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# Preparation for Electrolytic Dissolution at H-Canyon

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## INTRODUCTION

H-Canyon Operations involves the dissolution and recovery or disposition of nuclear material from Spent Nuclear Fuel (SNF). This dissolution of these materials is obtained by charging SNF bundles by crane to large dissolver vessels equipped with spacing inserts and filled with nitric acid. A combination of a mercury nitrate catalyst and boiling dissolves the fuel on a batch process. The current SNF is clad in aluminum which is readily able to dissolve in nitric acid. The Savannah River Site (SRS) has assumed responsibility of this fuel for disposition. A new mission has emerged to dissolve and dispose of the stainless steel clad, plutonium containing, Fast Critical Assembly (FCA) fuel plates. Unique challenges are involved in the dissolution process for this fuel as nitric acid is unable to dissolve stainless steel on its own. The disposal of these plates will need to incorporate the use of an electrolytic dissolver.

KENO-VI module with ENDF Version 7 238-group cross sections in SCALE 6.1 is used to simulate the neutron multiplication conditions.

## DESCRIPTION OF THE WORK

### Electrolytic Dissolution Process

The Savannah River Site H-Canyon facility has previously operated an electrolytic dissolver beginning in 1969 to dissolve power reactor fuel elements clad in stainless steel or zirconium but it has not seen operation since the mid-1980s. While these materials' resistance to corrosion under high-temperature irradiation is beneficial for a power reactor, neither stainless steel or zirconium are susceptible to direct chemical dissolution by nitric acid. The metal surfaces of these materials are protected by oxide films, however increasing the electrochemical potential at the metal-solution interface will destroy that film. This can be accomplished by applying an external source of electrical energy. Contact is established through the electrolyte in contact with the fuel to be dissolved suspended between two electrodes. Nitric acid serves as this electrolyte which is beneficial for both processing and disposal applications.

### Electrolytic Dissolver Description

Reference 1 details the description of the electrolytic dissolver. The electrolytic dissolver vessel is a jacketed pot constructed with 304L stainless steel. It is 8 ft tall and has a 7.5 ft inner diameter and an 8 ft diameter outer diameter with a slightly slanted bottom. The space between the tanks performs as a heat removal jacket and secondary containment. On top of the dissolver is a 12 ft charging chute where fuel is loaded into the dissolver as well as a condenser to recycle evaporated solution back into the dissolver. Two electrodes, a platinum-clad niobium anode and a niobium cathode, are suspended in the tank with the bottom of the cathode sloping towards the anode. The dissolver operates by inserting a cannister containing the fuel plates down a fuel charging chute into a niobium fuel basket which sits between the two electrodes (Figure 1) that will create a potential gradient within a nitric acid solution. Niobium was chosen as the material of construction due to its ability to withstand its acidic environment coupled with the intense electrical current of approximately 10,000 amps. This gradient enables the stainless steel cladding to dissolve and expose the rest of the material to dissolution. The dissolver is equipped with a three well basket insert 19" wide and 3.5 ft long. The basket insert follows the sloped form of the cathode and tapers from 5.5" to 1". Three sets of fifteen 2" cooling coils provide cooling as the power to operate is dispersed as heat in the bulk solution while heat is also generated by the decay of fission products. Cooling coils are required as acid boiling would greatly increase resistance which would decrease the amount of current that could be supplied for dissolution.

### FCA Fuel Description

The FCA fuel plates are plutonium-aluminum alloy plate clad in stainless steel. A number of stacked plates with a mass of metal compliant with 3013 shipping requirements (Reference 4) are contained in a stainless steel can which is 5" in diameter and 10" tall. The plutonium isotopic were conservatively assumed to contain 92.5 wt% Pu-239 and 7.5 wt% Pu-240. While other fuels which require the electrolytic process for dissolution could be dissolved in H-Canyon, the current mission scope only considers this FCA fuel.

### Modeling Plutonium Dissolution

Scoping Calculations were performed in Reference 1 to provide initial criticality safety calculations for the dissolution of the FCA plates in the electrolytic dissolver. A simple model of two FCA cans surrounded by an "infinite" (>60 cm radius) sphere of plutonium nitrate solution was constructed for the initial scoping. The plutonium nitrate solution was modeled at a conservative concentration of 3.7 g Pu/L calculated from the maximum fissile mass conservatively expected to be charged to the dissolver and the assumed minimum volume for electrolytic dissolver. A high concentrated boundary layer between the Pu metal and bulk solution was not modeled as "the dispersion of Pu by thermal currents indicates that minimal mixing will be

required to disperse dissolved Pu that descends away from the metal surface into the bulk dissolver solution" (Reference 2). Extra turbulence created from the electrolytic process in the dissolver further ensures that no boundary layer is formed, and the bulk solution is in direct contact with the metal. For the bulk solution, a minimum nitric acid concentration was assumed of 6M. While this could be considered non-conservative as nitric acid is an inherently less effective reflector/moderator than water as the nitrogen in the acid acts as a slight neutron poison, it is not realistic to assume no nitric acid is in the bulk solution. A minimum nitric acid concentration is needed to prevent plutonium precipitation/polymerization. The minimum nitric acid will also minimize corrosion of the platinum anode and hydrogen embrittlement of the niobium cathode. The nitric acid concentration was assumed to be this minimum when in reality the end nitic acid concentration in the dissolver will be higher than required to protect the minimum nitric acid concentration. Administrative controls on sampling the acid will be required before operation of the dissolver is allowed.

The fuel components charged were modeled as unclad cylinders of alpha-phase Pu with a height to diameter ratio of 1. Since the outer stainless steel will dissolve first and expose the fuel inside, the stainless steel can will be conservatively neglected. Two cans will be charged at the same time. Investigation was conducted to find the minimum spacing needed between the cans to prevent an inadvertent criticality. These cans are modeled in the bulk solution at the final bulk concentration for the batch which will address an abnormal condition of charging an additional charge in the batch.

The fully detailed electrolytic dissolver was also modeled as seen in Figures 2 and 3. The modeled cans were inserted in the dissolver basket as low as possible. This was done to maximize any reflection provided from the electrodes and insulators. The basket was modeled as roughly 57% aluminum oxide to simulate the insulators and roughly 25% bulk plutonium nitrate solution to simulate the solution flow through the basket. It should be noted that SCALE 6.1 does not include platinum cross sections so tungsten-184 was used as a surrogate. Tungsten-184 has comparable scattering and absorption cross section values per ENDF data. One of the cans was placed in the corner of the dissolver basket to further maximize reflection. The electrodes, basket, cooling coils, charging chute, and condenser are all included in the fully detailed model. H-Canyon uses gadolinium as a soluble neutron poison in its dissolvers for criticality safety. Initial calculations assumed no neutron poison added to the system, however later cases were run with the typical H-Canyon minimum of 0.25 Gd/L.

There were several potential upset conditions analyzed. Two cans were modeled as stacked on top of each other to simulate charging to the same well twice. Each can could potentially contain up to 50 g of Pu Oxide. The total 100 g of Pu Oxide was assumed to immediately fall out of the insert well and collect on the bottom of the tank as an optimally moderated hemisphere. This upset condition was modeled with a hemisphere of Pu oxide and water reflected by 1 inch of stainless steel and immediately followed by thick concrete on the flat face of the hemisphere. The stainless steel and concrete reflector simulate the tank bottom and concrete floor. The concrete reflection is considered conservative as in reality the concrete reflector is not immediately next to the tank bottom, but an air gap of a few inches exists between the floor and tank bottom. The fuel elements were placed a distance above the tank floor corresponding to a conservative estimate on how low they could fit in the basket insert and the distance between the basket and the bottom of the tank. An upset enrichment was also evaluated up to 100% Pu-239. This enrichment bounds any receipt of fuel over the currently analyzed 92.5% Pu-239. Finally, an overmass in the fuel cans could occur. Analysis was performed to evaluate up to 5kg of Pu per can. These upset conditions included Gd as a soluble neutron poison in the analysis. The SCALE model for these upset cases is presented in Figure 4.

**Simulation Tools**

Neutron multiplication simulation is carried out in the KENO-VI model of SCALE 6.1 using the ENDF-VII 238-group cross section library. Materials were defined by atom densities calculated using the atom density method or defined by the SRNS methods manual (stainless steel and concrete).

**Determination of $k_{SAFE}$**

An in-house validation for the SCALE 6.1 KENO-VI code for plutonium metal/solution systems with and without gadolinium poison established biased values with the lowest being 0.9887 (Reference 3). A conservative $k_{SAFE}$ assumed for this work was 0.965 which provides at least an additional 0.0237 $\Delta k$ subcritical margin.

## RESULTS AND FUTURE WORK

The results and analysis performed in this paper were a part of a scoping analysis to provide initial criticality safety calculations for the dissolution of FCA fuel plates in the electrolytic dissolver. Normal processing conditions modeled were determined to be safely subcritical with a minimum spacing of 3" between the charges in both the simple and fully detailed model. The predicted $k_{EFF}$ for the fully detailed model was slightly higher than the simple model but were within $2\sigma$ of each other. Results of two charges stacked on top of each other was determined to not be critically safe. Gadolinium nitrate poison up to 10 g Gd/L was evaluated for the "stacked" and "no separation" cases but did not reduce the $k_{EFF}$ value below a safe number. Results are presented in the Tables I, II, and III below as a k-best estimate or $k_{BE}$ which is the predicted $k_{EFF}$ value plus two times the uncertainty. Engineered or Administrative controls will need to be put in place to ensure no more than

one can is charged to the same well. These results did not require the use of gadolinium as a neutron poison. When gadolinium was included in the solution, a predicted decrease in the $k_{EFF}$ occurred further ensuring that the process remains subcritical.

Abnormal conditions incorporated poison into the calculations at 0.25 g Gd/L. All three upset conditions were simultaneously analyzed which will bound other upset conditions. Table III presents the keff data collected. The abnormal conditions analyzed were subcritical with a minimum spacing of 5" between the charges.

The results of this work conclude that dissolution of FCA fuel will require a minimum spacing between the fuel charges and a minimum soluble neutron poison concentration. These requirements will be accomplished by plugging the middle well of the insert which is slightly greater than 5" and the inclusion of a soluble neutron poison concentration of 0.25 g Gd/L.

Future work will involve developing a full nuclear criticality safety evaluation. All upset conditions will be determined based on a formal Hazards Analysis meeting with Engineering and Operations personnel in accordance with SRS guidelines. These potential upset scenarios will be analyzed for and controls will be developed to protect from these upsets occurring. A specific k-safe will be derived to reduce unnecessary conservatism. Future work will include other potential fuels that would be candidates for electrolytic dissolution in H-Canyon. In addition to the future nuclear criticality safety evaluation for dissolution of FCA fuel, a later evaluation is planned for fuel elements containing primarily Pu Oxide.

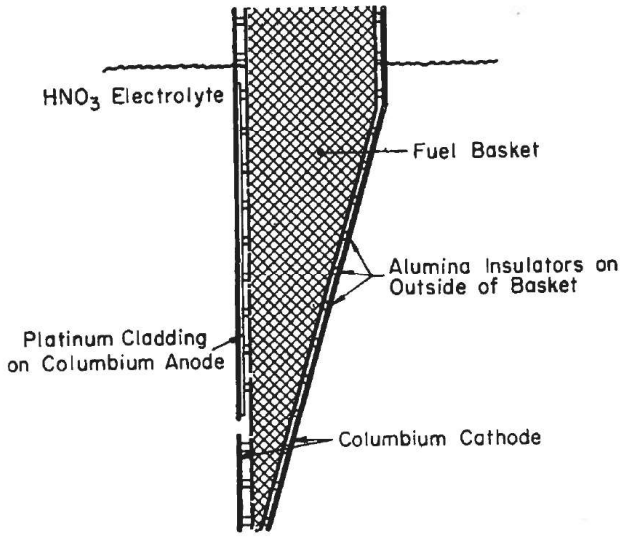


Figure 1: 6.3D Dissolver Fuel Basket (Reference 1)

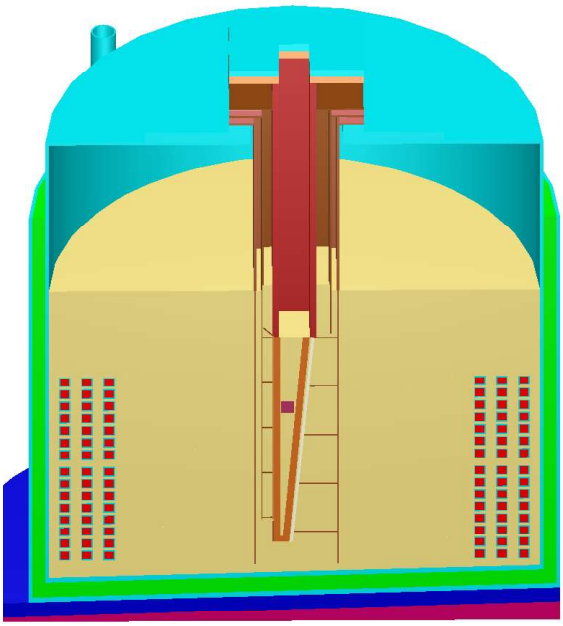

Figure 2: 6.3D dissolver vessel half cut away view with two FCA cans with 3" spacing.

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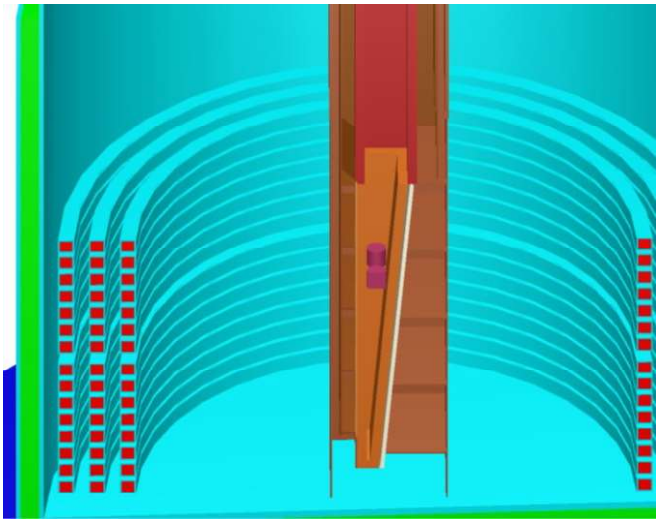

Figure 3: 6.3D dissolver view with bulk solution removed and two FCA cans with 3” spacing.

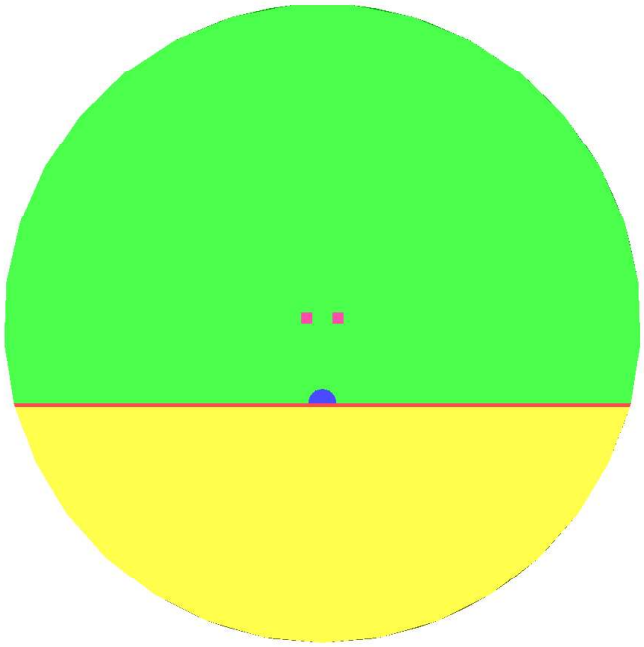

Figure 4: Simple spherical model used for upset cases with FCA cans with 5” spacing.

Table I. $k_{BE}$ values of initial scoping calculations

| Two Nominal Mass Pu Cylinders Reflected with Bulk Solution | Keff+2sigma |
|---|---|
| No Separation | 1.048 |
| 3” edge-to-edge separation | 0.950 |
| No Separation in 6.3D Model | 1.063 |
| Stacked in 6.3D Model | 1.078 |
| 3” edge-to-edge separation in 6.3D Model | 0.958 |

Table II. $k_{BE}$ values of initial scoping calculations with soluble neutron poison

| Two Nominal Mass Pu Cylinders Reflected with Bulk Solution with 0.25 g Gd/L | Keff+2sigma |
|---|---|
| No Separation in 6.3D Model | 1.038 |
| Stacked in 6.3D Model | 1.055 |
| 3” edge-to-edge separation in 6.3D Model | 0.926 |

Table III. $k_{BE}$ values of scoping calculations upset conditions

| Two Cylinders with 5” Spacing Reflected with Bulk Solution with 0.25 g Gd/L | Keff+2sigma |
|---|---|
| With 100 g Pu Oxide, 5kg Pu Charges, 100%Pu-239 | 0.963 |