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REACTIVITY EFFECTS AT THE MAYAK PRODUCTION ASSOCIATION, JANUARY 2, 1958 CRITICALITY ACCIDENT USING SERPENT 2 AND OPENFOAM

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ABSTRACT

The process nuclear criticality accident that occurred at the Mayak Production Association (Chelyabinsk-40) on January 2, 1958 involving a vessel of uranyl nitrate solution claimed the lives of three workers and left a fourth worker with continuing health problems. There are a myriad of uncertain parameters involved with this accident: *What was the molarity of the solution? How much solution was in the vessel at the time of the accident? In what position was the vessel and the solution when it went critical? How important was the impact of reflection due to the workers and/or the floor?* These uncertain parameters have made this accident particularly difficult to analyze in the past. This work aims to lower the uncertainty on some of these parameters. A most-probable solution composition is determined by comparing literature on the physical properties of uranyl nitrate solutions to those presented in LA-13638 [1], which describes the accident in question. Using this most-probable solution, the main contributions to the reactivity of the system and hence the eventual accident, are identified through Serpent 2 and OpenFOAM analyses. Serpent 2, a Monte Carlo software tool, is used to perform calculations of the reactivity effects of lowering the vessel toward the floor and the reactivity added by the close proximity of workers. OpenFOAM, a C++ partial differential equation solver toolkit, is used to simulate the fluid inside the vessel as the vessel is tipped. This is done by treating the solution and air inside the vessel as two incompressible, isothermal, and immiscible fluids using a volume of fluid (VoF) approach. The goal of this approach is simply to track the interface between the two fluids, and hence give an accurate description of the geometrical structure of the solution as the vessel is tipped. These two unique tools are then coupled to provide a time-dependent flow simulation to study the effect that the changing geometrical structure had on the criticality of the system, which is novel to the criticality safety field. This work provides a more accurate picture of the accident going forward.

Key Words: Serpent 2, OpenFOAM, multi-physics, prompt neutron excursion, nuclear criticality safety accident, process condition change

1 INTRODUCTION

The initial driver for this work has roots that go back over a decade, when the Monte Carlo criticality calculation of a freely moving fluid would have been difficult, if not impossible. It was thought that it would be valuable to demonstrate the impact on the multiplication factor for each change in

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a system parameter due to the process contingency of moving an experimental vessel filled with a fissile solution. An example would be to demonstrate the impact of a geometry change from a short, fat slab to a truncated hoof, caused by the vessel tilting. The benefit is that if one could show the various contributions to the reactivity for this process, then an in-depth deconstruction of an accident like the one that occurred at the Mayak Production Association on January 2, 1958 could be made that will help in future discussions on conduct of operations and process condition changes.

The advancements made in modern Monte Carlo particle transport codes have made modeling such a scenario far more feasible with interfaces that allow for coupling to other codes which produce fluid flow, density, and temperature fields. One example of such advancements is the recently implemented interface between the Serpent 2 code and the C++ partial differential equation solver toolkit OpenFOAM. Much of the work performed to date using this interface has focused on the simulations of transients with feedback from thermal expansion [2]. There are many cases of interest, such as the criticality accident being analyzed in this work, where the fissile material is in solution and is free to move within a vessel or apparatus. The movement of such a fluid can also be simulated using OpenFOAM, making coupled simulations of criticality accidents involving freely moving fissile solutions much more manageable. Other processes where fissile solutions are used, such as the extraction of medical isotopes can surely be modeled by similar methods as those presented in this study.

The accident of interest is described in detail in LA-13638 [1] along with the dimensions of the experimental vessel. This document is widely considered to be the preeminent resource for historical criticality accidents. The accident is unique in that it occurred as part of operations with a vessel used for critical experiments; however, it is considered as a process criticality accident due to the fact that it occurred after the cessation of an experiment and during the handling operations associated with transferring the fissile solution into bottles of favorable geometry. Even with the investigative work that took place regarding this accident, there are many parameters that still remain unknown. The authors acknowledge this fact and present logical explanations for the parameter evaluations within this work, such that these evaluations can be characterized as educated guesses.

The experimental setup was located in a separate room in the main processing building and had only been operational for a few months at the time of the accident. The experimental activities were set up as a response to a previous criticality accident which occurred in 1957, and had the goal of establishing measurements of the critical parameters of high concentration, highly enriched uranyl nitrate (UN) solution. The accident occurred on the first day of work after the New Year's holiday and a dedicated and knowledgeable team performed the activities. The previous series of experiments had focused on smaller vessels and had concluded prior to the end of the year. The equipment for this series of experiments had been assembled prior to the end of the year and was the first experiment conducted in this larger stainless steel vessel (750 mm inside diameter with a wall thickness of 2-4 mm). This vessel is considered representative of the vessels in common use at the facility at that time. The experimental vessel was bolted to a stand and sat atop an 8 mm thick steel support plate that was about 0.8 m above the concrete floor and at least 1.5 m from any walls. The vessel capacity was in excess of 400 L. After each experiment was completed, the written procedures called for the entirety of the solution to be drained through a line to favorable geometry 6 L bottles.

After filling some (the actual number is unknown) of these 6 L bottles, the experimenters judged that the remaining solution volume was highly subcritical and they decided to circumvent the prescribed, tedious draining process and manually pour the remaining solution from the vessel. They removed the neutron source and guide tube and unbolted the vessel from the stand. Then three experimenters began to move the vessel in order to directly pour the contents into the containers. They immediately noticed a flash and the fissile solution was violently ejected reaching the ceiling roughly 5 m above. The three experimenters, along with a fourth experimenter who was located about 2.5 m away, evacuated the room. Three experimenters died in five to six days and the fourth experimenter survived but experienced acute radiation sickness, followed by continuing health problems including cataracts and loss of sight in both eyes some years later.

2 SOLUTION CHARACTERIZATION

Many parameters surrounding this accident are uncertain, with the solution description being possibly most salient in modeling the criticality of the system. There are a myriad of factors that must be considered to properly characterize a UN solution. Density, excess nitric acid, molarity, percent hydration, uranium enrichment and concentration, and viscosity were the factors focused on within this work. Out of these parameters, only the uranium enrichment is given by LA-13638. The enrichment is quoted at 90 weight percent in ^{235}U, but any further characterization of the uranium assay was not provided. Any trace isotopes, such as ^{233}U, ^{234}U, and ^{236}U, were not reported.

The uranium concentration within the solution is quoted at two different values within LA-13638. In the text, it is reported at 418 gU/L, whereas the system figure and the diagram in the appendix show 376 gU/L. The authors believe the discrepancy is caused by assuming the uranium is enriched to 100%, (376 gU/L versus 418 gU/L for 90%) possibly for convenience. Since the solution is characterized as 90% enriched, the concentration is assumed to be 418 gU/L within this work.

The solution is described simply as a uranyl nitrate solution, without any mention of the solution's molarity. Furthermore, there is no mention of the existence of excess nitric acid or the percent hydration of the UN. Because of this, a complex three compound system consisting of all three materials, uranyl nitrate, water, and nitric acid, could have been in the vessel at the time of the accident. An exhaustive open source literature search for the physical properties of such a solution yielded few results. Many of the results lacked the viscosity correlations for certain solution characterizations. The viscosity of the fluid was necessary in the OpenFOAM modeling of the fluid behavior. If the viscosity was not representative of the solution, then the fluid would not react to the geometry change as it did during the accident. Therefore it was imperative that a viscosity correlation matched to a highly concentrated three compound system.

An addendum to a report by Krigens in 1968, ARH-724 [3], provided the necessary viscosity correlation for a three compound system. In this addendum the density, excess nitric acid, molarity, and compound concentrations are tabulated for 26 different solutions containing uranyl nitrate hexahydrate (UNH), water, and nitric acid. UNH was widely used at sites in the United States, therefore it is a reasonable assumption that the solution in the vessel could have been of this hydration. An argument could be made that the solution was in fact anhydrous, or a dihydrate, or even a trihydrate. However, any argument would be subject to the same uncertainties as the hexahydrate assertion. Furthermore, the increased hydrogen content provided by the extra water molecules increases neutron moderation and consequently the system multiplication factor. This is one of many conservative assumptions made herein.

The solution was therefore assumed to consist of

$$\underbrace{\mathrm{UO_2\left(NO_3\right)_2 \cdot 6H_2O}}_{\mathrm{UNH}} + \mathrm{HNO_3} \ + \ \mathrm{H_2O}, \tag{1}$$

of an unknown molarity. The density, UNH concentration (in g/L) and wt. %, HNO_3 concentration and molarity (in mol/L), and freezing points are reported for 26 UNH-HNO_3-H_2O systems. Using the data presented, the molar concentration for each compound was calculated. In addition,

$$\log\left(\eta\right) = 0.9509 + 0.2456 c_{\mathrm{UNH}} + 0.00854 c_{\mathrm{HNO_3}} + 0.00907 c_{\mathrm{UNH}}^2, \tag{2}$$

was used to calculate the dynamic viscosity for each solution. The correlation uses concentrations in moles per liter at a temperature of 25 °C and the resulting viscosity is given in millipoise (mP). It is important to note that the density and viscosity are both quoted at 25 °C. While the outside temperature was much lower than this during the time of the accident, the freezing point of many of these solutions indicate that the internal temperature must have been maintained much higher than the frigid conditions outside. Therefore, it is assumed that the solution was maintained near room temperature at 25 °C.

The results of these calculations are shown in Table I. The composition of each solution was determined by the data originally given on page 21 of ARH-724. With the composition known, the uranium concentration was calculated and a percent difference to the quoted solution concentration (418 gU/L) was performed for comparison. Four solutions were within ten percent and they are bolded in the table. While Solution 20 most closely represents the target solution, Solution 21 was selected as the best estimate solution because it is the most representative true three compound system since Solution 20 had no excess nitric acid. Furthermore, Solution 21 is more hydrogenous than Solution 20. This reinforced the Solution 21 selection.

The best estimate solution is consistently used in Serpent 2 and OpenFOAM to ensure realistic multi physics phenomena throughout coupling. Table II shows the physical properties and isotopic weight percentages of the best estimate solution. The weight percentages were calculated using NIST isotopic data [4] and were used to build a material card within Serpent 2. The density was used in both Serpent 2 and OpenFoam, while the viscosity was used exclusively within OpenFOAM. Any further reference to a solution refers to this best estimate solution and the properties outlined.

Table I. Solution characteristics and uranium concentration comparison.

Soln No.	ρ_{sol} (g/cc)	w_{UNH}	w_{HNO_3}	w_{H_2O}	M_{sol} (g/mol)	C_U (gU/L)	$\frac{\lvert 418 - C_U \rvert}{418}$	Viscosity (mP)
1	1.8707	0.704	0.0337	0.2623	60.60	620.57	0.4846	5.3128
2	1.8481	0.696	0.0273	0.2767	58.20	606.11	0.4500	5.2092
3	1.8733	0.78	0.0000	0.2200	72.62	688.52	0.6472	5.7377
4	1.7658	0.688	0.0000	0.3120	53.50	572.46	0.3695	4.9628
5	1.768	0.688	0.0000	0.3120	53.50	573.17	0.3712	4.9672
6	1.6988	0.6535	0.0000	0.3465	48.69	523.12	0.2515	4.6722
7	1.7491	0.6461	0.0276	0.3263	50.40	532.51	0.2740	4.7569
8	1.2357	0.299	0.0000	0.7010	25.32	174.10	0.5835	3.1191
9	1.2659	0.2907	0.0456	0.6637	26.22	173.40	0.5852	3.1412
10	1.0669	0.101	0.0000	0.8990	19.96	50.78	0.8785	2.7300
11	1.0994	0.0975	0.0569	0.8456	20.82	50.51	0.8792	2.7525
12	1.2579	0.086	0.1409	0.7731	22.07	50.98	0.8780	2.7970
13	1.3287	0.296	0.1343	0.5697	29.12	185.32	0.5566	3.2353
14	1.7644	0.626	0.0978	0.2762	55.15	520.46	0.2451	4.7674
15	1.8956	0.713	0.0741	0.2129	69.36	636.87	0.5236	5.4794
16	1.0758	0.1	0.0169	0.8831	20.21	50.69	0.8787	2.7365
17	1.1493	0.322	0.0154	0.6626	26.55	174.38	0.5828	3.1276
18	1.7092	0.648	0.0104	0.3416	48.97	521.89	0.2486	4.6765
19	1.7903	0.698	0.0102	0.2918	56.34	588.84	0.4087	5.0768
20	**1.5789**	**0.5778**	**0.0000**	**0.4222**	**40.67**	**429.88**	**0.0284**	**4.1778**
21	**1.5477**	**0.5491**	**0.0098**	**0.4411**	**38.86**	**400.45**	**0.0420**	**4.0435**
22	1.4699	0.4688	0.0412	0.4900	34.74	324.71	0.2232	3.7257
23	1.5317	0.4529	0.1250	0.4221	38.00	326.88	0.2180	3.8018
24	1.3862	0.333	0.1361	0.5309	30.97	217.51	0.4796	3.3576
25	**1.5628**	**0.529**	**0.0320**	**0.4390**	**38.57**	**389.56**	**0.0680**	**4.0112**
26	**1.5621**	**0.515**	**0.0426**	**0.4424**	**38.08**	**379.08**	**0.0931**	**3.9712**

Table II. Best Estimate Solution Properties

ρ	η	C_U	Isotopic Composition (wt. %)							
(g/cm^3)	(mP)	(gU/L)	^{1}H	^{2}H	^{14}N	^{15}N	^{16}O	^{18}O	^{235}U	^{238}U
1.5477	4.0435×10^{-4}	400.45	6.279	0.001	3.284	0.013	64.387	0.149	23.287	2.587

3 SERPENT 2 MODEL

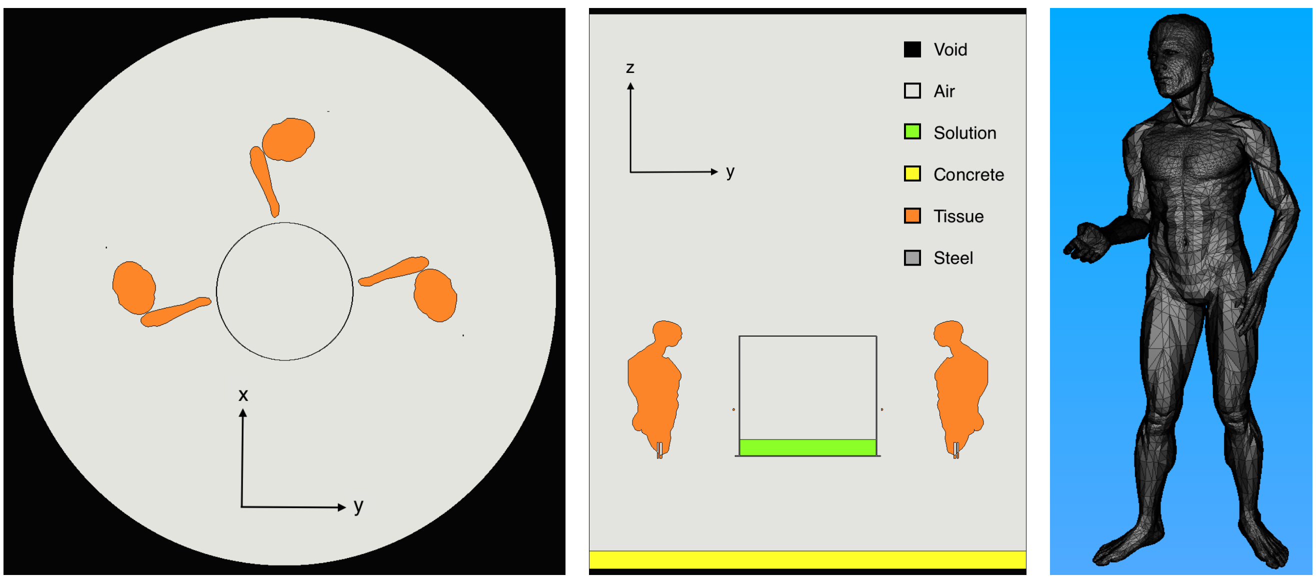
Serpent 2 is a beta-released extension of the recently popular, publicly available software Serpent. Serpent is a three-dimensional, continuous energy Monte Carlo neutral particle transport code commonly used for reactor physics and burnup, developed at VTT Technical Research Centre of Finland since 2004 [5]. Serpent employs a unionized energy grid format which results in a major speed-up in calculation at the expense of large memory use. Serpent has many similarities to the ubiquitous MCNP, such as universe-based combinatorial solid geometry modeling and ACE formatted cross section libraries.

The model is contained within a cylinder with a 1.5 m radius and a height of 5 m. These dimensions correspond to the assertion in LA-13638 that the cylinder was at least 1.5 m from any wall or other equipment and an appropriate ceiling height for a laboratory, respectively. This cylinder models the room in which the accident occurred and is filled with air. Anything outside of this room is modeled by void. A 6 inch thick slab of *Concrete, Los Alamos (MCNP)* is positioned in the bottom of the cylinder and represents the floor of the laboratory. The experimenters are modeled with anatomically correct phantoms consisting of *Tissue Equivalent, MS20*. A vessel support stand made of *Steel, Stainless 304* (8 mm thick cylinder with a 400 mm radius) was positioned 800 mm from the top of the floor in the center of the cylindrical room. The stand support structure, detectors, and other ancillary equipment used in the vicinity of the vessel were not modeled. This equipment was neglected because the exact details of the equipment are unknown. The vessel was modeled with a 750 mm inner diameter, 1000 mm inner height, and a 3 mm wall thickness consisting of *Steel, Stainless 304*. All of the aforementioned material names correspond to the material definitions outlined in PNNL-15870 [6].

There are three separate factors that could have contributed to the excursion: the additional neutron reflection caused by the three experimenters surrounding the vessel to lower and pour out the solution, the additional reflection caused by lowering the vessel toward the concrete floor, and the geometry of the fluid caused by tipping the vessel. The first two effects can be investigating by performing static calculations in Serpent 2. The last must be investigated by coupling OpenFOAM and Serpent 2 and will be discussed in the next section. Understanding how much solution would cause the system to go critical is also valuable because it allows for this study to set some bounding conditions. The experienced and knowledgable staff believed the vessel to be highly subcritical after draining the solution into some number of 6 L containers.

To determine the critical and subcritical solution volumes under accident conditions, the vessel was placed on the stand, without any reflectors nearby. The volume of the solution was changed, by changing the position of the air/solution interface in the vessel, and the system multiplication factor was calculated by averaging the absorption, analog, collision, and implicit estimates. The critical volume is defined as having $k_{avg} + 2\sigma = 1.00$ and the subcritical volume is defined as having $k_{avg} + 2\sigma = 0.98$. These definitions are simply used as a reference to show the changes in the system multiplication factor caused by the various reactivity effects. Serpent 2 reports the calculated multiplication factor out to five decimal places, but the volume was only changed in 0.1 liter increments. The volume that yielded results closest to the stated goals was accepted. All of the static simulations were performed using 50,000 particle histories per cycle, for 200 cycles (with the 50 initial cycles discarded) for a total of 7.5 million particles being used in the tallies. The averaged multiplication factor typically had an uncertainty of approximately 2×10^{-4}.

With the subcritical and critical volumes known, the reactivity effects of the concrete floor and the experimenters were studied. The subcritical volume is used to show how these effects may cause the system to become critical even though it was subcritical while isolated on the stand. The critical volume is used to compare the volume difference between it and the subcritical volume. The subcritical volume was then placed in the vessel, and surrounded by zero, one, two, and three anatomically-correct phantoms, and the vessel was lowered toward the floor in 100 mm increments. The system multiplication factor was calculated at each height for each configuration. Figure 1 shows the problem geometry including the three phantoms representing the experimenters.



(a) Bird's Eye View at z = 126 mm. **(b) yz Elevation View at x = 0 mm.** **(c) Phantom Model.**

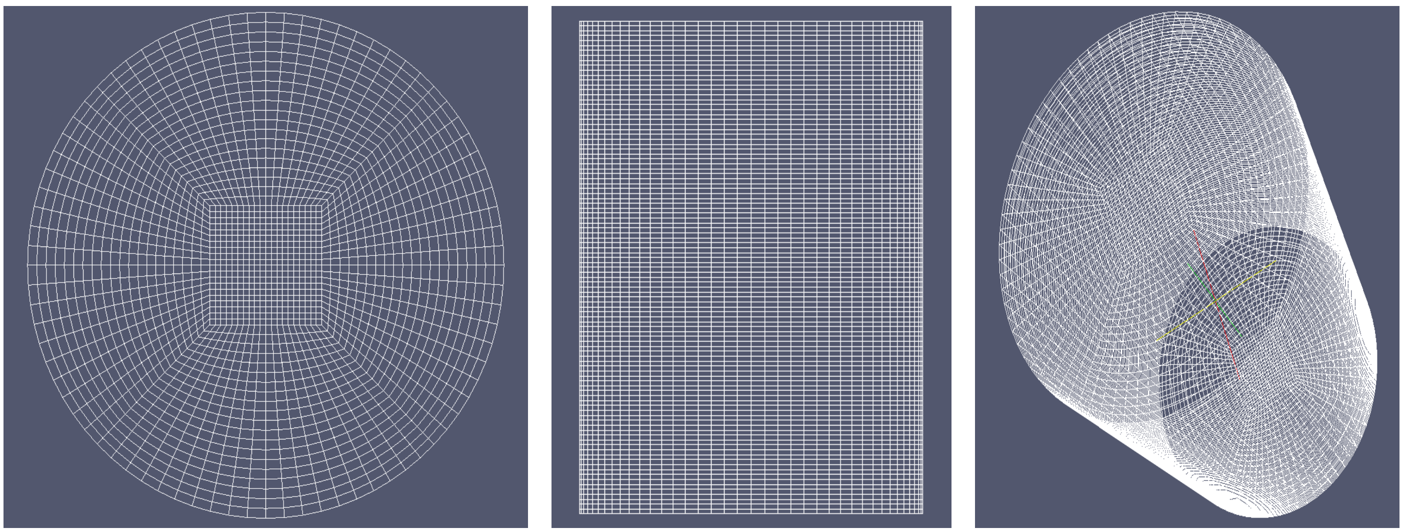
Figure 1. Serpent 2 Model of the Accident Environment.

4 OPENFOAM SIMULATION COUPLING

The OpenFOAM simulation uses the standard solver InterDymFOAM, which tracks the interface between two incompressible fluids using the volume of fluid (VoF) method. This method was first introduced by Hirt and Nichols in 1981 [7]. The basis of the method is a transport equation for the volume fraction for the m^{th} fluid $\alpha_m(\vec{r}, t)$:

$$\frac{\partial \alpha_m}{\partial t} + \vec{v} \cdot \nabla \alpha_m = 0 \tag{3}$$

where $\alpha_m = 1$ in cells filled completely with fluid m, and $\alpha_m = 0$ in cells completely devoid of fluid m. Cells in the domain with values in between these two extremes then contain the interface in a two fluid simulation. The necessary information required to solve for the flow field are the density and viscosity of the fluid, which were determined in Section 2, along with the density and viscosity of air which were taken to be 1 kg/m^3 and 1.48×10^{-5} m^2/s. The simulation begins with the fluid filled to a height of 132 mm as specified in LA-13638. The vessel is then tipped to an angle of $75°$ over the course of 1 s, at which point the vessel remained at this angle for the duration of the simulation. The length of the simulation was set to be 40 s with a time step size of 0.01 s. Although there is no data detailing how far the vessel was tipped, or how quickly, the authors judge these values to be reasonable for the purposes of this study. The grid used in the finite volume calculations is shown in Figure 2. This grid contained exactly 200,000 cells.



(a) Top view. **(b) Side view.** **(c) Tilted view.**

Figure 2. Grid used in the OpenFOAM Serpent 2 coupling.

The output of the simulation is the volume fraction in each cell at each timestep. The Serpent 2 - OpenFOAM interface was then used to import the OpenFOAM grid geometry into Serpent 2. If the volume fraction in a cell was less than 0.01, the cell in the Serpent 2 simulation was filled with air. Otherwise, it was filled with the uranyl nitrate solution specified in Section 2 at a density equal to the nominal density given in Table II times the volume fraction of the cell. The OpenFOAM grid geometry was modeled in Serpent 2 to be enclosed by the stainless steel vessel. The vessel was not modeled on any stand or in the vicinity of any phantoms or any concrete floors as the purpose of this simulation was to isolate the effect on the reactivity that the change in shape had on the fluid. The simulation can then be thought of as the vessel floating in air as it tips to an angle of $75°$ and then stops abruptly to simulate the sloshing of the fluid inside.

5 RESULTS

The purpose of this study has been to investigate the impact on the calculated system multiplication factor due to a change on a system parameter based on a process condition change. These changes include the additional reflection related to the workers, additional reflection related to the concrete floor, and the change in geometrical shape of the solution due to movement of the vessel. The first two of these are able to be modeled with static calculations using Serpent 2 alone. The last of these is modeled using dynamic calculations through the coupling of OpenFOAM and Serpent 2. As such, this section is broken into two sections for results of the static and dynamic calculations respectively.

5.1 Static Results

The best-estimate solution has a bare critical spherical volume of 17.5 L, and when reflected by the vessel (3 mm of SS-304) the critical volume is only 16.2 liters. When reflected by 60 cm of water, as a worst case scenario, the critical volume is reduced further to 7.1 L. The critical masses are 27.1 kg (∼60 lb), 25.1 kg (∼55 lb), and 11.1 kg (∼24 lb), accordingly. Simulations showed that with the vessel on the stand and no workers nearby, as the situation would be during a critical experiment, the critical volume of the solution was 61.2 L. The solution volume needed to meet the subcriticality goal of k = 0.98 was 59.4 liters. This resulted in a relatively small volume difference of 1.8 liters, with a reactivity change of $\Delta\rho = \$2.38$ using $\beta_{eff} \approx 0.0087$, which was also calculated using Serpent 2. The relatively large value of β_{eff} indicates the high leakage rate, causing a larger fraction of thermal neutrons in the system to be originally born delayed, since prompt neutrons emerge with higher energies and are thus more likely to leak. The vessel with these two volumes of solution in it would weigh 172 kg (∼379 lb) and 169 kg (∼373 lb), respectively. These masses are over six times larger than the bare critical mass for this solution.

To accurately simulate the lowering of the vessel by experimenters, the vessel must be moved off of, but still adjacent to, the stand support plate. By doing only this, with no experimenters in the vicinity, the multiplication factor dropped by 0.038 to k = 0.942. This 8 mm steel support plate caused a relatively large reactivity change, $\Delta\rho_{\text{plate}} = \$4.72$. For the effects of the floor, the vessel was lowered to the floor, in 100 mm increments, while in the presence of either zero, one, two, or three phantoms (experimenters). During all of the various elevation simulations, the vessel is off of, but adjacent to, the stand support plate. Figure 3 shows how the system multiplication factor changes as a function of both elevation and number of phantoms. The salient point of this plot is that, as expected, k is affected by the proximity of the floor much more than the proximity of phantoms due to a higher neutron leakage rate from the bottom and top of the short flat slab geometry than from the side.

To determine the reactivity worth of each experimenter individually, the multiplication factors for each phantom configuration, at every elevation, were compared to the multiplication factors without any phantoms. These Δk values were then averaged over the various elevations to determine an average experimenter reactivity worth. The reactivity worth of each phantom was determined to be statistically insignificant. Another measure of the experimenters' worth is their effect on the elevation at which the solution goes critical. With no experimenters present the critical height of the vessel was determined to be 152 mm. The critical height of the vessel in the presence of one, two, and three experimenters was 153, 154, and 155 mm, respectively. Thus, the experimenters only weakly influence the critical height of the vessel whereas lowering the vessel approximately 650 mm causes a average reactivity change of, $\Delta\rho_{\text{elevation}} = \$7.01$.

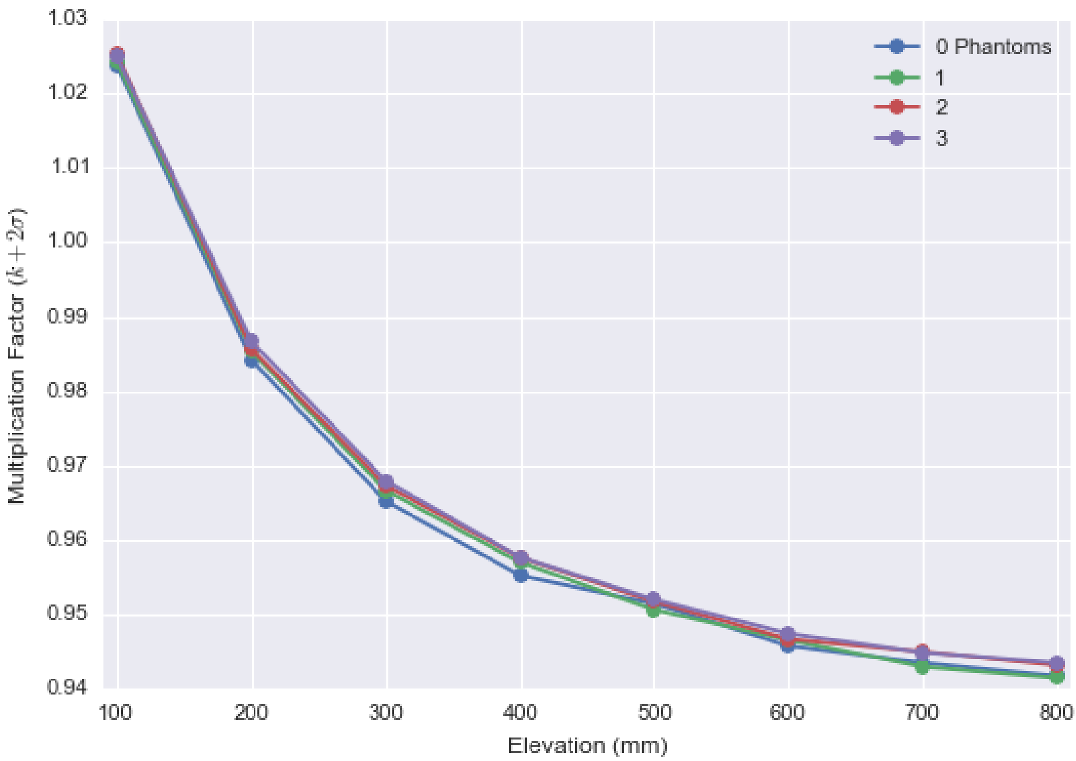
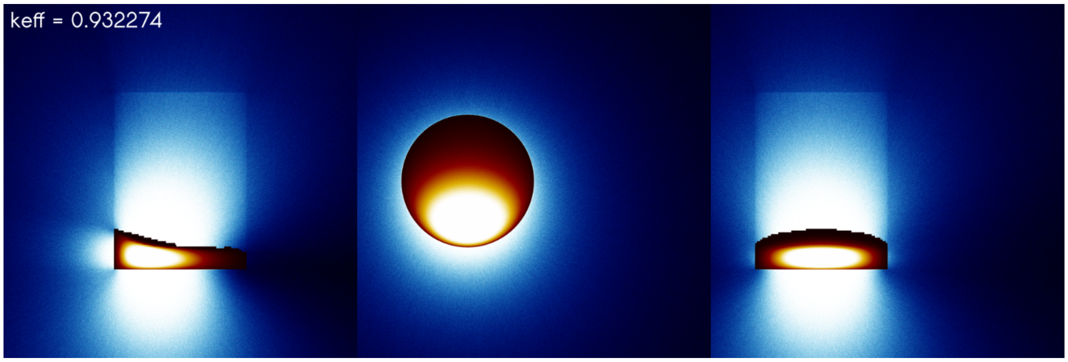


Figure 3. System multiplication factor as a function of elevation and phantoms.

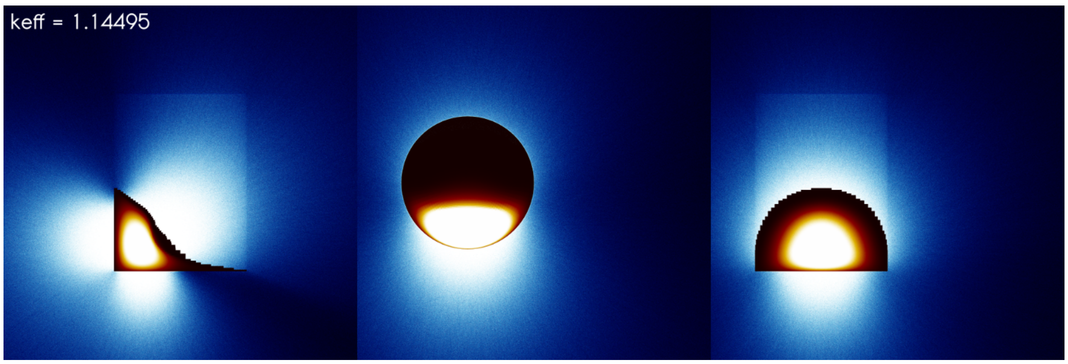
5.2 Dynamic Results

The results of the dynamic simulation are shown for 4 of the 21 time steps within the first second of the OpenFOAM simulation in Figure 4. The effect of the shape change on the multiplication factor is quite large, ranging from a minimum of $k = 0.906$ to a maximum of $k = 1.157$. This results in a reactivity change of $\Delta\rho_{\text{geometry}} = \$27.47$. For each criticality calculation, a total of 4×10^8 particle histories were run and the largest statistical standard deviation remained below $\sigma = 7 \times 10^{-5}$. The multiplication factors and reactivity changes for each time step and each tilt angle within the first second are given in Table III. All reported reactivity changes in this table are from the initial timestep before the vessel was tilted.

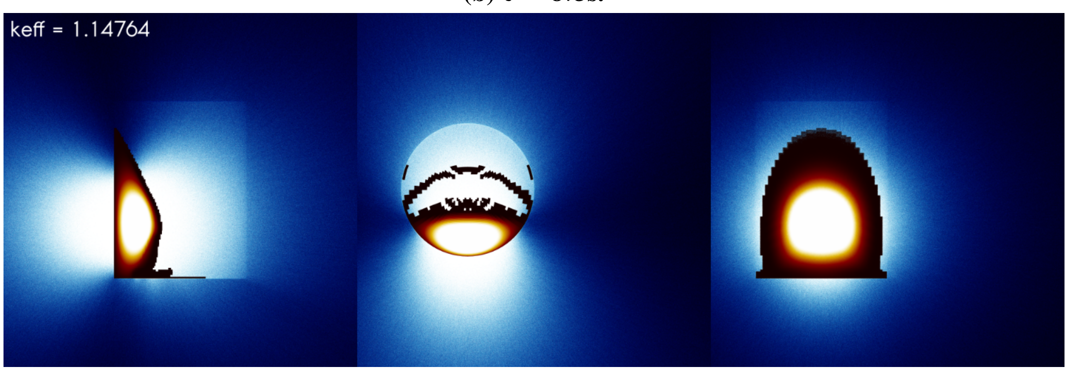
At this point, a few remarks are in order. It is acknowledged by the authors that the simulation results are meaningless as the system approaches $k \geq 1 + \beta$. In this accident, the prompt critical excursion resulted in the solution being ejected from the vessel, with some solution reaching the ceiling almost 5 m above. Regardless, the simulation is extremely valuable in that is shows just how drastic the reactivity effects could be from a geometrical shape change alone, without any added reflection. While it may be true that every nuclear engineer learns that geometry plays a significant role in the criticality of a system at an early point in their education, it is interesting to see this in the context of this scenario, and not for specific shapes like spheres, cylinders, and parallelepipeds.



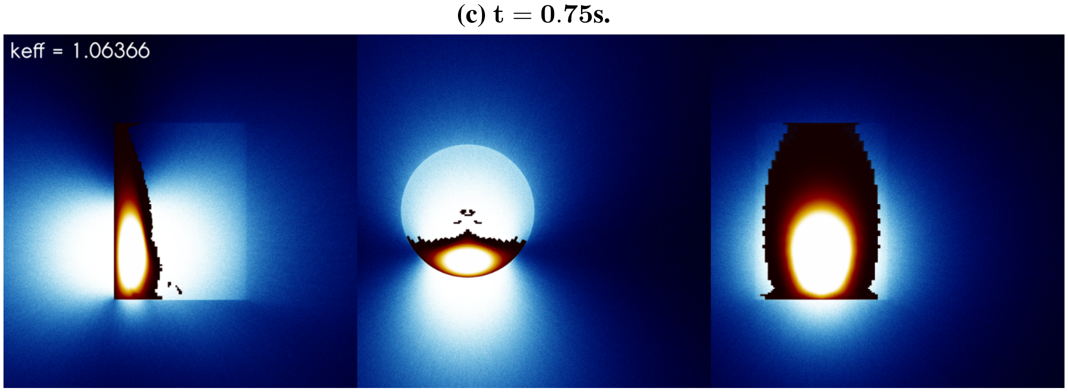
(a) $\mathbf{t = 0.25s.}$



(b) $\mathbf{t = 0.5s.}$



(c) $\mathbf{t = 0.75s.}$



(d) $\mathbf{t = 0.9s.}$

Figure 4. Thermal flux (in shades of bright white to dark blue in non-fissionable materials), fission rate density (in shades of bright yellow to dark brown in fissionable materials), and criticality (top left corner) at various time steps in the simulation.

Table III. Results for each time step of the dynamic simulation within the first second.

Time (s)	Tilt angle (degrees)	k_{eff}	$\Delta\rho$ (\$)
0.00	0.00	0.9142	0.000
0.05	3.75	0.9137	-0.072
0.10	7.50	0.9102	-0.554
0.15	11.25	0.9061	-1.118
0.20	15.00	0.9097	-0.615
0.25	18.75	0.9323	2.438
0.30	22.50	0.9748	7.817
0.35	26.25	1.0268	13.788
0.40	30.00	1.0764	18.945
0.45	33.75	1.1170	22.825
0.50	37.50	1.1450	25.340
0.55	41.25	1.1567	26.357
0.60	45.00	1.1530	26.037
0.65	48.75	1.1472	25.533
0.70	52.50	1.1476	25.570
0.75	56.25	1.1476	25.575
0.80	60.00	1.1360	24.552
0.85	63.75	1.1074	21.939
0.90	67.50	1.0637	17.667
0.95	71.25	1.0092	11.836
1.00	75.00	0.9485	4.542

6 CONCLUSIONS

It is clear from the static simulations that the experimenters had very little effect on the criticality of the system. Although they do add reactivity to the system, it is negligible compared to the proximity of the concrete floor and insignificant compared to the primary reactivity addition due to the change in solution geometry in the vessel. Because the quiescent solution takes the shape of a short and wide cylinder, the majority of the neutron leakage is from the top and bottom of the vessel. Thus, adding experimenters around the sides of the vessel adds little reactivity whereas the steel support stand and the floor add significant reactivity to the system.

It is important to note that by only performing these static simulations, it would appear that any solution volume in the vessel less than the critical volume of 61.2 L would be determined subcritical; however during a process condition change such as tipping, this is a substantial error as shown by the dynamic simulation and by noting that any solution volume in the vessel which is greater than the spherical reflected critical volume is not truly in a safe configuration. While the shape of the solution never truly attains a spherical form, it does become more compact reducing neutron leakage and increasing the multiplication factor of the system. It was determined that the change in shape of the fluid, as the experimenters tilted the vessel to pour the solution, had the most dominant effect of the system multiplication factor. A summary of the reactivity effects are shown in Table IV. It is important to understand that the reported reactivity worths border on the outlandish. Their exact values are not of interest because this accident cannot be reversed decades later. However, their relative magnitudes provide lessons learned. The authors believe this will be a valuable teaching tool when presenting this accident in the future to operations personnel as it clearly demonstrates a very large Δk due to something that is easy to relate to, understand, and visualize.

Table IV. Summary of Reactivity Effects

Effect	$\Delta\rho$ ($)
Solution Geometry	27.47
Vessel Elevation	7.01
Support Plate	4.72
Experimenter	< 0.08*

* Results statistically insignificant

In reality, this careful deconstruction of the accident does not change the fact that much of the details regarding the accident will never be known with certainty. For instance, even though it has been shown that tipping the vessel in the air before lowering to the floor would have been enough to cause an excursion, it is unknown whether the vessel was placed on the floor without rotation, in which case it would have gone critical without tipping as shown by the static calculations. The purpose of this study has been as much about demonstrating the ability of Serpent 2 and OpenFOAM as much as it has been about deconstructing the accident. The CAD geometry modeling of the human phantoms and the multi-physics coupling of the two codes are central to the work performed here; they have applications in modeling any system where more geometric accuracy is desired, or where the fissile material is in solution that is free to move within a vessel or apparatus.

The authors feel that the most probable course of events for this accident is that the experimenters unbolted the vessel and slid it to the edge of the stand then, due to the weight of the solution and vessel, used the edge to begin to tip the vessel before lowering it. The fact that solution was ejected onto the ceiling 5 m above indicates that the solution reached a prompt critical configuration at a relatively small tilt angle. In the authors' judgment, the main driver of this prompt critical excursion was the geometric change of the solution due to vessel movement. Future work includes a more rigorous investigation of the reactivity effects caused by the movement of the vessel. The work aims to show how varying the solution viscosity, solution volume, and tipping rate can effect the system multiplication factor. The simulation techniques presented can also be applied to other criticality accidents, in which a fissile solution is present such as the accident described on page 16 of LA-13638, as well as other applications such a Mo-99 extraction from an aqueous fissile solution.

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