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SAFE DISSOLUTION OF HIGH FLUX ISOTOPE REACTOR CORES

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ABSTRACT

The H-Canyon facility at the Savannah River Site is the U.S.A.'s only remaining full-scale chemical separations plant. H-Canyon maintains the capacity to safely and efficiently disposition a large inventory of excess nuclear material from across the Department of Energy (DOE) complex and used nuclear fuel from foreign and domestic research reactors. One such fuel that H-Canyon may dissolve is cores from the High Flux Isotope Reactor (HFIR). By employing realistic analysis, the need for administrative controls was eliminated.

A new criticality modeling approach has eliminated the need for administrative controls for HFIR dissolution operations in H-Canyon. The primary upsets of concern are an over-mass event and an over-concentration event. Several initiating events were considered for each type of upset. Over-concentration events were deemed to be not credible by taking credit for the chemistry of acid consumption, i.e. there are not enough moles of acid to dissolve enough moles of uranium in order to reach a critical configuration. Over-mass events were deemed to be not credible by taking credit for the design of the basket, in which fuel elements are placed. Because of the basket design and the annular design of the fuel elements, a critical configuration is not credible. By using realistic analysis, both types of upsets have been shown to be subcritical, and therefore administrative controls are not needed.

In this paper, the new methodology for performing criticality analyses for dissolving HFIR fuel elements will be presented, and the elimination of administrative controls will be discussed.

1. INTRODUCTION

H-Canyon is the only operating full-scale nuclear chemical separations plant remaining in the United States. It is located at the 310-square-mile Savannah River Site (SRS) in South Carolina and is owned by the U.S. Department of Energy. H-Canyon employs remote operations for most processes, including dissolving, separations cycles, and waste systems.

In the past, H-Canyon's missions have included recovering ^{235}U, ^{237}Np, and ^{238}Pu for use in defense, space, and commercial nuclear power purposes. Currently, H-Canyon maintains the capacity to safely and efficiently disposition a large inventory of excess nuclear material from across the Department of Energy (DOE) complex and used nuclear fuel from foreign and domestic research reactors.

H-Canyon processes involve large quantities of fissile solutions, including high enriched uranium and weapons grade plutonium, in geometrically unfavorable tanks. Therefore, criticality safety is of the

utmost importance. H-Canyon has an extensive criticality safety program, including criticality safety evaluations/double contingency analyses for all processes, monthly criticality assessments/walkdowns, and criticality safety training for all personnel with access to the facility.
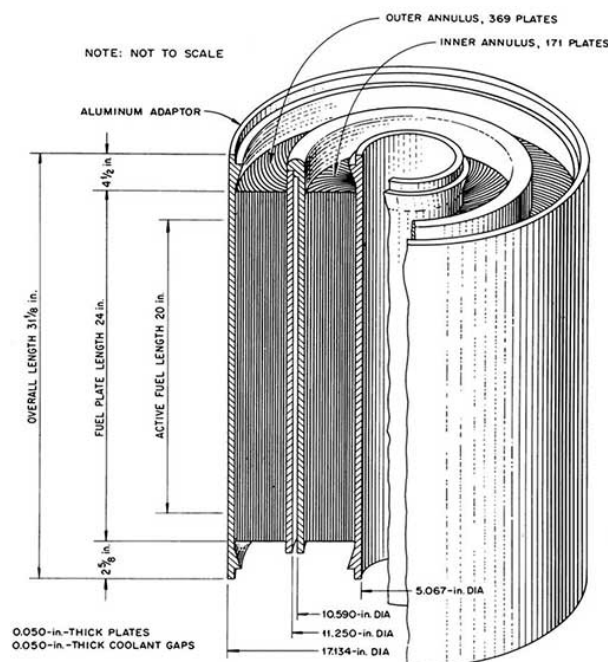
1.1. HFIR Fuel

High Flux Isotope Reactor (HFIR) fuel consists of two annular elements that are designed to be nested together with a beryllium reflector in the reactor configuration [1]. Figure 1 shows a photograph of the HFIR elements nested. HFIR cores are currently stored in L-Area at SRS as separate inner elements and outer elements, each on a unique aluminum carrier. HFIR elements will be shipped to H-Canyon in the 70-ton Cask Railcar and loaded into the dissolver with the HFIR insert installed.

Figure 1 HFIR Core

The inner element contains 171 fuel plates, and the outer element contains 369 fuel plates. The fuel plates are curved in the shape of an involute, thus providing a constant coolant channel width. The fuel (U_3O_8-Al cermet) is non-uniformly distributed along the arc of the involute to minimize the radial peak-to-average power density ratio. A burnable poison (^{10}B) is included in the inner fuel element primarily to flatten the radial flux peak providing a longer cycle for each fuel element. The mass of the inner element is 2,595.8 g ^{235}U. The mass of the outer element is 6,804.4 g ^{235}U. Figure 2 shows a cutaway drawing of the HFIR elements with general dimensions.

OUTER ANNULUS, 369 PLATES
INNER ANNULUS, 171 PLATES
NOTE: NOT TO SCALE
ALUMINUM ADAPTOR
OVERALL LENGTH 31⅛ in.
FUEL PLATE LENGTH 24 in.
ACTIVE FUEL LENGTH 20 in.
4½ in.
2⅝ in.
0.050-in.-THICK PLATES
0.050-in.-THICK COOLANT GAPS
5.067-in. DIA
10.590-in. DIA
11.250-in. DIA
17.134-in. DIA

Figure 2 HFIR Core Cutaway

In order to validate that the SCALE model provided by ORNL [2] is a reasonable and accurate model for criticality safety evaluations, SCALE was used to perform a Monte Carlo integration of the volumes of the many units in the model. Along with calculating the volume of each unit, SCALE also calculates the total mass of each unit and provides this data in a table in the output. SCALE also provides a "mixing table" in the output which gives the weight fraction of each nuclide for every material number. These two pieces of data (overall mass and nuclide-specific weight fractions) can be combined to determine how much mass is being modeled in the input.

When this technique was performed on the ORNL model of HFIR, the SCALE output mass calculations agree to within 1 gram ^{235}U based on the Appendix A mass values for both the inner and the outer element [3]. This indicates that the ORNL model is a reasonable and accurate model for criticality safety evaluations.
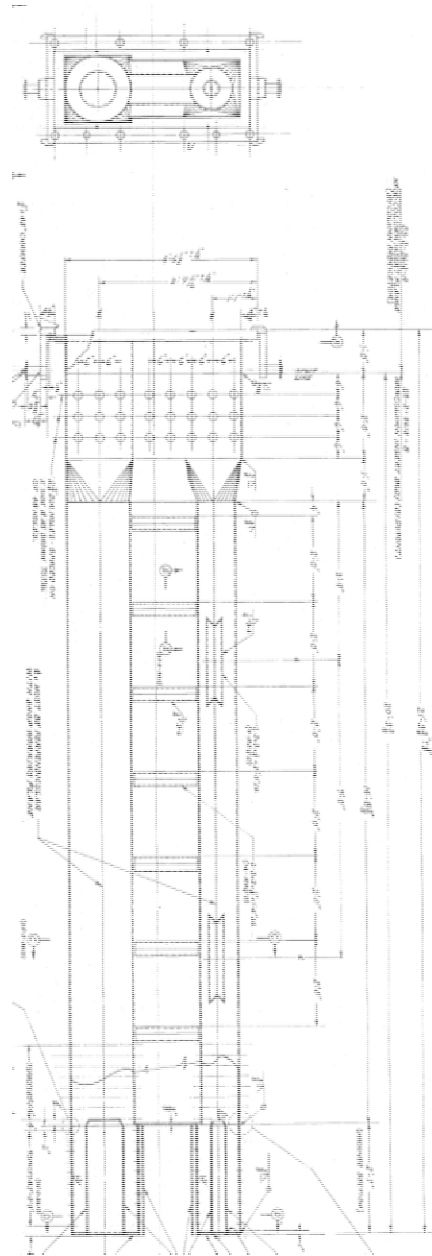
1.2. The Dissolving Process

Dissolving is the first step in processing material through H-Canyon. For each new type of material to be dissolved, the criticality safety of the operation must be established. A Nuclear Criticality Safety Evaluation (NCSE) must be developed that provides Criticality Safety Limits (CSLs) and controls to ensure all normal and credible abnormal conditions remain subcritical.

There are two dissolvers in use in H-Canyon. One is a 12-foot-diameter, 8-foot-tall, vertical, cylindrical tank. The other is an 8-foot-diameter, 8-foot-tall, vertical, cylindrical tank. The dissolving process utilizes nitric acid as the solvent with various catalysts, if needed.

An insert is placed in the dissolver pot to provide some control over geometry during the dissolution process. The HFIR insert is a basket-like device with two long, cylindrical, annual, vertical wells. The large well for the outer fuel element has an outer diameter of 18" and an inner diameter of 10". The small

well for the inner fuel element has an outer diameter of 12" and an inner diameter of 4". The insert is ~20 feet long, and has hundreds of holes to allow for flow of nitric acid in and nitric-acid-fissile solutions out into the bulk solution. Figure 3 shows several views of the HFIR insert.

Figure 3 HFIR Insert

2. NORMAL CONDITIONS

A basic knowledge of chemistry was used to bound the normal conditions based on aluminum solubility. The facility will strive to keep the dissolver solution below the aluminum solubility point, which is ~2 M Al in 2 M HNO_3 at 20° C. Therefore, it is assumed that the facility will not charge more than 5 cores to

the dissolver, because 6 cores would result in aluminum precipitation. Although aluminum precipitation does not cause a criticality concern, it can cause dip tube blockage and other operations concerns.

The mass of the inner element is 2,595.8 g ^{235}U. The mass of the outer element is 6,804.4 g ^{235}U. Also, there are 4,571.7 moles of Al in each HFIR core. A core consists of an inner element and an outer element. The preferred dissolver for HFIR dissolution is 6.4D (although either dissolver may be used), and a typical volume for HFIR dissolution is 13,500 L. It is assumed that one core (one inner element and one outer element) will be charged in each dissolution. The resulting ^{235}U concentration and Al molarity after each dissolution is shown in the Table I below.

Table I. Normal ^{235}U Concentration and Al Molarity

	Concentration (g ^{235}U/L)	Al Molarity (M)
After 1 core is dissolved	0.70	0.34
After 2 cores are dissolved	1.39	0.68
After 3 cores are dissolved	2.09	1.02
After 4 cores are dissolved	2.79	1.35
After 5 cores are dissolved	3.48	1.69
After 6 cores are dissolved	4.18	2.03

Assuming the facility will only charge up to 5 HFIR cores in a batch, which results in a concentration of 3.48 g ^{235}U/L, the concentration change was calculated at multiple points throughout the dissolution process of the 5th core. The dissolution process is divided into 10 phases (Phase 0 through Phase 9). The outer HFIR element is divided into 9 radial regions, and the inner HFIR element is divided into 8 radial regions. To simulate the dissolution process, a radial region (starting from the outside) is removed for each subsequent dissolution phase. For example, at Phase 0, the inner element and outer element are modeled intact inside the wells of the HFIR insert. At Phase 1, the outer radial region is removed from both elements and that mass is assumed to have entered the bulk solution. At Phase 2, the next outer radial region is removed from each element and that mass is assumed to have entered the bulk solution. This continues until each element is completely dissolved. Removing the material from the outermost radial region for each phase is representative of how the fuel will dissolve since there are holes in the outside of the wells to allow fresh acid to interact most with the outer surface of the fuel element. The Phase 0 configuration is shown in Figure 4 below.
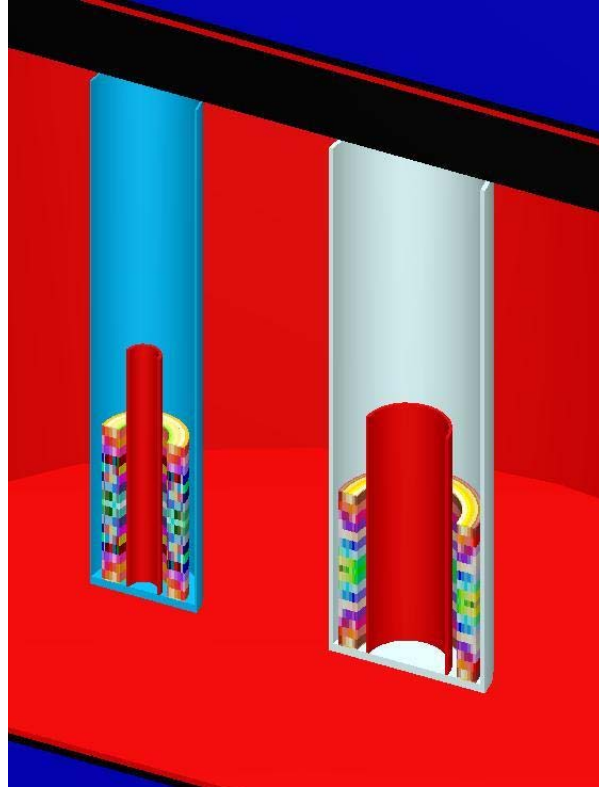
Figure 4 Normal HFIR Dissolution, 1 core, Phase 0

To account for the ^{235}U in the bulk solution between the fuel plates, the fuel ^{235}U atom density was increased by an amount equivalent to an upper bound concentration of 5.65 g ^{235}U /L. Using a constant value for the ^{235}U contribution minimized changes to the models for dissolution.

Table II shows the concentration change for each dissolution phase of dissolving the 5th HFIR core in a batch, assuming a normal volume of 13,500 L.

Table II. HFIR Dissolution Phases

Phase	Starting Concentration (g ^{235}U/L)	Starting Concentration (g U/L)	Inner Element Mass Lost (g ^{235}U)	Outer Element Mass Lost (g ^{235}U)	Concentration Increase (g ^{235}U/L)
0	2.79	2.94	341.65	333.17	0.05
1	2.84	2.99	305.40	427.07	0.05
2	2.89	3.05	623.16	509.43	0.08
3	2.97	3.14	558.15	1252.35	0.13
4	3.11	3.28	422.15	1457.32	0.14
5	3.25	3.42	155.60	1429.06	0.12
6	3.36	3.55	121.18	601.90	0.05
7	3.42	3.60	67.59	491.63	0.04
8	3.46	3.65	- *	298.46	0.02
9	3.48**	3.67**	-	-	-

*There is no inner element mass value for Phase 8 because there are only 8 radial regions in the inner element, and it is completely dissolved after Phase 7.
**These values are the final concentration values after all the material is dissolved.

Each of these phases was modeled in SCALE 5, and Table III provides the k_{eff}, sigma, and k_{eff} + 2*sigma values for the normal condition.

Table III. Normal HFIR Dissolution Results

Filename	k_{eff}	sigma	k_{eff} + 2*sigma
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE00	0.743	0.0017	0.747
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE01	0.732	0.0018	0.736
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE02	0.725	0.0014	0.728
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE03	0.715	0.0020	0.719
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE04	0.685	0.0015	0.689
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE05	0.638	0.0017	0.641
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE06	0.571	0.0015	0.574
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE07	0.512	0.0014	0.515
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE08	0.431	0.0003	0.432
ORNL2_HFIR_DISSOLVING_NORMAL_PHASE09	0.430	0.0003	0.431

Each phase is well below the k_{safe} value of 0.96, and therefore the normal condition has been shown to be subcritical.

3. ABNORMAL CONDITIONS

3.1 Over-Concentration

During dissolution, the fissile concentration is limited by the amount of acid available. Once all the acid is consumed, the dissolution process stops and the fissile concentration cannot increase further. Calculations were performed to determine the maximum fissile concentration based on acid consumption. First, the amount of Al and ^{235}U must be calculated. Based on the Appendix A Agreements, the inner elements contains 1,580.1 moles of aluminum, and the outer element contains 2,991.6 moles of aluminum. This results in 4,571.7 total moles of Al in a HFIR core. As discussed previously, the inner

element contains 2595.8 g ^{235}U, and the outer element contains 6804.4 g ^{235}U. ^{235}U has 235 g/mole. This results in 40.0 total moles of ^{235}U in a HFIR core. It is conservative to ignore all other elements and isotopes of uranium. In reality, moles of acid will be consumed by dissolving the other minor elements and isotopes and the real ^{235}U concentration will be lower than what is calculated here.

The total moles of acid available must also be calculated. Only 50% nitric acid is routed to the H-Canyon dissolvers. 50% nitric acid has a molarity of 10.351 M, and will be rounded up to 10.4 M for this analysis. It is assumed that 6.4D could be filled with 50% nitric acid up to the overflow, which is equivalent to 20,507 L. At 10.4 M acid and 20,507 L in 6.4D, there are a total of 213,275 moles of acid available.

The Savannah River National Laboratory study on spent fuel dissolution [4] indicates that 3.75 moles of acid are consumed for every mole of Al and 4 moles of acid are consumed for every mole of U. To dissolve one HFIR core that contains 4,571.7 moles of Al and 40.0 moles of ^{235}U, 17,303.8 moles of acid would be consumed.

$$\left(4{,}571.7 \; moles \; Al * 3.75 \frac{moles \; acid}{mole \; Al}\right) + \left(40.0 \; moles \; U^{235} * 4 \frac{moles \; acid}{moles \; U^{235}}\right) = 17{,}303.8 \; moles \; acid \tag{1}$$

If there are 213,275 moles of acid available, and it takes 17,303.8 to dissolve one HFIR core, then 12.33 cores (12.33 inner elements and 12.33 outer elements) could be dissolved before all the acid is consumed. Since there are 9400.2 g ^{235}U in each HFIR core, 12.33 cores, and an assumed volume of 20,507 L, this results in a concentration of 5.65 g ^{235}U/L.

Similar to how the normal condition cases were run, a series of cases were run that end at 5.65 g ^{235}U/L to ensure that an entire dissolution would be subcritical under acid consumption conditions. The results are given in Table IV.

Table IV. Acid Consumption Results

	k_{eff}	sigma	k_{eff} + 2*sigma
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE00	0.805	0.0021	0.810
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE01	0.796	0.0018	0.800
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE02	0.776	0.0038	0.784
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE03	0.779	0.0020	0.783
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE04	0.750	0.0018	0.754
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE05	0.718	0.0013	0.721
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE06	0.660	0.0016	0.664
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE07	0.645	0.0005	0.646
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE08	0.644	0.0006	0.646
ORNL2_HFIR_DISSOLVING_ACID-CONSUMPTION_PU_PHASE09	0.646	0.0006	0.647

Each phase is well below the k_{safe} value of 0.96, and therefore the acid consumption condition has been shown to be subcritical. This shows it is not possible to obtain a critical configuration with an over-concentration caused by full acid consumption.

Another potential cause for over-concentration is if the dissolver solution is allowed to evaporate due to steam heating. Several assumptions were made for this part of the analysis. They are listed below.

1. The facility does not operate tanks at a liquid level above the 10" limit, which is 10" below the overflow volume. For dissolvers, this means that the facility will not apply steam to the coils to begin dissolution if the liquid level is above the 10" limit. For tank 6.4D, the 10" limit is at 86.45", which equates to a volume of 18,152 L.
2. The facility will not operate tanks with any steam coils uncovered. The steam coil is covered at 51.08". This equates to a volume of 9,856 L.
3. The highest molarity nitric acid that is routed to the dissolver is 10.4 M (50% nitric acid). It is expected that 8-9 M acid will be used for HFIR dissolution.
4. The normal expected number of cores to be charged in a batch is 4 or 5.
5. The facility will not operate the dissolver without the total-reflux condenser being functional and operating.
6. Once the dissolver is shut down, it falls under the evaluation and controls of the H-Canyon Double Contingency Analysis (DCA), which has controls to track, trend, and maintain liquid level above a value set to protect the concentration CSL that is specified in the DCA. There are also controls to ensure that two steam valves to the dissolver are closed and that the low level alarm is functional in order to prevent evaporation due to a steam valve leak-through.

In order to approach an unsafe fissile concentration, the following upsets would have to occur:

1. 6.4D is filled with 10.4 M (50%) nitric acid up to the 10" limit (18,152 L).
2. 11 HFIR cores are charged (11 inner elements and 11 outer elements) without transferring any solution out of the dissolver. This would consume all the acid available and would result in a concentration of 5.65 g ^{235}U/L. This would also be well beyond the aluminum solubility limit.
3. After the 11th core is dissolved, steam would have to be applied to the dissolver until the volume was at 8,918 L. This would result in a loss of 9,234 L, indicating that the condenser was not functional. This would also expose some of the steam coils.
4. This would result in a concentration of 11.5 g ^{235}U/L, which is still subcritical.

Considering the assumptions listed above, a criticality due to over-concentrating the dissolver solution has been judge to be not credible.

3.2 Over-Mass

HFIR fuel consists of two large, discreet elements (an inner and an outer), and multiple elements will be shipped to H-Canyon for dissolution. Therefore, an evaluation has been performed to determine whether or not multiple elements loaded into the same well can pose a criticality risk. First, the number of elements that can fit in a well was determined. Each element is 50.8 cm tall. The maximum height of solution in the wells is ~162 cm. Therefore, only 3 elements could fit in each well and be submerged in solution. Also, in reality, the solution height at a normal volume of 13,500 L is ~112 cm, and only 2 cores would be submerged in solution. A SCALE model was created with 3 inner elements in the small well and 3 outer elements in the large well, as shown in Figure 5 below. To simplify the input, no post was modeled.
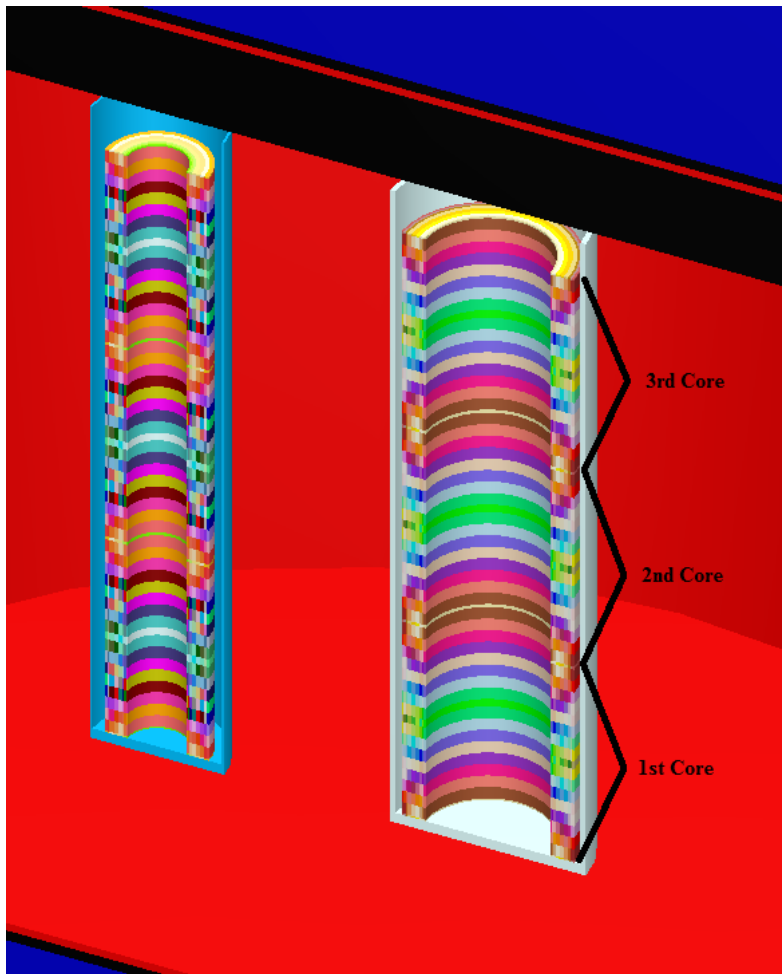
Figure 5 Over-Mass, 3 Cores

Since the results from the normal conditions show that the highest system multiplication (k_{eff}) occurs at Phase 0 (the beginning of dissolution), only this configuration was modeled. The results are given in Table V below.

Table V. Over-Mass, 3 Cores Results

Filename	k_{eff}	sigma	$k_{eff} + 2*sigma$
ORNL2_HFIR_DISSOLVING_3CORES	0.866	0.0024	0.871

These results show that charging multiple cores to the dissolver is more reactive than only one core. However, it is still below the k_{safe} value of 0.96. This is reasonable since the elements are annular and are designed to be subcritical unless they are nested together and surrounded by a beryllium reflector. Each element is already approaching an infinite height, from a neutron's perspective, and adding more height does not significantly increase the system multiplication.

This analysis also shows that charging an element on top of a partially dissolved core will not cause a criticality. Therefore, probing to ensure complete dissolution is not a required criticality control.

4. CONTROL ELIMINATION

Other similar operations in H-Canyon require many administrative controls to ensure criticality safety. These controls may include a detailed charge plan that identifies what material may be charged to the dissolver, a rigorous sampling program to ensure subcritical concentrations, probing to ensure complete dissolution, and soluble poisons to provide two-parameter control. None of these controls are required for HFIR dissolution.

A charge plan is not required because the bounding HFIR fuel elements have been evaluated and shown to be subcritical. It is assumed that only one core will be dissolved at a time. However, if multiple cores are mistakenly dissolved at the same time, it has been shown to be subcritical.

Sampling is not required for HFIR dissolution, because the chemistry limits the nature of the process and it is not possible to achieve a critical concentration. Similarly, probing is not required to show complete dissolution because analysis shows that many cores stacked on top of one another are subcritical.

Lastly, soluble poisons are not required to provide an independently-controlled parameter because there are no credible upsets that can lead to a criticality, with or without poison.

5. CONCLUSIONS

By using realistic analysis and taking credit for chemistry, administrative controls for HFIR were eliminated. The solubility of aluminum was recognized to limit the normal operations and created a reasonable bound to the analysis of normal conditions. The chemical process of acid consumption and using reasonable operational assumptions provided a basis to determine that there were no credible concentration upsets that could lead to a potential criticality. Finally, over-mass scenarios were modeled and shown to be subcritical, thereby eliminating the need for administrative controls.

ACKNOWLEDGMENTS

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