EPSRC
Engineering and Physical Sciences Research Council

Imperial College London

Centre for Nuclear Engineering

EPSRC/MoD Contractor PhD industrial CASE in Nuclear Engineering at Imperial College London 2019

Mathematical and computational modelling of transient nuclear criticality excursions in multi-fluid layered fissile aqueous-organic and emulsion systems for safety and risk assessment analysis

The aim of this proposed EPSRC/MoD contractor sponsored iCASE PhD studentship based at Imperial College London is to: develop novel mathematical and computational models to simulate nuclear criticality excursions in layered, multi-fluid systems such as mixtures of fissile aqueous-organic and emulsion solutions. The purpose of such models would be to perform nuclear criticality and risk assessment for such excursion as well as inform emergency planning procedures in the event of nuclear criticality accidents. These types of layered multi-fluid systems are very widespread within nuclear fuel processing facilities. Indeed many nuclear criticality excursions, including one at Windscale works on the 24th August 1970 (now Sellafield), have occurred in such systems. The Windscale works nuclear criticality excursion has been proposed as a potential validation benchmark for such complex, layered, multi-fluid systems at a recent Working Party on Nuclear Criticality Safety (WPNCS) meeting at the OECD/NEA headquarters in Paris on the 5th July 2018. The aim of this PhD project would be to develop the underpinning mathematical models and then implement these within a prototype code with associated verification and validation (V&V) through experimental data and the pre-existing database of such nuclear criticality excursions (four such nuclear criticality excursions have occurred world-wide). Imperial College London is recognised as the world leading authority in the modelling of such fissile solution systems involving very complex rheological properties of fluids. Imperial College London, over the last twenty years, have developed a number of sophisticated mathematical and computational models of fissile solution systems and these will provide the basis of this PhD project.

As discussed, this PhD project will involve the development of innovative mathematical and computational models for nuclear criticality safety assessment of fissile solutions involving layered systems of fluids with very complex rheologies. These mathematical model developments will include: the development of multi-point nuclear reactor kinetics and moving boundary methods for both nuclear reactor kinetics and thermal-hydraulic feedback. The moving boundary methods will be used to model the change in the emulsion interface between the fissile aqueous solution and the organic layer. The nuclear reactor kinetics will incorporate models for the rise and growth of emulsion globules through the different solutions via buoyancy effects; in addition to the associated radiolytic gas bubble growth and rise models. Finally, the ability to simulate injections and jets of solutions into the system must also be modelled which will affect the boundaries and heights of the layers of solution. These will require significant extensions and enhancements of pre-existing nuclear reactor kinetics and thermal-hydraulic feedback models. These extensions of the nuclear reactor kinetics models will include moving from point models to spatially-dependent models with moving boundaries. The improvements to the nuclear thermal-hydraulic models will include the addition of emulsion globule buoyancy rise and growth models in addition to those associated with radiolytic gas bubble movement. In addition, models will also need to be developed to take account of the movement of the solution boundaries between layers of fluids. This will be a significant extension and enhancement of pre-existing mathematical models of fissile solutions developed by Imperial College London.

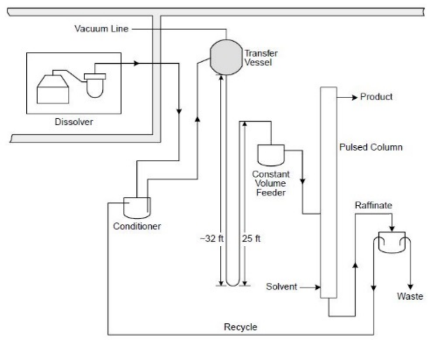
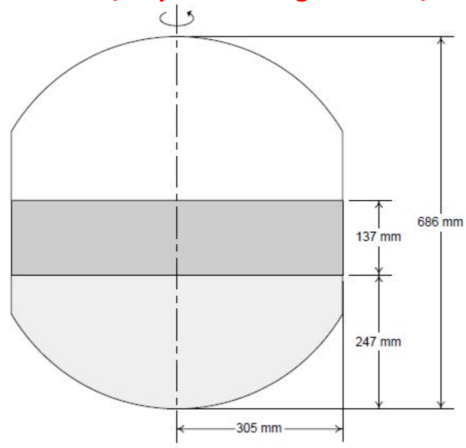


Figure 1: Process equipment related to the Windscale works nuclear criticality excursion that

As described previously there have been a number of nuclear criticality excursions involving layered fluid systems with very complex rheologies. From a UK perspective the most interesting of the fissile aqueous-organic layered solution systems is the Windscale works (now Sellafield) nuclear criticality excursion on the 24th August 1970. The Windscale system comprised a plutonium-organic solution in a transfer vessel. It led to one excursion with insignificant exposures to personnel. Below is a brief review of the Windscale works nuclear criticality excursion that occurred on the 24th August 1970. The following description of the nuclear criticality excursion accident is taken verbatim from Los Alamos National Scientific Laboratory Report: "*A Review of Criticality Accidents 2000 Revision LA-13638*" by Thomas P. McLaughlin et al viz: This particular criticality accident is one of the more interesting and complex because of the intricate configurations involved. The plant was used to recover plutonium from miscellaneous scrap, and



Vessel Volume	156.0 ℓ	Estimated Spherical Critical Mass	0.69 kg
Fissile Volume	40.0 ℓ	First Spike Yield	none
Fissile Mass	2.07 kg	Specific Spike Yield	none
Fissile Density	51.8 g/ℓ	Total Yield	0.01×10^{17}

Figure 2: Schematic of the transfer vessel in the Windscale works 1970

the processes used were thought to be subject to very effective nuclear criticality safety controls. Recovery operations started with a dissolver charge of about 300 g of plutonium. Following dissolution, the supernatant was transferred through a filter to a conditioner vessel, where the concentration was adjusted to between 6 and 7 g Pu/l, less than the minimum critical concentration. The solution was vacuum lifted from the conditioner to a transfer vessel (**see Figure 1**). When the transfer was completed the vessel contents were allowed to drain into a constant volume feeder that supplied a favourable geometry, pulsed, solvent extraction column. The connection from the transfer vessel to the constant volume feeder was through a trap 25 feet (7.6 m) in depth that prevented any potential backflow and thus controlled contamination. The excursion occurred on completion of the transfer of a 50 l batch of solution from the conditioner to the transfer vessel. The small size (10^{15} fissions) and brief duration (less than 10 s) of the excursion precluded the termination of the excursion due to any energy based shutdown mechanism. Radiation measurements indicated that the excursion occurred in the transfer vessel, but the solution from the conditioner was too lean to sustain criticality, and the total quantity of plutonium in the batch (300 g) was about 50% of the minimum critical mass. Thus, it was feared that the transfer vessel might contain large quantities of solids, perhaps tens of kilograms and that any disturbance of the system might cause another, possibly much larger, excursion. A 6 inch (150 mm) diameter hole was cut through the concrete roof, and the vacuum line to the transfer vessel was opened. The interior of the transfer vessel was inspected with a fibre-optics system (developed specifically for this recovery operation) and was found to contain liquid. A small diameter plastic line was inserted into the vessel and 2.5 l aliquots were siphoned to a collection point in an adjacent building. Inspection of the liquid revealed tributyl phosphate and kerosene with a specific gravity of 0.96 that contained 55 g Pu/l. Aqueous solution from the conditioner had a specific gravity of 1.3.

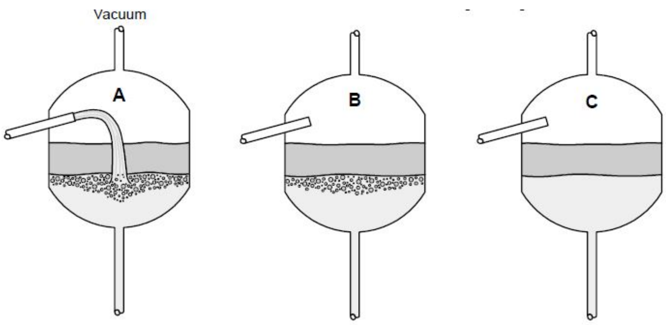


Figure 3: Solution transfer as reconstructed from the transparent plastic mock-up of the transfer vessel. Configuration (B) is the postulated state at the time of the accident.

A column 25 feet (7.6 m) in height of aqueous solution in one arm of the trap was sufficient to balance approximately 33.8 feet (10.3 m) of solvent in the other arm. Thus any solvent introduced into the transfer vessel was held in the arm and could accumulate until the volume of solvent corresponded to a height of 33.8 feet (10.3 m) above the bottom of the trap. Some 39 l, containing about 2.15 kg Pu, were present. Degradation of the solvent indicated it had been trapped in the transfer vessel for several months and perhaps for as long as 2 years. Each time a batch of aqueous solution was processed through the transfer vessel, the organic extractant would strip some plutonium from the aqueous solution. With each transfer, the plutonium concentration in the tributyl phosphate and kerosene increased. The operation that resulted in the excursion probably added about 30 g of plutonium to the solvent. Periodic plant cleanout by flushing nitric acid through the system presumably reduced the plutonium concentration in the trapped solvent. Thus, the concentration may have been slowly increased, then been abruptly reduced. Several such cycles could have been repeated before the system achieved criticality. The drain rate of the transfer vessel was not sufficient to account for the brief duration of the excursion. A transparent plastic mock-up of the transfer vessel (**Figure 2 presents a schematic of the transfer vessel**) was used to observe the configuration of the liquids during transfer. The situation existing during the transfer is shown in **Figure 3A**. Rich organic (55 g/ l) is floating on top of lean aqueous solution (6 to 7 g/ l). The aqueous solution stream pouring into the centre of the vessel provides a region of low reactivity. Between the organic and aqueous is a region of mixed phases, about 3 inches (7.6 cm) thick near the axis of the vessel. This configuration is sub-critical. Just after completion of the transfer (**see Figure 3B**) the central plug of aqueous solution has disappeared, the region of mixed phases is still present, and the configuration has reached the state of maximum reactivity. Separation of the two phases occurs within a few seconds of completing the transfer (**Figure 3C**). Monte Carlo calculations have indicated that the reactivity of **Figure 3B** is about 5 $ greater than that of **Figure 3A** and about 10 to 15 $ greater than **Figure 3C**. Apparently, there was sufficient time between nitric acid washes for the plutonium concentration to increase until the system became slightly supercritical at the conclusion of a transfer, tripping the criticality alarms. Two people were in the plant at the time of the accident. One received an estimated dose of 2 rad, the other less than 1 rad. This excursion illustrates the subtle ways in which accidents can occur during solution processing. Although the deep trap was considered a safety feature for the control of contamination, it contributed directly to the criticality accident. The difficulty of understanding what had happened, even after it was known in which vessel the excursion occurred, is an excellent example of the requirement of having computational simulation methods capable of modelling such complex processes as those that occurred in the 1970 Windscale works nuclear criticality excursion. Such modelling and simulation (M&S) methods also assist nuclear criticality safety assessment specialists in their emergency planning and preparedness procedures. In addition, applying such M&S methods also helps in determining the subsequent radiological consequences of any such nuclear criticality excursion should it occur in a nuclear fuel processing facility.

The successful candidate for this EPSRC/MoD contractor sponsored PhD industrial CASE studentship will join, and be supported by, a vibrant and dynamic group, led by Dr Matthew Eaton, with world class expertise in the numerical modelling of multi-physics phenomena for nuclear engineering and nuclear criticality safety assessment. In the course of four years of study they will be trained in state-of-the-art numerical methods as well as the physics of transient nuclear criticality safety assessment. They will also be trained in the use of industrial static nuclear criticality safety assessment software such as the Monte Carlo neutron transport code MCNP6. The successful candidate will be sent on a wide variety of training courses such as the international nuclear engineering summer school and the Frederic Joliot/Otto Hahn Summer School in reactor physics (FJOH) which are held in France and Germany (http://www.fjohss.eu). This is in addition to specific advanced courses at Imperial College London on nuclear reactor physics, numerical methods, object oriented and parallel programming techniques as well as an experimental nuclear reactor physics course. During their studies the successful candidate will be assigned an industrial supervisor by the sponsor (MoD contractor).

The successful candidate will have the opportunity to develop their career, transferable skills and profile by presenting at international conferences and publishing in high impact nuclear engineering and numerical analysis journals. Imperial College London also has a wide variety of professional development courses that PhD students must undertake as part of their studies in addition to all the technical training. The professional development courses that the successful candidate will undertake will help develop their non-technical transferable skills. This will help widen their recruitment appeal to both engineering/science and non-science/engineering based companies. The successful candidate will have the opportunity to work with engineers and scientists from the industrial sponsor (MoD contractor) during their PhD industrial CASE studentship to help broaden their industrial experience. Candidates for this PhD industrial CASE studentship should have a good mathematical background and a good degree (First Class or Upper Second Class honours) in an appropriate field such as physics, mathematics, computer science or engineering. Applications from candidates with an MSc in scientific computing or numerical modelling are particularly welcome. It cannot be over-emphasized that the candidate must have very good mathematical skills and the ability to put physical models into a mathematical form. The successful candidate must be willing, and able to achieve, developed (or enhanced) security vetting (DV) by the industrial sponsor (MoD contractor) as well as being eligible for EPSRC PhD studentship funding. To apply for this PhD industrial CASE studentship please email Dr Matthew Eaton (m.eaton@imperial.ac.uk) with a copy of your curriculum vitae (CV).