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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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Outline

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Accident overview

- Occurred during fissile material handling operations, after the cessation of a nuclear criticality safety experiment.
  - Categorized as a process criticality accident by LA-13638.
- Experiment was in response to a prior criticality accident on April 21, 1957.
  - The goal was to measure critical parameters of high concentration, highly enriched uranyl nitrate solutions for commonly used processing vessels.
- Prior experiments had used smaller vessels and this was the first performed in the larger vessel.
  - 750 mm inside diameter.
  - 2 to 4 mm wall thickness.
  - 1 m tall.

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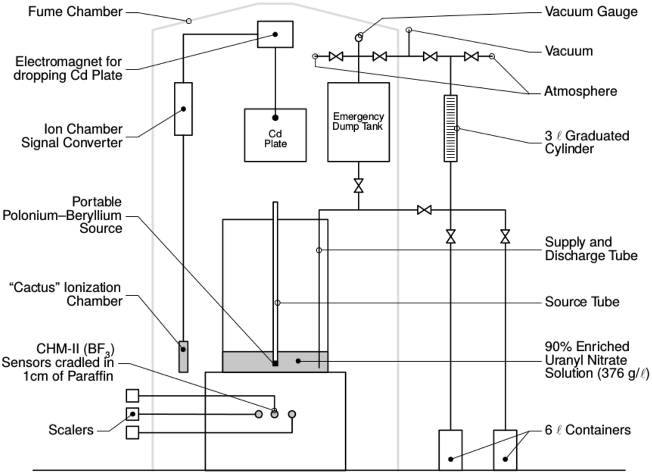
Experimental setup



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Accident description

- The vessel was bolted to an 8 mm thick steel support plate roughly 0.8 m ($\approx$ 31 in) above the concrete floor.
- Vessel capacity: $\approx$ 400 L.
- After each experiment, written procedures called for the draining of the uranyl nitrate solution to 6 L bottles of favorable geometry.
- Workers filled some of these before unbolting the vessel from the stand and removing the neutron source and guide tube in order to manually drain it.
  - The actual number of bottles filled is unknown.
  - The number of bottles filled was enough that the experimenters judged the remaining solution to be highly subcritical.
- Three experimenters were involved in moving/tipping the vessel to directly pour the contents into the 6 L bottles when the excursion occurred.

Casualties and contributing factors

- The three workers died within six days, and a fourth, who was $\approx$ 2.5 m away developed cataracts and loss of sight in both eyes.
- Estimated doses:
  - 6,000 $\pm$ 2,000 rad for the three workers.
  - 600 rad for the worker 2.5 m away.
- Criticality safety contributing factors:
  - Violation of the draining procedure.
  - Unbolting the vessel was not specifically permitted.
    - Draining process possibly encouraged work arounds.
    - Experiment design made it easy to unbolt and remove the vessel.
- Postulated contributing factors:
  - Reflection from the workers.
  - Reflection from the concrete floor.
  - Shape changes of the solution itself during tipping.

Purpose of this study

- There are too many unknown factors involved to accurately and realistically model the accident accurately.
- No amount of analysis is going to undo what has already been done.
- Instead, we aim to deconstruct the accident to determine the relative importance of the postulated contributing factors.
- Tools:

Serpent 2 beta

OpenFOAM

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Uncertain parameters

- Specific details are not known about the accident.
- Among the unknowns are:
  - How many 6 L bottles were removed prior to the accident?
  - How close were the experimenters' bodies to the outside of the vessel during vessel movement?
  - Did the experimenters set the vessel on the ground prior to tipping it?
  - What was the exact composition of the solution?
- The last of these is especially difficult to answer.
  - Was there excess nitric acid?
  - Was there excess water?
  - What was the molarity?
  - What was the concentration in gU/L?
  - What were the physical properties of the fluids(density, viscosity, etc. . .)?
- If we are to model the accident, we must make some assumptions.

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Solution assumptions

- All known data about this accident appears in LA-13638 by McLaughlin, *et al*.
- They give the concentration as 418 gU/L in the text and 376 gU/L in the appendix.
- The assumption was made that the solution was a uranyl nitrate hexahydrate (UNH), with excess water and nitric acid.

$$\underbrace{UO_2\,(NO_3)_2 \cdot 6H_2O}_{\text{UNH}} + HNO_3 + H_2O$$

- This was assumed because UNH was widely used in the United States at that time.
  - And because it was the only uranyl nitrate solution for which densities and viscosities could be found.

Solution candidates

Soln No.	ρ_{sol} (g/cc)	w_{UNH}	w_{HNO_3}	w_{H_2O}	M_{sol} (g/mol)	C_U (gU/L)	$\frac{\|418 - C_U\|}{418}$	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**

Final solution

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

- This is the solution composition used in all simulations presented for the remainder of this presentation.
- Knowledge of the density is necessary for both radiation transport in Serpent 2 and fluid flow simulation in OpenFOAM.
- Knowledge of the viscosity is necessary for the fluid flow simulation in OpenFOAM.

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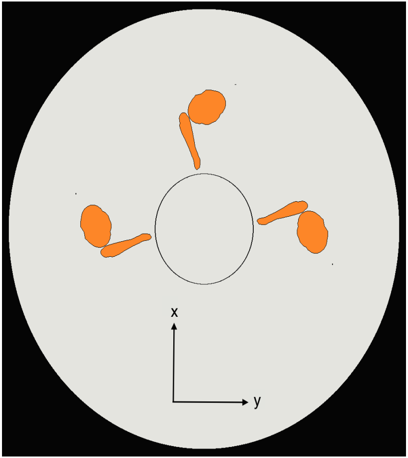
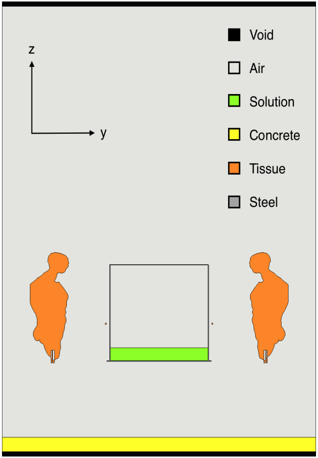
Serpent 2 Model details

- Room modeled as a cylinder of 1.5 m radius and 5 m height.
  - This is because LA-13638 states that the vessel was at least 1.5 m from any wall or other equipment.
- The floor is modeled as a 6 inch thick slab of *Concrete, Los Alamos (MCNP)*.
- The experimenters are modeled as anatomically correct phantoms made of *Tissue Equivalent, MS20*.
- The vessel and support stand are modeled as *Steel, Stainless 304*.
  - Vessel: 750 mm inner diameter, 3mm wall thickness, 1 m inner height.
  - Support stand: 8 mm thick, 400 mm radius, positioned 800 mm from the top of the floor.
- All material definitions above were obtained from PNNL-15870.

Serpent 2 Model geometry





Serpent 2 calculations

- Critical and subcritical quiescent fluid volumes in the vessel on the stand.
- Bare critical spherical volume.
- Reflected critical spherical volume.
- Criticality as a function of distance from the floor, with and without experimenters present.
- Total reactivity worth of the concrete floor.
- Total reactivity worth of the experimenters.
- Criticality as a function of solution geometry (coupled to OpenFOAM).

OpenFOAM description

- OpenFOAM is a C++ partial differential equation solving toolkit.
- InterDymFOAM is a solver within OpenFOAM that uses a volume of fluid method to track the interface between two fluids as the mesh moves dynamically.
- The basis of the method is a transport equation for the volume fraction for the m^{th} fluid $\alpha_m(\vec{r}, t)$:

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

  which is solved after obtaining the velocity field.
- The necessary information required to solve for the velocity field are the density and viscosity of the fluids.

OpenFOAM model

- Fluid properties:
  - UNH: $\rho = 1{,}547.7$ kg/m^3; $\nu = 2.61262 \times 10^{-11}$ m^2/s.
  - air: $\rho = 1$ kg/m^3; $\nu = 1.48 \times 10^{-5}$ m^2/s.
- Quiescent fluid height set to 132 mm as specified in the appendix of LA-13638.
- Vessel is tipped to an angle of 75° over the course of 1 s.
- Vessel remains at 75° for duration of simulation.
- Total simulation time: 40 s.
- Time step: 0.01 s.
- Grid size: 200,000 cells.

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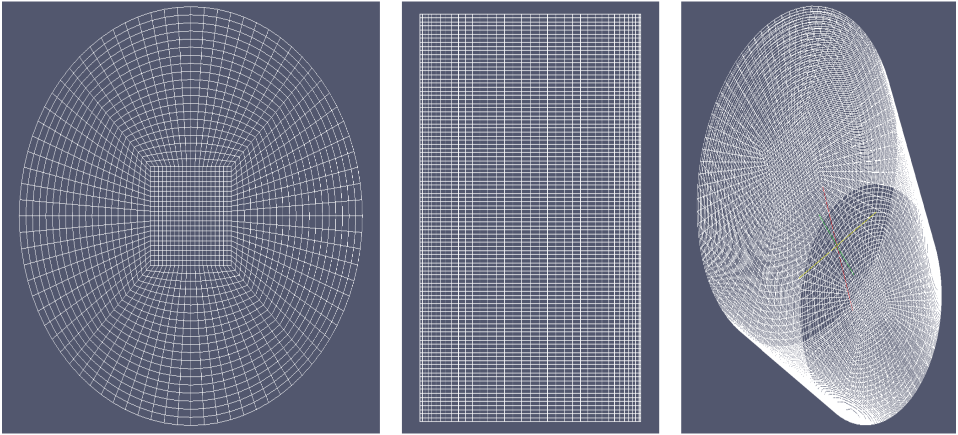
OpenFOAM Model geometry



Serpent 2 and OpenFOAM coupled calculations

- Output of OpenFOAM simulation is the volume fraction in each cell for each time step.
- From this volume fraction, we determine the material in each cell, and its density.
- This material and density field are fed to Serpent 2 for radiation transport simulation to calculate k_{eff}.
- End result: the k_{eff} of the solution as a function of time as the shape of the fluid changes, without reflection from either the experimenters or the floor.
- This isolates the reactivity effect due to the changing geometry of the fluid.

Serpent 2 static simulation results

- Critical quiescent volume in vessel on stand: 61.2 L.
- Subcritical quiescent volume ($k_{eff} = 0.98$): 59.4 L.
  - These values correspond to the vessel placed on the support stand with no experimenters present.
- Bare critical spherical volume: 17.5 L.
- Reflected critical spherical volume (reflected by 60 cm H_2O): 7.1 L.

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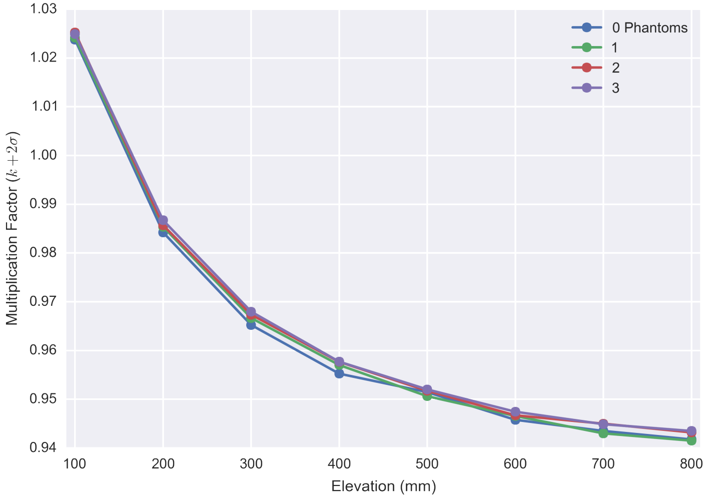
Reflection study



OpenFOAM simulation

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Coupled results

Coupled results

Conclusions

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

* Results statistically insignificant

- Experimenters have little (if any) affect on the system.
- Proximity to the floor and stand have a significant reactivity effect.
- Solution geometry has a dominant effect on the reactivity of the system.
- The results are as much about showcasing this novel multi-physics coupling, as they are about deconstructing the accident.
- Pertinent example for NCS classes going forward.