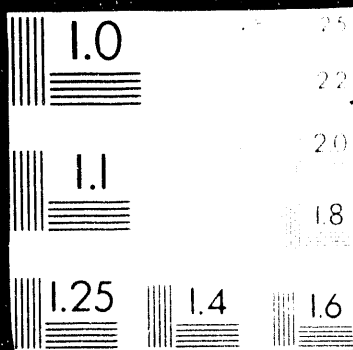


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**CRITICALITY SAFETY STRATEGY FOR THE FUEL CYCLE FACILITY  
ELECTROREFINER AT ARGONNE NATIONAL LABORATORY-WEST**

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### **ABSTRACT**

The Integral Fast Reactor being developed by Argonne National Laboratory (ANL) combines the advantages of metal-fueled, liquid-metal-cooled reactors and a closed fuel cycle. Presently, the Fuel Cycle Facility (FCF) at ANL-West in Idaho Falls, Idaho is being modified to recycle spent metallic fuel from Experimental Breeder Reactor II as part of a demonstration project sponsored by the Department of Energy. A key component of the FCF is the electrorefiner (ER) in which the actinides are separated from the fission products. In the electrorefining process, the metal fuel is anodically dissolved into a high-temperature molten salt and refined uranium or uranium/plutonium products are deposited at cathodes.

In this report, the criticality safety strategy for the FCF ER is summarized. FCF ER operations and processes formed the basis for evaluating criticality safety and control during actinide metal fuel refining. In order to show criticality safety for the FCF ER, the reference operating conditions for the ER had to be defined. Normal operating envelopes (NOEs) were then defined to bracket the important operating conditions. To keep the operating conditions within their NOEs, process controls were identified that can be used to regulate the actinide forms and content within the ER. A series of operational checks were developed for each operation that will verify the extent or success of an operation. The criticality analysis considered the ER operating conditions at their NOE values as the point of departure for credible and incredible failure modes. As a result of the analysis, FCF ER operations were found to be safe with respect to criticality.

### **I. INTRODUCTION**

The Integral Fast Reactor (IFR) being developed by Argonne National Laboratory (ANL) combines the advantages of metal-fueled, liquid-metal cooled reactors and a closed fuel cycle. Some of the primary advantages are passive safety for the reactor and resistance to proliferation for the heavy metal in the fuel cycle. In addition, the IFR pyroprocess recycles all the long-lived actinide activation products for casting into new fuel pins so that they may be burned in the reactor.

Presently, the Fuel Cycle Facility (FCF) at ANL-West near Idaho Falls, Idaho is being modified to recycle spent metallic fuel from EBR-II (Experimental Breeder Reactor-II) as part of the IFR demonstration sponsored by the Department of Energy. A key component of the recycling process in the FCF is the electrorefiner (ER) in which metal fuel is anodically dissolved into a high-temperature molten salt, and refined uranium or uranium/plutonium products are deposited at cathodes. Uranium will be deposited as a solid on a steel mandrel, and mixed plutonium/uranium products will be deposited in liquid cadmium cathodes. It is in the electrorefiner that the actinides are separated from the fission products. The actinides are collected as a minor constituents in the liquid cadmium cathodes, thus enabling them to be cast as recycled fuel. The FCF electrorefining process has been thoroughly described elsewhere [1,2].

Since the FCF ER is a complicated assembly of hardware, and the ER processes themselves are complex, the number of issues relevant to evaluation of the FCF ER process for criticality safety is substantial. The primary goal of this report is to summarize the criticality safety strategy for the FCF ER processes.

FCF ER operations and processes formed the basis for evaluating criticality safety and control during actinide metal fuel refining. The major processes for electrorefining metal fuel are anodic dissolution of the metallic fuel, uranium deposition onto solid cathodes, and mixed uranium/plutonium deposition into liquid cadmium cathodes. The significant operations for criticality hazards control are those that alter the ER fissile materials inventory or that alter their form and location. The ER fissile material inventory is enhanced when metallic fuel is added for anodic dissolution and is reduced when refined actinide deposits on cathodes are removed. Other forms and locations for fissile material in the ER are as chlorides in the molten salt phase, as metals dissolved in a liquid cadmium pool, and as insolubles (e.g., heavy metal oxides) dispersed in the ER.

The criticality safety strategy adopted incorporates five major features: 1) reference conditions and operating envelopes, 2) selection of abnormal events, 3) process controls, 4) operational checks, and 5) criticality analysis. In order to show criticality safety for the FCF ER, the reference operating conditions for the ER had to be defined with respect to the quantity of actinides in the ER, their possible forms, and their specific locations. Since the process is still at the research and development stage, normal operating envelopes (NOEs) were then defined that bracket the important operating conditions and allow latitude for process development. (See Table I.)

Experimental results with an engineering-scale ER were extensively used to develop the reference conditions and normal operations envelope. The process deviations and statistical fluctuations in the sample and analysis results figured prominently in the development of the NOEs. The NOE values subsequently provided the point of departure for credible and incredible failure modes in the criticality analysis.

The process controls for criticality safety were identified for the ER and for each kind of ER process item (e.g., solid cathode, liquid cathode, etc.). These are summarized in Table II. They were selected to provide multiple, independent controls so that the electrorefiner operated within the normal operations envelopes. Some of the process controls actively monitor the extent of the process. The passive process controls, such as the weights of process items, limit the extent of an operation from its inception.

Operational checks were developed that can monitor the ER operational state against the NOE values. When the checks for a given operation compare favorably with the expected result, follow-on processes that were pre-approved may continue unimpeded. Should an operational check indicate some process indicator did not fall within its specified tolerances, further operations would be suspended pending a technical review.

The criticality analysis for the electrorefiner is based on calculations using the KENO V.a code [3]. A summary of the calculational methodology and its validation has been given [4]. Values reported for  $k_{eff}$  in this document were calculated using the methodology described in this summary.

Abnormal events in the criticality analysis were categorized as 1) mass, 2) material (e.g., substitution), 3) density, 4) reflection, and 5) geometry. The criticality safety strategy for the ER is based on the criterion that nuclear criticality shall not result from the concurrent occurrence of one unlikely event from one category above and an extremely unlikely event from a different category. The ER criticality analysis typically considered abnormal configurations involving two or more extremely unlikely events.

In the absence of extensive statistical data on the operation of similar facilities, no formal probabilistic risk assessment was performed. Consequently, the definition and classification of abnormal events required careful examination of the design and operation of the ER and required application of sound technical judgement. The classification of individual events as unlikely or extremely unlikely was based on the following technical issues: a) equipment and container designs, b) physical limits and controls, c) administrative controls, d) criticality control limits, e) process variations, f) sampling and analysis uncertainties, g) distinguishability of different material forms and containers, and h) number of steps or length of time required for the event to develop without notice.

The margin of criticality safety that the ER processes offer was examined in a parametric criticality analysis. For example, the variation of  $k_{eff}$  with the actinide content of the molten salt phase was

determined. Since  $k_{\text{eff}}$  was still small even for actinide contents in large excess of the entire FCF ER NOE, the margin of safety was found to be large. With this approach, operations or processes that are sensitive ( $dk_{\text{eff}}/dx$  is large, where  $x$  is the ER parameter) could be identified.

In this report, each fissile material form and location and each ER operation will be discussed in terms of the five major features of the strategy: 1) the reference conditions and NOEs, 2) abnormal events, 3) the process controls, 4) the operational checks, and 5) the criticality analyses. With reference to the criticality analyses, the important aspects of the NOEs, process controls, and operational checks will be presented. The criticality safety of the ER for credible and incredible failure modes will thus be established.

This report assumes that the reader has a modest familiarity with ER operations and processes and makes frequent use of the NOE values in Table I. In the next section, the possibility and consequences of the entire ER actinide inventory locating in a single phase are examined (Section 2). Particular ER process items (Section 3) and operations (Section 4) are then considered. The concluding section summarizes the effectiveness of the adopted strategy to maintain criticality safety in the FCF ER.

## II. PHASE TRANSFERS OF ACTINIDES WITHIN THE ER

To assess the criticality safety of the ER process, the possibility of the entire ER NOE actinide inventory locating in each one of the possible states for the actinides was considered. As Table I indicates, there exist three possible states for actinides in the ER: the dissolved actinide chlorides in the salt phase, the soluble actinides in the cadmium phase, and the hold-up actinides in the cadmium phase. Hold-up actinides in the electrefiner are the actinide insolubles. Typically, the hold-up actinides will be actinide oxides that are produced from reaction of actinides with oxygen-bearing impurities such as water. Also indicated in Table I are the NOE values for these three phases and for the entire ER. Note however that the total ER inventory is less than the simple sum of the NOEs for the salt, cadmium, and hold-up. This nuance results from recognizing that process deviations and sampling and analysis uncertainties for individual phases do not simply sum for the entire ER.

In this section, complete transfers of all the ER NOE actinide inventory (82 kg) into one phase are discussed. First, the possibility and consequence of the entire ER NOE actinide inventory being present in the salt phase as the actinide chlorides is presented. Attention will be given to the criticality analyses that have been performed to demonstrate the safety margin that exists, to the process controls that will be used to maintain the actinide concentration within the NOE, and to the operational checks used to verify the heavy metal concentration in the salt phase. In addition, cooling the ER contents to room temperature with all the actinides in the salt is considered.

Subsequently, the cadmium phase will be discussed in an analogous manner, considering that the entire NOE actinide inventory (82 kg) has been reduced to the metal state. Lastly, the entire NOE actinide inventory will be considered to be present as actinide oxides resulting from reaction with water or oxygen. Specific ER operations (e.g., deposition) are presented in the next section (3.0).

### II.A. ER NOE Actinide Inventory Present as Actinide Chlorides at 500 C

The reference actinide content of the salt phase is 34 kg (i.e., 49 kg of actinide chlorides). The NOE for the salt phase is 44 kg of actinides (64 kg of actinide chlorides). In order for the entire ER NOE actinide inventory (82 kg) to be present as the actinide chlorides, sufficient oxidant ( $\text{CdCl}_2$ ) must be added to the ER to oxidize this quantity of actinide metals. Examination of the relevant phase diagrams indicates that  $\text{UCl}_3$  and  $\text{PuCl}_3$  would be the major actinide phases present with 82 kg of actinides dissolved in the salt phase [5-8].

At start-up,  $\text{CdCl}_2$  is added to establish the salt phase actinide content at about 34 kg (i.e., 49 kg of actinide chlorides). This amount of actinides requires 39 kg of  $\text{CdCl}_2$  to oxidize it. An additional 55 kg of  $\text{CdCl}_2$  would need to have been added to the ER to bring the actinide content of the salt phase up to 82 kg (assuming the actinides were available).

During routine operations,  $\text{CdCl}_2$  will be added to the ER to compensate for the active metal fission products and bond sodium. The active metal fission products in the fuel will reduce actinide chlorides in the salt phase thereby lowering the actinide concentration in the salt phase. Addition of  $\text{CdCl}_2$  will oxidize the actinides that have been reduced by the active metal fission products and bond sodium, thus restoring the actinides in the salt phase to their reference concentration. The amount of  $\text{CdCl}_2$  that will be necessary to compensate for the active metal fission products and bond sodium is about 3.2 kg  $\text{CdCl}_2$  per batch of irradiated fuel.

#### II.A.1. Process Controls

The amount of  $\text{CdCl}_2$  to be added to the ER at any time will be indicated on the current process data sheet. The process engineer decides the appropriate amount of  $\text{CdCl}_2$ , using process knowledge and operational objectives as a guide. The amount of  $\text{CdCl}_2$  to be added is reviewed independently. Note that the expected amounts for routine addition of  $\text{CdCl}_2$  and for an error that brings the salt actinide content to 82 kg are quite disparate (3.2 kg  $\text{CdCl}_2$  vs. 55 kg, although time constraints may make it undesirable to add the  $\text{CdCl}_2$  after every single batch in increments as small as 3.2 kg).

#### II.A.2. Operational Checks

The immediate check on the amounts of  $\text{CdCl}_2$  added to the ER is the net weight change for the process addition item. Furthermore, the amount that can be added at any time is constrained by the geometry and dimensions of the addition device. With the present design, one addition operation is limited to about 2 kg of  $\text{CdCl}_2$ .

The process model and sample analysis provide the checks on the actual salt phase actinide inventory. The inventory will be determined from the reported actinide concentration and the salt level determinations. The process model predicts the individual actinide contents of the salt phase, and ER salt sample analysis verifies the process model.

#### II.A.3. Criticality Analysis

A parametric criticality safety analysis has been performed that determines  $k_{\text{eff}}$  for varying salt concentrations. For 50 kg of actinides in the salt phase (12 inch depth) with the  $\text{PuCl}_3$  to  $\text{UCl}_3$  ratio at 6:1, the value of  $k_{\text{eff}}$  was found to be less than 0.14. Increasing the actinide content of the salt phase up to 250 kg has shown that  $k_{\text{eff}}$  is insensitive toward the actinide content of the salt phase for the range of contents evaluated. With 250 kg of actinides in the salt phase and a salt depth at 8 inches (2 inches below the NOE minimum depth),  $k_{\text{eff}}$  is less than 0.59. This illustrates the large criticality safety margin that the ER offers toward actinide chlorides dissolved in the salt phase at 500 C. (Note that values for the actinide salt content in excess of 82 kg are hypothetical in that some actinide chlorides may precipitate for higher concentrations. However, precipitation is discussed in the next section for the case where the ER cools to room temperature.)

### II.B. ER NOE Actinide Inventory Present as Chlorides at 20 C

If the ER lost power to the heaters, the ER contents could eventually cool to room temperature (20 C). This section considers the consequences of the cooling event with the entire ER NOE actinide inventory (82 kg) initially present in the salt phase. The conditions that could lead to 82 kg of actinides being present in the salt phase were presented in Section 2.1.

For 82 kg of actinides present in the salt phase at 500 C, uranium and plutonium will be present as  $\text{UCl}_3$  and  $\text{PuCl}_3$  dissolved in the ER salt. As the ER cools, solidification of the molten actinide and other chlorides will certainly occur. Some phases will precipitate earlier than others, since the ER salt will not likely be at the eutectic composition with regard to all phases present.

The densest phases possible, with regard to actinides, are  $\text{UCl}_3$  and  $\text{PuCl}_3$ . However, it is more likely that a significant portion or most of the actinides will precipitate as  $\text{K}_2\text{UCl}_5$  and  $\text{K}_3\text{PuCl}_6$ , as the phase diagrams indicate [5-8]. These phases will precipitate as small grains surrounded by the  $\text{LiCl/KCl}$  melt. As the temperature is lowered further, the  $\text{LiCl/KCl}$  melt will solidify to yield a lamellar structure of the actinide phases in the  $\text{LiCl/KCl}$  matrix. Should the density of the actinide

chlorides induce their settling, a  $\text{PuCl}_3$  (highest melting point, first to solidify) layer could result with a maximum density of about 50-60% theoretical.

#### II.B.1. Process Controls

The process controls and operational checks that guard against 82 kg of actinides being present in the salt phase have been discussed (Section 3.1.1). For the ER to cool to room temperature, all heat sources must be absent for an extended period of time. The ER is configured with three heater zones with a redundant set of heaters in each zone. Only the DBE (design basis earthquake) is viewed as possibly arresting the ER heating capability for an extended period of time. Note that fission product decay would extend considerably the time required for cooling the ER contents to room temperature.

#### II.B.2. Operational Checks

The operational checks used to maintain the ER actinide content of the salt phase within its NOE have been discussed (Sec. 3.1.2). The power to the heater banks will be used to monitor the performance of the heaters. Also, the temperature is monitored routinely at numerous locations in support of the heater performance.

#### II.B.3. Criticality Analysis

A criticality safety analysis has been performed for the case where 80 kg of actinides were in the salt phase, the plutonium to uranium ratio was 6 to 1, and the temperature was 20 C. The analysis was especially conservative in that the phases present were  $\text{UCl}_3$  and  $\text{PuCl}_3$  at 100% theoretical density, and were assumed to settle to the bottom of the salt phase. The value for  $k_{\text{eff}}$  under these conditions was less than 0.40.

Even for 160 kg of the binary actinide chlorides ( $\text{MCl}_3$ ) at 100% theoretical density in a layer between the salt and cadmium at 20 C,  $k_{\text{eff}}$  is less than 0.66. These conditions are substantially different from the expected behaviour, and the amount of actinides greatly exceeds the NOE, demonstrating the large criticality safety margin that the ER salt phase offers toward cooling events.

#### II.C ER NOE Actinide Inventory Present as Reduced Metals at 500 C

The reference actinide content of the cadmium phase is 20 kg, while the NOE is 30 kg. In order for the entire NOE actinide inventory (82 kg) to be present exclusively as reduced metals, all the actinides in the salt phase would have to be reduced with some reductant such as  $\text{LiCd}$  alloy. The phases that would possibly be present with 82 kg of reduced metal actinides in the ER are uranium and plutonium dissolved in cadmium, uranium metal, and  $\text{PuCd}_6$ .

The solubility of uranium is 2.35 wt% in cadmium. The reference cadmium content of the ER is about 1000 kg (6 inches of cadmium) which can dissolve about 24 kg of uranium. The lower NOE for the cadmium is 4.5 inches (750 kg) which can dissolve about 18 kg. Once the solubility limit is reached, uranium metal will precipitate as grains dispersed in molten cadmium.

The solubility of plutonium in cadmium is 3.62 wt%. Hence, the ER cadmium at its reference depth can dissolve about 38 kg of plutonium. At the lower ER NOE depth for cadmium of 4.5 inches, about 28 kg of plutonium can be dissolved. When plutonium reaches its solubility limit in cadmium,  $\text{PuCd}_6$  precipitates as grains dispersed in the molten cadmium. From the stoichiometry of the compound ( $\text{PuCd}_6$ ), a total of 267 kg of plutonium would be necessary to exhaust 750 kg of cadmium (4.5 inch layer) in making  $\text{PuCd}_6$  before pure plutonium could deposit. For comparison, the Criticality Hazards Control Statement (CHCS) limits the total ER plutonium content to 60.0 kg.

$\text{LiCd}$  alloy reductant may be used in a few different operations including drawdown and chemical reduction runs. These two operations require differing amounts of  $\text{LiCd}$  for the reduction of actinides in the ER. It is expected, however, that single additions of  $\text{LiCd}$  alloy will not exceed the amount corresponding to two solid cathodes (20 kg uranium). This amount of uranium is equivalent to 1.7 kg of lithium (30.1 kg of  $\text{LiCd}$  @ 1:1 mole ratio). By contrast, 6.97 kg of lithium (123 kg of  $\text{LiCd}$  @ 1:1 mole ratio) would be necessary to reduce the ER NOE actinide inventory of 82 kg.

### II.C.1 Process Controls

Any reductant to be added to the ER will be indicated on the process data sheet. The process engineer decides the appropriate amount of LiCd alloy to be added using process knowledge and operational goals as a guide. The amount of LiCd alloy to be added is peer-reviewed.

If excess reductant is added, then rare earth chlorides will be reduced to their metals, in addition to the actinides, and the rare earth metals will also dissolve in the cadmium. This is not expected to depress significantly the solubility of uranium or plutonium in cadmium. Even if the solubilities were depressed, uranium will still precipitate as uranium metal and plutonium as  $\text{PuCd}_6$ .

### II.C.2 Operational Checks

The most immediate check on the weight of reductant being added to the ER is its net weight. This can be established when the operation is being set-up and checked when the net weight change of the addition device is determined after the addition. The lithium content will have been determined prior to the addition by chemical analysis.

The process model and sample analysis are used to detail the cadmium pool's contents in the same way as for the salt phase. Sample analysis results and cadmium pool level measurements will be used to calculate the actinide content of the ER cadmium pool.

### II.C.3. Criticality Analysis

A parametric criticality safety analysis has been performed for the ER cadmium phase with the reduced metal actinide content ranging from 50 to 250 kg. At 50 kg of reduced metal actinides and a cadmium depth of 4.5 inches in the ER, the phases present would be uranium dissolved in cadmium, plutonium dissolved in cadmium, and  $\text{PuCd}_6$ . The plutonium to uranium ratio was taken as 6:1. The  $\text{PuCd}_6$  was assumed to form a layer on the bottom of the ER. For these conditions,  $k_{\text{eff}}$  was less than 0.21.

Even with 250 kg of reduced metal actinides,  $k_{\text{eff}}$  was less than 0.79. Again, the plutonium to uranium ratio was 6 to 1 and the cadmium depth was 4.5 inches. The phases that would be present for these amounts of cadmium and reduced metal actinides are uranium and plutonium dissolved in cadmium, uranium metal, and  $\text{PuCd}_6$ . The uranium was assumed to form the bottom layer in the ER, covered by a layer of  $\text{PuCd}_6$ , with a molten cadmium layer above saturated in uranium and plutonium. Thus, the criticality safety margin with regard to reduced actinide metals in the ER is seen to be large, primarily because of the formation of  $\text{PuCd}_6$  rather than plutonium metal when the cadmium becomes saturated in plutonium.

## II.D. ER NOE Actinide Inventory Present as Reduced Metals at 20 C

As mentioned previously, loss of ER heating capability could eventually result in the ER temperature reaching 20 C, albeit extremely unlikely. This section considers the consequences of the cooling event with the entire NOE actinide inventory (82 kg) present as reduced metal actinides. The conditions that could lead to 82 kg of actinides being present as reduced metals were discussed in the previous section.

With 82 kg of reduced metal actinides in the ER at 500 C, plutonium and uranium will be dissolved in the cadmium up to their saturation limits. As deduced from the appropriate phase diagrams [9,10], the remainder of the uranium will be precipitated as grains of solid uranium in the cadmium melt, and the remainder of plutonium as grains of  $\text{PuCd}_6$ . As the temperature is lowered to about 474 C, the solid uranium grains will convert to  $\text{UCd}_{11}$ , and any further precipitation of uranium from the cadmium solution will result in formation of  $\text{UCd}_{11}$ . On further cooling to 404 C, the  $\text{PuCd}_6$  will convert to  $\text{PuCd}_{11}$ . Below 321 C, the melting point of cadmium, all the metal phases will be solidified.

With the entire 82 kg of actinides existing as the  $\text{MCd}_{11}$  intermetallic compound, about 42% of the cadmium (at a depth of 6 inches) would have been consumed. The resulting microstructure would be crystals of  $\text{UCd}_{11}$  and  $\text{PuCd}_{11}$  dispersed in the cadmium matrix. Pure, solid plutonium or uranium would not obtain in this given system.

#### II.D.1. Process Controls

The process controls that guard against the entire ER NOE actinide inventory being present as reduced metal actinides have been discussed (Section 3.3.1). The heater banks for the ER were also mentioned with regard to cooling of the salt phase.

#### II.D.2. Operational Checks

The operational checks used to maintain the ER cadmium phase within its NOE with regard to its actinide concentration and to its depth have been presented (Sec. 3.3.2). The power to the heaters plus their temperatures will be used to monitor their performance.

#### II.D.3. Criticality Analysis

The criticality analysis for reduced actinide metals in the ER at 500 C (Section 3.3.3) bounds the case of 82 kg of reduced actinide metals in the ER at 20 C. The large criticality safety margin demonstrated at 500 C far outweighs any change in density of 82 kg of actinide solids on cooling. In fact, the number density of the plutonium and uranium *decreases* as uranium is converted to  $\text{UCd}_{11}$  and  $\text{PuCd}_6$  is converted to  $\text{PuCd}_{11}$  on cooling.

### II.E. ER NOE Actinide Inventory Present as Actinide Oxides at 500 C

Compromising the argon atmosphere of the ER can result in the formation of undesirable actinide by-products. The principal concern is the formation of actinide oxides upon contact with oxygen or water vapor in air. The ER atmosphere can be compromised by the DBE event, by mechanical failure of the ER argon supply-line, or by contamination of the ER high-purity argon supply.

The major actinide oxide phases that may form in the ER are  $\text{UO}_2$  and  $\text{PuO}_2$ . The particles that form are expected to be in the micron-size range, and a credible maximum density for the particles is about 40%. Even with some densification resulting from agglomeration over long time periods, densities for oxide particles above 60% theoretical are considered incredible [11]. Because of their density, the oxides can locate as a precipitate on top of the cadmium phase or as floccules in the salt phase.

When actinide oxides form in the ER, the actinides are obscured from the electrorefining process. Actinide oxides do not have any significant solubility in the  $\text{LiCl/KCl}$  eutectic salt; consequently, they cannot be deposited on the cathodes. Actinide oxides once formed represent a contribution to the hold-up in the ER. The NOE for actinide holdup in the ER is 12 kg of actinides.

If the ER atmosphere is greatly compromised, the entire actinide inventory can react to form oxides. The NOE for heavy metal in the ER is 82 kg. The CHCS limits in the ER are 60.0 kg for plutonium and 77.2 kg total fissile. If the ER contained the limit on fissile material and all of the heavy metal in the ER were converted to oxides, there would be 68.0 kg of  $\text{PuO}_2$ , 19.5 kg of  $^{235}\text{UO}_2$ , and 5.44 kg of  $^{238}\text{UO}_2$  in the ER. In the absence of convection or stirring, the oxide particles would settle on the cadmium pool; since gravity would be the only significant force acting on the particles, the layer should be rather uniform.

With stirring, the oxide particles may distribute non-uniformly in the salt phase or at the salt/cadmium interface. For example, some of the actinide oxide particles may accumulate close to the ER vessel walls in the vicinity of the ER stirrer baffles. Other possible sites for accumulation include a ring around the stirrer or a hemi-toroidal ring some radial distance from the stirrer, but located at the salt/cadmium interface because of the reduced particle density. The rotation of the solid cathode and anode assemblies may also influence the oxide particle distribution. In any case, if the ER atmosphere is greatly compromised, the cessation of all stirring will give the safest geometry of actinide oxides with respect to criticality.

#### II.E.1. Process Controls for Preventing Formation of Actinide Oxides

The ER argon supply line will be monitored for its water vapor content, its oxygen content, and its nitrogen content to help protect against unacceptable impurity levels in the argon purge gas. A separate, emergency ER argon supply line will be installed that is designed to withstand the DBE.

#### II.E.2. Operational Checks for Assessing Formation of Actinide Oxides



The results from ER salt and cadmium sample analysis will be compared to the ER actinide inventory as determined by the difference of actinides input and output. This comparison will permit the holdup actinide oxides to be assigned and maintained within its NOE (12 kg of actinides).

### II.E.3. Criticality Analysis for Actinide Oxides

The criticality analysis examined the formation of both a uniform and a non-uniform layer of the actinide oxides. For the case of the uniform layer of actinide oxides, calculations were performed for 84 and 168 kg of actinides at both 50% and 100% of the theoretical density. The Pu/U ratio was taken as 6:1. The layer of oxides was assumed to settle to the bottom of the ER for the most conservative analysis, since cadmium is a better reflector than the eutectic salt. At 50% theoretical density for 84 kg of actinides as oxides,  $k_{eff}$  is less than 0.46; at 100% theoretical density,  $k_{eff}$  is less than 0.49. Even for 168 kg of actinides as oxides at 100% theoretical density,  $k_{eff}$  is less than 0.80 for a uniform layer.

To be conservative, the uncertainty with respect to location or geometry of particles of actinide oxide led to modeling a non-uniform layer in a very reactive configuration, a hemisphere of actinide oxides on the base of the ER surrounded by cadmium, in the criticality safety analysis. For 40 kg of actinides as oxides at 50% theoretical density, the computed  $k_{eff}$  was less than 0.91. However, given the forces at work in the ER, the assembly of scraper brackets on the ER floor, and the density of the oxides being less than the cadmium (preventing oxide accumulation on the ER floor), it is incredible that a single hemisphere (15.6 cm radius) of actinide oxides can form on the ER floor.

### II.F. ER NOE Actinide Inventory Present as Actinide Oxides at 20 C

If the ER is cooled to room temperature with all of the heavy metal as oxides on the ER floor, the only change from the above conditions will be a very slight increase in the density of the oxide precipitate. There is no driving force for altering the phases present, their location, or their geometry. Even though the density increases, it will still not come close to 60% of theoretical density of the oxides for the reasons given above. The additional effect of cooling on criticality safety evaluations is then negligible if the heavy metal contents of the ER have been converted to the oxides at operating temperature.

## III. ER PROCESS ITEMS

Several operations together comprise the electrorefining process, and different pieces of equipment are associated with each operation. This section summarizes each process item, its process controls, operational checks, and criticality analysis. ER operations and their failure modes are discussed in Section 4.

The fuel dissolution baskets are used to introduce fissile material into the ER. The fissile material can take the form of chopped fuel or fissile material other than chopped fuel such as cathode processor heels. The solid cathode is used to collect uranium deposits. The liquid cathode (alternatively, the chemical cathode) is used to collect plutonium/uranium deposits. Each of these process items are discussed in turn.

### III.A. Fuel Dissolution Baskets With Chopped Fuel

Chopped fuel segments are added to the ER in the fuel dissolution baskets (FDBs). The FDBs have a box-like shape with a rectangular opening at the top. The top-opening measures approximately 3.02 cm x 7.0 cm. They are about 23.7 cm deep. The sides of the FDBs are perforated to permit the salt phase in the ER to mix with the chopped fuel segments.

When using chopped fuel, the CHCS limits the fissile material content of an anode assembly (four FDBs) to 12.7 kg total fissile and 4.4 kg plutonium. The NOE for the FDBs is 15 kg of heavy metal per anode (3.75 kg of heavy metal per basket). The nominal lengths for chopped fuel will be 1/4 inch.

### III.A.1. Process and Administrative Controls for Chopped Fuel in the FDBs

The number and type of elements per basket will be specified on a process data sheet by the process engineer. The chopping recipe will indicate the length of fuel to be chopped and the segment length. The chopping recipe for the fuel will be approved by a peer-reviewer. Chopped fuel segments of nominal dimensions other than 1/4 inch diameter by 1/4 inch length will not be loaded into the FDBs without determining their packing fraction.

The FDB volume and the packing fraction of the chopped fuel segments also provide process control. With the maximum packing fraction, the maximum fissile content of the FDBs can be determined for a given fuel type from the maximum number of segments in an FDB. The maximum fissile content is also the densest configuration for the FDBs which corresponds to a maximum net mass for each fuel type. The packing fraction is an experimentally determined quantity. The theoretical maximum packing for infinite cylindrical segments is 91%.

### III.A.2. Operational Checks for FDBs With Chopped Fuel

The net weight of scrap material from the chopping plus the net weight of the FDBs will be compared to the weight of the pins before chopping. The net weights of the FDBs will be compared to the modeled weights of the FDBs. The process model assesses the actinide mass in the FDBs. Destructive sample analysis of select chopped fuel segments will be performed to corroborate the actinide mass assessed by the process model.

### III.A.3. Criticality Analysis for FDBs With Chopped Fuel

Criticality safety analysis has been performed for the FDBs with the NOE actinide mass per anode assembly and two anode assemblies in the ER (packing fraction is 60%, fuel is unirradiated 26 Pu-64U-10Zr). The  $k_{eff}$  for this configuration is less than 0.41.

For the extreme case of 91% packing fraction (maximum for cylinders), 20 kg of chopped fuel per anode assembly, two anode assemblies in the ER, concrete reflection on three sides, spherical electrode assemblies (incredible), and fuel at 35Pu-55U-10Zr @ 20°C,  $k_{eff}$  is less than 0.84. Hence, the analysis demonstrates the large safety margin that exists for the FDBs in the ER.

Another kind of mass error involving the FDBs results from loading the ER with a second pair of anode assemblies without any cathode deposits having been taken. This error has been referred to as double-batching the ER. The worst possible initial conditions for the double-batching event would have the ER at its NOE of 82 kg of actinides when fully loaded anode assemblies are inserted into the ER.

A criticality analysis has been performed for a case that bounds the double-batching error for the ER. To examine the margin of safety toward criticality, the analysis considered unreasonable actinide loadings of both the ER and the anode assemblies. The ER contained 250 kg of actinides dissolved in the salt phase with a Pu/U ratio of 6 to 1. This represents plutonium and total fissile material contents well in excess of the ER NOE. Each of two anode assemblies contained 40.68 kg of 35Pu-55U-10Zr alloy with 10.17 kg per FDB. This mass represents a packing fraction of 91%, the theoretical maximum for infinite cylinders. For this case that bounds the double-batching event,  $k_{eff}$  was less than 0.81, demonstrating criticality safety for the double-batching event.

### III.B. Fuel Dissolution Baskets With Fissile Material Other Than Chopped Fuel

The FDBs may be used to input fissile material other than chopped fuel into the ER. The fissile material loaded into the FDBs may be uranium; heels or ingots of plutonium/uranium alloys from the cathode processor; heels from casting binary and ternary alloy heels; make-up uranium or plutonium, etc. The CHCS limits the fissile material content of an electrode (anode) assembly (four FDBs) to 8.5 kg total fissile and 3.0 kg plutonium for operations with fissile material other than chopped fuel.

### III.B.1. Process Controls for FDBs with Fissile Material Other Than Chopped Fuel

Generally, the FDBs are loaded with fissile material other than chopped fuel in a zone separate from where fuel is being chopped. The process data sheet gives the recipe describing input types, their compositions, and weights of materials to be loaded. The process engineer prescribes the recipe using process knowledge and operational goals as guides. The recipe is peer-reviewed before being assembled. A complete recipe is assembled into a single container and weighed before loading the material into an FDB. Only one FDB is loaded at a time, then the FDB is transferred to the zone for assembling the anode.

For fissile material other than chopped fuel, the weight of fissile material in the FDBs will be determined three separate times before the FDBs are immersed in the ER. The fissile material will be weighed container-wise as it is prepared according to a recipe, it will be weighed after the recipe is assembled into the FDBs, and it will be weighed upon arrival to the ER preparation zone where the FDBs will be assembled as the anode. These multiple weighings are the process controls that protect against the mass error when loading the FDBs with fissile material other than chopped fuel. Mass errors are administratively protected by limiting the mass of material in a given fissile container.

### III.B.2. Operational Checks for FDBs With Fissile Material Other Than Chopped Fuel

The actinide composition of the fissile material will have been chemically assayed, and the weight changes in the fissile containers will be checked against the weight change in FDB. The net weight of the loaded FDB after transfer to the anode assembly zone will also be compared to its weight prior to transfer.

### III.B.3. Criticality Analysis for FDBs With Fissile material Other Than Chopped Fuel

Criticality safety analysis for FDBs with fissile material other than chopped fuel has been performed for many fissile material feedstocks. These include pure plutonium, pure uranium, low Pu/U alloy, and U-Pu-Zr ternary alloy. The criticality analysis for pure plutonium is the only feedstock summarized here, since plutonium case is the most reactive material.

For the unlikely event that 3.75 kg of plutonium is loaded in one FDB, a total of 4.25 kg plutonium per electrode assembly and two electrode assemblies in the ER,  $k_{eff}$  is less than 0.82. This  $k_{eff}$  also represents the extremely unlikely events of 1) the plutonium at 100% theoretical density at 20°C, 2) concrete reflection on three sides of the ER, and 3) spherical electrode assemblies.

Even with the above extremely unlikely conditions, and 5.0 kg plutonium in one FDB, 5.5 kg plutonium per electrode assembly, and two electrode assemblies in the ER,  $k_{eff}$  is less than 0.89. These analyses demonstrate the criticality safety margin offered by FDB operations with the CHCS limit of 3.0 kg pure plutonium per electrode assembly for fissile material other than chopped fuel. Alternate feedstocks of fissile material other than chopped fuel gave lesser values of  $k_{eff}$  for similar conditions.

When loading the FDBs with fissile material other than chopped fuel, the worst substitution event possible is substitution of plutonium for a less reactive material. The criticality analysis considered a scenario involving a substitution error whose initial premise was a mass error. In the analysis, a single FDB is overloaded with 4.88 kg of low Pu/U alloy by mistake. On top of this mass error for loading low Pu/U alloy, the entire mass is substituted for with pure plutonium. With 4.88 kg of plutonium in a FDB, 9.38 kg of total fissile material per electrode assembly, and two electrode assemblies in the ER, the analysis showed that  $k_{eff}$  is less than 0.79. This analysis was even more conservative in that the plutonium was considered a single piece at 100% theoretical density and that the ER was reflected by concrete on three sides.

### III.C. Solid Cathodes

Solid cathodes are used to collect uranium metal in the FCF ER. The solid cathode consists of a steel or uranium rod that is placed in the ER salt phase. The uranium that electrochemically deposits typically has a dendritic appearance similar to a bottle-brush. Dendritic deposits are typical for metals with rapid deposition kinetics, and deposition processes in molten salts typically have fast kinetics. The density of the deposits are naturally low because of the voids left by a dendritic structure.

Rotation of the solid cathode during the deposition process constrains the deposit's diameter to 10 inches because of the action of a scraper mounted on the ER side-wall. Another scraper mounted on the floor of the ER and protruding into the salt phase prevents the dendrites from growing down to the cadmium pool (whereupon the electrorefining process would be short-circuited). The uranium dendrites that are knocked off the deposit by the scrapers drop to the cadmium pool and dissolve in it.

Plutonium metal does not deposit on the solid cathode because of the presence of  $\text{UCl}_3$  in the ER salt phase. Plutonium metal is unstable in the presence of  $\text{UCl}_3$  and would be oxidized to form  $\text{PuCl}_3$  unless the  $\text{PuCl}_3$  to  $\text{UCl}_3$  ratio in the salt phase is very large. For a system at thermodynamic equilibrium, the  $\text{PuCl}_3$  to  $\text{UCl}_3$  ratio would need to be about  $10^5$  before plutonium metal becomes stable [2]. Experimentally, it has been demonstrated that plutonium metal does not deposit with the  $\text{PuCl}_3$  to  $\text{UCl}_3$  ratio less than 13 [12].

The reference uranium mass for a solid cathode deposit is 10 kg of uranium. The NOE mass is 15 kg of uranium. The reference density of the deposit is 4% theoretical [13]; however, for conservatism, the NOE density was assumed to be 40% theoretical.

### III.C.1. Process Controls for Solid Cathodes

The mass of uranium that may be deposited on the solid cathode is limited by the integrated current (number of coulombs passing) during the deposition process. The deposition process typically has an efficiency less than 100%. If a low efficiency is assumed, and a higher efficiency is observed, then the mass of the deposit will be greater than expected. Notwithstanding, the mass of the deposit cannot exceed the mass corresponding to 100% efficiency. The integrated current will be actively monitored through both legs of the power supply (anode and cathode leads) simultaneously, and the two values will be compared.

The mass of the deposit will also be gauged by actively monitoring the power required to rotate the SC, since it has been shown that the power increases in time as a result of the action of the scrapers on the growing deposit. The relationship for the mass of actinides on the SC to the power required for rotation will be empirically established in the FCF ER. If the side scraper fails during operation, the power required to rotate the SC will be obviously different from the expected behavior. If the bottom scraper fails, the deposit will electrically short with the cadmium pool. This event will be detected by a voltage surge toward zero volts arising from a dramatically lower resistance.

Process Data Sheets will indicate both the maximum integrated current and the maximum power required to rotate the solid cathode for each solid cathode operation. They will be specified by a process engineer and reviewed by a peer.

### III.C.2. Operational Checks for Solid Cathodes

The operational checks for the solid cathode depend on its net weight initially and at a later time on the sample analysis results from the cathode processor products. The cathode processor produces actinide metal ingots in a retort operation that takes the solid cathode ER products as input. Since the content of solid cathodes is around 85% uranium metal and 15% adhering salt for routine electrorefining operations [1], the net weight provides a good indication of the uranium being removed from the ER, pending analysis of the cathode processor products. Sampling and analysis of the ER salt phase will be used to determine the ratio of  $\text{PuCl}_3$  to  $\text{UCl}_3$ .

### III.C.3. Criticality Safety Analysis of Solid Cathodes

A criticality safety analysis has been performed for solid cathodes. For 20 kg of actinides per solid cathode at 40% theoretical density, two solid cathodes in the ER, a plutonium to uranium ratio of 1 to 6 on the deposit, and 100 kg of actinides in the salt phase,  $k_{\text{eff}}$  is less than 0.57.

Even for the incredible case of 30 kg of actinides per solid cathode at 100% theoretical density with the most reactive geometry for a cylinder (height to diameter ratio is one),  $k_{\text{eff}}$  is less than 0.95. This case also considered that two solid cathodes are in the ER, the plutonium to uranium ratio on the deposit is 1 to 6, and 150 kg of actinides are dissolved in the salt.

Despite the large value for  $k_{\text{eff}}$  in this latter case, it is incredible that two cathodes can collect 30 kg each of actinides because of the reference ER inventory of actinides and because of the process controls. It is even more incredible that the deposit would form with an optimal cylindrical geometry

with a diameter to height ratio of unity. Finally, the typical density of the deposits taken from the engineering scale ER is about 4% theoretical compared to 100% theoretical used in the analysis [13]. Thus a large margin of safety toward criticality for the solid cathodes is demonstrated.

### III.D. Liquid Cathodes

Liquid cathode operations are the principal means to collect plutonium/uranium products along with small quantities of the minor actinides. A liquid cathode consists of molten cadmium in a ceramic crucible that is placed in the ER salt phase. Both the plutonium and uranium are electrochemically deposited in the liquid cathode cadmium. Uranium also has a tendency to form dendrites on the surface of the cadmium in the liquid cathode. A mechanical device such as an impeller or pounder is used to force the uranium dendrites below the surface of the cadmium in the liquid cathode. As with the solid cathode, any actinides that are knocked out of the liquid cathode by the impeller or pounder do not increase the ER inventory so that a criticality issue is not posed by this event.

The reference cadmium content of a liquid cathode is 30 kg. The reference deposit for a liquid cathode is 2.8 kg of plutonium and 0.6 kg of uranium. The NOE for a liquid cathode is 6.0 kg of plutonium and 1.0 kg of uranium. The possible phases in the liquid cathode after solidifying are uranium and plutonium as fine grains dispersed in solid cadmium,  $\text{PuCd}_6$ , but mostly  $\text{PuCd}_{11}$  and  $\text{UCd}_{11}$ .

#### III.D.1. Process Controls for Liquid Cathodes

The process controls for the liquid cathode are analogous to Those used for the solid cathode. The maximum mass of the actinides depositing in the liquid cathode is limited by the integrated current. The integrated current and the power required to rotate the impeller are actively monitored in exactly the same way as for the solid cathode.

Process Data Sheets will indicate both the maximum integrated current and the maximum power required to rotate the impeller for each liquid cathode operation in a similar fashion to the solid cathodes.

#### III.D.2. Operational Checks for the Liquid Cathodes

As for the solid cathodes, the operational checks for the liquid cathodes depend on their net weight and on the sample analysis results for the cathode processor products. Since the volume of cadmium input to a liquid cathode is known, the volume and corresponding weight of salt free-boarding can be used to calculate the actinide content. Sampling and analysis of the ER salt phase will be used to determine the ratio of  $\text{PuCl}_3$  to  $\text{UCl}_3$ .

#### III.D.3. Criticality Analysis of the Liquid Cathodes

Criticality safety analysis has been performed for the liquid cathodes. The analysis considered that two liquid cathodes were in the ER salt phase, each containing 14.0 kg of actinides with a Pu:U ratio of 6 to 1. The ER salt phase was assumed to have 250 kg of actinide chlorides with a  $\text{PuCl}_3$  to  $\text{UCl}_3$  ratio of 6 to 1.

With 30 kg of cadmium per liquid cathode, 10.6 kg of the 12 kg of plutonium available would form  $\text{PuCd}_6$ . The liquid cathode deposit in the analysis consisted of a 2 kg bottom layer of uranium, covered by a 1.4 kg layer of plutonium, followed by a top layer of  $\text{PuCd}_6$ . The analysis gave a  $k_{\text{eff}}$  less than 0.65.

The resulting small value of  $k_{\text{eff}}$  for the analyzed configuration and the large difference between the actinide content of the analyzed configuration and the NOE actinide content of a liquid cathode (6.0 kg Pu, 1.0 kg U) together illustrate the criticality safety margin that operations with the liquid cathodes offer.

### III.E. Chemical Cathodes

A chemical cathode operation is the alternate method for removing a refined plutonium product from the electrorefiner. This operation involves placing a chemical cathode containing LiCd alloy in the salt phase of the ER. The lithium in the LiCd alloy chemically reacts with the actinides in the ER salt phase. The lithium metal is oxidized and transfers to the ER salt phase while the actinides in the ER salt phase are reduced and transfer to the cadmium phase of the chemical cathode.

The reference and NOE values for actinides in the chemical cathode are identical to those for the liquid cathode. The operational checks and the criticality analysis are also identical. The only substantial difference between the two process items is in the process controls.

#### III.E.1. Process Controls for the Chemical Cathode

The process controls for chemical cathodes are the weight of LiCd alloy and the fractional composition of lithium in the alloy. In addition, the ingots of the alloy will be uniform in size and shape, so that the number of ingots added provides process control. The amount of LiCd alloy to be added in a given chemical cathode will be indicated on the Process Data Sheet.

### IV. ER OPERATIONS

The ER criticality analysis primarily treats the failure mechanisms for ER operations categorically as mass, material, density, reflection and geometry. However, some of the off-normal situations needed to be handled as a separate case (e.g., not characterized as one of the five event categories). In this section, the off-normal configurations for ER operations are highlighted.

The off-normal configurations are 1) reversing the power supply polarity during electrotransport, 2) hardware failure, and 3) depositing uranium on the ER side-walls during anodic dissolution operations. Case 1 actually is a processing error while case 2 pertains to failure of the electrode assemblies. Case 3 is a secondary reaction in the anodic dissolution process [1]. It should be emphasized that the two failure mechanisms, 1 and 2, are considered unlikely or extremely unlikely.

#### IV.A. Reversal of Polarity on the Power Supply

In the event the polarity is erroneously reversed on the power supplies, the anode will act as a cathode, and the cathode as the anode. Two conditions are possible for polarity reversal, depending on the planned operation. Either the anode assemblies will erroneously become the cathodes, or the cadmium pool will erroneously become the cathode. It should be clear that if the cadmium pool erroneously becomes the cathode, this is a benign event because criticality safety has been shown for the case where the entire ER NOE inventory (82 kg) is present as reduced metals, much of which would be dissolved in the cadmium (Section 3.3.3).

When the anode assemblies are erroneously wired as cathodes, uranium deposits on the FDBs. The FDBs will act like a solid cathode, i.e., collect uranium but not plutonium. The morphology of the deposit will be the same as that for the solid cathode--dendritic. The worst credible initial conditions would be that the FDBs were filled with actinides. The worst credible result would be that 10 kg actinides deposits were taken on each of the two anode assemblies.

Process diagnostics are available that will detect polarity reversal of the power supply. These include voltage, resistance, and power required to rotate the electrode assemblies. Their essential features will be the departure of the diagnostics from their characteristic behavior when the power supply is correctly connected. Additionally, the behavior of each diagnostic will depend on what electrodes are connected to the power supply.

Criticality analysis has been performed for the case where the anode assemblies are erroneously connected as the cathodes. The analysis considered three cases, two of which are summarized here. In one case, the FDBs are loaded with chopped fuel, in the second case with fissile material other than chopped fuel.

The chopped fuel case considered that the FDBs contained 15 kg of actinides in each of two anode assemblies. The chopped fuel was 35Pu-55U-10Zr at a packing fraction of 91%. Each anode assembly collected 15 kg of actinides at 40% theoretical density. For conservatism, the ER was assumed to be 20 C with concrete reflection on three sides and the electrode assemblies were assumed

to be spherical. The evaluated  $k_{eff}$  was less than 0.92 for this case. Even with one liquid and one solid cathode (with their NOE actinide contents) simultaneously present in the ER,  $k_{eff}$  increases only slightly and is still less than 0.92.

While this value for  $k_{eff}$  is large, the conditions leading to it are incredible. The FDBs were loaded at the theoretical packing fraction for infinite cylinders, and the deposits on the FDBs were abnormally large at an exceptionally high density. Furthermore, it is incredible that such an operation may proceed for the required length of time without being detected by some process diagnostic. Indeed, the dendrites will soon grow to such a length that they will short electrically at the cadmium pool, since a scraper does not exist under the anode ports. This is only one example of several diagnostics that will indicate polarity reversal.

The case of fissile material other than chopped fuel considered that each of two anode assemblies contains 17.7 kg of 21Pu-69U-10Zr ternary alloy. However, to be conservative, in one of the FDBs 6.0 kg of plutonium was erroneously substituted for 6.0 kg of the ternary alloy. The packing fraction of the FDBs' contents was assumed to be 100%. The anode assemblies were assumed to collect 15 kg each at 40% theoretical density. The  $k_{eff}$  for this case was less than 0.88. The analysis was additionally conservative in that the ER was assumed to be at 20 C with concrete reflection on three sides.

These analyses demonstrate the criticality safety of the ER in the event of reversing the polarity from the power supply.

#### IV.B. Failure of the Electrode Assemblies

In the event the electrode assemblies break apart while in the ER, the electrodes with their contents can drop to the ER bottom. If the metal actinides in the electrodes dissolve in the cadmium, a criticality safety issue is not posed because criticality safety has been demonstrated should the entire ER NOE actinide content (82 kg) be reduced to the metal state. The issue to examine is the effect of the geometry of the actinides constrained by the electrodes when immersed in the cadmium pool. The criticality analysis that has been performed for each type of electrode assembly (anode, liquid cathode, and solid cathode) will be summarized.

The configurations analyzed were chosen to bound all credible configurations of actinides in the ER for this failure mechanism. The electrode assemblies in each case are assumed to be completely immersed in 10 inches of cadmium. The cadmium level was raised above the NOE (7.5 inch upper limit) for the analysis because cadmium is a better reflector than the electrolyte, thus rendering the analysis more conservative. The cadmium contained 100 kg of actinides (18 kg above the ER NOE) uniformly distributed with a Pu/U ratio of 6:1.

##### IV.B.1. Failure of the Anode Assemblies

One configuration was examined for anode assemblies in the cadmium pool. Each of two anode assemblies contained 40.68 kg of 35Pu-55U-10Zr at a packing fraction of 91%. The computed  $k_{eff}$  for this configuration is less than 0.81.

##### IV.B.2. Failure of the Solid Cathode Assemblies

Two configurations for solid cathodes were examined with regard to failure of the electrode assemblies. In case I, the two solid cathodes each contained 20 kg of actinides with a Pu/U ratio of 1:6 at a theoretical density of 100%. The computed  $k_{eff}$  was less than 0.93. In case II, the two solid cathodes each contained 30 kg of actinides at a Pu/U ratio of 1:6 at a theoretical density of 0.60. This second case yielded a  $k_{eff}$  of less than 0.88.

Even though the  $k_{eff}$  for case I is high, the conditions leading to this result are incredible. The deposits were abnormally large (40 kg total) and nearly incredible given the assumed ER inventory of actinides (100 kg). The analysis also assumed a) 10 inches of cadmium in the ER (2 inches above the NOE) so that the entire deposits are immersed, and b) the incredible condition of deposits at theoretical density.

#### IV.B.3. Failure of the Liquid Cathode Assemblies

One configuration for liquid cathodes in the cadmium pool was analyzed. Each of two liquid cathodes contained 14.0 kg of actinides at Pu/U ratio of 6:1. Each liquid cathode also contained 30 kg of cadmium. Because the solubility limit of the 30 kg of cadmium in the liquid cathode was exceeded, the liquid cathode contents were modeled as follows. The bottom layer of the liquid cathode consisted of 2.0 kg of uranium. On top of this layer, 1.36 kg of plutonium was placed followed by 10.64 kg of plutonium as  $\text{PuCd}_6$ . All of the 30 kg of cadmium was consumed in forming the  $\text{PuCd}_6$ . The computed  $k_{\text{eff}}$  for this case is less than 0.59.

#### IV.C. Uranium Deposition on the ER Sidewalls

When uranium is being electrorefined first to the cadmium pool and then to the solid cathode, some of the uranium deposits on the side-wall of the ER. This secondary reaction is possible only during the first step when the fuel dissolution baskets are the anode and the cadmium pool is the cathode. This observation results from the ER vessel being an electronic conductor in contact with the cadmium pool, and its being basically at the same potential as the cadmium pool. Consequently, uranium that is electrotransporting to the cadmium pool can take an alternate route to deposit on the ER sidewall as well as in the cadmium because they are at the same potential. (This secondary reaction is not possible during direct transport operation, without an electrical short circuit between the vessel and solid cathode, since the vessel and cadmium pool are electrically isolated from the fuel dissolution baskets (anode) and the solid cathode.)

Note that only uranium can deposit on the ER sidewall for the same reason that only uranium deposits on the solid cathode: plutonium metal is unstable in the presence of  $\text{UCl}_3$  in the salt phase. In practice, any uranium that deposits on the ER sidewall will form dendrites as it does on the solid cathode at nominally the same theoretical density (about 5%). It will be distributed on the ER sidewall near the fuel dissolution baskets because this provides the path of least resistance (the shortest path) through the salt.

Uranium on the ER sidewall is subsequently recovered when a solid cathode deposit is taken with the cadmium pool as the anode. Since the cadmium pool is basically at the same potential as the ER, the uranium on the ER sidewall, as well as the uranium in the cadmium pool, will anodically dissolve and transport to the solid cathode.

The case of reverse polarity bounds the situation where uranium deposits on the ER sidewall because of their similar configurations. By comparison, reverse polarity has uranium deposit directly on the FDBs-where anodic dissolution operations have uranium deposit on the ER sidewall in the vicinity of the FDBs. For reverse polarity the deposit will be concentric with the anode assemblies, while anodic dissolution operations will yield a deposit more spread out. Reverse polarity also assumes that 15 kg deposits are taken on each anode assembly, but anodic dissolution operations result in only a fraction of the actinides depositing on the ER sidewall with the remainder transported to the cadmium pool [1]. In the comparison of uranium deposition on the ER side-walls and reverse polarity operation of the ER, the similarity of the mass and geometry features demonstrates the criticality analysis for the cases of reverse polarity bounds the case for uranium deposition on the ER sidewall. Hence, criticality safety is assured for the secondary electrorefining reaction involving uranium deposition on the ER sidewall.

#### V. CONCLUSION

The literature on the criticality safety strategy for the FCF ER has been summarized. The ER criticality safety strategy accounts for the reference conditions and the NOEs by establishing the methodology for maintaining the actinide levels within their NOEs. Process controls will limit the extent of an operation, and operational checks will verify that an operation has performed as expected. The process control parameters and operational checks will be specified on the Electro-refiner Process Data Sheet by a process engineer and approved by a peer reviewer. Provided operational checks are satisfied, pre-approved follow-on operations may continue. In the event an operational check is not satisfied, operations will be suspended until the discrepancy is resolved.



The ER criticality safety strategy accounts for credible failure modes (such as hardware, operator, etc.) by combining two or more unlikely and extremely unlikely events. The credible failure modes were found to be safe toward criticality. In addition, some incredible failure modes were considered and shown to be safe toward criticality. A handful of incredible failure modes were found to be marginally subcritical. For those situations that gave rise to analyses yielding  $k_{\text{eff}}$  greater than 0.90, the reasons that make these failure modes incredible were stated. The remoteness of the required combinations of unlikely and extremely unlikely events that were necessary to produce the incredible scenarios that are marginally subcritical demonstrates nonetheless that overall the ER is safe toward criticality for the strategy adopted.

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Table I. Reference States and Normal Operations Envelopes  
for Routine Operation of the Electrorefiner.

| Description            | Units   | Reference State | Normal Operations Envelope |
|------------------------|---------|-----------------|----------------------------|
| <b>Salt Phase</b>      |         |                 |                            |
| HM Concentration       | weight% | 7               | 9.5                        |
| HM Content             | kg      | 34              | 46                         |
| Pu:U Ratio             |         | 2 to 1          | 6 to 1                     |
| <b>Cadmium Phase</b>   |         |                 |                            |
| Soluble HM             | kg      | 20              | 40.5                       |
| Hold-up HM             | kg      | 7               | 12                         |
| <b>ER HM Inventory</b> |         |                 |                            |
| HM Content             | kg      | 61              | 82                         |
| <b>Anode Baskets</b>   |         |                 |                            |
| HM Content/batch       | kg      | 20              | 30                         |
| HM Content/anode       | kg      | 10              | 15                         |
| HM Content/basket      | kg      | 2.5             | 3.75                       |
| Void fraction          |         | 0.43            | 0.39                       |
| <b>Solid Cathode</b>   |         |                 |                            |
| HM Content/batch       | kg      | 20              | 27.8                       |
| HM Content/cathode     | kg      | 10              | 15                         |
| Pu:U ratio             |         | 1 to 100        | 1 to 6                     |
| Void Fraction          |         | 0.96            | 0.60                       |
| <b>Liquid Cathode</b>  |         |                 |                            |
| Pu, U Content/batch    | kg      | 5.6 Pu, 1.2 U   | 11.2 Pu, 2.4 U             |
| Pu, U Content/cathode  | kg      | 2.8 Pu, 0.6 U   | 5.6 Pu, 1.2 U              |

TABLE II. Process Control Parameters and Operational Checks for Assuring Normal Operations

| Process Operation                                         | Process Control Parameter                                                                   | Operational Check                                           |
|-----------------------------------------------------------|---------------------------------------------------------------------------------------------|-------------------------------------------------------------|
| Electrorefiner Inventory                                  | Input and Output Weights                                                                    | Sample Analysis<br>Level Measurements                       |
| Fuel Dissolution Baskets with Fuel Segments               | Number of Elements<br>Chipped Fuel Element Length<br>Element Composition                    | Net Weight<br>Sample Analysis                               |
| Fuel Dissolution Baskets with Actinides in Any Other Form | Two Step Weighing Operation                                                                 | Net Weight<br>Sample Analysis                               |
| Solid Cathode                                             | Total Integrated Current<br>Cathode Configuration<br>Power Required to Rotate Solid Cathode | Net Weight<br>Sample Analysis of Cathode Processor Products |
| Liquid Cathode                                            | Total Integrated Current<br>Power Required to Rotate Liquid Cathode Impeller                | Net Weight<br>Sample Analysis of Cathode Processor Products |
| Chemical Cathode                                          | Weight of Alloy Reductant<br>Number of Reductant Ingots<br>Composition of Reductant Ingots  | Net Weight<br>Sample Analysis of Cathode Processor Products |

# END

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