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Zirconium Sludge Criticality Calculations in Large Process Tanks

Brindley Wade, Nathan Devine

Nuclear and Criticality Safety Engineering

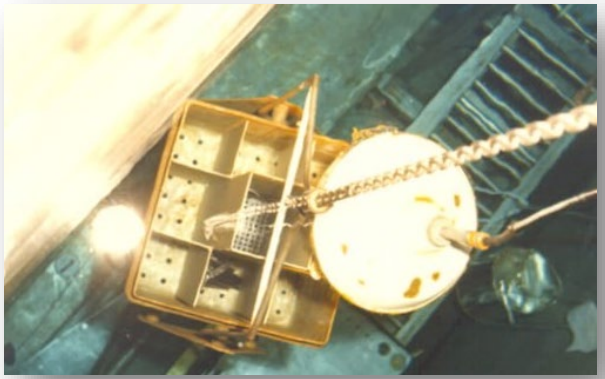
American Nuclear Society 2024 Annual Meeting, June 16-19, 2024.

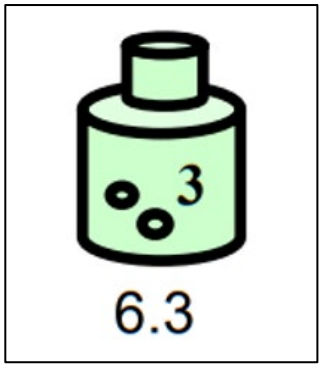
Overview

- **The Savannah River Site's H-Canyon facility has been tasked with accelerating the disposition of used research reactor fuels.**
- **Future missions will involve dissolution of "Non-Aluminum" clad fuels**
  - not easily dissolved
  - Nitric acid is not capable of dissolving stainless steel, Zirconium, and Hastelloy, clad fuels by itself
- **The electrolytic dissolver has been (re) selected as a disposition path for these fuels.**

Project Introduction

- 6.3D is the dissolver tank designed to dissolve ‘NA-SNF’ through electrolytic dissolution.
- Zr clad fuel is the concern, with ‘sludge’ creation happening historically for Zr fuels.
- The tank is approximately 8 ft. tall with an 8 ft. diameter.



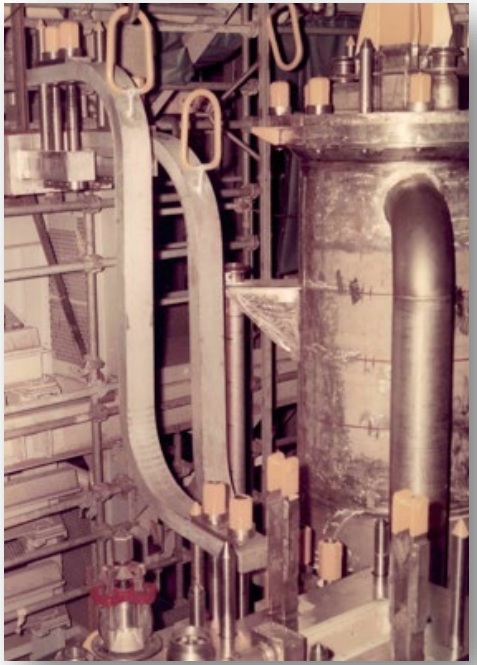


Inputs and Assumptions

From Savannah River National Laboratory Literature Review:

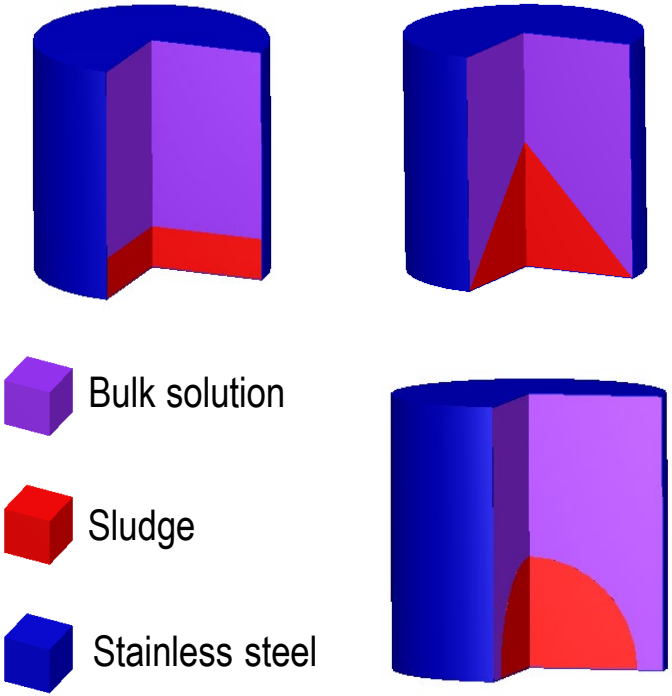
- 85% of the zirconium is converted to an oxide resulting in about 0.33 gallons of sludge settling to the tank bottom per kilogram of zirconium dissolved.
- Reports from the 1960s show a 7% holdup of uranium in the sludge, but further rinsing the sludge reduces this to 0.5%.
- 40kg of U-235 per batch is the conservative maximum, assuming 40 kg of beginning of life U-235 fuel.
- Use only pre-defined “Campaign 1” fuels.

Evaluate the k-effective and feasibility for these scenarios.



Project Setup

- One 40kg U-235 batch will be initially evaluated.
  - Vary the percentages of uranium holdup.
- Three different geometries representing potential sludge buildup.
  - Actual sludge geometry is unknown.
- Assume 95% U-235 enrichment (conservative)
- Bulk solution is uranyl nitrate.
- SCALE 6.1 will be used for all calculations.
  - ENDF-VII 238 cross-section library.



Material Compositions

Material	SCALE 6.1 Composition Title	Density (g/cc)	Purpose in Model
Stainless steel	ss304	7.94	Vessel material
Sludge	wtptsludge	Dependent on holdup	Byproduct of dissolving

Uranyl Nitrate (bulk solution)	SCALE 6.1 Composition Title	Number Density (g/cc)	Purpose in Model
U-235	u-235	1.458E-5	Byproduct of dissolving
Hydrogen	h	0.054	Byproduct of dissolving
Nitrogen	n	0.004	Byproduct of dissolving
Oxygen	o	0.036	Byproduct of dissolving

Hold Up Percentages

All of Campaign 1	Dissolved zirconium (kg)	Starting uranium (kg)	Uranium in sludge (kg)	U-235 in sludge (kg)	Total mass (kg)	Total volume (liters)	Density (g/cc)
7% Uranium Holdup	1,607.00	12,917.00	904.19	858.98	2,748.33	2,027.73	1.36
0.5% Uranium Holdup	1,607.00	12,917.00	64.59	61.36	1,908.73	2,027.73	0.94

- Rinsing = reducing the uranium holdup by using a dilute acid spray and agitating the sludge mixture.
  - The holdup for unrinsed sludge is typically 7%, but it can be reduced to 0.5% by rinsing.
  - However, some reports showed even higher losses of uranium to sludge (60% or higher).

Percentage vs. Batch Size

40kg U-235 batch	Dissolved zirconium (kg)	Starting uranium (kg)	Uranium in sludge (kg)	U-235 in sludge (kg)	Total mass (kg)	Total volume (liters)	Density (g/cc)
7% Uranium Holdup	4.98	40.00	2.80	2.66	8.51	6.28	1.36
0.5% Uranium Holdup	4.98	40.00	0.20	0.19	5.91	6.28	0.94

- Holdup percent directly impacts the density and fissile mass.
  - The volume stays the same as holdup increases.
  - Volume is only dependent on the batch size (dissolved zirconium) and not the holdup percentage of uranium.

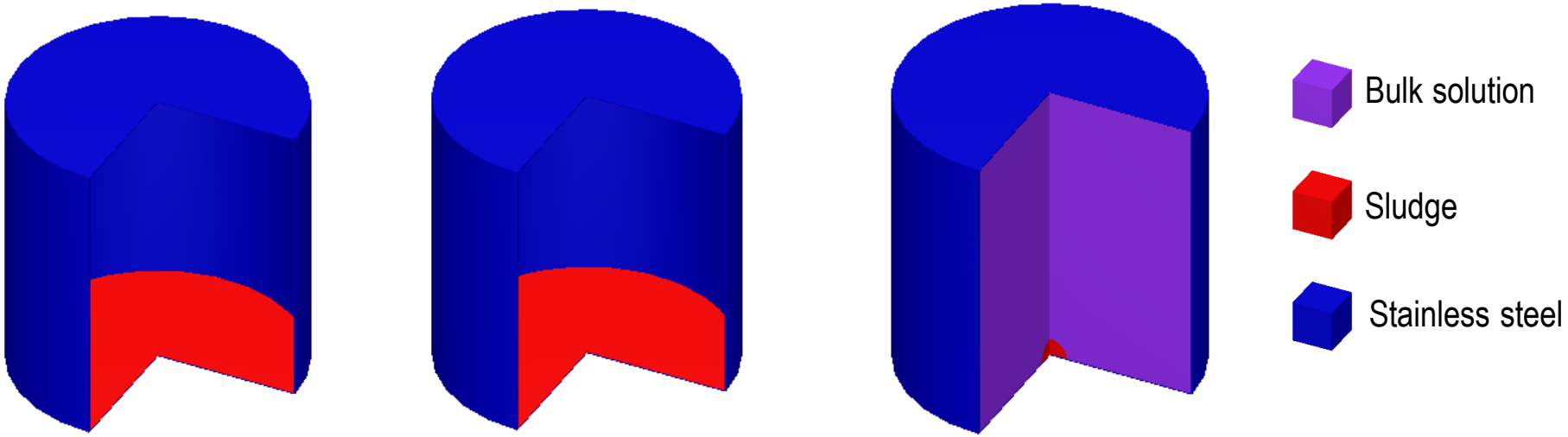
Campaign 1 and 40kg Batch K-effective

	Cone K-eff	Cylinder K-eff	Hemisphere K-eff
Campaign 1 7% holdup	0.8510 ± 0.00025	0.7040 ± 0.00029	0.8706 ± 0.00026
Campaign 1 0.5% holdup	0.7665 ± 0.00029	0.6316 ± 0.00018	0.7850 ± 0.00027
40kg U-235 batch 7% holdup	0.5667 ± 0.00004	0.5668 ± 0.00004	0.5668 ± 0.00005
40kg U-235 batch 0.5% holdup	0.5667 ± 0.00004	0.5667 ± 0.00004	0.5667 ± 0.00004

Campaign 1 and 40kg Batch K-effective

	Cone K-eff	Cylinder K-eff	Hemisphere K-eff
Campaign 1 60% holdup	1.4680 ± 0.00012	1.2248 ± 0.00010	1.49890 ± 0.00010
Campaign 1 100% holdup	1.6852 ± 0.00011	1.4818 ± 0.00010	1.70635 ± 0.00011
40kg U-235 batch 60% holdup	0.5668 ± 0.00003	0.5669 ± 0.00003	0.5734 ± 0.00008
40kg U-235 batch 100% holdup	0.5668 ± 0.00003	0.5669 ± 0.00003	0.6336 ± 0.00010

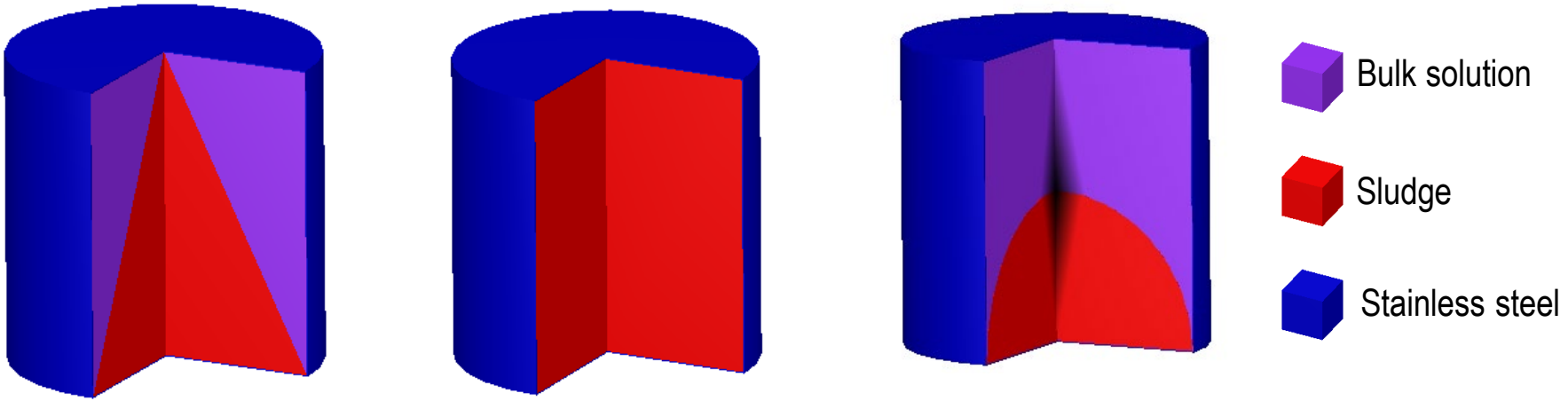
Realistic Visualization For 40kg Batch



Filling Up the Entire Tank With Sludge

Material	Cone	Cylinder	Hemisphere
Dissolved zirconium (kg)	3,008.00	9,024.30	3,008.00
Uranium (kg)	1,692.47	5,077.58	1,692.47
U-235 (kg)	1,607.85	4,823.70	1,607.85
Total volume (liters)	3,795.52	11,386.90	3,795.52
K-eff of 7% holdup	0.9700 ± 0.00027	0.9181 ± 0.00026	0.9263 ± 0.00025
K-eff of 60% holdup	1.6237 ± 0.00034	1.7685 ± 0.00032	1.6105 ± 0.00036

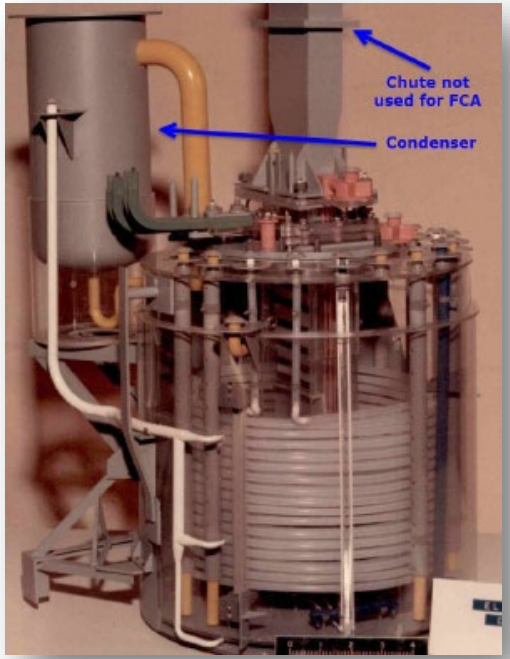
Filling Up the Entire Tank (1.87 times Campaign 1 Volume)



Results

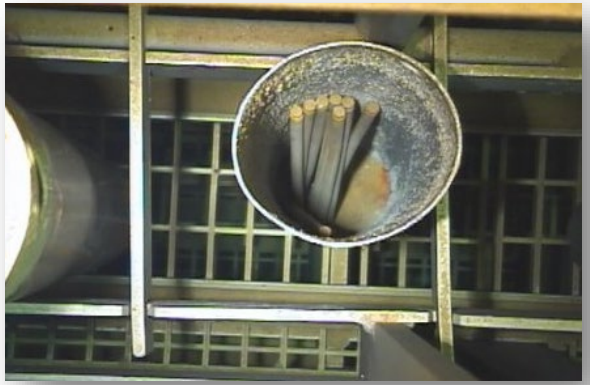
The results indicate that:

- **Criticality <u>should not</u> be a concern for 40kg batch sizes at any holdup percentage.**
  - Even at 100% holdup, k-effective ≈ 0.64 for the least conservative model.
- **The <u>entire tank</u> could be filled with sludge at the predicted 7% holdup, and it still would remain subcritical.**
  - This is 1.87 times the total amount for Campaign 1.
- **If <u>all of campaign 1 was dissolved at or below 19.16%</u> holdup as a single batch, there should not be a criticality concern.**
  - This would go against procedure anyways.
  - Rinsing/washing and batch processing further mitigates any risk.



Future Work

- Evaluate the number of batches - How many batches remain subcritical?
- Evaluation of sludge washing procedures/upsets
- Other modeling options geometries (angle of repose?)
- Mixing the sludge with the bulk solution at different amounts
- Vary the uranyl nitrate uranium concentration in the bulk solution



References

- SRNL-TR-2020-00313, Revision 0, NASNF Processing and Packaging Technical Study, Savannah River National Laboratory, Aiken, SC.

Acknowledgements

This work was funded by the Savannah River Nuclear Solutions, LLC, Summer Internship Program,

Questions??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