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# Criticality Safety of Processing Salt Solution at SRS

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## INTRODUCTION

High level radioactive liquid waste generated as a result of the production of nuclear material for the United States defense program at the Savannah River Site has been stored as 36 million gallons in underground tanks. About ten percent of the waste volume is sludge, composed of insoluble metal hydroxides primarily hydroxides of Mn, Fe, Al, Hg, and most radionuclides including fission products. The remaining ninety percent of the waste volume is saltcake, composed of primarily sodium (nitrites, nitrates, and aluminates) and hydroxides. Saltcakes account for 30% of the radioactivity while the sludge accounts for 70% of the radioactivity.

A pilot plant salt disposition processing system has been designed at the Savannah River Site for interim processing of salt solution and is composed of two facilities: the Actinide Removal Process Facility (ARPF) and the Modular Caustic Side Solvent Extraction Unit (MCU). Data from the pilot plant salt processing system will be used for future processing salt at a much higher rate in a new salt processing facility.

Saltcake contains significant amounts of actinides, and other long-lived radioactive nuclides such as strontium and cesium that must be extracted prior to disposal as low level waste. The extracted radioactive nuclides will be mixed with the sludge from waste tanks and vitrified in another facility. Because of the presence of highly enriched uranium in the saltcake, there is a criticality concern associated with concentration and/or accumulation of fissionable material in the ARP and MCU.

## DISCUSSION AND RESULTS

Once the sludge is settled in the waste tanks, a layer composed of water and dissolved salts, commonly named "supernate" collects above the sludge layer. To reduce the volume and the chances of escaping to the environment in the event of a tank leak, the supernate in the storage tanks were decanted and concentrated in the evaporators. The concentrated supernate crystallized to form the saltcake when returned to storage tanks. Saltcake is composed of salt crystals, interstitial liquid, gas, and insoluble solids, including uranium and plutonium.

Saltcakes will be dissolved using inhibited (0.02 M NaOH) water and the solution sodium ion concentration will be adjusted to around 5 M prior to processing. The dissolved saltcake solution contains some insoluble compounds, including fissionable material. These compounds were formed in the evaporators during concentration process and are essentially insoluble in water. The solubility of uranium and plutonium in the dissolved saltcake solution is 25 mg/L and 1.68 mg/L, respectively [1].

ARPF uses two 6000-gallon tanks to strike the salt solution with monosodium titanate (MST) at a concentration of 0.4 – 0.6 g/L. MST ($NaHTi_2O_5$) adsorbs soluble actinides and strontium in the salt solution. The resultant slurry is agitated for approximately 24 hours before it is transferred to a 6000-gallon concentration tank. The MST/solids slurry from processing several salt solution batches will be accumulated and concentrated to approximately 5 weight percent solids by circulating the salt solution with MST/solids through a crossflow filter. The concentrated MST/solids will be mixed with the sludge from waste tanks to be vitrified in another facility. The filtrate, containing cesium and a small amount of uranium and plutonium, will be transferred to MCU for cesium removal.

MCU, as shown in Figure 1, uses a solvent extraction process to remove cesium from the filtrate. The stream with the cesium is mixed with the MST/solids/sludge stream from ARPF. The decontaminated salt solution with a trace amount of radionuclides is routed for disposal as low level waste.

In MCU, an organic solvent called "calixarene" is contacted with the salt solution stream in a series of seven countercurrent centrifugal contactors. This results in the extraction of cesium and nitrate ions into the solvent phase. The cesium is held by calixarene molecules while the modifier molecules stabilize the nitrate ions. Downstream from the contactors, the resulting decontaminated salt solution which is the raffinate from the extraction process is decanted to remove entrained organics. After organic removal, the decontaminated salt solution is routed for disposal as a low level waste.

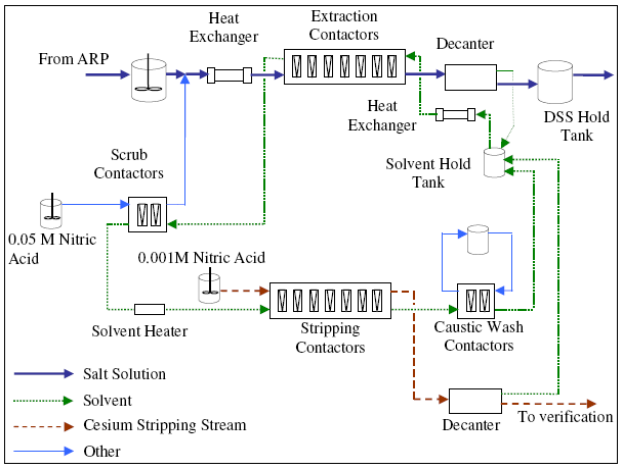


Fig. 1. MCU Process Flow Diagram

The organic stream from the extraction contactors is routed to a pair of scrub contactors where a dilute nitric acid stream scrubs salts other than cesium salts from the organic stream. The organic stream then flows to another bank of seven centrifugal contractors where a stream of very weak nitric acid strips the cesium from the organic stream. The organic stream then flows to a pair of caustic wash contactors that remove contaminants from the organic, then to a solvent hold tank. The aqueous strip solution is decanted to remove entrained organics, and transferred to the verification facility.

Uranium and plutonium loading onto MST has been determined to be 14 and 2.79 weight percent, respectively, after nearly two weeks of contact time [2]. The maximum uranium enrichment in the saltcakes to be proceed is 66 weight percent.

In ARPF, because of the presence of highly enriched uranium in the salt solution, concentration of MST/solids slurry in the concentration tank or in the crossflow filter pose a criticality concern. In MCU facility, there is a potential for accumulation of fissionable material in the extraction columns or accumulation of fissionable material in the decanter or in the decontaminated salt solution hold tank, due to the potential change in the salt solution pH caused by the nitric acid used in the scrub contactors [3].

The Department of Energy has approved interim processing of salt solution in the pilot plant ARP/MCU facility provided no more than a single subcritical fissionable material can be allowed to be within the boundary of the ARP/MCU at a time. Data from the pilot plant facility will be used to process salt solution at a much higher rate in a new salt processing facility.

Restricting the fissionable material mass within the ARP/MCU boundary to a single subcritical mass limit or downblending the uranium in the saltcakes to less than the subcritical uranium enrichment of 0.93 weight percent [4] is a cost prohibitive option to process the saltcake without a criticality safety concern.

An alternative option is to take credit for the presence of titanium in MST in the concentration tank and the crossflow filter. Titanium is a weak neutron absorber with a thermal absorption cross section of 6.1 barns. Scoping calculations have demonstrated that accounting for the presence of titanium in the MST laden with fissionable material, saltcakes with uranium enrichments of up to 10 weight percent can safely be processed. Due to the lack of any benchmark experiments involving titanium, an additional safety margin of 3 % was included in determining the $k_{safe}$ for the scoping calculations.

Efforts are underway for evaluation of titanium cross sections. Using the obtained data for validation of titanium cross sections could allow for elimination of the additional safety margin, thereby increasing the maximum uranium enrichment that can be safely processed in the actinide removal processing section of the new salt processing facility.

## CONCLUSION

Use of the validated cross section data for titanium combined with the data obtained from the pilot plant salt processing system will be used to maximize the uranium enrichment in the saltcake that can be safely processed in the new salt processing facility.

## REFERENCES

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