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Neutron Poisoning of Sodium Reactor Experiment Material for Disposition

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INTRODUCTION

The Sodium Reactor Experiment (SRE) was an experimental sodium cooled, fast spectrum breeder reactor meant to demonstrate the technology for electricity production. It was conceived in 1954 (Ref. 1) and was built and in operation by 1957 (Ref. 2).

In July 1959, the reactor suffered a severe failure due in part to core design and in part to mechanical design (Ref. 3). During power ascension, several pulsating thermal excursions were experienced. From Ref. 1 it can be seen that during heat up or cool down the reactor core would go through a region where the temperature of the coolant and fuel design resulted in positive reactivity feedback (increase in temperature increases power). As this only occurred during a small region of the temperature profile between cold and hot operations, it was expected to be manageable. However, as investigation of the accident revealed, degradation products partially blocked several coolant channels in multiple fuel assemblies. When the temperature of the core reached this region of operation, a power excursion from ~4 MW to ~14 MW occurred. Insufficient coolant flow exacerbated and prolonged the excursion, resulting in severe damage to approximately 1/3 of the fuel assemblies. The reactor was shut down for a over a year for repairs and restarted in September 1960. It was shut down in 1964 but deactivation and decommissioning activities lasted until 1983.

The aluminum clad fuel contained both thorium meant for U-233 production and enriched U-235 as a driver. Since the fuel was irradiated it also contained quantities of plutonium and the usual cadre of fission products as well. In 2013, the damaged core fuel stored at the Savannah River Site (SRS) was received at the H-Canyon facility and dissolved (Ref. 4, 5). There it was to be held in storage until such time that it could be dispositioned through the high-level waste tanks at SRS and vitrified for permanent disposal. To reduce the enrichment of the material in storage, the dissolved SRE fuel solution was cut with another dissolved high aluminum, ~20 wt.% enriched uranium research reactor fuel.

Due to its fissile content, the waste is required to be accompanied by a neutron poison for criticality control. Because the waste goes through several significant pH changes (acid solution to caustic solution back to acid solution then to dried particulate), there is concern that all of the poison may not stay with (precipitate or not precipitate) with the fissile material. Conservative poisoning requirements using manganese already exist for fissile material discards from the H-Canyon, however due to its special nature, a 5-fold higher poisoning ratio was suggested to account for partial precipitation of the fissile or poison. The necessity of this poisoning was assessed in this work.

POISONING SIMULATION

Tank Inventory

SRE fuel was dissolved in six batches along with a limited amount of high aluminum fuel to provide aluminum salting. A portion of the SRE material was promptly transferred to the discard facility in 2014. The remainder of the SRE fuel inventory was stored in a single segregated bi-cylindrical tank (Figs. 1 and 2). Accountability samples taken indicated the solution in each side of the tank to have the composition given in Table I. The important thing to note here is that the large side of the tank contains less fissile material (~52 kg) than the small side (~77 kg); fissile being the equivalenced sum of U-233, U-235, and Pu-239. The approximate volumes of the small and large sides of the tank are 35,700 L and 42,000 L, respectively. However, this volume is not achievable as mechanical design includes an overflow outlet at 33,400 L and 38,700 L, respectively. Note the thorium concentration in comparison to the uranium concentration.

Table I: SRE Hold Tank Contents (April 2019)

	Small Side	Large Side	Units
Density	1.2966	1.3006	g/ml
Acidity (HNO_3)	1.5	3.942	M
U concentration	6.924	3.934	g/L
U-233	0.33	0.362	wt.%
U-234	0.412	0.577	wt.%
U-235	37.184	51.871	wt.%
U-236	1.81	1.965	wt.%
U-238	60.264	45.225	wt.%
Pu concentration	1.59E+07	7.26E+06	dpm/ml
Pu-239	40.7	37.47	Alpha%
Pu-238	59.3	62.53	Alpha%
Al concentration	1.1379	0.8325	M
Th concentration	26.74	22.73	g/L
Tank level (at time of sample)	84.1	62.7	% of instrument range

Simulation Approach

SCALE 6.1 using KENO-VI with the ENDF-B VII 238-group cross section library was used for simulation of the system as it is the currently validated version and library in use at SRS (Ref. 6).

While in storage, the criticality concern is related to the over concentration of the solution due to long term evaporation. During neutralization the criticality concern is the precipitation of fissile material. Models of the bi-cylindrical tank were constructed that simulated the range between the physically most dilute and the theoretically precipitated conditions. For dilution, the current contents of the tank were diluted with water up to the physical inner top of the tank. This is not physically achievable as overflow mechanical designs prevent such filling but is conservative to model. The precipitated case removes all water leaving behind only the chemical species at their theoretical densities. Interstitial points were run as needed to capture the behavior of the multiplication factor.

A total of fourteen data sets are calculated to examine the behavior of the system. The first six examine the behavior of the system without any additional neutron poisoning. Thorium is itself a poison in this system. Those data sets are:

1. Large side of tank held constant at maximum volume, small side of tank varied in concentration
2. Large side of tank held constant at minimum volume, small side of tank varied in concentration
3. Large side of tank held constant at optimally moderated volume, small side of tank varied in concentration
4. Small side of tank held constant at maximum volume, large side of tank varied in concentration
5. Small side of tank held constant at minimum volume, large side of tank varied in concentration
6. Small side of tank held constant at optimally moderated volume, large side of tank varied in concentration

Next, sets 1, 2, 4, and 5 were run with an additional manganese neutron poison at a ratio of 14 g Mn per g of equivalent U-235 normally used by the facility. Equivalent U-235 was conservatively set at the mass of U-235 plus 1.6 times the mass of both U-233 and Pu-239. Note this is a procedural facility conservatism that rounds up the ANSI/ANS 8.1 Pu-239 to U-235 equivalency and treats U-233 as Pu-239. Finally, sets 1, 2, 4, and 5 were run using the 70:1 Mn to Equivalent U-235 ratio requested by the discard facility.

Note the absence of an optimally moderated condition in the poisoned cases. The large volumes of neutron poison added results in a new minimum volume and substantially reduced multiplication. The four condition sets span the moderating conditions achievable in the poisoned systems.

Determination of k-safe

As the results will bear out, setting a k-safe is rather arbitrary in this system. Normally, the Ref. 5 validation for highly enriched uranyl nitrate solutions results in a biased k-safe of 0.976 from which an additional administrative margin of 0.01Δk is commonly taken. Considering that U-233 and thorium are not part of this validation one could take an additional 0.02 Δk margin, setting a k-safe of 0.946. All results will be k-best estimate (k-calculated + 2 times the Monte Carlo uncertainty) compared to k-safe.

RESULTS

A base model of the system was generated with the tank concentrations and levels at the values from the sample data in Table I. This resulted in a k-best estimate of 0.319.

All results are plotted versus moderating ratio (H/fissile) ratio which is more useful to the analyst. Tank level is the alternative but is useful only to the facility. The H/fissile only extends as far as the tank can be diluted within its physical dimension. The low end of the H/fissile value is the point at which only the dry precipitants remain. At this condition, residual H is provided by the acid in the system, not water.

Fig. 3 shows the results of the unpoisoned cases 1 through 3 in which the large side of the tank is held at one of three constant volumes while the small side varies in concentration. These cases resulted in the largest multiplication calculated for the system at 0.592. This indicates that the small side of the tank is also driving multiplication, which stands to reason as it has the higher concentration and higher enrichment of fissile uranium.

Fig. 4 shows the results of the unpoisoned cases 4 through 6 in which the small side of the tank is held at one of three constant volumes while the large side varies in concentration. The reactivity impacts of the larger side can only be seen when the smaller side is at its most dilute (high level). Otherwise the small side is driving the reactivity.

Fig. 5 and 6 show cases 1 and 2 and cases 4 and 5, respectively, repeated using the two different poisoning ratios. The optimal unpoisoned cases are also presented for reference. The trending of the behaviors is similar to the unpoisoned cases but damped and significantly reduced in multiplication.

FACIILTY APPROACH

Ultimately, to meet the discard facility waste acceptance criteria, the manganese to fissile ratio of 70 was chosen. An option exists to convert to gadolinium poisoning pending further analysis by chemistry and criticality safety. A portion of the material was discarded in May 2020 to a tank in the discard facility containing sufficient neutron poison to not require the H-Canyon facility to add additional manganese.

Manganese poisoning of the remaining material will require approximately 80,000 pounds 50 wt.% concentrated manganous nitrate solution to be added to the waste stream.

This solution is added manually from 55-gallon drums to a head tank before transfer to the mixing tank inside the shielded facility. The material is an oxidizer and carries associated chemical hazards. It is a material the facility normally handles, albeit in smaller quantities.

As if this was not sufficient, an additional plan was initially proposed by the SRS Tank Farm to reduce the effective uranium enrichment to 4.2 wt.% U-235 by addition of depleted uranium (0.15 wt.%). While this would have helped disposition some of SRS's legacy depleted uranium stockpile, it also would have increased the volume of waste to discard without significant benefit to criticality safety. This plan was concluded to not be necessary by the discard facility after analysis of the waste batch that would receive the material (Ref. 7).

The waste was also neutralized from acid to caustic, further increasing volume, but this has been the required practice since SRS was built due to the construction material of the tanks. Dilution is also required due to the rheology of the material, primarily due to the precipitation of thorium and manganese during the neutralization process. Large amounts of depleted uranium were also considered in the rheology in the event it was needed. The solution is required to be less than 1.33 g/cm^3 and less than 5% undissolved solids for discard. The manganese (as $Mn(OH)_2$) and thorium (as $Th(OH)_4$) are two of the biggest contributors to the undissolved solids. Additional water is added as necessary to meet this requirement, further diluting the fissile material.

CONCLUSIONS

The SRE material was conservatively over poisoned. Discards to the SRS tank farm began in May 2020 and are currently scheduled to be completed by February 2021.

ACKNOWLEDGEMENTS

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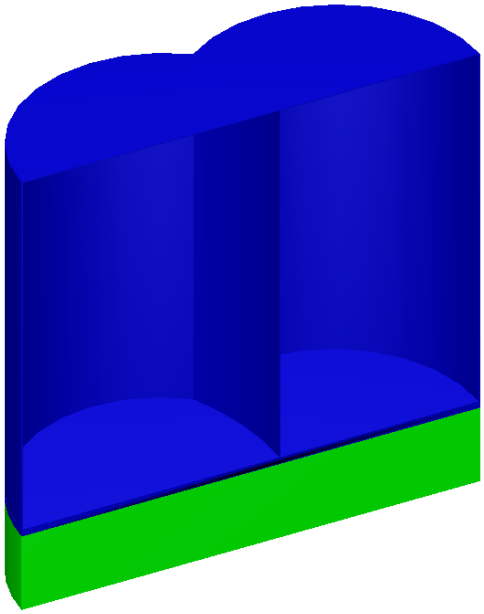

Fig. 1. SCALE rendering of segmented bi-cylindrical tank, empty.

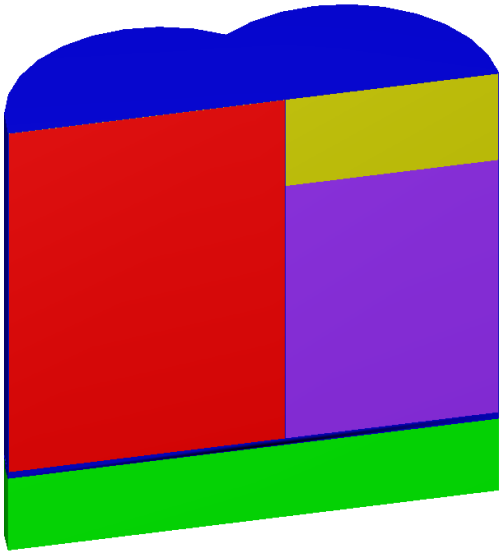

Fig. 2. SCALE rendering of segmented bi-cylindrical tank; large side full, small side at sampled level, balance filled with dry air.

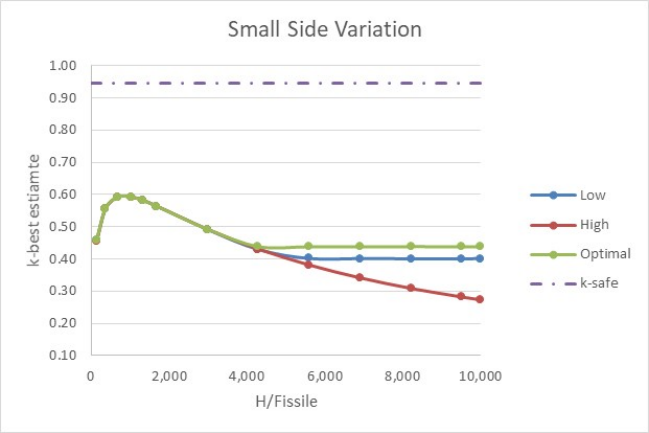

Fig. 3. Unpoisoned tank response, small side allowed to vary.

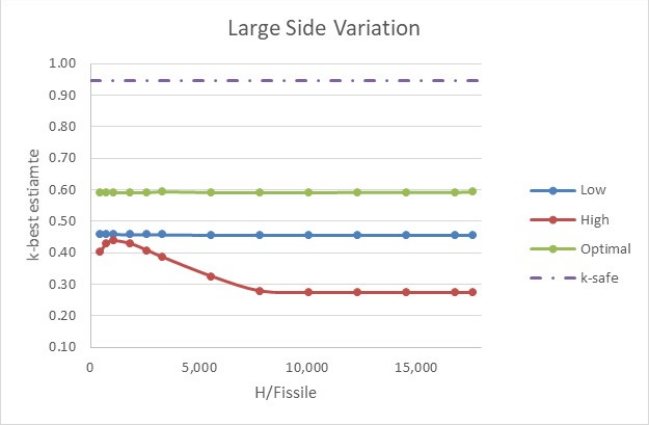

Fig. 4. Unpoisoned tank response, large side allowed to vary.

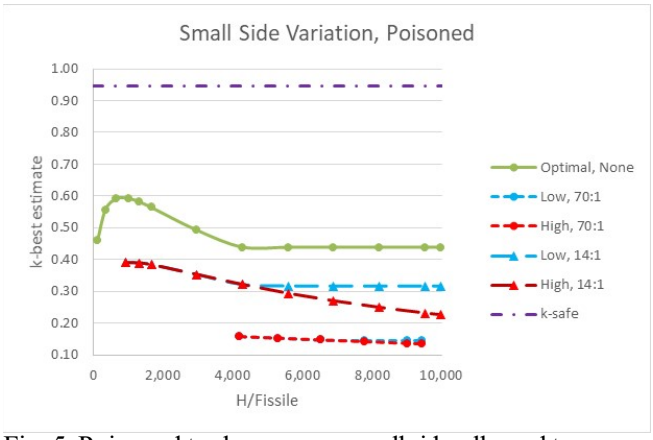

Fig. 5. Poisoned tank response, small side allowed to vary.

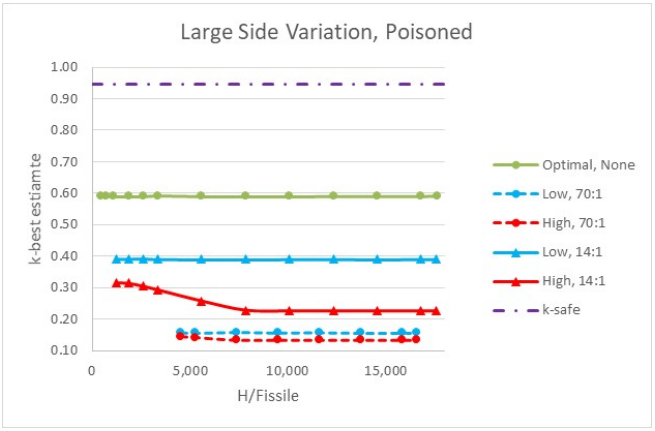

Fig. 6. Poisoned tank response, large side allowed to vary.