueous and organic layers can be determined. As described in Section 3.4.2, the viscosity of an emulsion tends to be relatively high due to non-Newtonian behaviour. As such, the emulsion will be the phase treated as a solid flat plate.

In both instances, natural convection dominated over forced convection, even at liquid speeds greater than the  $15 \text{ cm s}^{-1}$  assumed in Section 5.2. Thus, the HTCs between the aqueous and emulsion, and emulsion and organic, phases can be calculated using Eqs. (97), (98) and the same calculation route as Section 5.3.3.

$$\frac{1}{h_{sur,aq \rightarrow em}} = \frac{1}{h_{int,em}} + \frac{1}{h_{nat,aq \rightarrow em}} = \frac{1}{\alpha h_{int,aq} + (1 - \alpha) h_{int,or}} + \frac{1}{h_{nat,aq \rightarrow em}} \quad (97)$$

$$\frac{1}{h_{sur,em \rightarrow or}} = \frac{1}{h_{int,em}} + \frac{1}{h_{nat,em \rightarrow or}} = \frac{1}{\alpha h_{int,aq} + (1 - \alpha) h_{int,or}} + \frac{1}{h_{nat,em \rightarrow or}} \quad (98)$$

Finally, it is possible that the aqueous and organic phases are in contact. In this case, as the organic phase is the most viscous, it will

be treated as the flat plate. By once again following the methodology and calculation route detailed in Section 5.3.3, Eq. (99) can be used to determine the HTC between the aqueous and organic phases, resulting in  $h_{sur,aq \rightarrow or}$  calculated to be  $96.200 \text{ W m}^{-2} \text{ K}^{-1}$ .

$$\frac{1}{h_{sur,aq \rightarrow or}} = \frac{1}{h_{int,or}} + \frac{1}{h_{nat,aq \rightarrow or}} \quad (99)$$

#### 5.4. Radiolytic gas generation coefficient

##### 5.4.1. Plutonium nitrate

In aqueous solutions, high LET radiations produce high yields of  $H_2$ ,  $O_2$  and  $H_2O_2$ , whereas low LET radiations produce high yields of free radicals  $H^\bullet$   $HO^\bullet$ . These free radicals very quickly recombine in dense fission tracks, and therefore are not considered in the proposed model (Lane et al., 1958).

Experiments to determine the production of radiolytic gas due to the fission of  $^{239}\text{Pu}$  have been performed by Kazanjian and Horrell (1972) and Sheppard (1968). Fig. 9 shows the data from these experiments, and the equation of the fitted curve is presented in Eq. (100).  $G_{total}$  is the radiolytic gas generation coefficient of aqueous plutonium nitrate, and  $C_{HNO_3}$  is the concentration of nitric acid in the solution. Kazanjian and Horrell (1972) notes that varying plutonium concentration has a minimal effect on  $G_{total}$  for aqueous plutonium nitrate. There is insufficient data in the available literature to determine the effect of temperature on  $G_{total}$ .

$$G_{total} = 5.318896 \times 10^{-1} - 1.769539 \times 10^{-1} C_{HNO_3} + 2.163226 \times 10^{-2} C_{HNO_3}^2 - 8.877015 \times 10^{-4} C_{HNO_3}^3 \quad (100)$$

The acid nitric acid present in the 1970 Windscale nuclear criticality incident was  $7\text{M HNO}_3$ , which, using Eq. (100), will result in an aqueous phase radiolytic gas generation coefficient,  $G_{aq}$ , of  $4.871143 \times 10^{-2}$  molecules per 100 eV of fission energy.

##### 5.4.2. Organic phase

In the 1970 Windscale criticality incident, the organic phase was 24% TBP diluted with kerosene. Kerosene is a mixture of hydrocarbons, with structure  $C_{12}H_{26}$  to  $C_{15}H_{32}$ . In addition, the phase will contain plutonium complexes, and will be in contact with the aqueous phase. As the organic phase consists of multiple components, competing reactions will occur, including radiolysis and hydrolysis. Blake et al. (1963) notes that, for short term, moderate-temperature conditions, TBP hydrolysis can be safely neglected, and will be done so in this work. In the

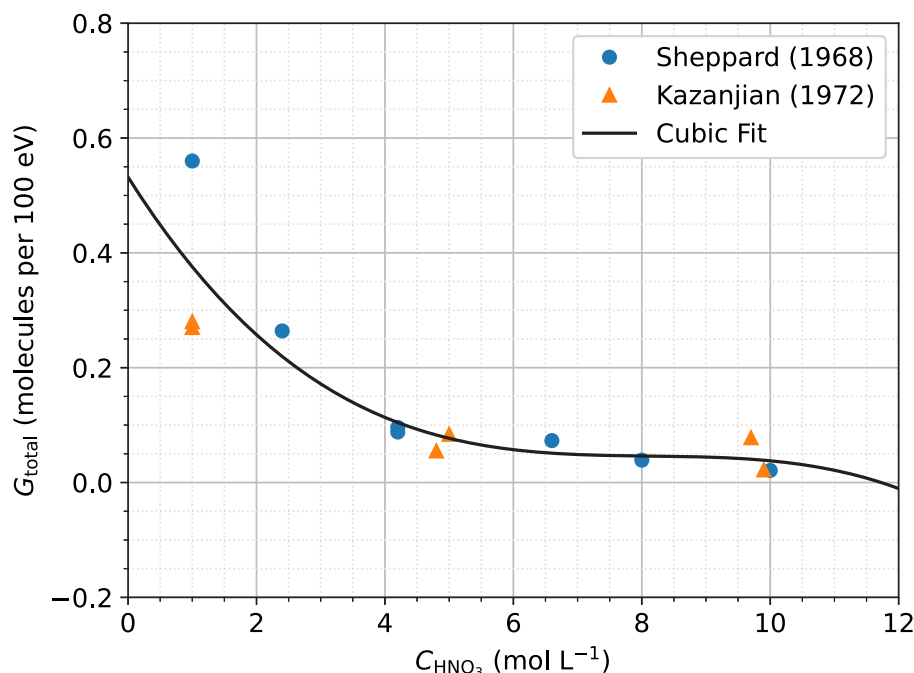


Fig. 9. Experimental data from Kazanjian and Horrell (1972) and Sheppard (1968), with  $G_{\text{total}}$  shown as a function of  $C_{\text{HNO}_3}$ .

system investigated, TBP would have been a complex with water, nitric acid, and plutonium. These complexes react at different rates with the reactive species produced from radiolysis, making exact product yields hard to determine (Mincher et al., 2009).

For both neat and diluted TBP, dibutylphosphoric acid (HDBP) is the predominant radiolysis product (Mincher et al., 2009; Mincher and Mezyk, 2009), with the yield decreasing in the presence of  $\text{HNO}_3$  (Brodda and Heinen, 1977; Mincher and Mezyk, 2009). Other radiolysis products include hydrogen, methane, monobutylphosphoric acid ( $\text{H}_2\text{MBP}$ ) and phosphoric acid (Mincher et al., 2009; Mincher and Mezyk, 2009). Nitrated phosphates are also produced in the presence of  $\text{HNO}_3$  (Mincher et al., 2009).

An adequate number of studies are available for low LET electron or gamma ( $\gamma$ ) radiation of TBP, for example (Burr, 1958; Holland et al., 1978; United States Nuclear Regulatory Commission, 2013; Williams et al., 1957). A conservative estimate from these reports indicates  $G_{\text{gas,TBP}} \approx 1.1$  molecules per 100 eV. However, there are limited studies available on high LET radiation of TBP, which will be the major contributor to radiolysis, in Section 3.4.1. Mincher et al. (2009) notes that there is very little difference in HDBP yield for  $\alpha$  and  $\gamma$  radiation of TBP. Thus, it will be assumed that the same can be said for the gaseous yield, and  $G_{\text{gas,TBP}} = 1.1$  molecules per 100 eV will be used in the presented work.

Using electron radiation, a conservative estimate of  $G_{\text{gas,C}_{12}\text{H}_{26}} = 4.4$  molecules per 100 eV can be obtained based on the experiments of Spinks and Woods (1990) and Mincher et al. (2009). In the absence of available data for  $\alpha$  radiation, this value will be used in the presented work.

Combining the obtained values for  $G_{\text{gas,TBP}}$  and  $G_{\text{gas,C}_{12}\text{H}_{26}}$  in the proportions that each constituent was present in the organic phase leads to an assumed value of  $G_{\text{or}}$ , the radiolytic gas generation coefficient of the organic phase in the 1970 Windscale nuclear criticality incident, of 3.5 molecules per 100 eV of fission energy. It is worth noting the high uncertainties associated with this value; and is worth noting how high  $G_{\text{or}}$  is compared to  $G_{\text{aq}}$ . It is unclear how the presence of plutonium in the organic phase will affect the yield of radiolytic gas; for the aqueous phase, an increase in plutonium decreased the gaseous yield.

Table 6

Constant case study kinetics data.

| $i$ | $\beta_i$ | $\lambda_i$ ( $\text{s}^{-1}$ ) |
|-----|-----------|---------------------------------|
| 1   | 0.00011   | 0.01327                         |
| 2   | 0.00067   | 0.03088                         |
| 3   | 0.00050   | 0.11337                         |
| 4   | 0.00093   | 0.29250                         |
| 5   | 0.00049   | 0.85749                         |
| 6   | 0.00014   | 2.72973                         |

#### 5.4.3. Emulsion

It will be assumed that the radiolytic gas generation coefficient of the emulsion,  $G_{\text{em}}$ , can be calculated as the volume-weighted average of the aqueous and organic radiolytic gas generation coefficients:

$$G_{\text{em}} = \alpha G_{\text{aq}} + (1 - \alpha) G_{\text{or}} \quad (101)$$

## 6. Kinetics and reactivity feedback data

MCNP was used to determine the reactivity feedback terms in Eq. (4), alongside the required kinetics parameters. For five prescribed values for  $\alpha$  in the range 0.54 to 0.60, the mass of the emulsion layer was varied from 0.0 to 25.252167 kg. Then, the aqueous and organic masses were varied within their possible ranges. Nine different masses for each phase were used. For each combination of volumes, the average temperature,  $\bar{T}$ , or void fraction,  $\bar{\theta}$ , of each layer was varied in turn.

### 6.1. Kinetics data

The results from these MCNP simulations were analysed: it was observed that the parameters  $\nu$ ,  $\beta_{\text{eff}}$ ,  $\beta_i$  and  $\lambda_i$  were independent of time, and the temperature or void fraction of any layer. The resulting averaged parameters are presented in Table 6, with  $\beta_{\text{eff}} = 0.00284 \pm 0.00000$ , and  $\nu = 2.868$ . Due to averaging the parameters across all simulations, the standard deviations of each value in Table 6 are a maximum of  $5.0 \times 10^{-6}$ .

Fig. 10 depicts the variation of the mean neutron generation time,  $\Lambda$ , with the average temperature and void fraction of each material.

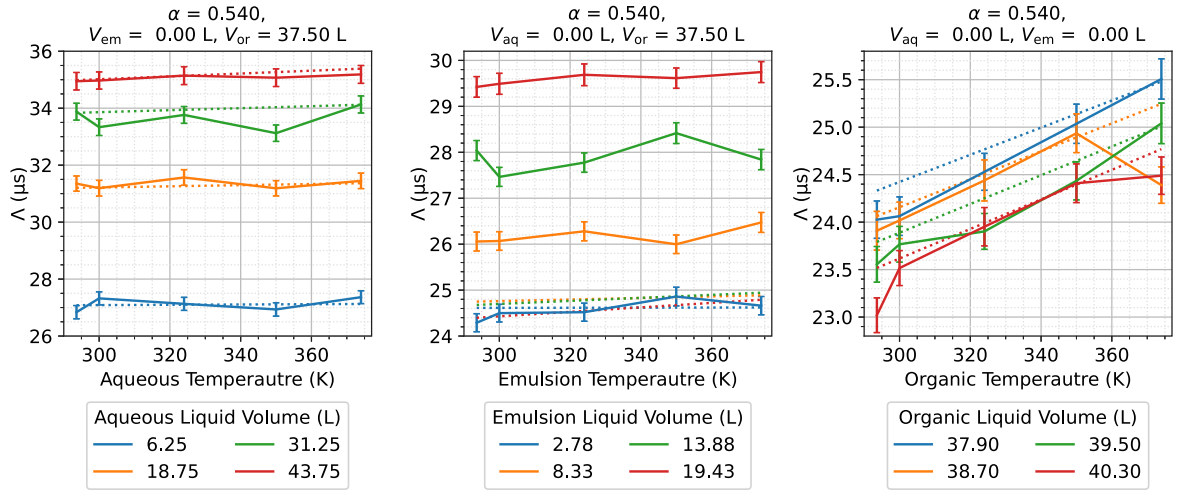
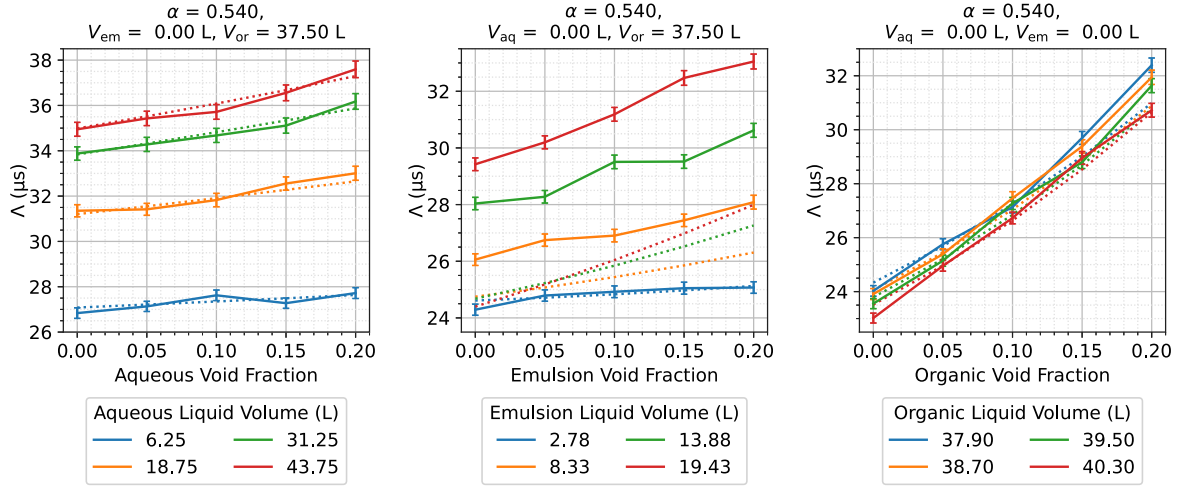
(a)  $\Lambda$  as a function of  $\bar{T}$  for each material.(b)  $\Lambda$  as a function of  $\bar{\theta}$  for each material.

Fig. 10. Graphs depicting the dependency of  $\Lambda$  on the temperatures and void fractions of each of the three regions, for  $\alpha = 0.540$  and selected region volumes. Solid lines represent data calculated using MCNP, and dotted lines represent data calculated using Eq. (102).

In both Figs. 10(a) and 10(b), it can be seen that increasing the temperature or void fraction in any phase will result in an increase in  $\Lambda$ , with the exact relationship being unclear due to oscillations and uncertainties in the results. This is because an increase temperature and/or void fraction will decrease the density of the fissile material, decreasing the probability of neutrons colliding with fissile isotopes, and thus increasing the time it takes for neutrons to collide with fissile isotopes and generate more neutrons. Increasing the temperature or void fraction of the aqueous or emulsion phases increases  $\Lambda$  by up to  $\approx 10\%$ , whereas increasing the temperature or void fraction of the organic phase increases  $\Lambda$  by up to  $\approx 25\%$ . This difference is, in part, due to the higher plutonium concentration in the organic phase, which increases its relative neutronic importance compared to the aqueous and emulsion phases. Additionally, the results in the right-hand graphs of Fig. 10, showing variations  $\Lambda$  with organic temperature or void fraction, were obtained without any aqueous or emulsion phases in the vessel, enhancing the effect of the organic temperature on  $\Lambda$ .

By inspecting  $\Lambda$ 's dependencies on the relevant parameters,  $\Lambda$  variations for each  $\alpha$  were fitted, using non-linear least squares, to the following Equation:

$$\Lambda[V_{\text{aq}}(t), V_{\text{em}}(t), V_{\text{or}}(t), \bar{T}_{\text{aq}}(t), \bar{T}_{\text{em}}(t), \bar{T}_{\text{or}}(t), \bar{\theta}_{\text{aq}}(t), \bar{\theta}_{\text{em}}(t), \bar{\theta}_{\text{or}}(t)] =$$

$$A_1 + A_2 + A_3 + A_4 + A_5 + A_6 + A_7 + A_8 + A_9 + A_{10} \quad (102)$$

where

$$A_1 = a_1 + a_2 V_{\text{aq}} + a_3 V_{\text{em}} + a_4 V_{\text{or}} + a_5 \bar{T}_{\text{aq}} + a_6 \bar{T}_{\text{em}} + a_7 \bar{T}_{\text{or}} + a_8 \bar{\theta}_{\text{aq}} + a_9 \bar{\theta}_{\text{em}} + a_{10} \bar{\theta}_{\text{or}}, \quad (103)$$

$$A_2 = a_{11} V_{\text{aq}}^2 + a_{12} V_{\text{em}}^2 + a_{13} V_{\text{or}}^2 + a_{14} V_{\text{aq}} V_{\text{em}} + a_{15} V_{\text{aq}} V_{\text{or}} + a_{16} V_{\text{em}} V_{\text{or}}, \quad (104)$$

$$A_3 = a_{17} V_{\text{aq}}^2 V_{\text{em}} + a_{18} V_{\text{aq}}^2 V_{\text{or}} + a_{19} V_{\text{em}}^2 V_{\text{aq}} + a_{20} V_{\text{em}}^2 V_{\text{or}} + a_{21} V_{\text{or}}^2 V_{\text{aq}} + a_{22} V_{\text{or}}^2 V_{\text{em}} + a_{23} V_{\text{aq}} V_{\text{em}} V_{\text{or}}, \quad (105)$$

$$A_4 = a_{24} V_{\text{aq}}^2 V_{\text{em}}^2 + a_{25} V_{\text{aq}}^2 V_{\text{or}}^2 + a_{26} V_{\text{em}}^2 V_{\text{or}}^2 + a_{27} V_{\text{aq}}^2 V_{\text{em}} V_{\text{or}} + a_{28} V_{\text{aq}} V_{\text{em}}^2 V_{\text{or}} + a_{29} V_{\text{aq}} V_{\text{em}} V_{\text{or}}^2, \quad (106)$$

$$A_5 = a_{30} V_{\text{aq}}^2 V_{\text{em}}^2 V_{\text{or}} + a_{31} V_{\text{aq}}^2 V_{\text{em}} V_{\text{or}}^2 + a_{32} V_{\text{aq}} V_{\text{em}}^2 V_{\text{or}}^2 + a_{33} V_{\text{aq}}^2 V_{\text{em}}^2 V_{\text{or}}^2, \quad (107)$$

$$A_6 = a_{34} \bar{\theta}_{\text{aq}}^2 + a_{35} \bar{\theta}_{\text{em}}^2 + a_{36} \bar{\theta}_{\text{or}}^2, \quad (108)$$

$$A_7 = a_{37} V_{\text{aq}}^2 \bar{\theta}_{\text{aq}} + a_{38} V_{\text{em}}^2 \bar{\theta}_{\text{em}} + a_{39} V_{\text{or}}^2 \bar{\theta}_{\text{or}} + a_{40} \bar{\theta}_{\text{aq}}^2 V_{\text{aq}} + a_{41} \bar{\theta}_{\text{em}}^2 V_{\text{em}} + a_{42} \bar{\theta}_{\text{or}}^2 V_{\text{or}}, \quad (109)$$

**Table 7**

A reference table detailing where coefficients of Eq. (102) can be found, for each  $\alpha$  investigated. The corresponding values of  $\bar{R}^2$  describing the suitability of these coefficients are given.

| $\alpha$ | Table | $\bar{R}^2$ |
|----------|-------|-------------|
| 0.540    | C.18  | 0.99067     |
| 0.555    | C.19  | 0.99053     |
| 0.570    | C.20  | 0.99072     |
| 0.585    | C.21  | 0.99071     |
| 0.600    | C.22  | 0.99072     |

$$A_8 = a_{43}V_{aq}^2\bar{\theta}_{aq}^2 + a_{44}V_{em}^2\bar{\theta}_{em}^2 + a_{45}V_{or}^2\bar{\theta}_{or}^2, \quad (110)$$

$$A_9 = a_{46}V_{aq}\bar{T}_{aq} + a_{47}V_{em}\bar{T}_{em} + a_{48}V_{or}\bar{T}_{or} + V_{aq}V_{em}V_{or} \times (a_{49}\bar{T}_{aq} + a_{50}\bar{T}_{em} + a_{51}\bar{T}_{or}), \quad (111)$$

$$A_{10} = a_{52}V_{aq}\bar{\theta}_{aq} + a_{53}V_{em}\bar{\theta}_{em} + a_{54}V_{or}\bar{\theta}_{or} + V_{aq}V_{em}V_{or} \times (a_{55}\bar{\theta}_{aq} + a_{56}\bar{\theta}_{em} + a_{57}\bar{\theta}_{or}). \quad (112)$$

For each  $\alpha$ , the values of the coefficients in Eq. (102) have been detailed in Tables C.18–C.22, as described in Table 7. Table 7 also presents the  $\bar{R}^2$  value for each fit. All equations fit the MCNP data very well, and consistently, with  $0.99053 \leq \bar{R}^2 \leq 0.99072$ .

The results of using Eq. (102) to calculate  $\Lambda$  are demonstrated by the dashed lines in Fig. 10. Fig. 10(a) shows the approximated linear relationship between  $\Lambda$  and  $\bar{T}$  for each material. The most notable difference between the MCNP results and results from Eq. (102) in the graph for  $\Lambda$  as a function of emulsion temperature for various emulsion volumes and aqueous and organic phase volumes of 0.0 L and 37.5 L respectively. Here, it can be seen that  $\Lambda$  is significantly underestimated by Eq. (102). Similarly, in Fig. 10(b), it can be seen that  $\Lambda$  is also underestimated for various emulsion volumes and void fractions (when the aqueous and organic phase volumes are 0.0 L and 37.5 L respectively).

Due to the nature of aqueous ingress and draining, and emulsion formation and dissipation, it is highly unlikely that high volumes of emulsion will exist without an aqueous layer existing underneath it. Thus, this difference is expected to have a minimal effect on further analysis.

## 6.2. Reactivity feedbacks

Similarly to  $\Lambda$ , the changes in reactivity,  $\Delta\xi$ , due to the effects of temperature and void fraction can be determined by creating best-fit equations from the MCNP results. Fig. 11 provides examples of how  $\Delta\xi$  varies with temperature and void fraction.

Fig. 11(a) depicts the effect of average phase temperature on  $\Delta\xi$ . For the organic phase,  $\Delta\xi_{th,or}$  decreases linearly with increasing  $\bar{T}_{or}$ . The relationship between  $\Delta\xi_{th,em}$  and  $\bar{T}_{em}$  is similar, though small deviations from linear behaviour are observed, and the overall gradients of the relationships are smaller. Finally, for the three highest aqueous volumes,  $\Delta\xi_{th,aq}$  is seen to increase slightly with  $\bar{T}_{aq}$ . For the lowest aqueous volume, 6.25 L,  $\Delta\xi_{th,aq}$  decreases with increasing  $\bar{T}_{aq}$ .

Fig. 11(b) depicts the effect of average phase void fraction on  $\Delta\xi$ . For all three liquid phases, it can be seen that  $\Delta\xi$  decreases with increasing void fraction. For most cases, this relationship is weakly second order, but for  $\Delta\xi_{th,aq}$  and  $\Delta\xi_{th,em}$ , small fluctuations in the results make the relationship order difficult to observe. Voids present in the organic phase will have the most significant effect on  $\Delta\xi$ , with  $\Delta\xi_{rg,or}$  decreasing to around  $-30$  \$ at an organic void fraction of 0.2.

As with  $\Lambda$ , increasing the temperature or void fraction of the organic phase has the greatest effect on reactivity when compared to equal increases in either the aqueous or emulsion phases. Again, this

difference is mostly due to the higher plutonium concentration in the organic phase, with the absence of aqueous or emulsion phases in the vessel also contributing to this phenomenon.

Again, equations were fitted (using non-linear least squares) to the available data to determine how  $\Delta\xi_{xx,yy}$  could be calculated. These equations take the form:

$$\Delta\xi_{xx,yy} [V_{aq}(t), V_{em}(t), V_{or}(t), \bar{X}_{yy}(t)] = V_{yy} (\bar{X}_{yy}(B_1 + B_2 + B_3 + B_4 + B_5) + \bar{X}_{yy}^2(B_6 + B_7 + B_8 + B_9 + B_{10})) \quad (113)$$

where

$$B_1 = b_1 + b_2V_{aq} + b_3V_{em} + b_4V_{or}, \quad (114)$$

$$B_2 = b_5V_{aq}^2 + b_6V_{em}^2 + b_7V_{or}^2 + b_8V_{aq}V_{em} + b_8V_{aq}V_{or} + b_{10}V_{em}V_{or}, \quad (115)$$

$$B_3 = b_{11}V_{aq}^2V_{em} + b_{12}V_{aq}^2V_{or} + b_{13}V_{em}^2V_{aq} + b_{14}V_{em}^2V_{or} + b_{15}V_{or}^2V_{aq} + b_{16}V_{or}^2V_{em} + b_{17}V_{aq}V_{em}V_{or}, \quad (116)$$

$$B_4 = b_{18}V_{aq}^2V_{em}^2 + b_{19}V_{aq}^2V_{or}^2 + b_{20}V_{em}^2V_{or}^2 + b_{21}V_{aq}^2V_{em}V_{or} + b_{22}V_{aq}^2V_{em}V_{or} + b_{23}V_{aq}V_{em}V_{or}^2, \quad (117)$$

$$B_5 = b_{24}V_{aq}^2V_{em}^2V_{or} + b_{25}V_{aq}^2V_{em}V_{or}^2 + b_{26}V_{aq}V_{em}^2V_{or}^2 + b_{27}V_{aq}^2V_{em}^2V_{or}^2, \quad (118)$$

$$B_6 = b_{28} + b_{29}V_{aq} + b_{30}V_{em} + b_{31}V_{or}, \quad (119)$$

$$B_7 = b_{32}V_{aq}^2 + b_{33}V_{em}^2 + b_{34}V_{or}^2 + b_{35}V_{aq}V_{em} + b_{36}V_{aq}V_{or} + b_{37}V_{em}V_{or}, \quad (120)$$

$$B_8 = b_{38}V_{aq}^2V_{em} + b_{39}V_{aq}^2V_{or} + b_{40}V_{em}^2V_{aq} + b_{41}V_{em}^2V_{or} + b_{42}V_{or}^2V_{aq} + b_{43}V_{or}^2V_{em} + b_{44}V_{aq}V_{em}V_{or}, \quad (121)$$

$$B_9 = b_{45}V_{aq}^2V_{em}^2 + b_{46}V_{aq}^2V_{or}^2 + b_{47}V_{em}^2V_{or}^2 + b_{48}V_{aq}^2V_{em}V_{or} + b_{49}V_{aq}V_{em}^2V_{or} + b_{50}V_{aq}V_{em}V_{or}^2, \quad (122)$$

$$B_{10} = b_{51}V_{aq}^2V_{em}V_{or} + b_{52}V_{aq}^2V_{em}V_{or}^2 + b_{53}V_{aq}V_{em}^2V_{or}^2 + b_{54}V_{aq}^2V_{em}^2V_{or}^2. \quad (123)$$

The subscript 'yy' in Eq. (113) can refer to any of the three materials: aqueous ('aq'), emulsion ('em') or organic ('or'). Similarly, the subscript 'xx' in Eq. (113) can refer to either thermal 'th' or voidage 'rg'. If 'xx' = 'rg',  $\bar{X}_{yy}$  refers to the average void fraction of region 'yy'; if 'xx' = 'th',  $\bar{X}_{yy}$  refers to the average temperature of region 'yy', minus 293.6 K (i.e.  $\bar{X}_{th,yy} = \bar{T}_{yy} - 293.6$ ).

For each  $\alpha$  and  $\Delta\xi_{xx,yy}$ , the values of the coefficients in Eq. (113) have been detailed in Appendix D: Table 8 gives a summary of in exactly which table the coefficients for each reactivity component can be located. In addition, Table 8 details the  $\bar{R}^2$  values describing the goodness of each fit, and Fig. 11 demonstrates the results of using these equations.

As demonstrated by the  $\bar{R}^2$  values in Table 8, the equations for  $\Delta\xi_{xx,yy}$  have signification variation in their goodness of fit. The equations fitted to  $\Delta\xi_{th,aq}$  and  $\Delta\xi_{rg,aq}$  are the least suitable, with  $0.51789 \leq \bar{R}^2 \leq 0.54590$  and  $0.91527 \leq \bar{R}^2 \leq 0.91654$  respectively. Conversely, equations for  $\Delta\xi_{th,or}$  and  $\Delta\xi_{rg,or}$  are the most suitable, with  $\bar{R}^2 \geq 0.99139$ .

The poorness of Eq. (113)'s fit to the data for  $\Delta\xi_{th,aq}$  and  $\Delta\xi_{rg,aq}$  can be observed in Figs. 11(a) and 11(b). The poorness of these fits can be attributed to data fluctuations and the weaker dependencies of  $\Delta\xi_{th,aq}$  and  $\Delta\xi_{rg,aq}$  on temperature and void fraction, as shown in Fig. 11. As Eq. (113) is only second order with respect to  $\bar{T}_{aq}$ , it is unable to recreate the fluctuations seen in the data presented in Fig. 11. However, as the dependency of  $\Delta\xi_{th,aq}$  on  $\bar{T}_{aq}$  is much weaker than that of the other two phases, the results of Eq. (113) are considered suitable for future use.

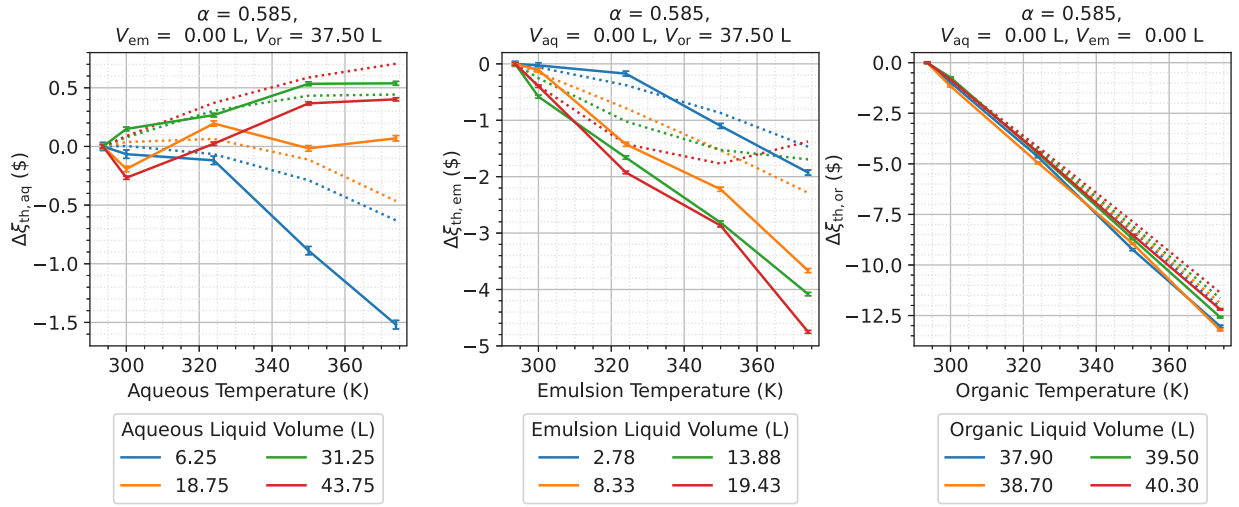
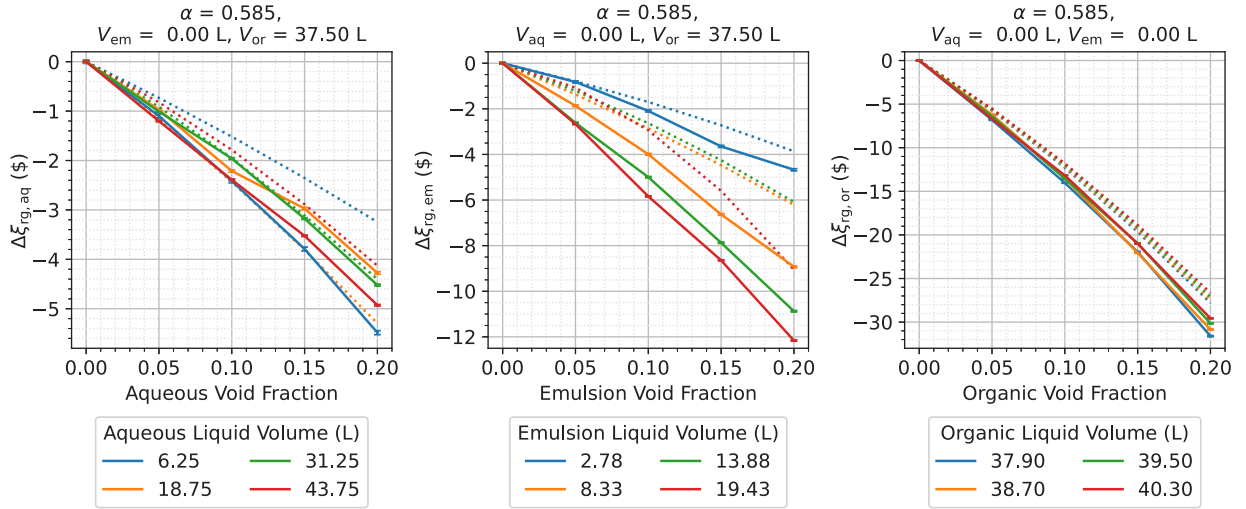
(a) Thermal reactivity feedbacks as a function of each region's  $\bar{T}$ .(b) Voidage reactivity feedbacks as a function of each region's  $\bar{\theta}$ .

Fig. 11. Graphs depicting the dependency of  $\Delta\xi$  on the temperatures and void fractions of each of the three regions, for  $\alpha = 0.585$  and selected region volumes. Solid lines represent data calculated using MCNP, and dotted lines represent data calculated using Eq. (113).

Though the equations fitted to data for  $\Delta\xi_{rg,em}$  have relatively high  $\bar{R}^2$  values (no less than 0.95887), indicating good fits, some deviation from the MCNP data is observed. For example, with aqueous, emulsion and organic volumes of 0.00, 37.50 and 19.43 L respectively, the equation for  $\Delta\xi_{th,em}$  has a turning point at the higher temperatures, whereas the MCNP data shows a continuous decrease in  $\Delta\xi_{th,em}$  with temperature. In addition, when  $\bar{\theta}_{em} = 0.2$ ,  $\Delta\xi_{rg,em}$  should be around  $-12$  \$, whereas Eq. (113) calculates a value of around  $-9$  \$. As it is unlikely that a large volume of emulsion will be present without the presence of the aqueous region, the effect of these inaccuracies will be considered negligible.

## 7. Investigation into the 1970 Windscale nuclear criticality incident configuration

As described in Section 2.2, there are several uncertainties and unknowns associated with the 1970 Windscale nuclear criticality incident. Those investigated in this section are detailed in Table 9, along with the minima and maxima of the investigated ranges.

In this Section, the “effective thickness”,  $\zeta$ , is defined such that a phase “yy” has a mass  $M_{yy} = \pi R^2 \zeta_{yy} \rho_{yy,293.6}$ , where  $R$  is the radius of the cylindrical section of the vessel (30.5 cm) and  $\rho_{yy,293.6}$  is the density of the phase at 293.6 K.

As such, the ranges taken for  $\zeta_{or}(0)$  and  $\zeta_{em,max}$  in Table 9 correspond to initial organic volumes in the range 38.0 L to 41.5 L and maximum emulsion volumes in the range 11.7 L to 22.2 L, at temperatures of 293.6 K. Though these ranges go below the minimum cited in the literature, the previously obtained results indicate that these values may be conducive to a yield of  $\sim 10^{15}$  fissions, as seen during the 1970 Windscale nuclear criticality incident.

### 7.1. System characteristics

Many of the intrinsic phase and system properties were kept constant throughout the investigation; these are presented in Table 10. The details of the properties of each phase are presented in Table 11, where, if a value is given instead of an equation, it is held constant throughout all simulations. Additionally, SOURCES-4C was used to determine the intrinsic neutron sources of the each phase.

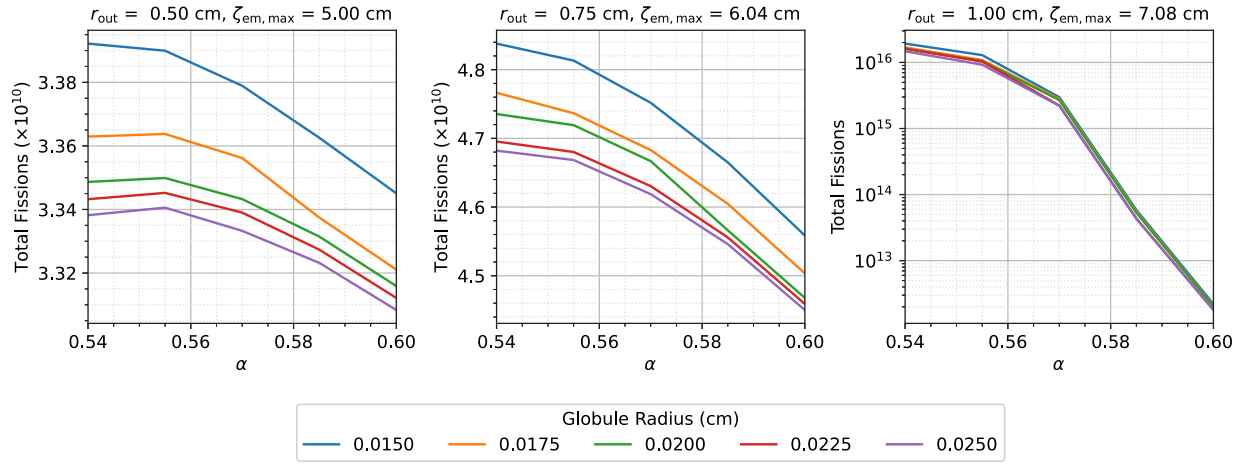


Fig. 12. Three example graphs presenting the variation in total fissions with  $\alpha$  (volume fraction of aqueous solution in the emulsion region), with  $\zeta_{or,max} = 13.34$  cm.

Table 8

A reference table detailing where coefficients of Eq. (113), can be found, for each of the six reactivity feedback components, and for each  $\alpha$  investigated. The corresponding values of  $\bar{R}^2$  describing the suitability of these coefficients are given.

| $\alpha$ | Material | Thermal feedback         |       |             | Voidage feedback         |       |             |
|----------|----------|--------------------------|-------|-------------|--------------------------|-------|-------------|
|          |          | Symbol                   | Table | $\bar{R}^2$ | Symbol                   | Table | $\bar{R}^2$ |
| 0.540    | Aqueous  | $\Delta\epsilon_{th,aq}$ | D.23  | 0.53740     | $\Delta\epsilon_{rg,aq}$ | D.28  | 0.91555     |
|          | Emulsion | $\Delta\epsilon_{th,em}$ | D.23  | 0.96451     | $\Delta\epsilon_{rg,em}$ | D.28  | 0.98260     |
|          | Organic  | $\Delta\epsilon_{th,or}$ | D.23  | 0.99139     | $\Delta\epsilon_{rg,or}$ | D.28  | 0.99254     |
| 0.555    | Aqueous  | $\Delta\epsilon_{th,aq}$ | D.24  | 0.54590     | $\Delta\epsilon_{rg,aq}$ | D.29  | 0.91560     |
|          | Emulsion | $\Delta\epsilon_{th,em}$ | D.24  | 0.96436     | $\Delta\epsilon_{rg,em}$ | D.29  | 0.98312     |
|          | Organic  | $\Delta\epsilon_{th,or}$ | D.24  | 0.99153     | $\Delta\epsilon_{rg,or}$ | D.29  | 0.99260     |
| 0.570    | Aqueous  | $\Delta\epsilon_{th,aq}$ | D.25  | 0.53436     | $\Delta\epsilon_{rg,aq}$ | D.30  | 0.91527     |
|          | Emulsion | $\Delta\epsilon_{th,em}$ | D.25  | 0.95953     | $\Delta\epsilon_{rg,em}$ | D.30  | 0.98171     |
|          | Organic  | $\Delta\epsilon_{th,or}$ | D.25  | 0.99143     | $\Delta\epsilon_{rg,or}$ | D.30  | 0.99250     |
| 0.585    | Aqueous  | $\Delta\epsilon_{th,aq}$ | D.26  | 0.52001     | $\Delta\epsilon_{rg,aq}$ | D.31  | 0.91535     |
|          | Emulsion | $\Delta\epsilon_{th,em}$ | D.26  | 0.95924     | $\Delta\epsilon_{rg,em}$ | D.31  | 0.98300     |
|          | Organic  | $\Delta\epsilon_{th,or}$ | D.26  | 0.99170     | $\Delta\epsilon_{rg,or}$ | D.31  | 0.99251     |
| 0.600    | Aqueous  | $\Delta\epsilon_{th,aq}$ | D.27  | 0.51789     | $\Delta\epsilon_{rg,aq}$ | D.32  | 0.91654     |
|          | Emulsion | $\Delta\epsilon_{th,em}$ | D.27  | 0.95887     | $\Delta\epsilon_{rg,em}$ | D.32  | 0.98286     |
|          | Organic  | $\Delta\epsilon_{th,or}$ | D.27  | 0.99200     | $\Delta\epsilon_{rg,or}$ | D.32  | 0.99247     |

Table 9

Parameters in the 1970 Windscale nuclear criticality incident with uncertainties investigated in the presented research.

| Property                             | Symbol           | Minimum | Maximum | Unit |
|--------------------------------------|------------------|---------|---------|------|
| Aqueous volume fraction in emulsion  | $\alpha$         | 0.540   | 0.600   | –    |
| Maximum emulsion effective thickness | $\zeta_{em,max}$ | 4.0     | 7.6     | cm   |
| Initial organic effective thickness  | $\zeta_{or}(0)$  | 13.0    | 14.2    | cm   |
| Outflow pipe radius                  | $r_{out}$        | 0.5     | 1.0     | cm   |
| Emulsion globule radius              | $r_{glob}$       | 0.15    | 0.25    | mm   |

## 7.2. Results

### 7.2.1. Variation of emulsion composition, emulsion globule size, and outlet pipe radius

Figs. 12–14 depict the variation in total fissions with varying  $\alpha$ ,  $r_{glob}$  and  $r_{out}$ , respectively.

In Fig. 12, it can be seen that, when  $\zeta_{or,max} = 13.34$  cm, the total number of fissions tends to decrease with increasing  $\alpha$ . When  $\zeta_{em,max} = 7.08$  cm, this decrease is significant, with the total fissions decreasing by four orders of magnitude. This indicates that the emulsion composition has a greater effect on the fissions produced during a transient when the emulsion and organic contents are relatively high and low respectively.

Fig. 13 shows that the total number of fissions decreases slightly with increasing  $r_{glob}$ . This is because larger globule radii correspond to

faster emulsion separation times. For example, when  $\zeta_{or,max}$  and  $\zeta_{em,max}$  are 14.03 and 7.08 cm respectively, the emulsion is 60.0% aqueous solution by volume, and the vessel has an outlet radius of 0.5 cm, increasing the globule radius from 0.15 to 0.25 mm decreases the total number of fissions from  $2.942 \times 10^{17}$  to  $2.922 \times 10^{17}$ .

Similarly, as shown in Fig. 14, increasing the outlet radius slightly decreases the total number of fissions, as larger radii correspond to faster aqueous draining rates (and thus faster ejection of fissile material from the system). For example, when  $\zeta_{or,max}$  and  $\zeta_{em,max}$  are 13.34 and 5.00 cm respectively, the emulsion is 60.0% aqueous solution by volume, and the emulsion globule radii are 0.15 mm respectively, increasing the vessel's outlet radius from 0.5 to 1.0 cm decreases the total number of fissions from  $3.34 \times 10^{10}$  to  $3.03 \times 10^{10}$ . In addition, it can be seen that increasing the quantity of emulsion and organic phases in the system reduces the effect of outlet radius on total fission yield.

### 7.2.2. Variation of total reactivity, fission yield and organic temperature

The left hand graph of Fig. 15 describes how the total fission yield varies with the maximum reactivity reached during the transient. As shown, the fission yield remains below  $10^{11}$  fissions for maximum reactivities below 0 \$. For maximum reactivities between 0 and 1 \$ (for which the system becomes delayed critical but never prompt supercritical) the fission yield increases exponentially.

Fig. 15 depicts the variation in the maximum temperature of the organic phase with the total fission yield of the transient. As shown, the temperature increases exponentially with increasing fission yield.

In Section 5, equations used to determine the organic phase's density and dynamic viscosity were valid for specified temperature ranges, both of which had an upper limit of 333.15 K. From 15, it can be seen that when the total fission yield exceeds  $10^{17}$ , the maximum organic temperature exceeds 333.15 K. As such, the behaviour of transients with a total yield of over  $10^{17}$  fissions may be simulated inaccurately. For example, the effects of boiling will not be accounted for within the presented model.

However, as the 1970 Windscale nuclear criticality incident is known to yield less than  $10^{17}$  fissions, using the presented mathematical and computational models to simulate cases similar to this can be considered appropriate, though care must be taken when interpreting results which exceed a fission yield of  $10^{17}$ , or a maximum reactivity  $\geq 1$  \$.

### 7.2.3. Variation of maximum emulsion and organic volumes

Fig. 16 depicts the total fission yield and maximum reactivity of transients with various  $\zeta_{em,max}$  and  $\zeta_{or,max}$ , with  $\alpha = 0.570$ ,  $r_{glob} = 0.20$  mm and  $r_{out} = 0.75$  cm.

**Table 10**

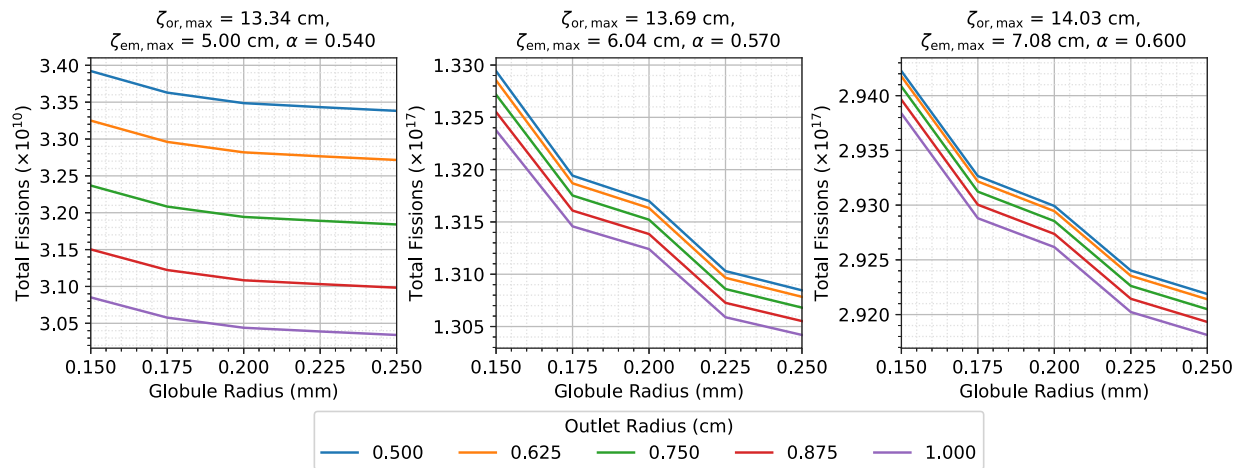
Basic system properties for the presented case study.

| Property                                                   | Symbol             | Value    | Unit                |
|------------------------------------------------------------|--------------------|----------|---------------------|
| Pu concentration in the aqueous phase                      | $c_{Pu}$           | 7.0      | $\text{g L}^{-1}$   |
| Free nitric acid concentration in the aqueous phase        | $c_{HN}$           | 7.0      | $\text{mol L}^{-1}$ |
| Mass fraction of $\text{HNO}_3$ in the aqueous phase       | $m_{HN}$           | 0.34959  | –                   |
| Mass fraction of $\text{H}_2\text{O}$ in the aqueous phase | $1 - m_{HN}$       | 0.65041  | –                   |
| Mass fraction of TBP in the organic phase                  | $m_{TBP}$          | 0.28433  | –                   |
| Mass fraction of OK in the organic phase                   | $m_{C_{11}H_{20}}$ | 0.71567  | –                   |
| External temperature                                       | $T_{ext}$          | 293.6    | K                   |
| System pressure                                            | $p$                | 0.101325 | MPa                 |
| Maximum aqueous phase mass                                 | $M_{aq,max}$       | 63.800   | kg                  |
| Time taken to reach $M_{em,max}$                           | $t_{form}$         | 240.0    | s                   |
| Time taken for aqueous vacuum-lifted                       | $t_{fill}$         | 242.0    | s                   |

**Table 11**

Thermophysical and intrinsic properties of each phase.

| Property                                  | Symbol    | Phase                 | Value or Equation(s)                                 |
|-------------------------------------------|-----------|-----------------------|------------------------------------------------------|
| Density                                   | $\rho$    | Aqueous               | Eq. (72)                                             |
|                                           |           | Emulsion              | Eq. (33)                                             |
|                                           |           | Organic               | Eq. (73)                                             |
|                                           |           | Air                   | $1.191 \text{ kg m}^{-3}$                            |
|                                           |           | Steel                 | $7930.0 \text{ kg m}^{-3}$                           |
| Dynamic viscosity                         | $\mu$     | Aqueous               | Eqs. (76), (74) and (75)                             |
|                                           |           | Emulsion              | Eq. (32)                                             |
|                                           |           | Organic               | Eq. (77)                                             |
|                                           |           | Air                   | $18.14 \text{ mPa s}$                                |
| Surface tension                           | $\sigma$  | Aqueous               | Eq. (81)                                             |
|                                           |           | Emulsion              | Eq. (81)                                             |
|                                           |           | Organic               | Eqs. (82), (83) and (84)                             |
| Specific heat capacity                    | $C_p$     | Aqueous               | $2965 \text{ J kg}^{-1} \text{ K}^{-1}$              |
|                                           |           | Emulsion              | Eq. (80)                                             |
|                                           |           | Organic               | $1568 \text{ J kg}^{-1} \text{ K}^{-1}$              |
|                                           |           | Steel                 | $497 \text{ J kg}^{-1} \text{ K}^{-1}$               |
| HTCs between the vessel and liquid phases | $h_{int}$ | Aqueous               | $440 \text{ W m}^2 \text{ K}^{-1}$                   |
|                                           |           | Emulsion              | Eq. (90)                                             |
|                                           |           | Organic               | $110 \text{ W m}^2 \text{ K}^{-1}$                   |
| HTCs between fluid phases                 | $h_{sur}$ | Aqueous to emulsion   | Eq. (97)                                             |
|                                           |           | Aqueous to organic    | $96.200 \text{ W m}^2 \text{ K}^{-1}$                |
|                                           |           | Emulsion to organic   | Eq. (98)                                             |
|                                           |           | Organic to plenum gas | $6.470 \text{ W m}^2 \text{ K}^{-1}$                 |
| Radiolytic gas generation coefficient     | $G_{rg}$  | Aqueous               | $1.21565 \times 10^{-10} \text{ m}^3 \text{ J}^{-1}$ |
|                                           |           | Emulsion              | Eq. (101)                                            |
|                                           |           | Organic               | $8.63915 \times 10^{-9} \text{ m}^3 \text{ J}^{-1}$  |
| Intrinsic neutron source                  | $s'''_n$  | Aqueous               | $1.244 \text{ neutrons cm}^{-3} \text{ s}^{-1}$      |
|                                           |           | Emulsion              | Table 12                                             |
|                                           |           | Organic               | $11.000 \text{ neutrons cm}^{-3} \text{ s}^{-1}$     |

**Fig. 13.** Three example graphs presenting the variation in total fissions with  $r_{glob}$  (emulsion globule radius).

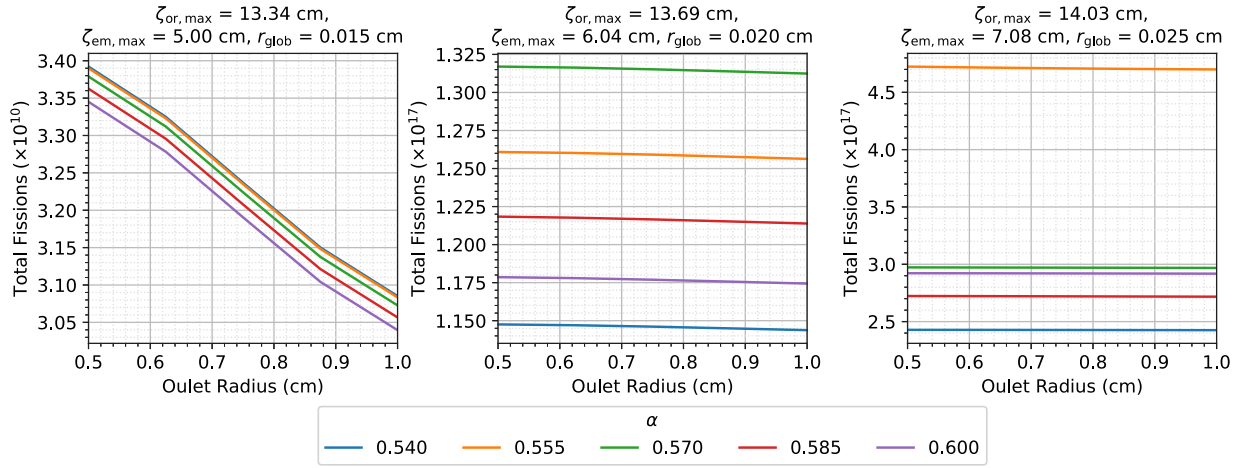


Fig. 14. Three example graphs presenting the variation in total fissions with  $r_{out}$  (outlet pipe radius).

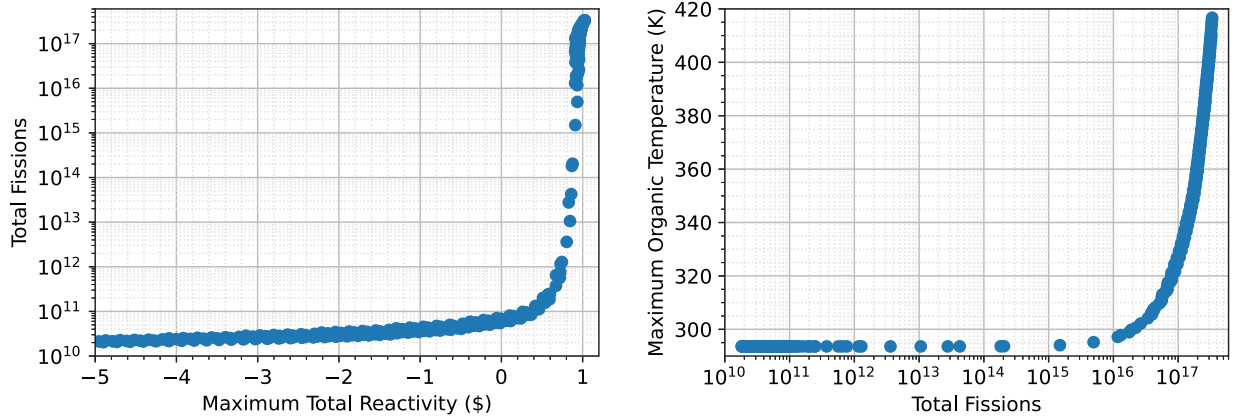


Fig. 15. The variation in total number of fissions with maximum reactivity (left) and maximum organic temperature with total number of fissions (right).

Table 12

Emulsion phase intrinsic neutron sources for the five compositions investigated.

| $\alpha$ | $s'''_{n,em}$ (neutrons $\text{cm}^{-3} \text{s}^{-1}$ ) |
|----------|----------------------------------------------------------|
| 0.540    | 5.431                                                    |
| 0.555    | 5.288                                                    |
| 0.570    | 5.145                                                    |
| 0.585    | 5.002                                                    |
| 0.600    | 4.860                                                    |

The left-hand plot of Fig. 16 shows the narrow window in which the total number of fissions is comparable to the  $10^{15}$  produced during the 1970 Windscale nuclear criticality incident. A maximum effective organic thickness of 13.9 cm (corresponding to a volume of 40.62 L at 293.6 K) or above will result in at least  $6.6 \times 10^{16}$  fissions, even with emulsion thicknesses as low as 4.0 cm.

The right-hand plot of Fig. 16 shows part of the reason for this narrow window: there is a large range of configurations that result in a maximum reactivity  $\geq 1.0$  \$, i.e. the system, at some point, becomes prompt critical. When a system is prompt critical, neutron losses are balanced by the production of prompt neutrons alone, and thus the neutron population increases exponentially in time. Thus, the configurations with a maximum reactivity  $\geq 1.0$  \$ are also those for which the most fissions ( $\geq 10^{17}$ ) occur, as also demonstrated by Fig. 15.

For reactivities between 0.0 and 1.0 \$, the fissile system is in the delayed critical state: neutron losses are not balanced by the production of prompt neutrons, and delayed neutrons are needed to sustain the chain reaction. For these states, the neutron population increases at

Table 13

Details of possible configurations of the 1970 Windscale nuclear criticality incident.

| Effective thickness (cm) |          | Total fission yield   |                       |
|--------------------------|----------|-----------------------|-----------------------|
| Organic                  | Emulsion | Min.                  | Max.                  |
| 13.4                     | 6.6–6.8  | $3.61 \times 10^{12}$ | $2.23 \times 10^{16}$ |
| 13.5                     | 6.0–6.1  | $4.24 \times 10^{13}$ | $1.17 \times 10^{16}$ |
| 13.6                     | 5.4–5.5  | $2.04 \times 10^{14}$ | $1.75 \times 10^{16}$ |

a slower rate. As can be seen in Figs. 15 and 16, the systems with comparable fission yield to the 1970 Windscale nuclear criticality incident are those with a maximum reactivity between 0.8 and 1.0 \$.

### 7.3. Possible configurations of the 1970 Windscale nuclear criticality incident

As detailed in Section 2, the 1970 Windscale nuclear criticality incident yielded around  $10^{15}$  fissions, with 39.0 to 41.5 L of organic solvent in the vessel, and a maximum emulsion band 5.0 to 7.6 cm thick.

From Fig. 16, it can be seen that, for vessel contents within the above ranges, the combinations of effective emulsion and organic volumes detailed in Table 13 result in a fission yield comparable to that of the 1970 Windscale nuclear criticality incident.

Using these combinations, further simulations with varying  $\alpha$ ,  $r_{glob}$  and  $r_{out}$  re-enforce the observations of Section 7.2.1: the radius of the vessel's outlet pipe has a negligible effect on fission yield, the globule

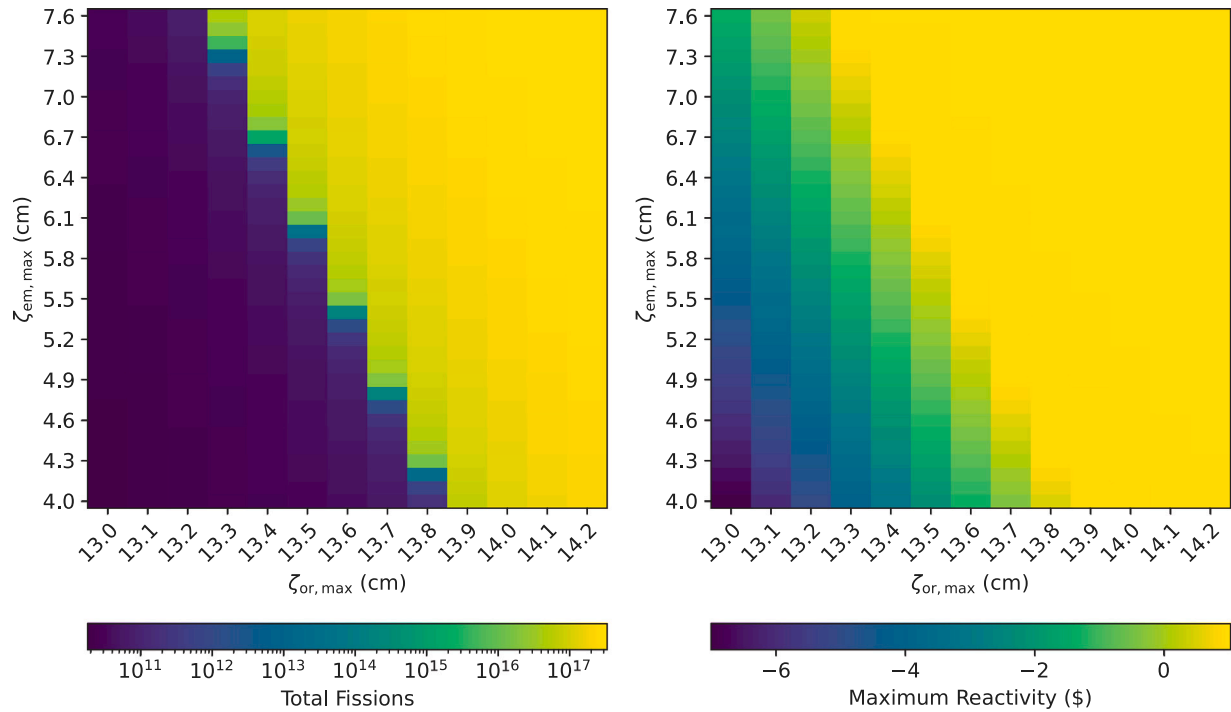


Fig. 16. Pseudocolour plots presenting the total number of fissions (left) and maximum reactivity (right) for varying effective organic and emulsion thicknesses.

**Table 14**  
Details of possible configurations of the 1970 Windscale nuclear criticality incident.

| Effective thickness (cm) |          | $\alpha$ | $r_{\text{glob}}$ (mm) |       | $r_{\text{out}}$ (cm) |      | Total fission yield   |                       |
|--------------------------|----------|----------|------------------------|-------|-----------------------|------|-----------------------|-----------------------|
| Organic                  | Emulsion |          | Min.                   | Max.  | Min.                  | Max. | Min.                  | Max.                  |
| 13.4                     | 6.7      | 0.555    | 0.150                  | 0.250 | 0.50                  | 1.00 | $5.73 \times 10^{15}$ | $8.85 \times 10^{15}$ |
| 13.4                     | 6.7      | 0.570    | 0.150                  | 0.250 | 0.50                  | 1.00 | $1.19 \times 10^{15}$ | $1.70 \times 10^{15}$ |
| 13.4                     | 6.8      | 0.600    | 0.150                  | 0.175 | 0.50                  | 1.00 | $5.51 \times 10^{14}$ | $6.24 \times 10^{14}$ |
| 13.5                     | 6.1      | 0.585    | 0.150                  | 0.250 | 0.50                  | 1.00 | $3.42 \times 10^{15}$ | $4.60 \times 10^{15}$ |
| 13.6                     | 5.4      | 0.540    | 0.150                  | 0.250 | 0.50                  | 1.00 | $6.36 \times 10^{14}$ | $9.19 \times 10^{14}$ |
| 13.6                     | 5.4      | 0.555    | 0.150                  | 0.250 | 0.50                  | 1.00 | $5.32 \times 10^{14}$ | $7.63 \times 10^{14}$ |
| 13.6                     | 5.5      | 0.600    | 0.150                  | 0.250 | 0.50                  | 1.00 | $3.55 \times 10^{15}$ | $4.66 \times 10^{15}$ |

radius had a minor effect on the fission yield, and  $\alpha$  could significantly effect the fission yield.

From these further simulations, Table 14 gives the system properties which are conducive to a fission yield comparable to the 1970 Windscale nuclear criticality incident.

In Table 14, effective organic thicknesses of 13.4, 13.5 and 13.6 cm correspond to volumes of 39.16, 39.45 and 39.75 L (at 293.6 K) respectively. Thus the total fission yield of transients of this nature is very sensitive to the volume of the organic phase: for a fission yield of  $\sim 10^{15}$ , there is a 0.6 L range of suitable organic volumes. Due to the high plutonium content of the organic phase, this is to be expected: a 0.6 L increase in organic volume would correspond to a 33 g increase in plutonium content in the vessel.

As detailed in Sections 3 and 5, there is a large degree of uncertainty associated with many of the parameters involved in the modelling and calculation of this transient. It is also possible that the ranges described in Section 2.2 are inaccurate or imprecise. In addition, the possibility of stochastic effects have not been considered. Thus, exact specifications of the emulsion and organic volumes present in the 1970 Windscale nuclear criticality incident are impossible to determine using the analysis presented in this paper.

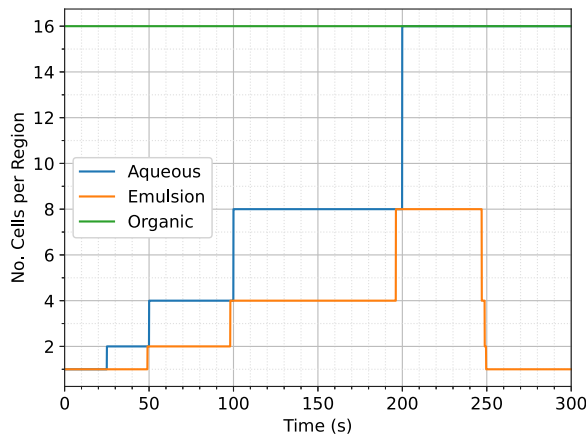
Nevertheless, the research presented in this paper has lead to a handful of possible configurations of the 1970 Windscale nuclear criticality incident, as demonstrated by the results of Table 14.

In particular, the system in which the vessel had an outlet pipe radius of 0.5 cm, and contained 13.6 cm (39.75 L) of organic solvent, that underwent a transient that formed an emulsion that was 54% aqueous by volume, contained organic globules of radii 0.15 mm, and reached a maximum effective thickness of 5.4 cm, yielded a total of  $9.19 \times 10^{14}$  fissions, which is the closest result to the  $10^{15}$  reported to have occurred during the 1970 Windscale nuclear criticality incident.

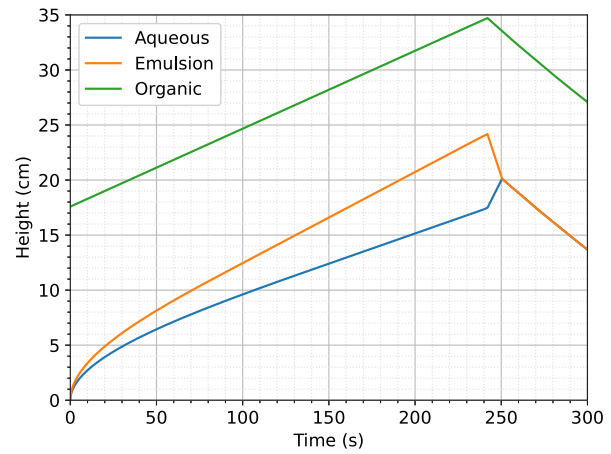
## 8. Selected case study parameters and results

In this section, two case studies are presented. The first aims to give an understanding and insight into the 1970 Windscale nuclear criticality incident, and the second presents the behaviour of a transient yielding a total of  $\sim 10^{17}$  fissions.

For both case studies, Table 10 details the basic system properties, Table 15 gives the case specific parameters, and Table 11 provides information on the calculation of the aqueous, emulsion and organic phase properties.



(a) The number of axial cells in the discretisation of the thermal energy and radiolytic gas ODEs.



(b) The heights of each fissile liquid as a function of time.

Fig. 17. Discretisation and geometry of the Case Study 1 transient.

Table 15

Parameters in the 1970 Windscale nuclear criticality incident with uncertainties investigated in the presented research.

| Property                             | Symbol           | Value        |              | Unit |
|--------------------------------------|------------------|--------------|--------------|------|
|                                      |                  | Case Study 1 | Case Study 2 |      |
| Aqueous volume fraction in emulsion  | $\alpha$         | 0.570        | 0.570        | –    |
| Maximum emulsion effective thickness | $\zeta_{em,max}$ | 6.7          | 5.0          | cm   |
| Initial organic effective thickness  | $\zeta_{or}(0)$  | 13.4         | 13.8         | cm   |
| Outflow pipe radius                  | $r_{out}$        | 0.75         | 0.75         | cm   |
| Emulsion globule radius              | $r_{glob}$       | 0.20         | 0.20         | mm   |

### 8.1. Case Study 1: Results

Fig. 17 depicts the axial discretisation of the thermal energy and radiolytic gas ODEs, alongside the heights of each of the fissile liquids. It can be seen that as the heights (and thus thicknesses) of the aqueous and emulsion layers increase, so do the number of cells in each of the discretised layers. Additionally, Fig. 17(b) gives an overall depiction of the aqueous entry and drainage, and emulsion formation and dissipation.

As shown in Fig. 18(a), the total fission energy produced during the transient was 49.56 kJ, as a result of  $1.49 \times 10^{15}$  fissions. This is predominantly due to a power spike of 47.76 kW at 242.00 s, which is a result of the system reaching criticality at around 223.35 s (as shown in Fig. 18(b)). The system becomes critical due to the changing geometry of the system (increased aqueous content and emulsion formation) acting as a positive feedback mechanism. A maximum reactivity of 0.90957 \$ is reached before the emulsion dissipation and aqueous drainage begins, causing a sharp decrease in reactivity.

As shown in Fig. 19(a), there is only a slight temperature increase, which occurs at around 240.00 s, coinciding with the time at which the maximum reactivity is reached. Due to the lower specific heat capacity of the organic phase, the temperature of this region increases the most, but only by around 0.47 K. After around 244 s, heat transfer between the three phases can be seen, before the temperatures of the aqueous and organic phases reach a constant level.

Similarly, Fig. 19(b) shows that there is only a slight increase in void fraction across the three phases. Due to the differences in radiolytic gas generation coefficient, the organic phase experiences the highest void fraction, peaking at 0.00105 at 242.00 s. Generation of radiolytic gas is noticeable from around 239.00 s, when the total reactivity reaches around 0.86 \$. After 242.00 s, the advection of radiolytic gas bubbles

dominates over their generation, reducing the void fractions in the three regions.

As shown in 18(b), the slight increases in system temperature and void fraction from around 240.00 s slightly decreases the total reactivity of the system.

### 8.2. Case Study 2: Results

The variation of the axial discretisation of the thermal energy and radiolytic gas ODEs, and the heights of each of the fissile liquids, is very similar to that of the first case study. However, as shown in Fig. 20, the behaviour of the transient is notably different.

Fig. 20(a) shows that the transient yields a total of 3.69 MJ of fission energy, due to the occurrence of  $1.11 \times 10^{17}$  fissions.

In Fig. 20(b), it can be seen that, in this Case, the criticality is reached at an earlier time of 195.55 s, due to the configuration containing a higher plutonium content (due to more of the organic phase being present in the vessel) than the first case study.

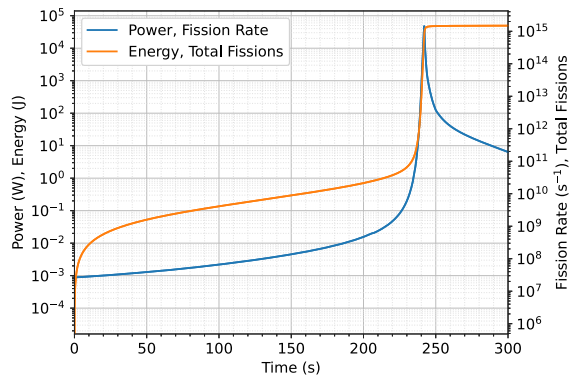
As shown in Fig. 20(b), between 195.60 and 214.25 s, the total reactivity increases to a maximum of 0.93689 \$, due to the continued addition of aqueous solution to the system. After 214.25 s, reactivity starts to decrease to 0.0 \$, as the increase in negative reactivity feedback due to the increase in system temperature and void fraction.

Fig. 20(b) also shows that the onset of emulsion dissipation and aqueous drainage at 242.00 s acts as the dominant negative feedback mechanism, terminating criticality.

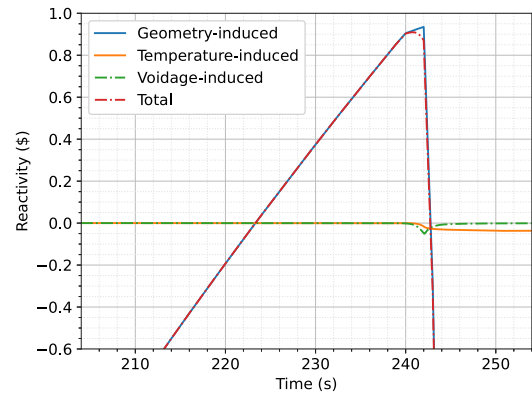
Due to the maintenance of criticality between 195.60 and 242.00 s, the power, which spikes to 0.25 MW at 217.36 s, remains relatively high (above 67 kW) until 242.00 s, as shown in Fig. 20(a).

The period of criticality between 214.25 s and 242.00 s causes a steady increase in temperature in the three phases, as depicted in Fig. 21(a). As with Case Study 1, the organic phase reaches the highest temperature of 332.12 K at 242.00 s. From 242.00 s (as the reactivity decreases below critical), heat transfer between the three phases becomes noticeable.

In Fig. 21(b), it can be seen that the void fractions within the three liquid phases also increase from 214.25 s. Again, the highest void fraction, 0.00309, is observed in the organic phase. As criticality is maintained between 214.25 s and 242.00 s, the void fractions decrease more gradually when compared to Case Study 1. This is because the maintenance of criticality results in the constant generation of voids, such that their advection out of the system is not as dominant as it is in Case Study 1.

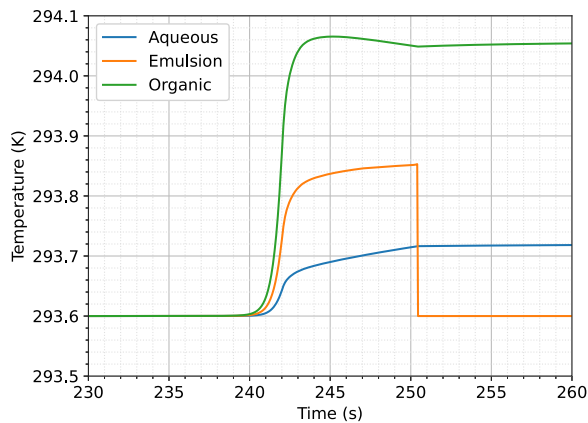


(a) The fission power and fission energy profiles during the case study transient.

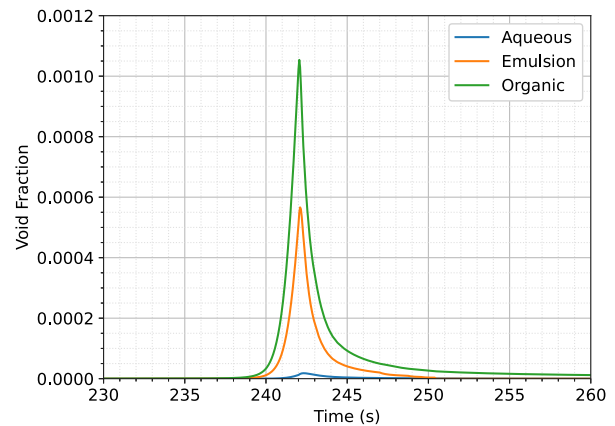


(b) The total reactivity of the system, and its constituents, as a function of time.

**Fig. 18.** The variation in fission power, fission energy, and reactivity components over time during the Case Study 1 transient.

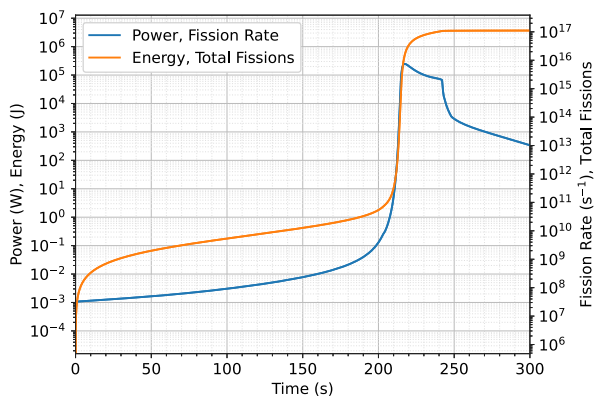


(a) The temperature variation of the fissile liquids during the transient.

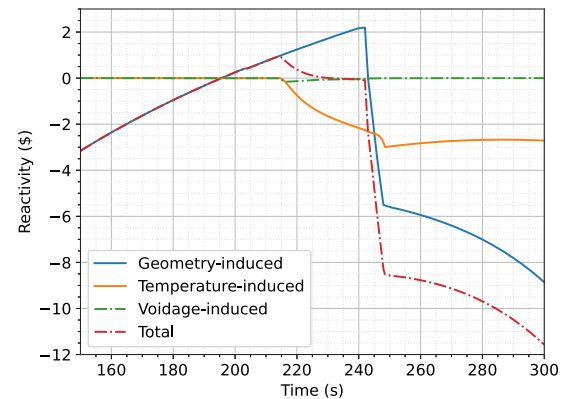


(b) The void fraction of each fissile liquid layer during the transient.

**Fig. 19.** The variation in temperature and void fraction during the Case Study 1 transient.

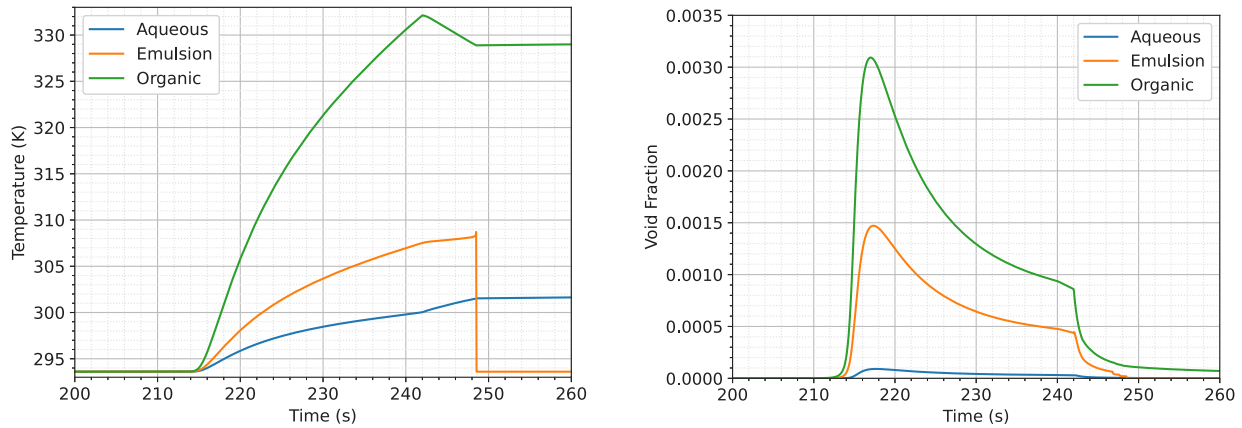


(a) The fission power and fission energy profiles during the case study transient.



(b) The total reactivity of the system, and its constituents, as a function of time.

**Fig. 20.** The variation in fission power, fission energy, and reactivity components over time during the Case Study 2 transient.



(a) The temperature variation of the fissile liquids during the transient.

(b) The void fraction of each fissile liquid layer during the transient.

Fig. 21. The variation in temperature and void fraction during the transient of the Case Study 2.

## 9. Conclusion

This paper presented a novel methodology for the analysis of transient nuclear criticality excursions in layered aqueous-emulsion-organic plutonium nitrate systems, and the application of this methodology to describe the nuclear criticality incident that occurred at the Windscale Works in 1970.

A thorough description of the 1970 Windscale criticality incident was given in Section 2, in which it was noted that there were numerous parametric and phenomenological uncertainties associated with this transient. For example, the exact volume of organic solvent trapped in the transfer vessel, and the properties of the emulsion that formed between the aqueous and organic phases.

In Section 3, transient model phenomenology for a layered aqueous-emulsion-organic fissile liquid system was described. Point kinetics equations were coupled with 1D thermal hydraulics and radiolytic gas models, and equations governing the mass flow rates of each of the layers were presented. The CP method was used to determine the scalar neutron flux distribution and geometry-induced reactivity of the system (via the calculation of  $k_{eff}$ ) at prescribed time intervals. The Richardson-Zaki equations were used to approximate the emulsion dissipation speed. Heat transfer between the fissile layers, the fissile layers and the vessel, and from the fissile system to the plenum gas was modelled. The generation, advection and mixing of radiolytic gas were considered, as was turbulent mixing of the three fissile layers.

Section 5 discussed the determination of the variables required to solve the system of equations described in Section 3, including the thermophysical properties of aqueous, emulsion and organic fissile liquids, heat transfer coefficients and radiolytic gas generation coefficients. It was noted that many approximations and assumptions were required, meaning there is some degree of uncertainty associated with these variables and thus in the developed model.

In Section 6, the effect of increased system temperature and void fraction on the mean neutron generation time and reactivity was investigated.

Increasing the average temperature and void fraction of the system increased the mean neutron generation time. It was determined that increasing the average temperature of any of the three phases from 293.6 to 374.0 K had less of an effect on the mean neutron generation time than increasing the void fraction from 0.0 to 0.2. In addition, variations in the average temperature and void fraction in the organic phase had a larger effect on the mean neutron generation time than the same increases in either the emulsion or the aqueous phases.

Similarly, increasing the average temperature of any of the three phases from 293.6 to 374.0 K had less of an effect on the system's reactivity than increasing the void fraction from 0.0 to 0.2. Again, variations in the average temperature and void fraction in the organic phase had a larger effect on the reactivity than the same increases in either the emulsion or the aqueous phases. For all phases, increasing the void fraction decreased the system's reactivity. Increasing the temperature of the emulsion and organic phases reduced reactivity, however increasing the temperature of a large volume of aqueous solution could increase the system's reactivity.

Section 7 presented the overall results (e.g. total fission yield, maximum reactivity) from using the transient phenomenological model to simulate criticality transients similar to that of the 1970 Windscale nuclear criticality incident. Variations in properties such as organic and emulsion volume, emulsion composition and globule radius, and vessel outlet pipe radius were made: it was determined that the radius of the outlet pipe had a negligible effect on the fission yield of a transient, whereas fission yield was extremely sensitive to the volumes of the emulsion and organic phases.

It was determined the more energetic transients yielding over  $10^{17}$  fissions, would result in the temperature of the organic phase exceeding the range of validity in the equations for this phase's density and dynamic viscosity. In these cases, the presented model is likely to produce erroneous results as phenomena such as liquid boiling has not been accounted for in the presented model.

Using the results from Section 7, it was postulated that the vessel of the 1970 Windscale nuclear criticality incident contained 39.75 L of organic solvent and an outlet radius of 0.5 cm, and during the transient the emulsion was 54% aqueous phase by volume, contained organic globules that had radii of 0.15 mm, and reached a maximum thickness of 5.4 cm. Using these parameters, the presented model calculated a total fission yield of  $9.19 \times 10^{14}$  fissions.

In Section 8, the results of two case study transients were presented in more detail. The case with a total yield of  $1.49 \times 10^{15}$  fissions had a maximum total reactivity of 0.90957 \$, and saw minor increases in system temperature and void fraction. Due to the differences in the properties of the aqueous and organic phases, the organic phase experienced the highest temperature (294.07 K) and void fraction (0.00105).

Conversely, the second case, which yielded  $1.11 \times 10^{17}$  fissions, saw a maximum reactivity of 0.93689 \$. The organic temperature and void fraction reach 332.12 K and 0.00309 respectively, due to a sustained period of criticality, lasting around 47 s. In this example, the effect

of increased temperature and void fraction on system reactivity was observed: at around 210 s, the overall reactivity was decreased 1 to 0 \$ due to the temperature- and voidage-induced reactivity feedback mechanisms.

## CRediT authorship contribution statement

**J.R. Daniels:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing. **M.M.R. Williams:** Conceptualization, Methodology, Supervision, Writing – review & editing. **M.D. Eaton:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

In accordance with EPSRC funding requirements, all supporting data used to create figures in this paper may be accessed at the following DOI: <https://doi.org/10.17632/vg5gn73yhh.1> which will be activated upon acceptance.

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## Appendix A. Emulsion dynamics

Most references (e.g. [Tadros \(2016\)](#)) define types of emulsion based on droplet size, as shown in [Table A.16](#). The largest type of emulsion is a macroemulsion, with a maximum size of the order 5000 nm. [Evans \(1984a\)](#), reports that the emulsion formed during the 1970 Windscale criticality excursion was “coarse”, and [Ganguli and Ganguli \(2003\)](#) reports that, for a coarse dispersion, droplets are visible to the naked eye. In the available literature regarding the 1970 Windscale nuclear criticality incident ([British Nuclear Fuels Limited, no date](#); [Chalmers, 1971](#); [Daniels et al., 1971](#); [Evans, 1984a,b](#); [Hughes, 1972](#); [McLaughlin et al., 2000](#)), there is no indication of the exact (or estimated) emulsion globule size or size distribution. Thus, an average globule radius will be assumed, and uncertainty quantification in this area will be performed.

The concentration of an emulsion can be described by  $\psi$ , the volume fraction of the dispersed phase within a continuous phase (equivalent to the packing fraction of the globules in the emulsion). As detailed in [Section 7](#), it was likely that the transfer vessel contained 39 L of organic liquid (and 50 L of aqueous liquid), with the organic phase being dispersed in the continuous aqueous phase. Thus, in this section, the volume fraction of the emulsion that existed during the 1970 Windscale nuclear criticality incident is assumed to be 0.43820.

**Table A.16**

A comparison of different emulsion length scales.

| Emulsion type     | Typical length scales (nm) |
|-------------------|----------------------------|
| Micellar or Micro | 5–50                       |
| Nano              | 20–100                     |
| Macro             | 100–5000                   |

### A.1. Creaming

Creaming is the process by which emulsion globules rise due to a difference in density between the globules and the continuous phase. For very dilute macroemulsions ( $\psi < 0.01$ ), droplets can be considered “isolated”: there will be no globule–globule interaction, and, assuming droplets are rigid spheres, Stoke’s law can be used to obtain an expression for the terminal velocity,  $U_0$ , of an isolated emulsion droplet, as shown in [Eq. \(A.1\)](#) ([Chanamai and McClements, 2000](#); [Robins, 2000](#); [Tadros, 2013](#)). Alternatively, [Eq. \(A.2\)](#) has been used to describe the velocity,  $U$ , of the globule if it is not rigid (i.e. has a viscosity). However, [Tadros \(2016\)](#) notes that this prediction does not agree well with experimental data, perhaps due to the “neglect of interfacial viscosity contribution”.

$$U_0 = \frac{2}{9} \frac{(\rho_{\text{cont}} - \rho_{\text{glob}})gr_{\text{glob}}^2}{\mu_{\text{cont}}} \quad (\text{A.1})$$

$$U = \frac{2}{3} \frac{(\rho_{\text{cont}} - \rho_{\text{glob}})gr_{\text{glob}}^2}{\mu_a} \frac{\mu_{\text{cont}} + \mu_{\text{glob}}}{3\mu_{\text{cont}} + 2\mu_{\text{glob}}} \quad (\text{A.2})$$

In the above equations, subscripts ‘cont’ and ‘glob’ refer to the continuous phase and discontinuous phase (globules), respectively.  $\rho$  and  $\mu$  represent density and dynamic viscosity, respectively,  $g$  is the acceleration due to gravity, and  $r_{\text{glob}}$  is the globule radius.

As the emulsion becomes more concentrated, the hydrodynamic force between separate droplets will increase, and needs to be taken into account. [Tadros \(2013\)](#) notes that, for  $0.1 < \psi < 0.15$ , the velocity of the droplets can be expressed using [Eq. \(A.3\)](#):

$$U = U_0(1 - 6.55\psi). \quad (\text{A.3})$$

For  $\psi > 0.155$ , the rate of creaming becomes a more complex function ([Chanamai and McClements, 2000](#); [Tadros, 2013](#)), and, at present, no empirical correlations exist to determine the creaming rate of a concentrated emulsion. A review of the literature has revealed no correlations for creaming in concentrated emulsions, which would be the most applicable to the 1970 Windscale emulsion.

### A.2. Coalescence and flocculation

Coalescence is the process by which individual globules combine into a larger particle. It involves the interaction between particles of the dispersed phase with themselves, as well as with the continuous phase ([Yeoh et al., 2014](#)). [Tadros \(2016\)](#) describes a simplified method for calculating the combined rate of coalescence and flocculation of emulsion globules: [Eq. \(A.4\)](#) can be used to describe the number of globules,  $n$ , which have not yet coalesced or combined into flocs at a time  $t$ :

$$n(t) = \frac{n_0}{(1 + kn_0t)^2} \quad (\text{A.4})$$

$$\text{where } k = \frac{4}{3} \frac{k_B T_B}{\mu} \quad (\text{A.5})$$

where  $n_0$  is the initial number of globules per cubic metre,  $k_B$  is the Boltzmann constant ( $1.38 \times 10^{-23} \text{ J K}^{-1}$ ),  $T_B$  is the temperature of the system and  $\mu$  is the dynamic viscosity of the continuous phase.

Using  $\mu = 9.35 \times 10^{-4} \text{ Pa s}$  and  $T_B = 293.6 \text{ K}$ ,  $k$  is calculated to be  $5.78 \times 10^{-18} \text{ m}^3 \text{ s}^{-1}$ . Assuming a globule radius of 0.025 cm and  $\psi$  equal to 0.43820, it is calculated that there are  $6.70 \times 10^9$  globules per

cubic metre of emulsion ( $n_0$ ). The time taken to decrease the bubble population by 0.01% can be evaluated by substituting  $n(t) = 0.9999n_0$  into Eq. (A.4), rearranging, and solving for time  $t$ . Using the previously calculated  $k$  and  $n_0$ , it is calculated that it would take around 1291 s (or 21.5 min) for  $n$  to decrease to  $0.9999n_0$ , which two orders of magnitude higher than the observed emulsion separation time during the 1970 Windscale criticality incident. Using this simplistic model, it can be assumed that the effects of coalescence and flocculation are negligible.

Yeoh et al. (2014) provides a more in-depth description of droplet coalescence. Collisions due to turbulence, wake entrainment and velocity gradients are considered, along with the “efficiency” of the collisions (i.e. how likely it is that a collision will result in coalescence). Yeoh et al. (2014) derived equations and correlations to describe the coalescence rate for each of the potential coalescence mechanisms. These equations vary in complexity, and when combined they require detailed knowledge of the system at hand, including droplet size and surface tension, and turbulent length scales and kinetic energy dissipation. As there is limited data available regarding the 1970 Windscale nuclear criticality incident, these models are deemed too detailed for use in the presented research.

Flocculation is the aggregation of emulsion globules, without the rupture of the interfacial layer (Tadros, 2016). Assuming droplets act as rigid spheres, the velocity of flocs in a dilute, flocculated emulsion can be expressed in a similar manner to Eq. (A.1):

$$U_{\text{floc}} = \frac{2}{9} \frac{r_{\text{floc}}^2 (\rho_{\text{floc}} - \rho_{\text{cont}}) g}{\mu_{\text{cont}}}, \quad (\text{A.6})$$

where variables and subscripts retain their previously-defined meaning. The radius and density of the floc,  $r_{\text{floc}}$  and  $\rho_{\text{floc}}$  respectively, will depend on parameters such as the volume fraction of the droplets in the floc, and the number of droplets present in the floc (Chanamai and McClements, 2000). The actual process of flocculation is a contribution of many surface forces, including van der Waals and electrostatic forces (Ivanov et al., 1999), which, similar to coalescence, require a detailed knowledge of the system to evaluate, and thus will not be considered in the presented research.

### A.3. Ostwald ripening

Regarding emulsions, Ostwald ripening is a diffusive transfer process: smaller droplets dissolve and redeposit onto larger droplets, a phenomenon which is observed in dilute emulsions (Langevin, 2020; Leal-Calderon et al., 2007), and thus will not be considered as a possible mechanism which drove emulsion separation during the 1970 Windscale criticality excursion.

### A.4. Particle sedimentation models

In lieu of a suitable correlation that specifically describes the separation of a concentrated emulsion with relatively large globule size, particle sedimentation models have been considered. For the purposes of the presented research, it will be assumed that emulsion globules dissipate in a similar manner to that in which particles undergo sedimentation, with the caveat that the emulsion globules will rise instead of sink.

The most widely-used empirical equation for describing the speed of sedimentation of solid-liquid homogeneous suspensions is the Richardson–Zaki Equation (Richardson and Zaki, 1954, 1997). To determine this correlation, Richardson and Zaki (1954, 1997) determined the drag force on a constituent particle in a suspension, and accounted for this force by applying a correction factor to Stoke’s velocity. The resulting Richardson–Zaki Equation is given in Eq. (A.7):

$$U_{\text{RZ}} = U_0(1 - \psi)^n \quad (\text{A.7})$$

where  $U_{\text{RZ}}$  is the hindered settling velocity of a suspension,  $U_0$  is Stoke’s velocity (calculated using Eq. (A.1)),  $\psi$  is the volume fraction of particles in the suspension, and  $n$  is an experimentally-determined constant,

**Table B.17**

The discretisation (energy group boundaries) used for the 30-group energy structure.

| Group | Upper energy boundary (eV) | Group | Upper energy boundary (eV) |
|-------|----------------------------|-------|----------------------------|
| 1     | $1.96 \times 10^7$         | 16    | $9.91 \times 10^0$         |
| 2     | $6.70 \times 10^6$         | 17    | $5.35 \times 10^0$         |
| 3     | $2.23 \times 10^6$         | 18    | $3.30 \times 10^0$         |
| 4     | $9.07 \times 10^5$         | 19    | $2.36 \times 10^0$         |
| 5     | $3.02 \times 10^5$         | 20    | $1.93 \times 10^0$         |
| 6     | $8.23 \times 10^4$         | 21    | $1.59 \times 10^0$         |
| 7     | $2.93 \times 10^4$         | 22    | $1.37 \times 10^0$         |
| 8     | $1.11 \times 10^4$         | 23    | $1.17 \times 10^0$         |
| 9     | $3.53 \times 10^3$         | 24    | $1.10 \times 10^0$         |
| 10    | $1.43 \times 10^3$         | 25    | $1.02 \times 10^0$         |
| 11    | $6.77 \times 10^2$         | 26    | $9.10 \times 10^{-1}$      |
| 12    | $1.49 \times 10^2$         | 27    | $6.25 \times 10^{-1}$      |
| 13    | $5.56 \times 10^1$         | 28    | $3.91 \times 10^{-1}$      |
| 14    | $3.73 \times 10^1$         | 29    | $2.48 \times 10^{-1}$      |
| 15    | $2.26 \times 10^1$         | 30    | $1.34 \times 10^{-1}$      |

dependent on the Reynolds number of a single droplet (Richardson and Zaki, 1954, 1997).

Walstra and Oortwijn (1975), who investigated the effect of fat globule size and concentration on creaming in pasteurised milk, fitted their experimental data to the Richardson–Zaki Equation, with  $n = 8.6$ . Using data from Buscall and White (1987) and Davis and Birdsell (1988), Barker and Grimson (1990) reported that a value of  $n$  in the range  $5 < n < 7$  is suitable for either semi-dilute or concentrated, non-flocculated suspensions. Frising et al. (2008) reported that, for concentrated water-in-oil emulsions, a suitable range for  $n$  was  $2.3 < n < 5.0$ . Rowe (1987) formed an empirical relation for determining  $n$ , as shown in Eq. (A.8):

$$n = 2.35 \frac{2 + 0.175 Re^{3/4}}{1 + 0.175 Re^{3/4}} \quad (\text{A.8})$$

where  $Re$ , the Reynolds number for an isolated suspended particle, is defined as:

$$Re = \frac{2\rho_{\text{cont}} U_0 r_{\text{glob}}}{\mu_{\text{cont}}} \quad (\text{A.9})$$

In the above,  $U_0$  is as defined in Eq. (A.1), and all other symbols are as previously defined.

The Richardson–Zaki equation, combined with the correlation of Rowe (1987), can now be applied to the emulsion expected to be present in the 1970 Windscale nuclear criticality incident. For a globule of radius 0.025 cm,  $U_0$  is calculated to be  $4.68 \text{ cm s}^{-1}$ . With a continuous phase of density  $1276 \text{ kg m}^{-3}$ , and viscosity  $9.35 \times 10^{-4} \text{ Pa s}$ , the Reynolds number of an isolated particle is calculated, using Eq. (A.9), to be 31.89, giving a value of  $n$  equal to 3.05. Thus,  $U_{\text{RZ}}$  of this example system is  $0.80 \text{ cm s}^{-1}$ : an emulsion of thickness 6.8 cm will take 8.45 s to dissipate. As this result is within the 5–10 s dissipation time recorded to have occurred during the 1970 Windscale nuclear criticality incident, the Richardson–Zaki equation, combined with the correlation of Rowe (1987) will be used to model the emulsion separation in the presented research.

## Appendix B. Energy group structures

See Table B.17.

## Appendix C. Coefficients and parameters for determining $\Lambda$ using Eq. (102)

See Tables C.18–C.22.

## Appendix D. Coefficients for determining $\xi_{\text{xx,yy}}$ using Eq. (113)

See Tables D.23–D.32.

**Table C.18**For  $\alpha = 0.540$ , the values of  $a_i$  in Eq. (102). These coefficients result in an  $\bar{R}^2$  of 0.99067.

| $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        |
|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| 1   | 3.61250E+01  | 13  | -1.96788E+03 | 25  | -2.14961E+07 | 37  | -5.30902E+03 | 49  | -6.39037E+00 |
| 2   | 3.61026E+03  | 14  | -9.94956E+04 | 26  | -3.11304E+07 | 38  | 2.31107E+03  | 50  | 2.55995E+01  |
| 3   | 1.01420E+03  | 15  | -1.43424E+05 | 27  | -2.38432E+07 | 39  | -1.48167E+04 | 51  | 1.50937E+02  |
| 4   | -3.40338E+02 | 16  | -4.37450E+04 | 28  | -7.97178E+06 | 40  | 3.11408E+01  | 52  | 4.74801E+02  |
| 5   | -2.16714E-04 | 17  | 1.10111E+06  | 29  | 7.70945E+05  | 41  | 1.09455E+03  | 53  | 7.34143E+02  |
| 6   | -6.82740E-04 | 18  | 1.90493E+06  | 30  | 7.05528E-06  | 42  | 4.02812E+03  | 54  | 1.88385E+03  |
| 7   | -5.69221E-03 | 19  | 8.80421E+05  | 31  | 2.13258E-05  | 43  | 1.84991E+03  | 55  | -2.45053E+05 |
| 8   | -1.98954E-01 | 20  | 2.17501E+06  | 32  | 1.85440E-05  | 44  | -1.78408E+04 | 56  | 1.57620E+04  |
| 9   | -2.35829E-01 | 21  | 1.55141E+06  | 33  | -4.98088E-07 | 45  | -3.48357E+04 | 57  | -9.98740E+04 |
| 10  | -2.51510E+01 | 22  | 4.29952E+05  | 34  | 7.82339E-01  | 46  | 1.19249E-01  |     |              |
| 11  | -4.59635E+04 | 23  | 2.00410E+06  | 35  | 8.50507E-01  | 47  | 2.86113E-01  |     |              |
| 12  | -4.11761E+04 | 24  | -5.94751E+06 | 36  | -6.11221E+01 | 48  | 5.27312E-01  |     |              |

**Table C.19**For  $\alpha = 0.555$ , the values of  $a_i$  in Eq. (102). These coefficients result in an  $\bar{R}^2$  of 0.99053.

| $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        |
|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| 1   | 5.59381E+01  | 13  | 1.14529E+04  | 25  | -7.67201E+06 | 37  | -5.15811E+03 | 49  | 3.39454E-02  |
| 2   | 2.11343E+03  | 14  | -7.66390E+04 | 26  | -2.35391E+07 | 38  | -3.05098E+03 | 50  | 3.15074E+01  |
| 3   | 6.77887E+02  | 15  | -6.43965E+04 | 27  | -1.81444E+07 | 39  | -3.26255E+04 | 51  | 1.45021E+02  |
| 4   | -1.36926E+03 | 16  | -3.30948E+04 | 28  | -7.10849E+06 | 40  | -7.66550E+00 | 52  | 4.71965E+02  |
| 5   | -6.37174E-04 | 17  | 9.08025E+05  | 29  | -7.04544E+05 | 41  | 1.58675E+02  | 53  | 8.32853E+02  |
| 6   | -6.99113E-04 | 18  | 8.80597E+05  | 30  | 2.41422E-05  | 42  | -2.16789E+03 | 54  | 3.09174E+03  |
| 7   | -6.21797E-03 | 19  | 7.75190E+05  | 31  | 2.40863E-05  | 43  | 2.17410E+03  | 55  | -2.46455E+05 |
| 8   | -1.50974E-01 | 20  | 1.69883E+06  | 32  | 3.14444E-05  | 44  | 2.70101E+04  | 56  | 3.55706E+04  |
| 9   | -7.29893E-01 | 21  | 5.06629E+05  | 33  | 0.00000E+00  | 45  | 5.70763E+04  | 57  | -9.26831E+04 |
| 10  | -4.54181E+01 | 22  | 3.95309E+05  | 34  | 9.15594E-01  | 46  | 1.41464E-01  |     |              |
| 11  | -2.70947E+04 | 23  | 1.43555E+06  | 35  | 3.77502E+00  | 47  | 2.73414E-01  |     |              |
| 12  | -3.25563E+04 | 24  | -6.18919E+06 | 36  | 4.19210E+01  | 48  | 5.41632E-01  |     |              |

**Table C.20**For  $\alpha = 0.570$ , the values of  $a_i$  in Eq. (102). These coefficients result in an  $\bar{R}^2$  of 0.99072.

| $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        |
|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| 1   | 9.06671E+01  | 13  | 3.40553E+04  | 25  | 3.96678E+07  | 37  | -4.69416E+03 | 49  | -1.85940E+01 |
| 2   | -1.60612E+03 | 14  | 2.43692E+04  | 26  | -1.61847E+07 | 38  | 3.77098E+03  | 50  | -1.61455E+00 |
| 3   | -2.52662E+02 | 15  | 1.23536E+05  | 27  | 2.79645E+07  | 39  | -1.09392E+03 | 51  | 1.18500E+02  |
| 4   | -3.14972E+03 | 16  | -6.81371E+03 | 28  | -4.24039E+06 | 40  | 1.55109E+02  | 52  | 4.52829E+02  |
| 5   | -7.77597E-05 | 17  | -9.25904E+05 | 29  | 8.14998E+06  | 41  | 1.06277E+03  | 53  | 6.95782E+02  |
| 6   | -3.69569E-04 | 18  | -2.86933E+06 | 30  | 9.04907E-06  | 42  | 1.11761E+04  | 54  | 8.45691E+02  |
| 7   | -3.03674E-03 | 19  | 1.35084E+05  | 31  | 2.31343E-05  | 43  | -9.08157E+02 | 55  | -2.68791E+05 |
| 8   | 2.91965E-01  | 20  | 1.23697E+06  | 32  | 2.20268E-05  | 44  | -1.81958E+04 | 56  | 3.72678E+04  |
| 9   | -7.33191E-02 | 21  | -1.86564E+06 | 33  | 1.53166E-06  | 45  | -1.28373E+05 | 57  | -8.97365E+04 |
| 10  | -5.60261E+00 | 22  | 3.27312E+05  | 34  | -1.97630E+00 | 46  | 1.37674E-01  |     |              |
| 11  | 4.71434E+04  | 23  | -1.43303E+06 | 35  | 1.00836E-01  | 47  | 2.74283E-01  |     |              |
| 12  | -2.00272E+04 | 24  | 5.24035E+06  | 36  | -1.96459E+02 | 48  | 4.64585E-01  |     |              |

**Table C.21**For  $\alpha = 0.585$ , the values of  $a_i$  in Eq. (102). These coefficients result in an  $\bar{R}^2$  of 0.99071.

| $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        |
|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| 1   | 5.57290E+01  | 13  | 1.11170E+04  | 25  | 3.31624E+06  | 37  | -5.82139E+03 | 49  | -2.28963E+01 |
| 2   | 1.24553E+03  | 14  | -4.74721E+04 | 26  | -2.69278E+07 | 38  | -5.29160E+03 | 50  | -2.82011E+01 |
| 3   | 1.14265E+03  | 15  | -2.15014E+04 | 27  | -4.29445E+06 | 39  | -5.52134E+02 | 51  | 1.31582E+02  |
| 4   | -1.36622E+03 | 16  | -5.92681E+04 | 28  | -7.17787E+06 | 40  | -1.88369E+02 | 52  | 5.23415E+02  |
| 5   | 2.26612E-05  | 17  | 3.51986E+05  | 29  | 1.30066E+06  | 41  | 7.75770E+01  | 53  | 8.53904E+02  |
| 6   | 2.13544E-04  | 18  | -1.00378E+04 | 30  | 2.03946E-05  | 42  | 1.26356E+04  | 54  | 8.04724E+02  |
| 7   | -4.90390E-03 | 19  | 5.99260E+05  | 31  | 2.58534E-05  | 43  | 3.87318E+03  | 55  | -2.63553E+05 |
| 8   | -4.66047E-01 | 20  | 2.13420E+06  | 32  | 4.31975E-05  | 44  | 3.16538E+04  | 56  | 3.21948E+04  |
| 9   | -3.52136E-01 | 21  | -1.87643E+04 | 33  | 2.32991E-06  | 45  | -1.51385E+05 | 57  | -9.92166E+04 |
| 10  | -4.93546E+00 | 22  | 7.69915E+05  | 34  | 2.20606E+00  | 46  | 1.22333E-01  |     |              |
| 11  | -9.07030E+03 | 23  | 6.59885E+05  | 35  | 2.40674E+00  | 47  | 2.51998E-01  |     |              |
| 12  | -4.22729E+04 | 24  | -2.44158E+06 | 36  | -2.18229E+02 | 48  | 5.18970E-01  |     |              |

**Table C.22**For  $\alpha = 0.600$ , the values of  $a_i$  in Eq. (102). These coefficients result in an  $\bar{R}^2$  of 0.99072.

| $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        | $i$ | $a_i$        |
|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| 1   | 6.96616E+01  | 13  | 1.95326E+04  | 25  | 3.65572E+07  | 37  | -4.40257E+03 | 49  | -2.00128E+01 |
| 2   | -6.08621E+02 | 14  | -3.84241E+04 | 26  | -7.16396E+06 | 38  | -8.16159E+03 | 50  | 2.87116E+01  |
| 3   | 1.21202E+03  | 15  | 7.29601E+04  | 27  | 2.02741E+07  | 39  | -7.13191E+03 | 51  | 1.68786E+02  |
| 4   | -2.03758E+03 | 16  | -6.87795E+04 | 28  | -1.66236E+07 | 40  | 2.04327E+02  | 52  | 4.34544E+02  |
| 5   | -2.29816E-04 | 17  | -6.12197E+05 | 29  | -1.89280E+07 | 41  | -2.93494E+02 | 53  | 9.47526E+02  |
| 6   | -7.47214E-04 | 18  | -2.61406E+06 | 30  | 1.71633E-05  | 42  | 8.87711E+03  | 54  | 1.26069E+03  |
| 7   | -6.43832E-03 | 19  | 7.34267E+05  | 31  | 2.98850E-05  | 43  | -2.48157E+03 | 55  | -2.57725E+05 |
| 8   | 2.62033E-02  | 20  | 9.97393E+05  | 32  | 3.63452E-05  | 44  | 4.13032E+04  | 56  | 2.61958E+04  |
| 9   | -9.73263E-01 | 21  | -1.22183E+06 | 33  | 0.00000E+00  | 45  | -9.52101E+04 | 57  | -8.08879E+04 |
| 10  | -1.30616E+01 | 22  | 9.74476E+05  | 34  | -1.29104E-01 | 46  | 1.24595E-01  |     |              |
| 11  | 4.19006E+04  | 23  | 1.19848E+06  | 35  | 4.61700E+00  | 47  | 2.45001E-01  |     |              |
| 12  | -2.52273E+04 | 24  | 2.23784E+06  | 36  | -1.55870E+02 | 48  | 5.42982E-01  |     |              |

**Table D.23**For  $\alpha = 0.540$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta\epsilon_{\text{th},yy}^{\text{e}}$ .

| yy | $i$ | $b_i$        | $i$ | $b_i$        | $i$ | $b_i$        | $i$ | $b_i$        | $i$ | $b_i$        |
|----|-----|--------------|-----|--------------|-----|--------------|-----|--------------|-----|--------------|
| aq | 1   | -1.45184E+02 | 12  | 5.26024E+06  | 23  | -3.51825E+08 | 34  | 1.16864E+03  | 45  | -2.13375E+05 |
|    | 2   | 7.53718E+03  | 13  | 4.68772E+06  | 24  | -9.50538E+09 | 35  | 8.21841E+03  | 46  | 5.71386E+05  |
|    | 3   | 1.64750E+04  | 14  | 1.22405E+07  | 25  | 2.83453E+09  | 36  | 3.76060E+03  | 47  | 1.20227E+06  |
|    | 4   | 7.43944E+03  | 15  | 4.81149E+06  | 26  | -5.27616E+09 | 37  | 9.34750E+03  | 48  | 4.03284E+06  |
|    | 5   | -1.05045E+05 | 16  | 9.88342E+06  | 27  | 2.02246E+11  | 38  | -9.01923E+04 | 49  | 1.22183E+06  |
|    | 6   | -3.01866E+05 | 17  | 3.20954E+07  | 28  | 1.70591E+00  | 39  | -4.43751E+04 | 50  | 4.48617E+06  |
|    | 7   | -9.51615E+04 | 18  | 7.51347E+07  | 29  | -7.14463E+01 | 40  | -8.43145E+04 | 51  | 6.71086E+07  |
|    | 8   | -7.03821E+05 | 19  | -6.59873E+07 | 30  | -1.89274E+02 | 41  | -1.42043E+05 | 52  | -4.18458E+07 |
|    | 9   | -3.80569E+05 | 20  | -1.06883E+08 | 31  | -8.99227E+01 | 42  | -4.87585E+04 | 53  | 3.39016E+07  |
|    | 10  | -8.12562E+05 | 21  | -3.12307E+08 | 32  | 8.49078E+02  | 43  | -1.12558E+05 | 54  | -1.72611E+09 |
|    | 11  | 7.67447E+06  | 22  | 6.34350E+07  | 33  | 3.50685E+03  | 44  | -3.92031E+05 |     |              |
| em | 1   | -9.30819E+01 | 12  | 6.16050E+05  | 23  | -2.95126E+08 | 34  | 1.55410E+03  | 45  | 1.07410E+07  |
|    | 2   | 2.73163E+03  | 13  | 2.45413E+07  | 24  | 3.47587E+10  | 35  | 1.97782E+04  | 46  | 5.72874E+04  |
|    | 3   | 1.47527E+04  | 14  | 2.45679E+07  | 25  | 1.20145E+10  | 36  | 5.48610E+03  | 47  | 5.81575E+06  |
|    | 4   | 4.29954E+03  | 15  | 4.92069E+04  | 26  | 1.42356E+10  | 37  | 1.69730E+04  | 48  | 1.73498E+07  |
|    | 5   | -3.88401E+04 | 16  | 9.92628E+06  | 27  | -5.39904E+11 | 38  | -3.66189E+05 | 49  | 2.56127E+07  |
|    | 6   | -4.70001E+05 | 17  | 2.65008E+07  | 28  | 2.13716E+00  | 39  | -4.65078E+04 | 50  | 1.17010E+07  |
|    | 7   | -5.32750E+04 | 18  | -6.21463E+08 | 29  | -1.19684E+02 | 40  | -5.17954E+05 | 51  | -5.44334E+08 |
|    | 8   | -6.28586E+05 | 19  | -1.10484E+06 | 30  | -3.14753E+02 | 41  | -4.34215E+05 | 52  | -2.08182E+08 |
|    | 9   | -6.14868E+04 | 20  | -3.26007E+08 | 31  | -1.15968E+02 | 42  | -5.80418E+04 | 53  | -3.15301E+08 |
|    | 10  | -7.54546E+05 | 21  | -7.58727E+08 | 32  | 1.54638E+03  | 43  | -2.28114E+05 | 54  | 7.20728E+09  |
|    | 11  | 1.44010E+07  | 22  | -1.15944E+09 | 33  | 8.09444E+03  | 44  | -9.69698E+05 |     |              |
| or | 1   | -4.39827E+01 | 12  | -3.79763E+05 | 23  | 7.92284E+07  | 34  | 1.97824E+02  | 45  | 5.03318E+05  |
|    | 2   | 1.91372E+03  | 13  | 2.60050E+06  | 24  | -1.06197E+09 | 35  | 2.19260E+03  | 46  | 1.14470E+05  |
|    | 3   | 2.96852E+03  | 14  | -2.45902E+05 | 25  | -2.94043E+08 | 36  | 6.53129E+02  | 47  | -4.56553E+05 |
|    | 4   | 5.62054E+02  | 15  | 1.60959E+06  | 26  | -4.94858E+09 | 37  | 1.64691E+03  | 48  | 6.98116E+05  |
|    | 5   | -2.24610E+04 | 16  | -1.62677E+06 | 27  | 6.17562E+10  | 38  | -2.86652E+04 | 49  | 1.37334E+06  |
|    | 6   | -5.26726E+04 | 17  | -2.32540E+06 | 28  | 4.91664E-01  | 39  | -5.54553E+03 | 50  | -4.92720E+05 |
|    | 7   | -2.47027E+04 | 18  | -3.28205E+07 | 29  | -2.58931E+01 | 40  | -3.82555E+04 | 51  | -1.43367E+07 |
|    | 8   | -1.42890E+05 | 19  | -3.54358E+07 | 30  | -4.18002E+01 | 41  | -3.12283E+04 | 52  | 9.75839E+06  |
|    | 9   | 6.41901E+03  | 20  | 8.52855E+07  | 31  | -1.66180E+01 | 42  | -8.50328E+03 | 53  | 3.63737E+07  |
|    | 10  | 1.31977E+03  | 21  | 5.11460E+07  | 32  | 3.38551E+02  | 43  | 2.07865E+03  | 54  | -6.26229E+08 |
|    | 11  | 1.76726E+06  | 22  | 5.16131E+07  | 33  | 7.24846E+02  | 44  | -7.01164E+04 |     |              |

**Table D.24**For  $\alpha = 0.555$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta \xi_{\text{th},yy}^z$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | -1.84167E+02 | 12 | 6.30527E+06  | 23 | 4.34763E+08  | 34 | 1.30956E+03  | 45 | -6.64589E+06 |
|    | 2  | 1.04496E+04  | 13 | -3.55157E+07 | 24 | -3.70635E+10 | 35 | -8.45698E+02 | 46 | 8.67004E+05  |
|    | 3  | 1.76563E+02  | 14 | -3.02591E+07 | 25 | -7.81078E+09 | 36 | 5.77937E+03  | 47 | -6.45780E+06 |
|    | 4  | 9.38325E+03  | 15 | 6.61851E+06  | 26 | -3.58723E+10 | 37 | -5.14935E+02 | 48 | -3.33465E+06 |
|    | 5  | -1.26595E+05 | 16 | -3.78212E+06 | 27 | 5.69755E+11  | 38 | 3.44678E+04  | 49 | -2.76164E+07 |
|    | 6  | 4.48038E+05  | 17 | -2.28280E+07 | 28 | 1.96737E+00  | 39 | -6.85465E+04 | 50 | -3.55698E+06 |
|    | 7  | -1.19372E+05 | 18 | 5.83235E+08  | 29 | -1.12451E+02 | 40 | 4.09610E+05  | 51 | 4.38826E+08  |
|    | 8  | 2.41283E+05  | 19 | -7.86599E+07 | 30 | -2.12274E+01 | 41 | 3.85130E+05  | 52 | 6.48995E+07  |
|    | 9  | -5.25655E+05 | 20 | 4.87757E+08  | 31 | -1.02105E+02 | 42 | -7.35990E+04 | 53 | 4.48500E+08  |
|    | 10 | 1.36742E+05  | 21 | 4.50605E+08  | 32 | 1.34314E+03  | 43 | 3.05267E+04  | 54 | -7.02019E+09 |
|    | 11 | -5.84047E+06 | 22 | 2.30155E+09  | 33 | -5.49752E+03 | 44 | 1.53767E+05  |    |              |
| em | 1  | -2.16572E+02 | 12 | 4.09638E+07  | 23 | -1.56337E+09 | 34 | 1.62737E+03  | 45 | 1.70435E+07  |
|    | 2  | 2.29404E+04  | 13 | 2.92554E+07  | 24 | 8.52946E+10  | 35 | 1.94367E+04  | 46 | 6.85728E+06  |
|    | 3  | 6.71118E+03  | 14 | 1.13763E+06  | 25 | 6.02619E+10  | 36 | 1.41781E+04  | 47 | 1.33647E+06  |
|    | 4  | 1.18569E+04  | 15 | 1.95327E+07  | 26 | 3.26830E+10  | 37 | 5.64483E+03  | 48 | 4.63638E+07  |
|    | 5  | -6.82431E+05 | 16 | 6.18000E+06  | 27 | -1.36848E+12 | 38 | -7.31038E+05 | 49 | 2.68859E+07  |
|    | 6  | 6.63106E+02  | 17 | 9.86799E+07  | 28 | 1.81745E+00  | 39 | -4.50754E+05 | 50 | 1.94047E+07  |
|    | 7  | -1.67819E+05 | 18 | -1.31614E+09 | 29 | -2.34474E+02 | 40 | -4.09757E+05 | 51 | -1.09834E+09 |
|    | 8  | -1.57585E+06 | 19 | -6.27752E+08 | 30 | -8.83632E+01 | 41 | -7.70701E+04 | 52 | -7.29055E+08 |
|    | 9  | -1.32164E+06 | 20 | -3.72437E+07 | 31 | -1.09494E+02 | 42 | -2.12862E+05 | 53 | -4.34142E+08 |
|    | 10 | -3.97595E+05 | 21 | -3.80386E+09 | 32 | 7.36987E+03  | 43 | -8.84918E+04 | 54 | 1.74912E+10  |
|    | 11 | 6.02543E+07  | 22 | -1.96664E+09 | 33 | 1.04798E+03  | 44 | -1.23427E+06 |    |              |
| or | 1  | -9.32777E+01 | 12 | -7.44415E+05 | 23 | 1.99774E+08  | 34 | -1.60003E+02 | 45 | 8.03131E+05  |
|    | 2  | 4.43606E+03  | 13 | 5.20481E+06  | 24 | -2.70971E+08 | 35 | 4.06671E+03  | 46 | -3.59314E+05 |
|    | 3  | 7.02395E+03  | 14 | 2.19324E+06  | 25 | -2.91972E+09 | 36 | 8.50624E+02  | 47 | -7.39720E+05 |
|    | 4  | 1.16971E+03  | 15 | -1.75872E+06 | 26 | -5.11479E+09 | 37 | 2.26057E+03  | 48 | 3.47820E+05  |
|    | 5  | -5.47216E+04 | 16 | -2.29266E+06 | 27 | 8.32555E+10  | 38 | -5.24664E+04 | 49 | 1.14504E+06  |
|    | 6  | -1.03306E+05 | 17 | 1.78944E+06  | 28 | 1.01309E+00  | 39 | -3.95368E+03 | 50 | -2.59630E+06 |
|    | 7  | 2.28966E+04  | 18 | -6.63122E+07 | 29 | -5.23298E+01 | 40 | -6.18425E+04 | 51 | -5.24156E+06 |
|    | 8  | -3.50868E+05 | 19 | 1.80531E+07  | 30 | -7.87041E+01 | 41 | -3.58634E+04 | 52 | 4.78112E+07  |
|    | 9  | 4.95237E+03  | 20 | 6.16781E+07  | 31 | -2.68276E+01 | 42 | 1.99287E+04  | 53 | 6.60844E+07  |
|    | 10 | -1.25745E+05 | 21 | 3.01088E+07  | 32 | 6.73708E+02  | 43 | 2.47203E+04  | 54 | -1.19255E+09 |
|    | 11 | 4.43363E+06  | 22 | -4.27833E+07 | 33 | 1.18901E+03  | 44 | -7.24108E+04 |    |              |

**Table D.25**For  $\alpha = 0.570$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta \xi_{\text{th},yy}^z$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | -2.67779E-01 | 12 | -2.18753E+06 | 23 | 3.14163E+08  | 34 | -7.00002E+01 | 45 | -1.44382E+06 |
|    | 2  | -1.57627E+03 | 13 | -9.54771E+06 | 24 | -6.57167E+09 | 35 | -4.30079E+03 | 46 | -7.36156E+05 |
|    | 3  | -4.85157E+03 | 14 | -3.43008E+06 | 25 | -4.16004E+09 | 36 | -2.57557E+03 | 47 | -7.95367E+05 |
|    | 4  | -1.79680E+02 | 15 | -1.43592E+06 | 26 | -4.68791E+09 | 37 | -3.15245E+03 | 48 | -3.29207E+06 |
|    | 5  | 3.77895E+04  | 16 | -4.40898E+06 | 27 | 6.69733E+10  | 38 | 6.46525E+04  | 49 | -5.38877E+06 |
|    | 6  | 8.51384E+04  | 17 | -2.21890E+07 | 28 | -7.36621E-02 | 39 | 5.59863E+04  | 50 | -3.19978E+06 |
|    | 7  | 4.72353E+03  | 18 | 1.35329E+08  | 29 | 4.91245E+01  | 40 | 1.05847E+05  | 51 | 7.01673E+07  |
|    | 8  | 3.78317E+05  | 19 | 3.08961E+07  | 30 | 4.94925E+01  | 41 | 6.81721E+04  | 52 | 4.16928E+07  |
|    | 9  | 9.72485E+04  | 20 | 1.39785E+07  | 31 | 4.10904E+00  | 42 | 3.42788E+04  | 53 | 6.39328E+07  |
|    | 10 | 2.99807E+05  | 21 | 3.02167E+08  | 32 | -1.07283E+03 | 43 | 4.87993E+04  | 54 | -7.98447E+08 |
|    | 11 | -5.31227E+06 | 22 | 4.64556E+08  | 33 | -1.27492E+03 | 44 | 2.34906E+05  |    |              |
| em | 1  | -5.34914E+01 | 12 | 1.45498E+07  | 23 | 1.43754E+07  | 34 | 1.08861E+03  | 45 | -2.11533E+06 |
|    | 2  | 6.50784E+03  | 13 | 3.85054E+06  | 24 | -1.77649E+10 | 35 | 6.16049E+03  | 46 | 2.31727E+06  |
|    | 3  | 1.12789E+04  | 14 | 9.79242E+06  | 25 | -8.06479E+09 | 36 | 6.56539E+03  | 47 | 1.81566E+06  |
|    | 4  | 2.35973E+03  | 15 | 5.73622E+06  | 26 | -3.11284E+09 | 37 | 9.37724E+03  | 48 | -5.40972E+06 |
|    | 5  | -2.32449E+05 | 16 | 6.05489E+06  | 27 | 3.00465E+11  | 38 | 3.55390E+04  | 49 | 9.49919E+05  |
|    | 6  | -2.19303E+05 | 17 | 6.48476E+06  | 28 | 1.50450E+00  | 39 | -1.63104E+05 | 50 | 7.44320E+05  |
|    | 7  | -3.03299E+04 | 18 | 2.16552E+08  | 29 | -1.23294E+02 | 40 | -7.36805E+04 | 51 | 1.89008E+08  |
|    | 8  | -3.09708E+05 | 19 | -2.40954E+08 | 30 | -1.89974E+02 | 41 | -1.54622E+05 | 52 | 1.10683E+08  |
|    | 9  | -3.67130E+05 | 20 | -1.09448E+08 | 31 | -8.11198E+01 | 42 | -8.82240E+04 | 53 | 2.45839E+07  |
|    | 10 | -5.17596E+05 | 21 | 4.72998E+08  | 32 | 2.94649E+03  | 43 | -1.14803E+05 | 54 | -3.34891E+09 |
|    | 11 | -4.67681E+06 | 22 | 3.38330E+07  | 33 | 3.24653E+03  | 44 | -1.89285E+05 |    |              |
| or | 1  | 6.98076E+00  | 12 | -1.64363E+06 | 23 | 9.61353E+07  | 34 | -2.56126E+02 | 45 | 6.49951E+05  |
|    | 2  | -7.44274E+02 | 13 | 2.86240E+06  | 24 | 2.50043E+09  | 35 | 2.65776E+03  | 46 | -1.68233E+05 |
|    | 3  | 2.99535E+03  | 14 | 4.35792E+06  | 25 | -1.55789E+09 | 36 | -3.07917E+02 | 47 | 8.81453E+04  |
|    | 4  | -2.04653E+03 | 15 | -1.43970E+06 | 26 | 1.55238E+08  | 37 | 2.99926E+03  | 48 | 1.45004E+06  |
|    | 5  | 1.21364E+04  | 16 | -8.28993E+05 | 27 | 1.03864E+10  | 38 | -3.42194E+04 | 49 | 3.08363E+06  |
|    | 6  | -5.81488E+04 | 17 | 4.71429E+06  | 28 | -1.19399E-02 | 39 | 4.53759E+03  | 50 | -1.18736E+06 |
|    | 7  | 3.69796E+04  | 18 | -3.59497E+07 | 29 | 6.40952E-01  | 40 | -5.01128E+04 | 51 | -3.61909E+07 |
|    | 8  | -1.42504E+05 | 19 | 3.88624E+06  | 30 | -5.15139E+01 | 41 | -6.47337E+04 | 52 | 2.99778E+07  |
|    | 9  | 1.23452E+05  | 20 | -2.47585E+07 | 31 | 5.03191E+00  | 42 | 1.37579E+04  | 53 | 1.64755E+07  |
|    | 10 | -1.29345E+05 | 21 | -4.31162E+07 | 32 | -1.01279E+01 | 43 | 1.35670E+03  | 54 | -5.15670E+08 |
|    | 11 | 1.74095E+06  | 22 | -2.05590E+08 | 33 | 9.64502E+02  | 44 | -1.34099E+05 |    |              |

**Table D.26**For  $\alpha = 0.585$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta_{\text{th},yy}^{\varepsilon}$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | 2.12989E+02  | 12 | -1.30017E+07 | 23 | 6.32157E+08  | 34 | -2.33321E+03 | 45 | -5.55411E+06 |
|    | 2  | -1.53802E+04 | 13 | -1.27465E+07 | 24 | -3.91803E+09 | 35 | -2.02037E+04 | 46 | -2.57093E+06 |
|    | 3  | -1.74643E+04 | 14 | -1.93582E+06 | 25 | -8.82708E+09 | 36 | -1.26697E+04 | 47 | -2.71887E+06 |
|    | 4  | -1.10724E+04 | 15 | -1.04186E+07 | 26 | -1.33243E+09 | 37 | -1.48669E+04 | 48 | -1.43536E+07 |
|    | 5  | 2.49687E+05  | 16 | -9.42049E+06 | 27 | -1.10892E+09 | 38 | 3.02072E+05  | 49 | -1.75795E+07 |
|    | 6  | 1.38515E+05  | 17 | -5.51026E+07 | 28 | -3.54146E+00 | 39 | 1.99561E+05  | 50 | -1.17135E+07 |
|    | 7  | 1.43862E+05  | 18 | 1.62315E+08  | 29 | 2.46719E+02  | 40 | 3.81778E+05  | 51 | 2.52314E+08  |
|    | 8  | 1.18065E+06  | 19 | 1.68881E+08  | 30 | 3.06564E+02  | 41 | 2.39040E+05  | 52 | 1.70182E+08  |
|    | 9  | 8.01437E+05  | 20 | -4.36638E+07 | 31 | 1.81278E+02  | 42 | 1.63210E+05  | 53 | 2.11188E+08  |
|    | 10 | 8.18179E+05  | 21 | 7.92799E+08  | 32 | -3.88167E+03 | 43 | 1.80980E+05  | 54 | -3.01281E+09 |
|    | 11 | -1.73809E+07 | 22 | 3.71992E+08  | 33 | -5.38464E+03 | 44 | 9.72186E+05  |    |              |
| em | 1  | -2.49907E+02 | 12 | -1.70355E+06 | 23 | 4.13719E+08  | 34 | 2.83305E+03  | 45 | -1.93786E+07 |
|    | 2  | 8.36072E+03  | 13 | -3.43584E+07 | 24 | -9.05296E+10 | 35 | -2.20081E+03 | 46 | -2.48565E+06 |
|    | 3  | 1.09913E+04  | 14 | -6.72142E+06 | 25 | -3.82384E+10 | 36 | 4.97400E+03  | 47 | -1.12977E+06 |
|    | 4  | 1.28751E+04  | 15 | 3.25651E+06  | 26 | -3.19173E+10 | 37 | 8.99868E+03  | 48 | -3.37367E+07 |
|    | 5  | -1.60252E+04 | 16 | 7.18531E+06  | 27 | 1.40481E+12  | 38 | 4.83562E+05  | 49 | -2.75677E+07 |
|    | 6  | 1.06963E+05  | 17 | -1.89726E+07 | 28 | 4.08623E+00  | 39 | 1.08641E+05  | 50 | -6.83248E+06 |
|    | 7  | -1.68060E+05 | 18 | 1.47074E+09  | 29 | -1.30908E+02 | 40 | 4.45012E+05  | 51 | 1.20559E+09  |
|    | 8  | 1.66385E+05  | 19 | 5.31474E+07  | 30 | -1.72515E+02 | 41 | 6.73564E+04  | 52 | 5.83067E+08  |
|    | 9  | -3.42275E+05 | 20 | 1.04540E+08  | 31 | -2.16930E+02 | 42 | -3.34363E+04 | 53 | 4.31928E+08  |
|    | 10 | -5.63394E+05 | 21 | 2.26610E+09  | 32 | -7.68769E+02 | 43 | -1.13165E+05 | 54 | -1.89243E+10 |
|    | 11 | -3.32449E+07 | 22 | 2.08845E+09  | 33 | -1.04037E+03 | 44 | 2.88189E+05  |    |              |
| or | 1  | 6.35309E+01  | 12 | -3.02299E+06 | 23 | -1.37981E+08 | 34 | -9.77741E+01 | 45 | -1.07180E+06 |
|    | 2  | -3.60301E+03 | 13 | -7.55883E+06 | 24 | -2.74441E+09 | 35 | -4.23823E+03 | 46 | 2.69287E+05  |
|    | 3  | -7.25908E+03 | 14 | -7.14495E+06 | 25 | 4.24741E+09  | 36 | -1.67687E+03 | 47 | 1.36358E+06  |
|    | 4  | -5.28797E+03 | 15 | -2.49050E+05 | 26 | 6.59981E+09  | 37 | -3.51549E+03 | 48 | -1.19114E+06 |
|    | 5  | 4.82318E+04  | 16 | -5.77294E+05 | 27 | -1.36555E+11 | 38 | 5.34636E+04  | 49 | -1.93451E+06 |
|    | 6  | 1.46781E+05  | 17 | -1.95402E+07 | 28 | -8.43461E-01 | 39 | 1.63519E+04  | 50 | 2.97121E+06  |
|    | 7  | 3.66325E+04  | 18 | 9.64273E+07  | 29 | 4.27977E+01  | 40 | 8.50538E+04  | 51 | 1.20731E+07  |
|    | 8  | 3.77922E+05  | 19 | -2.45759E+07 | 30 | 8.39516E+01  | 41 | 5.66885E+04  | 52 | -5.77016E+07 |
|    | 9  | 2.60497E+05  | 20 | -5.65615E+07 | 31 | 4.04117E+01  | 42 | -8.73205E+03 | 53 | -1.09756E+08 |
|    | 10 | 4.35847E+05  | 21 | 2.05183E+08  | 32 | -5.43944E+02 | 43 | -2.57297E+04 | 54 | 1.89649E+09  |
|    | 11 | -4.85646E+06 | 22 | 2.95920E+08  | 33 | -1.68603E+03 | 44 | 1.36838E+05  |    |              |

**Table D.27**For  $\alpha = 0.600$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta_{\text{th},yy}^{\varepsilon}$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | -2.11955E+01 | 12 | 4.11839E+05  | 23 | 7.08914E+07  | 34 | -8.51289E+02 | 45 | -7.23629E+05 |
|    | 2  | 1.25446E+03  | 13 | 1.42422E+06  | 24 | 8.45890E+09  | 35 | -5.05978E+03 | 46 | -9.46950E+05 |
|    | 3  | -3.21774E+03 | 14 | -4.03359E+06 | 25 | 2.84865E+08  | 36 | -4.59410E+03 | 47 | -2.83338E+06 |
|    | 4  | 1.07403E+03  | 15 | 7.47136E+05  | 26 | 2.66942E+09  | 37 | -6.03080E+03 | 48 | -1.92043E+06 |
|    | 5  | -9.20182E+03 | 16 | -2.65978E+06 | 27 | -1.36497E+11 | 38 | 4.91050E+04  | 49 | -6.91369E+06 |
|    | 6  | 8.53905E+04  | 17 | -4.68025E+06 | 28 | -1.32628E+00 | 39 | 7.43690E+04  | 50 | -2.77019E+06 |
|    | 7  | -1.35723E+04 | 18 | -1.33089E+08 | 29 | 9.09546E+01  | 40 | 1.30385E+05  | 51 | 3.33532E+07  |
|    | 8  | 7.54756E+04  | 19 | -4.77516E+06 | 30 | 1.19703E+02  | 41 | 1.98362E+05  | 52 | 1.82730E+07  |
|    | 9  | -6.07017E+04 | 20 | 4.47087E+07  | 31 | 6.67046E+01  | 42 | 5.85392E+04  | 53 | 1.00814E+08  |
|    | 10 | 1.84939E+05  | 21 | -2.19656E+07 | 32 | -1.46943E+03 | 43 | 7.82230E+04  | 54 | -5.04316E+08 |
|    | 11 | 4.44635E+05  | 22 | -1.29015E+08 | 33 | -3.64924E+03 | 44 | 2.33338E+05  |    |              |
| em | 1  | 4.25181E+02  | 12 | -7.76858E+06 | 23 | 1.64357E+08  | 34 | -3.28344E+03 | 45 | 1.91602E+06  |
|    | 2  | -2.46979E+04 | 13 | -1.07961E+07 | 24 | 8.67098E+09  | 35 | -7.72454E+03 | 46 | -1.62385E+06 |
|    | 3  | -2.71588E+04 | 14 | -2.75687E+07 | 25 | 7.12188E+09  | 36 | -1.42262E+04 | 47 | -3.34033E+06 |
|    | 4  | -2.26459E+04 | 15 | -1.30583E+07 | 26 | 2.46029E+08  | 37 | -1.36052E+04 | 48 | 2.84891E+06  |
|    | 5  | 2.57892E+05  | 16 | -1.73844E+07 | 27 | -1.02697E+11 | 38 | -9.29628E+03 | 49 | -7.17389E+05 |
|    | 6  | 5.48451E+05  | 17 | -2.70597E+07 | 28 | -4.79618E+00 | 39 | 1.66152E+05  | 50 | -1.56789E+06 |
|    | 7  | 2.96745E+05  | 18 | -1.04947E+08 | 29 | 2.95235E+02  | 40 | 8.12258E+04  | 51 | -1.34172E+08 |
|    | 8  | 7.93404E+05  | 19 | 1.50402E+07  | 30 | 2.73484E+02  | 41 | 2.70523E+05  | 52 | -4.34751E+07 |
|    | 9  | 1.16472E+06  | 20 | 3.44224E+08  | 31 | 2.50503E+02  | 42 | 1.70461E+05  | 53 | -2.77731E+07 |
|    | 10 | 1.38179E+06  | 21 | -3.70790E+08 | 32 | -4.04343E+03 | 43 | 1.70074E+05  | 54 | 1.50878E+09  |
|    | 11 | 1.74911E+06  | 22 | 2.56867E+08  | 33 | -5.50391E+03 | 44 | 2.49347E+05  |    |              |
| or | 1  | 4.51585E+01  | 12 | -1.00336E+06 | 23 | -4.95914E+08 | 34 | 7.73511E+02  | 45 | -5.81873E+05 |
|    | 2  | -2.65585E+03 | 13 | -4.68529E+06 | 24 | 1.99314E+09  | 35 | -2.18149E+03 | 46 | 7.24152E+05  |
|    | 3  | -4.86953E+03 | 14 | 5.88247E+05  | 25 | 8.59444E+09  | 36 | 5.65634E+02  | 47 | 2.21841E+06  |
|    | 4  | -2.23214E+03 | 15 | 3.42757E+06  | 26 | 1.27659E+10  | 37 | 1.15458E+03  | 48 | 1.68763E+06  |
|    | 5  | 3.60516E+04  | 16 | 6.92674E+06  | 27 | -2.03197E+11 | 38 | 2.74947E+04  | 49 | 2.70592E+06  |
|    | 6  | 8.95552E+04  | 17 | -2.07455E+06 | 28 | -3.45982E-01 | 39 | -1.07904E+04 | 50 | 5.59523E+06  |
|    | 7  | -3.77550E+04 | 18 | 6.04060E+07  | 29 | 1.72849E+01  | 40 | 4.63121E+04  | 51 | -4.53908E+07 |
|    | 8  | 2.58593E+05  | 19 | -7.01472E+07 | 30 | 4.32842E+01  | 41 | -3.68588E+04 | 52 | -8.25017E+07 |
|    | 9  | 1.03245E+05  | 20 | -1.97454E+08 | 31 | -5.72633E+00 | 42 | -4.83025E+04 | 53 | -1.32191E+08 |
|    | 10 | 8.76038E+04  | 21 | -1.45125E+07 | 32 | -2.17244E+02 | 43 | -9.10660E+04 | 54 | 1.89485E+09  |
|    | 11 | -3.36957E+06 | 22 | -8.69614E+07 | 33 | -9.21117E+02 | 44 | -9.49041E+04 |    |              |

**Table D.28**For  $\alpha = 0.540$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta_{\text{rg,yy}}^{\varepsilon}$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$       |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|-------------|
| aq | 1  | 5.11301E+03  | 12 | -1.25820E+08 | 23 | 9.82145E+09  | 34 | -5.45213E+06 | 45 | 3.70866E-04 |
|    | 2  | -3.24877E+05 | 13 | -1.03359E+08 | 24 | 0.00000E+00  | 35 | -3.44249E+07 | 46 | 2.52285E-05 |
|    | 3  | -2.51372E+05 | 14 | 9.30596E+07  | 25 | 0.00000E+00  | 36 | -5.56516E+07 | 47 | 1.25814E-04 |
|    | 4  | -5.53813E+05 | 15 | -4.62751E+08 | 26 | 0.00000E+00  | 37 | 4.36606E+07  | 48 | 9.67725E-04 |
|    | 5  | -3.98175E+05 | 16 | -1.34774E+08 | 27 | 0.00000E+00  | 38 | 4.70758E+07  | 49 | 1.25814E-04 |
|    | 6  | -4.70925E+05 | 17 | -9.26451E+08 | 28 | -2.16064E+04 | 39 | 2.02616E+08  | 50 | 1.10990E-04 |
|    | 7  | 9.03855E+06  | 18 | -1.74406E+09 | 29 | 1.40213E+06  | 40 | 1.98486E+08  | 51 | 0.00000E+00 |
|    | 8  | 1.43833E+07  | 19 | 2.64266E+09  | 30 | -2.64585E+05 | 41 | -4.28915E+08 | 52 | 0.00000E+00 |
|    | 9  | 2.92375E+07  | 20 | -4.17761E+09 | 31 | 7.25236E+05  | 42 | 5.33744E+08  | 53 | 0.00000E+00 |
|    | 10 | 1.64552E+07  | 21 | -8.37556E+08 | 32 | -8.36196E+06 | 43 | -8.76208E+08 | 54 | 0.00000E+00 |
|    | 11 | 9.90221E+07  | 22 | 5.77803E+09  | 33 | 7.63582E+06  | 44 | 8.02601E+08  |    |             |
| em | 1  | 2.11075E+04  | 12 | -6.63979E+08 | 23 | 3.40744E-05  | 34 | -9.77143E+07 | 45 | 0.00000E+00 |
|    | 2  | -1.38341E+06 | 13 | 1.98001E+08  | 24 | 0.00000E+00  | 35 | -6.78690E+05 | 46 | 0.00000E+00 |
|    | 3  | 1.23297E+06  | 14 | 1.38629E+09  | 25 | 0.00000E+00  | 36 | -8.67498E+07 | 47 | 0.00000E+00 |
|    | 4  | -1.70316E+06 | 15 | -9.51078E+08 | 26 | 0.00000E+00  | 37 | -5.64505E+06 | 48 | 0.00000E+00 |
|    | 5  | 1.34404E+07  | 16 | 5.98901E+08  | 27 | 0.00000E+00  | 38 | -4.84535E-06 | 49 | 0.00000E+00 |
|    | 6  | -2.91240E+07 | 17 | 1.20566E+07  | 28 | -1.54255E+05 | 39 | 1.59525E-04  | 50 | 0.00000E+00 |
|    | 7  | 2.51437E+07  | 18 | -1.36356E-03 | 29 | 3.44708E+06  | 40 | 5.98646E-05  | 51 | 0.00000E+00 |
|    | 8  | -2.28690E+07 | 19 | 8.65544E-05  | 30 | 1.41943E+04  | 41 | -1.03913E+08 | 52 | 0.00000E+00 |
|    | 9  | 8.74270E+07  | 20 | -1.97334E+10 | 31 | 7.69680E+06  | 42 | 4.20982E-04  | 53 | 0.00000E+00 |
|    | 10 | -4.59382E+07 | 21 | 7.44146E-04  | 32 | -1.97615E+07 | 43 | 1.47772E+08  | 54 | 0.00000E+00 |
|    | 11 | 3.20609E+08  | 22 | 0.00000E+00  | 33 | 3.90591E+06  | 44 | 1.08623E-04  |    |             |
| or | 1  | -1.17852E+04 | 12 | -2.20481E+08 | 23 | -6.19075E-05 | 34 | 9.49087E+05  | 45 | 0.00000E+00 |
|    | 2  | 3.46593E+05  | 13 | 3.56827E+07  | 24 | 0.00000E+00  | 35 | -1.25997E+07 | 46 | 0.00000E+00 |
|    | 3  | 2.26417E+05  | 14 | 6.82954E+07  | 25 | 0.00000E+00  | 36 | 6.62759E+06  | 47 | 0.00000E+00 |
|    | 4  | 1.53319E+05  | 15 | -1.63999E+08 | 26 | 0.00000E+00  | 37 | -4.71193E+06 | 48 | 0.00000E+00 |
|    | 5  | -2.92231E+06 | 16 | 1.38993E+08  | 27 | 0.00000E+00  | 38 | 7.06087E+07  | 49 | 0.00000E+00 |
|    | 6  | -2.48304E+06 | 17 | 1.27667E+08  | 28 | 7.30526E+03  | 39 | 3.75951E-05  | 50 | 0.00000E+00 |
|    | 7  | 4.20280E+05  | 18 | 7.00994E-06  | 29 | -7.41308E+05 | 40 | 1.49696E+08  | 51 | 0.00000E+00 |
|    | 8  | -3.64354E+06 | 19 | -8.55794E-05 | 30 | 4.36714E+05  | 41 | 8.60299E+07  | 52 | 0.00000E+00 |
|    | 9  | 9.02936E+06  | 20 | -6.19075E-05 | 31 | -1.27530E+05 | 42 | 1.93871E-05  | 53 | 0.00000E+00 |
|    | 10 | -1.18717E+07 | 21 | 3.76444E-04  | 32 | 1.15894E+07  | 43 | 4.61432E-06  | 54 | 0.00000E+00 |
|    | 11 | -3.44554E+06 | 22 | 3.95887E-04  | 33 | -6.45773E+06 | 44 | 2.78769E-05  |    |             |

**Table D.29**For  $\alpha = 0.555$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta_{\text{rg,yy}}^{\varepsilon}$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$       |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|-------------|
| aq | 1  | -5.32415E+04 | 12 | 1.50273E+09  | 23 | 1.02147E+10  | 34 | 6.97156E+07  | 45 | 5.00858E-04 |
|    | 2  | 2.72563E+06  | 13 | 9.15058E+07  | 24 | 0.00000E+00  | 35 | 6.61889E+07  | 46 | 8.80108E-06 |
|    | 3  | 9.11121E+05  | 14 | -2.28839E+08 | 25 | 0.00000E+00  | 36 | 1.57991E+08  | 47 | 1.64161E-03 |
|    | 4  | 2.37371E+06  | 15 | 1.43837E+09  | 26 | 0.00000E+00  | 37 | 1.37304E+08  | 48 | 7.36716E-04 |
|    | 5  | -3.31749E+07 | 16 | -1.53999E+08 | 27 | 0.00000E+00  | 38 | -8.29949E+07 | 49 | 1.14347E-03 |
|    | 6  | 7.84572E+04  | 17 | 5.45478E+08  | 28 | 1.16213E+05  | 39 | -2.75357E+08 | 50 | 1.44621E-03 |
|    | 7  | -2.75264E+07 | 18 | -5.46130E+09 | 29 | -3.41935E+06 | 40 | -2.74698E+08 | 51 | 0.00000E+00 |
|    | 8  | -4.50313E+07 | 19 | -1.74909E+10 | 30 | -3.68936E+06 | 41 | -5.02372E+08 | 52 | 0.00000E+00 |
|    | 9  | -1.23380E+08 | 20 | -1.32089E+09 | 31 | -5.76358E+06 | 42 | -1.74683E+09 | 53 | 0.00000E+00 |
|    | 10 | -1.21196E+07 | 21 | -1.59165E+10 | 32 | 9.83878E+06  | 43 | -1.08833E+09 | 54 | 0.00000E+00 |
|    | 11 | 7.07403E+08  | 22 | 1.07288E+10  | 33 | 2.53098E+07  | 44 | -1.57381E+09 |    |             |
| em | 1  | -3.84667E+04 | 12 | -5.00982E+08 | 23 | 4.29305E-07  | 34 | 3.76781E+07  | 45 | 4.05016E-05 |
|    | 2  | 4.59494E+05  | 13 | 1.95403E+08  | 24 | 0.00000E+00  | 35 | 3.33459E+07  | 46 | 4.05016E-05 |
|    | 3  | 2.71022E+06  | 14 | 1.40329E+09  | 25 | 0.00000E+00  | 36 | 5.49300E+07  | 47 | 1.85460E-08 |
|    | 4  | 1.31409E+06  | 15 | -5.92351E+08 | 26 | 0.00000E+00  | 37 | 9.40828E+07  | 48 | 4.05016E-05 |
|    | 5  | 1.23343E+06  | 16 | 1.46736E+09  | 27 | 0.00000E+00  | 38 | 5.11225E-06  | 49 | 4.05016E-05 |
|    | 6  | -3.02512E+07 | 17 | 9.19478E+08  | 28 | 6.37249E+04  | 39 | 3.21844E-05  | 50 | 4.05016E-05 |
|    | 7  | -1.25853E+07 | 18 | 9.86445E-07  | 29 | -2.01594E+06 | 40 | 3.26305E+07  | 51 | 0.00000E+00 |
|    | 8  | -5.68373E+07 | 19 | 4.29305E-07  | 30 | -1.95185E+06 | 41 | -2.94475E+08 | 52 | 0.00000E+00 |
|    | 9  | 2.51584E+07  | 20 | -1.90162E+10 | 31 | -3.23242E+06 | 42 | 3.99336E-05  | 53 | 0.00000E+00 |
|    | 10 | -1.18819E+08 | 21 | 0.00000E+00  | 32 | 1.07493E+07  | 43 | -1.00898E+09 | 54 | 0.00000E+00 |
|    | 11 | 4.76036E+08  | 22 | 0.00000E+00  | 33 | 8.95032E+06  | 44 | -1.10310E+09 |    |             |
| or | 1  | -1.32344E+04 | 12 | -2.00362E+08 | 23 | 1.94561E-04  | 34 | -1.85712E+06 | 45 | 0.00000E+00 |
|    | 2  | 4.23949E+05  | 13 | 5.25366E+07  | 24 | 0.00000E+00  | 35 | 1.45866E+06  | 46 | 0.00000E+00 |
|    | 3  | 6.06332E+05  | 14 | 8.36424E+07  | 25 | 0.00000E+00  | 36 | -4.49618E+06 | 47 | 0.00000E+00 |
|    | 4  | 1.78050E+05  | 15 | -1.56326E+08 | 26 | 0.00000E+00  | 37 | -2.22739E+06 | 48 | 0.00000E+00 |
|    | 5  | -3.94799E+06 | 16 | 1.80894E+08  | 27 | 0.00000E+00  | 38 | -1.04370E+08 | 49 | 0.00000E+00 |
|    | 6  | -3.16119E+06 | 17 | 3.13475E+08  | 28 | -5.94113E+03 | 39 | 8.20675E-05  | 50 | 0.00000E+00 |
|    | 7  | 2.82784E+05  | 18 | -2.50503E-05 | 29 | -8.97154E+04 | 40 | 9.15405E+07  | 51 | 0.00000E+00 |
|    | 8  | -2.25702E+07 | 19 | 1.10904E-04  | 30 | 1.50418E+05  | 41 | 1.77014E+07  | 52 | 0.00000E+00 |
|    | 9  | 7.63200E+06  | 20 | 2.56741E-04  | 31 | 3.02999E+05  | 42 | 2.40394E-05  | 53 | 0.00000E+00 |
|    | 10 | -1.97675E+07 | 21 | 1.69629E-04  | 32 | 3.58376E+06  | 43 | 7.90789E-06  | 54 | 0.00000E+00 |
|    | 11 | 2.32306E+08  | 22 | 6.91511E-05  | 33 | -4.06646E+06 | 44 | 1.04081E-05  |    |             |

**Table D.30**For  $\alpha = 0.570$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta \xi_{rg,yy}^z$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | 1.14155E+04  | 12 | -2.69318E+08 | 23 | 2.49253E+10  | 34 | -2.48333E+06 | 45 | 9.29248E-04  |
|    | 2  | -1.89145E+05 | 13 | -3.67996E+08 | 24 | 0.00000E+00  | 35 | 4.05738E+07  | 46 | 2.10771E-05  |
|    | 3  | -1.06484E+06 | 14 | 1.84569E+08  | 25 | 0.00000E+00  | 36 | 6.30255E+07  | 47 | 9.88038E-04  |
|    | 4  | -9.58461E+05 | 15 | -4.66056E+08 | 26 | 0.00000E+00  | 37 | -1.61392E+06 | 48 | -3.33723E-04 |
|    | 5  | 1.54633E+06  | 16 | -7.65305E+08 | 27 | 0.00000E+00  | 38 | -4.97175E+07 | 49 | 1.18082E-03  |
|    | 6  | 2.98244E+06  | 17 | -1.96446E+09 | 28 | 2.73990E+03  | 39 | -1.14724E+08 | 50 | -6.13894E-06 |
|    | 7  | 1.52814E+07  | 18 | -1.62767E+09 | 29 | -1.37688E+06 | 40 | -2.98717E+08 | 51 | 0.00000E+00  |
|    | 8  | 2.99884E+07  | 19 | 5.04922E+09  | 30 | -3.58227E+05 | 41 | -9.79867E+07 | 52 | 0.00000E+00  |
|    | 9  | 2.58563E+07  | 20 | -1.19239E+10 | 31 | -1.92876E+04 | 42 | -6.76656E+08 | 53 | 0.00000E+00  |
|    | 10 | 6.30559E+07  | 21 | 1.98449E+09  | 32 | 3.96958E+06  | 43 | 2.58142E+08  | 54 | 0.00000E+00  |
|    | 11 | 8.02209E+06  | 22 | 1.64979E+10  | 33 | 9.10643E+06  | 44 | -9.35199E+08 |    |              |
| em | 1  | 2.64642E+04  | 12 | -9.01624E+08 | 23 | 1.71005E-04  | 34 | -7.51032E+07 | 45 | 0.00000E+00  |
|    | 2  | -1.98144E+06 | 13 | 2.09187E+08  | 24 | 0.00000E+00  | 35 | -2.95426E+06 | 46 | 0.00000E+00  |
|    | 3  | 1.79458E+06  | 14 | 1.21028E+09  | 25 | 0.00000E+00  | 36 | -5.03790E+07 | 47 | 0.00000E+00  |
|    | 4  | -1.97119E+06 | 15 | -1.38974E+09 | 26 | 0.00000E+00  | 37 | -6.00272E+07 | 48 | 0.00000E+00  |
|    | 5  | 2.21597E+07  | 16 | 8.23127E+08  | 27 | 0.00000E+00  | 38 | -9.10226E-06 | 49 | 0.00000E+00  |
|    | 6  | -2.75994E+07 | 17 | 5.38632E+08  | 28 | -1.19325E+05 | 39 | 1.24737E-04  | 50 | 0.00000E+00  |
|    | 7  | 2.89571E+07  | 18 | -1.50067E-02 | 29 | 2.26340E+06  | 40 | 1.73571E-05  | 51 | 0.00000E+00  |
|    | 8  | -4.24565E+07 | 19 | 4.17758E-04  | 30 | 9.13158E+05  | 41 | -2.52096E+08 | 52 | 0.00000E+00  |
|    | 9  | 1.18170E+08  | 20 | -1.59151E+10 | 31 | 5.86527E+06  | 42 | 2.95174E-04  | 53 | 0.00000E+00  |
|    | 10 | -6.99848E+07 | 21 | 0.00000E+00  | 32 | -1.56103E+07 | 43 | 1.00087E+09  | 54 | 0.00000E+00  |
|    | 11 | 4.13069E+08  | 22 | 0.00000E+00  | 33 | 8.55346E+06  | 44 | 1.10935E-04  |    |              |
| or | 1  | -3.81315E+01 | 12 | -3.10819E+08 | 23 | -2.48875E-05 | 34 | -4.79688E+06 | 45 | 0.00000E+00  |
|    | 2  | -2.54438E+05 | 13 | 6.41210E+07  | 24 | 0.00000E+00  | 35 | -1.09399E+07 | 46 | 0.00000E+00  |
|    | 3  | 2.45797E+05  | 14 | 8.02969E+07  | 25 | 0.00000E+00  | 36 | -1.74357E+07 | 47 | 0.00000E+00  |
|    | 4  | -2.34102E+05 | 15 | -2.15590E+08 | 26 | 0.00000E+00  | 37 | -2.89337E+06 | 48 | 0.00000E+00  |
|    | 5  | 4.76501E+06  | 16 | 1.48577E+08  | 27 | 0.00000E+00  | 38 | 1.00302E+08  | 49 | 0.00000E+00  |
|    | 6  | -3.59278E+06 | 17 | 1.41268E+08  | 28 | -3.23151E+04 | 39 | 7.30441E-05  | 50 | 0.00000E+00  |
|    | 7  | 3.57585E+06  | 18 | -8.28786E-06 | 29 | 1.27044E+06  | 40 | 3.22587E+07  | 51 | 0.00000E+00  |
|    | 8  | -3.52734E+06 | 19 | 5.98471E-06  | 30 | 3.27607E+05  | 41 | 2.62662E+07  | 52 | 0.00000E+00  |
|    | 9  | 2.24418E+07  | 20 | 1.29606E-04  | 31 | 8.23287E+05  | 42 | 1.89556E-05  | 53 | 0.00000E+00  |
|    | 10 | -1.28436E+07 | 21 | -1.44699E-04 | 32 | -1.39509E+07 | 43 | -3.71580E-06 | 54 | 0.00000E+00  |
|    | 11 | -1.93817E+07 | 22 | 8.08842E-05  | 33 | -1.84920E+06 | 44 | 1.28381E-05  |    |              |

**Table D.31**For  $\alpha = 0.585$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta \xi_{rg,yy}^z$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | 4.11717E+03  | 12 | 1.51191E+10  | 23 | -1.36584E+08 | 34 | -3.84959E+07 | 45 | -1.50716E-04 |
|    | 2  | 1.36027E+05  | 13 | 0.00000E+00  | 24 | -2.63061E+08 | 35 | -8.06600E+06 | 46 | 4.69680E-05  |
|    | 3  | -6.30017E+05 | 14 | 0.00000E+00  | 25 | 3.76730E+08  | 36 | 3.88418E+06  | 47 | -1.63111E-04 |
|    | 4  | -5.91738E+05 | 15 | 0.00000E+00  | 26 | -2.97566E+08 | 37 | 6.34868E+06  | 48 | -1.10469E-03 |
|    | 5  | -1.66522E+06 | 16 | 0.00000E+00  | 27 | -3.69508E+08 | 38 | -6.99605E+07 | 49 | -2.29519E-04 |
|    | 6  | -1.56071E+06 | 17 | -5.35935E+04 | 28 | -1.32205E+09 | 39 | -2.61223E+08 | 50 | -1.05942E-03 |
|    | 7  | 1.07460E+07  | 18 | -3.18276E+05 | 29 | -1.17034E+09 | 40 | 1.41901E+08  | 51 | 0.00000E+00  |
|    | 8  | 1.97491E+07  | 19 | 7.17563E+05  | 30 | 3.83020E+09  | 41 | -3.92251E+08 | 52 | 0.00000E+00  |
|    | 9  | 1.07611E+07  | 20 | 2.81885E+06  | 31 | -1.19225E+10 | 42 | 1.64100E+08  | 53 | 0.00000E+00  |
|    | 10 | 3.63414E+07  | 21 | 9.18904E+06  | 32 | 2.98565E+09  | 43 | -6.15992E+08 | 54 | 0.00000E+00  |
|    | 11 | -2.30111E+07 | 22 | 1.13556E+06  | 33 | 1.12957E+10  | 44 | 2.94345E+08  |    |              |
| em | 1  | 2.38275E+04  | 12 | 4.80699E+08  | 23 | 1.52702E+11  | 34 | 7.29610E+07  | 45 | -1.84607E+12 |
|    | 2  | -1.16432E+06 | 13 | -9.73340E+09 | 24 | -2.64015E+13 | 35 | 4.09328E+08  | 46 | -4.73307E+11 |
|    | 3  | -2.26137E+06 | 14 | -4.36399E+09 | 25 | -9.30227E+12 | 36 | 2.37754E+08  | 47 | 4.49307E+10  |
|    | 4  | -1.77501E+06 | 15 | -4.35675E+08 | 26 | -8.75022E+12 | 37 | 6.56121E+08  | 48 | -3.39297E+12 |
|    | 5  | 1.46127E+05  | 16 | -2.16930E+09 | 27 | 4.13369E+14  | 38 | 4.29008E+10  | 49 | -1.56041E+12 |
|    | 6  | 6.90611E+07  | 17 | -1.10430E+10 | 28 | 1.03974E+05  | 39 | 2.16386E+10  | 50 | -1.84820E+11 |
|    | 7  | 2.56980E+07  | 18 | 4.21955E+11  | 29 | -6.31208E+06 | 40 | 2.09985E+10  | 51 | 1.25487E+14  |
|    | 8  | 1.83259E+08  | 19 | -2.01495E+10 | 30 | -1.24288E+07 | 41 | -5.43386E+09 | 52 | 6.60255E+13  |
|    | 9  | 6.07480E+07  | 20 | 6.46631E+10  | 31 | -5.85405E+06 | 42 | -4.97323E+08 | 53 | 2.90702E+13  |
|    | 10 | 1.49334E+08  | 21 | 5.86049E+11  | 32 | -2.31144E+08 | 43 | -7.96079E+09 | 54 | -2.14507E+15 |
|    | 11 | -9.12678E+09 | 22 | 5.88953E+11  | 33 | 1.22504E+08  | 44 | -8.77310E+09 |    |              |
| or | 1  | -7.52158E+03 | 12 | -4.20955E+08 | 23 | -8.84594E-03 | 34 | -5.21575E+05 | 45 | 0.00000E+00  |
|    | 2  | 1.35110E+05  | 13 | 6.62435E+07  | 24 | 0.00000E+00  | 35 | 3.06391E+06  | 46 | 0.00000E+00  |
|    | 3  | 3.35116E+05  | 14 | 8.06251E+07  | 25 | 0.00000E+00  | 36 | 7.03258E+05  | 47 | 0.00000E+00  |
|    | 4  | -2.87169E+05 | 15 | -2.76398E+08 | 26 | 0.00000E+00  | 37 | -2.00238E+06 | 48 | 0.00000E+00  |
|    | 5  | -2.91135E+05 | 16 | 1.38673E+08  | 27 | 0.00000E+00  | 38 | -8.40059E+07 | 49 | 0.00000E+00  |
|    | 6  | -3.64231E+06 | 17 | 1.34366E+08  | 28 | 8.19963E+03  | 39 | 3.63536E-05  | 50 | 0.00000E+00  |
|    | 7  | 5.18288E+06  | 18 | -3.43976E-06 | 29 | -8.18096E+05 | 40 | -3.25282E+06 | 51 | 0.00000E+00  |
|    | 8  | -8.67490E+06 | 19 | -2.47341E-05 | 30 | 6.44297E+04  | 41 | -9.93585E-06 | 52 | 0.00000E+00  |
|    | 9  | 2.81757E+07  | 20 | 5.04993E-06  | 31 | 9.36792E+04  | 42 | 8.43973E-06  | 53 | 0.00000E+00  |
|    | 10 | -1.24527E+07 | 21 | 2.21316E-04  | 32 | 1.29201E+07  | 43 | -2.20546E-06 | 54 | 0.00000E+00  |
|    | 11 | 5.27706E+07  | 22 | 3.69789E-04  | 33 | -5.12559E+05 | 44 | -2.73280E-06 |    |              |

**Table D.32**For  $\alpha = 0.600$ , the values of  $b_i$  in Eq. (113) used to determine  $\Delta \varepsilon_{\text{rg},yy}^{\varepsilon}$ .

| yy | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        | i  | $b_i$        |
|----|----|--------------|----|--------------|----|--------------|----|--------------|----|--------------|
| aq | 1  | 1.38504E+04  | 12 | 3.23360E+08  | 23 | 1.02855E+10  | 34 | -9.70687E+07 | 45 | 1.32775E-03  |
|    | 2  | -2.22037E+04 | 13 | -2.05776E+08 | 24 | 0.00000E+00  | 35 | -7.62467E+07 | 46 | 6.86265E-05  |
|    | 3  | -2.39012E+05 | 14 | 2.86767E+08  | 25 | 0.00000E+00  | 36 | -1.90579E+08 | 47 | -4.27670E-04 |
|    | 4  | -1.04310E+06 | 15 | -3.61685E+08 | 26 | 0.00000E+00  | 37 | 7.97887E+07  | 48 | 1.39579E-03  |
|    | 5  | -1.02937E+07 | 16 | 7.24896E+07  | 27 | 0.00000E+00  | 38 | -6.45822E+07 | 49 | 2.29416E-03  |
|    | 6  | -2.36319E+06 | 17 | -9.57741E+08 | 28 | -1.45443E+05 | 39 | -2.31091E+08 | 50 | 3.16167E-03  |
|    | 7  | 1.59612E+07  | 18 | -3.03383E+09 | 29 | 3.51460E+06  | 40 | 4.27084E+08  | 51 | 0.00000E+00  |
|    | 8  | 1.33419E+07  | 19 | -2.26847E+09 | 30 | 2.92185E+05  | 41 | -1.02878E+09 | 52 | 0.00000E+00  |
|    | 9  | 1.72506E+07  | 20 | -7.92635E+09 | 31 | 7.45312E+06  | 42 | 2.63506E+09  | 53 | 0.00000E+00  |
|    | 10 | 8.56608E+06  | 21 | -3.63452E+09 | 32 | 7.93245E+06  | 43 | -2.18840E+09 | 54 | 0.00000E+00  |
|    | 11 | 2.32862E+08  | 22 | 1.05597E+10  | 33 | 1.60925E+07  | 44 | 2.01372E+09  |    |              |
| em | 1  | 8.50148E+03  | 12 | -6.58814E+08 | 23 | -4.07957E+10 | 34 | -8.71254E+06 | 45 | -3.04247E-09 |
|    | 2  | -5.83156E+05 | 13 | -1.37177E+09 | 24 | 0.00000E+00  | 35 | 2.31393E+07  | 46 | -3.04247E-09 |
|    | 3  | -1.42257E+06 | 14 | -4.24215E+09 | 25 | 0.00000E+00  | 36 | -6.14417E+07 | 47 | -1.05958E-08 |
|    | 4  | -9.11738E+05 | 15 | -2.73008E+08 | 26 | 0.00000E+00  | 37 | 1.09264E+08  | 48 | -3.04247E-09 |
|    | 5  | 1.27256E+07  | 16 | -1.12455E+09 | 27 | 0.00000E+00  | 38 | -2.15333E-07 | 49 | -8.02569E-09 |
|    | 6  | 7.76935E+07  | 17 | 1.38719E+09  | 28 | -2.15680E+04 | 39 | -6.35739E-07 | 50 | -8.02569E-09 |
|    | 7  | 1.32535E+07  | 18 | -2.60174E-08 | 29 | 1.31975E+06  | 40 | 3.78064E+08  | 51 | 0.00000E+00  |
|    | 8  | -2.21035E+07 | 19 | -6.44503E-08 | 30 | -1.57066E+06 | 41 | 6.08216E+08  | 52 | 0.00000E+00  |
|    | 9  | 4.00324E+07  | 20 | 5.38374E+10  | 31 | 7.61062E+05  | 42 | 9.61763E+08  | 53 | 0.00000E+00  |
|    | 10 | 9.01572E+07  | 21 | -2.31111E-08 | 32 | -5.80740E+06 | 43 | -1.69958E+09 | 54 | 0.00000E+00  |
|    | 11 | 3.71851E+08  | 22 | 4.52389E+10  | 33 | -2.48404E+07 | 44 | -1.15420E+09 |    |              |
| or | 1  | 9.95825E+02  | 12 | -4.03386E+08 | 23 | 1.52879E-04  | 34 | -4.38673E+06 | 45 | 0.00000E+00  |
|    | 2  | -3.00194E+05 | 13 | 4.72065E+07  | 24 | 0.00000E+00  | 35 | 8.18456E+05  | 46 | 0.00000E+00  |
|    | 3  | 2.66522E+05  | 14 | 7.39123E+07  | 25 | 0.00000E+00  | 36 | -1.83466E+07 | 47 | 0.00000E+00  |
|    | 4  | -4.12127E+05 | 15 | -2.55189E+08 | 26 | 0.00000E+00  | 37 | -1.27471E+06 | 48 | 0.00000E+00  |
|    | 5  | 5.26593E+06  | 16 | 1.36428E+08  | 27 | 0.00000E+00  | 38 | -3.44087E+07 | 49 | 0.00000E+00  |
|    | 6  | -2.92490E+06 | 17 | 1.22460E+08  | 28 | -3.02166E+04 | 39 | 2.59590E-05  | 50 | 0.00000E+00  |
|    | 7  | 5.26670E+06  | 18 | -1.03112E-05 | 29 | 1.17324E+06  | 40 | 1.10147E+07  | 51 | 0.00000E+00  |
|    | 8  | -5.46564E+06 | 19 | 3.75364E-04  | 30 | 7.39590E+04  | 41 | -1.68656E-06 | 52 | 0.00000E+00  |
|    | 9  | 3.05756E+07  | 20 | 1.48281E-05  | 31 | 8.34286E+05  | 42 | 1.73342E-05  | 53 | 0.00000E+00  |
|    | 10 | -1.18248E+07 | 21 | 1.30428E-04  | 32 | -1.28618E+07 | 43 | 1.28868E-05  | 54 | 0.00000E+00  |
|    | 11 | 1.63724E+07  | 22 | 8.57813E-05  | 33 | -1.02240E+06 | 44 | -2.45455E-06 |    |              |

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