ANL/FC/CP-- 79/15 Conf-930907--10

Criticality Safety Evaluation of the Fuel Cycle Facility Electrorefiner

R.M. Lell, R. D. Mariani, E.K. Fujita, R.W. Benedict, R.B. Turski

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

SEP 07 1533

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free ficense to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes

MASTER

de

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

1.0 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-loop fuel cycle. Some of the primary advantages are passive safety for the reactor and resistance to diversion for the heavy metal in the fuel cycle. In addition, the IFR pyroprocess recycles all the long-lived 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 in Idaho Falls, Idaho is being modified to recycle spent metallic fuel from EBR-II (Experimental Breeder Reactor II) as part of a demonstration project sponsored by the Department of Energy. A key component in the FCF recycling process is the electrorefiner (ER) in which the actinides are separated from the fission products. In the process, the metal fuel is electrochemically dissolved into a high-temperature molten salt, and electrorefined uranium or uranium/plutonium products are deposited at cathodes.

This report addresses the new and innovative aspects of the criticality analysis ensuing from processing metallic fuel, rather than metal oxide fuel, and from processing the spent fuel in batch operations. In particular, the criticality analysis employed a mechanistic approach as opposed to a probabilistic one. A probabilistic approach was unsuitable because of a lack of operational experience with some of the processes, rendering the estimation of accident event risk factors difficult. The criticality analysis also incorporated the uncertainties in heavy metal content attending the process items by defining normal operations envelopes (NOEs) for key process parameters. The goal was to show that reasonable process uncertainties would be demonstrably safe toward criticality for continuous batch operations provided the key process parameters stayed within their NOEs. Consequently the NOEs became the point of departure for accident events in the criticality analysis.

1.1 Process Description

The electrorefiner (ER) is a steel vessel 40 inches in diameter and 31 inches high. Normally it will contain a cadmium pool at the bottom that is about 6 inches deep (Fig. 1). Above the

cadmium pool will be a 12 inch layer of molten salt initially containing 44 wt% LiCl and 56 wt% KCl (the eutectic composition). The salt phase will be loaded to about 7 wt% actinides; and as spent fuel is processed, fission products will accumulate in the salt phase or in the cadmium pool. The ER is the heart of the refining process because the electrodeposited metal is considerably separated from the fission products in the ER.

The separation of the actinides from the fission products results from some fission products being more active metals than the actinides and some fission products being less active, or more noble, than the actinides (Ref. 1). The metals that are more active than the actinides will partition mostly or exclusively into the salt phase as the corresponding metal chlorides. The metals that are less active, or more noble, than the actinides will partition into the cadmium pool.

In order to get the spent metal fuel into the electrorefiner, the fuel elements are chopped into segments and loaded into fuel dissolution baskets. After positioning the baskets in the salt phase, the spent fuel is electrochemically dissolved by connecting the anode baskets to the positive terminal of a power supply. Simultaneously the negative terminal of the power supply is connected to the cathode where the actinides will be electrochemically deposited. Since the actinides are deposited at the same rate as they are dissolved, the actinide concentration in the salt phase is effectively unaltered and serves only to support the electrorefining process. Three different cathodes for depositing the actinides are possible depending on the electrorefining scheme and on the actinides desired in the deposit.

Two of the cathodes possible for use are the solid cathode and liquid cathode. The solid cathode is used when uranium is the desired product, and the liquid cathode is used when a plutonium/uranium mixture with the minor actinides is the desired product. Since uranium is a significantly less active metal than plutonium, uranium will electrochemically deposit on the solid cathode without plutonium. Plutonium metal is not stable in the presence of U+3 in the salt phase. If plutonium metal did deposit, the U+3 in the salt would exchange with it. Plutonium metal will deposit on the solid cathode only if the U+3 concentration in the salt is sufficiently low or the Pu+3/U+3 ratio is large enough (Ref 2).

The liquid cathode is effective for taking mixed deposits of plutonium and uranium with the minor actinides because the liquid cathode contains liquid cadmium. At 500 C, plutonium and the minor actinides form compounds with cadmium when they reach their solubility limits in cadmium (Ref. 1). On the other hand, uranium does not form a compound with cadmium when it reaches its solubility limit; instead, uranium precipitates as the pure metal. These plutonium and minor actinide compounds represent a more stable thermodynamic state than the pure metals (otherwise the pure metals would precipitate), and this increased stability renders them less active as metals. In fact, plutonium dissolved in cadmium is only slightly more active than uranium dissolved in cadmium, thus enabling the co-deposition of uranium and plutonium. Furthermore, a similar situation exists for the minor actinides, enabling their co-deposition with uranium in the liquid cadmium cathode.

Two other features of the liquid cathode are worth noting. As discussed, the minor actinides are expected to behave similarly to plutonium. Consequently, the minor actinides can be burned within the refined fuel rather than isolated as TRU waste. Additionally, the rare earth metal fission products form compounds with cadmium that are thermodynamically stable enough to allow some of the rare earths to partition into the liquid cathodes (Ref. 1). The presence of these rare earth fission products in the liquid cathode products renders the plutonium products resistant to diversion.

The third possible cathode is the cadmium pool at the bottom of the electrorefiner. In some schemes for electrorefining, the cadmium pool may be used in a two-step operation in which the spent fuel is refined first from the fuel dissolution baskets to the cadmium pool, and second from the cadmium pool to the solid or liquid cathode. While this scheme is more time-consuming than refining the spent fuel directly from the fuel dissolution baskets to the solid or liquid cathode, it may offer some advantages.

Up to this point, four primary forms for the actinides in the electrorefiner have been identified: actinide metal in the fuel dissolution baskets, actinide chlorides dissolved in the salt

phase, actinide metals dissolved in the cadmium pool, and actinide metals deposited in the solid or liquid cathode. In addition, two secondary forms for actinides are significant. If any of the actinides exceed their solubilities in the cadmium pool at the bottom of the ER, then they will precipitate as the appropriate cadmium compounds or as the pure metal in the case of uranium. The other secondary form of actinides is actinide oxides. Actinide oxides can form upon reaction with impurities such as water and oxygen in the ER environment. Because the density of the actinide oxides will be considerably less than theoretical, they will locate somewhere above the cadmium phase, either as floccules in the salt phase or as a precipitate near the bottom of the salt phase. The criticality analysis and the NOEs for the ER included these forms of actinides.

2.0 Mechanistic Approach to Criticality Accidents for the ER

A number of factors prompted the use of a mechanistic approach, as opposed to a probabilistic one, for assessing the criticality safety of the ER. For one thing, the electrorefining processes for irradiated fuel are novel. Secondly, experience with engineering-scale electrorefiner operations spans only a couple of years, and that experience was with depleted uranium, not with irradiated fuel. A lack of experience also exists for the FCF in connection with ER operations. Finally, some of the process uncertainties are not well-defined. These factors contributed to making probabilistic risk assessment numbers very uncertain, and led to the use of a mechanistic accident formalism rather than a probabilistic one.

The mechanisms for criticality accidents for the ER were developed in the context of credible accident configurations that involved some combination of abnormal events. Abnormal events were divided into five categories such as mass and density, and were further distinguished as unlikely or extremely unlikely events. A contingency matrix was then formed in order to combine abnormal events from the different categories. At least one unlikely event from one category was combined with an extremely unlikely event from a different category, and a particular combination of abnormal events defined an accident configuration. Efforts then were directed toward identifying similar accident configurations so that the criticality analysis could be simplified by addressing the bounding cases.

The abnormal event categories are 1) mass, 2) material, 3) density, 4) reflection and 5) geometry. Abnormal mass events include inventory errors, weighing errors, or inadvertent hold-up accumulation. An abnormal material event would involve substitution/misidentification of a given item or an inopportune transfer. If the density of an item is outside of an acceptable range, an abnormal density event results. Abnormal events in reflection amount to changes in reflection in and around the ER. Abnormal reflection events could arise from equipment or facility failures or from the design basis earthquake. Should fissile material be rearranged into unanticipated or non-standard configurations, an abnormal geometry event would occur.

3.0 Reference Conditions and Normal Operations Envelopes in the FCF ER

In order to show criticality safety for the FCF ER, the reference conditions for several key ER process parameters had to be defined with respect to the quantity of actinides in the ER, their possible forms, and their specific locations. These forms and locations have been previously discussed in the process description. A summary of these actinide forms, their locations, and their amounts is contained in Table 1.

The reference conditions for the key process parameters were established in view of equipment limitations and prior experience. For example the ports in the top of the ER limit the sizes of process items that can be inserted and removed. Consequently, the diameter of a uranium deposit that can be withdrawn from the ER is limited to about 10 inches. Experimental results with an engineering-scale ER were extensively used to define the reference conditions. For example, setting the actinide concentration in the salt phase at 7 wt% was strongly influenced by the past performance of the process at this concentration.

Since the process is still at the research and development stage, NOEs were then defined (Table 1) that bracket the key process parameters and allow latitude for process development. Batch throughput economics obviously made it desirable to define some of the NOEs to be as large as possible. For example, a denser uranium deposit is consistent with less salt entrainment and a simpler retort operation for the final purification step.

Another concern in batch operations that can influence throughput is the number of sample and hold points required. A system of operational checks was devised that verified process items contained the expected amount of actinides within some uncertainty level. With the operational checks satisfied, each of the possible forms of actinides would be verified to be within their NOEs. Consequently, the need for sample and hold points would be reduced.

An example of an operational check would be weighing a uranium deposit on a solid cathode. The uncertainty about the uranium content stems from the amount of entrained salt. This uncertainty is used to infer the maximum amounts of actinides removed on the deposit and the maximum left behind in the ER. Since planning successive batch operations assumes the process item is within its uncertainty level, the next operation, regardless of its nature, is coordinated so that it does not compromise the NOEs. Consequently, sample analysis results for the salt and cadmium phases of the ER are not required prior to performing the next batch operation as long as the weight of the solid cathode is within its uncertainty level (i.e., the operational check is satisfied).

The process deviations and statistical fluctuations in the sample and analysis results for the engineering-scale ER also figured prominently in the development of the NOEs. When possible, a 2-sigma uncertainty was derived in order that the majority of operations do not invoke sample and hold points. Otherwise, uncertainties represent conservative estimates that often reflect a maximum deviation rather than a multiple of sigma. As data becomes available for the estimated uncertainties, they will be included in refining the uncertainty levels of the operational checks. It is expected that the uncertainties will be reduced, and could permit fewer sample and hold points. These NOE values subsequently provided the point of departure for defining abnormal events.

4.0 Calculational Methodology and Validation

The criticality safety evaluation of the electrorefiner is based on calculations using the KENO V.a code (Ref. 3). For those calculations, a standard 27 energy group neutron cross section library based on ENDF/B-IV data was used (Ref. 4). KENO V.a and the chosen cross section

library were validated for this purpose by using them to compute k for several sets of critical experiments and by comparison of KENO V.a with VIM (Ref. 5), a continuous energy Monte Carlo code with ENDF/B-V cross sections, for a number of electrorefiner configurations. The validation is necessary to demonstrate that KENO V.a and the chosen cross section library correctly predict k for appropriate configurations and spectra and to determine any calculational bias.

For the first series of calculations, nine experimental critical assemblies were selected from the literature (Ref. 6). These assemblies were spheres or cylinders of either plutonium or highly-enriched uranium with graphite reflectors. These criticals were selected because they had neutron spectra and configurations that are representative of processes and equipment in FCF, e.g., crucibles in the electrorefiner. KENO V.a was used to compute the neutron multiplication factors for these criticals. The minimum and maximum computed values of k for this series were 0.99381 and 1.00763 respectively. The average computed k for these nine cases was 1.00203.

For the second series of calculations, fifteen additional criticals were selected for analysis. Eleven of these configurations consisted of a sphere of plutonium or highly-enriched uranium reflected by uranium, iron, or copper. Two configurations consisted of alternating disks of plutonium and stainless steel or depleted uranium with a depleted uranium reflector. The final two configurations consisted of alternating plates of highly-enriched uranium and natural uranium. These assemblies were chosen because their configurations and neutron spectra are representative of various limiting cases identified during the preliminary criticality analysis of the electrorefiner. The minimum and maximum computed values of k for this series were 0.99441 and 1.01619 respectively. The average computed k for these fifteen criticals was 1.00515.

A series of benchmark critical experiments was carried out for several mixes of IFR fuel materials in the Zero Power Physics Reactor. These configurations consisted of mixtures of uranium, plutonium, stainless steel, zirconium, and void surrounded by a graphite reflector. Starting with a plutonium core, six critical configurations were constructed by systematically substituting enriched uranium for plutonium on the basis of matching reactivity. These assemblies

are representative of the fuel enrichments, diluent materials, and process equipment in FCF processes. For the six cores calculated with KENO V.a, the average C/E was 1.00960.

The fourth set of calculations tested the ability of KENO V.a to correctly predict k for epithermal and thermal spectra. Nine critical experiments were analyzed for this series. Three experiments consisted of uranium metal reflected by water or polyethylene. The other six configurations consisted of solutions of uranium or plutonium in water. These cases are relevant to the possible presence of limited amounts of moderators in various places in the facility. The minimum and maximum values of k for this series were 0.99759 and 1.01256 respectively. The average computed k for these cases is 1.00445.

To complete the validation of KENO V.a for this analysis, comparisons were made between KENO V.a and the VIM Monte Carlo code for several electrorefiner configurations identified during preliminary criticality analysis as representing the limiting cases for the electrorefiner analysis. This comparison procedure was necessary because lithium, cadmium, and chlcrine, important materials in the electrorefiner, are not commonly used in critical experiments in the configurations and spectra typical of the electrorefiner. In order to provide a meaningful test of the KENO V.a cross sections for these materials in the correct spectrum and configuration, KENO V.a and VIM were used to compute k for a number of hypothesized accident configurations in the electrorefiner. VIM and its cross section library are completely independent of KENO V.a and the cross sections used with KENO V.a for this analysis. VIM and KENO V.a generally agreed to within 0.3% k; the difference was 0.8% k in the worst case.

5.0 Criticality Analysis Methodology

The criterion adopted as the basis of the criticality analysis is that criticality shall not result from the concurrent occurrence of one unlikely event and one extremely unlikely event from two different classes of abnormal events described above.

The contingency matrix is a logical and systematic way to organize and present the possible combinations of unlikely and extremely unlikely events for a particular operation. A table which

shows all of the unlikely and extremely unlikely mass, material, density, reflection, and geometry events for a given operation is created in a matrix format. Use of this table provides assurance that no credible event combinations have been overlooked and demonstrates that, at the minimum, the nuclear criticality safety criterion has been met. In addition, when the values of k for the various event combinations are arranged in the matrix format, the important safety parameters are readily identified. This allows efforts to be concentrated on developing controls for the important factors that affect criticality safety.

There are five basic categories of unlikely events and five basic categories of extremely unlikely events. The unlikely event in each category is paired with the extremely unlikely event in each of the five categories, leading to a 5 x 5 matrix of possible combinations of one unlikely event and one extremely unlikely event. The event pairs for the same category, e.g., the combination of the unlikely and extremely unlikely density events, are eliminated for several reasons.

- Some pairs are self-contradictory. For example, it is not possible to have the unlikely
 density event and the extremely unlikely density event or the unlikely geometry event and the
 extremely unlikely geometry event in the same material or location simultaneously.
- 2. In some cases, the extremely unlikely event is actually a repeated occurrence of the unlikely event. For example, the unlikely material event during preparation of the fuel dissolution charge for the electrorefiner is substitution of one container of plutonium for one container of lower worth material while the corresponding extremely unlikely material event is substitution of two containers of plutonium for an equal mass of lower worth material during preparation of the fuel dissolution charge.
- 3. In some cases, the unlikely event in a category is either a subset of the extremely unlikely event or is clearly bounded by some other event combination. Changes in reflection and most abnormal mass events in the electrorefiner fall into this category.

4. In some cases, it is not credible for two events from the same category to occur within the limited scope of a single operation.

The NOE was taken as the basis for the definition and classification of unlikely and extremely unlikely events. Using the NOE as the basis for event classification is very conservative because the NOE values for electrorefiner parameters do not represent the normal operating conditions in the electrorefiner. The NOE values include large margins to account for process deviations, measurement uncertainties, etc.; the NOE parameter values are not ordinarily approached during routine operations.

There may be several paths to similar events or several very similar events in a category. It is important to define a single event with sufficient conservatism to cover all of the similar events and event paths if possible. This approach produces a conservative analysis, reduces the volume of required calculations, and eliminates the need to address every possible event path in conjunction with every other possible event from another category.

Because there is insufficient data for a meaningful probabilistic risk analysis, the actual definition and classification of abnormal events as unlikely or extremely unlikely was made on the basis of engineering judgement after considering the following factors for each operation.

- 1. The chemistry and physics of electrorefiner operation impose limits on possible events in the electrorefiner. For example, the chemistry and physics of the deposition process limit the composition of a solid cathode.
- 2. Equipment and container designs restrict the range of credible events. For example, there are vertical and horizontal scrapers in the electrorefiner which constrain the size and shape of the actinide deposit on a solid cathode. Similarly, the design of the fuel dissolution basket limits the shape and quantity of its contents.

- 3. Previous experience acquired during developmental work in an engineering-scale version of the electrorefiner provides an indication of the normal values and expected deviations for some process parameters, e.g., the bulk density of the deposit on a solid cathode.
- 4. The availability of real-time diagnostic information during operation of the electrorefiner allows an operation to be stopped as soon as an abnormal condition is detected. For example, the diagnostics would reveal an error in the electrical connections for an electrode assembly (reverse polarity event).
- 5. There are multiple administrative controls over each operation in the electrorefiner. These controls include independent peer review of the proposed operation, strict adherence to approved written procedures for the conduct of the operation, and inspection of materials and equipment prior to commencement of the operation.
- 6. Expected process variations and sampling and analysis uncertainties are factored into the specifications for and the conduct of each operation, e.g., proposed actinide additions to or collections in the electrorefiner.
- 7. Electrorefiner operations require multiple steps and/or long time periods which offer repeated opportunities to discover and rectify an abnormal situation.
- 8. The number of persons involved in the conduct and oversight of an operation, the number of procedural errors required for an event, and the number of equipment failures required for an event affect the likelihood of that event.

The resulting event definitions include very conservative safety margins to account for both the uncertainties in the basic electrorefiner processes and the uncertainties in the probabilities for the occurrence of the various events.

In general, the criticality analysis considered abnormal events with regard to mass, material, density, reflection, and geometry. These five event categories lead to a 5 x 5 matrix. For a given operation, the dimensions of this matrix may increase or decrease. The matrix dimensions will decrease if there are no credible events in a particular category. For example, a liquid cathode consists of a cadmium-filled crucible attached to supporting hardware. Actinides deposit in the liquid cathode as dissolved uranium and plutonium up to the solubility limits of the cadmium. Any additional uranium and plutonium deposit as uranium metal and PuCd6 respectively. All of these materials are incompressible, so there are no density events for a liquid cathode.

The dimensions of the matrix can increase if there is more than one possible abnormal event in a particular category. For example, a variety of different materials may be added to the electrorefiner to establish the required cadmium and electrolyte depths and the required actinide concentrations for normal operations. The range of materials added leads to a number of possible mass overloading and material substitution events during the initial preparation of the electrorefiner.

For a complex piece of equipment such as the electrorefiner, some operational errors may result in an event that can be placed in more than one category. For example, it is possible to reverse the polarity of the electrical connections during preparations for dissolution of fuel in the electrorefiner. If the polarity is reversed, the fuel dissolution baskets can function as solid cathodes and collect uranium. The reverse polarity event can be categorized as either a mass event or a material event.

There are a small number of special situations and events which do not readily fit into the contingency matrix formalism. Examples of such events are freezing of the electrorefiner due to a prolonged loss of heating and formation of actinide oxides due to a breach in the electrorefiner structure. The contingency matrix could be expanded to accommodate these events, but it proved more convenient to analyze such events as special cases.

6.0 Application of the Contingency Matrix Methodology

The contingency matrix methodology was applied to the electrorefiner criticality analysis. The entire electrorefiner criticality analysis is too long for inclusion here; the cathode deposition operation will be used to demonstrate the methodology.

Table 2 shows the events which define the contingency matrix for solid and liquid cathode deposition operations in the electrorefiner. The first column lists the values or specifications for mass, material, density, reflection, and geometry in the cathodes and electrorefiner at the NOE limit. The center column lists the values that correspond to the various unlikely events for the cathode deposition operation; the third column lists the parameter values for the extremely unlikely events. All event classifications were made on the basis of the number of physical and administrative barriers to the occurrence of the event.

The reference and NOE actinide contents of a solid cathode are 10.0 kg and 15.0 kg respectively; the reference and NOE actinide contents of a liquid cathode are 3.5 kg and 7.0 kg respectively. These values demonstrate the large margin between the nominal cathode depositions and the NOE limits. The unlikely and extremely unlikely mass events for a solid cathode are actinide depositions of 20.0 kg and 30.0 kg respectively. The unlikely and extremely unlikely mass events for a liquid cathode are actinide depositions of 10.5 kg and 14.0 kg respectively.

There are many barriers to the occurrence of abnormal mass events during the deposition process. Only the principal ones are summarized here. First, the total actinide deposition is limited by the amount of actinides in the chemically reduced state that are available for deposition. Second, the amount of actinides that can be collected by electrotransport is limited by the total charge (time-integrated current) applied to the cathode. Third, the electrorefiner diagnostics provide a real-time indication of the state of the deposition process. Fourth, horizontal and vertical scrapers in the electrorefiner constrain the size and shape of a solid cathode. Fifth, if a solid cathode grows too large, it will contact either the vessel wall or the cadmium. In either case, an electrical short circuit will occur, that will be obvious in real time. Sixth, there are a number of administrative and procedural controls prior to and during the deposition process.

In the reference case, the Pu/U ratio in a solid cathode deposit is 1:100 because of the chemical thermodynamics in the electrorefiner. At the NOE limit, the Pu/U ratio in a solid cathode is 1:6, principally because of PuCl₃ entrained in the actual deposit. For a liquid cathode at the NOE composition limit, the Pu/U ratio is 6:1. Because of the conservative NOE compositions, no unlikely material event was defined for the deposition process. The extremely unlikely material event for both types of cathode was defined to be collection of pure plutonium on the cathode rather than the appropriate actinide mixture.

There are three principal barriers to the occurrence of abnormal material events during the deposition process. First, the composition of the deposit is limited by the composition of the actinides in the chemically reduced state. The amount of reduced actinides in the electrorefiner determines the amount of actinides that can be collected by electrotransport. Second, plutonium will not deposit on a solid cathode to any significant degree unless the Pu/U ratio in the electrolyte far exceeds the envelope for normal electrorefiner operations. Third, collection of pure plutonium (or any composition beyond the NOE limit) requires violation of a number of administrative controls governing previous fuel dissolution and cathode deposition operations.

Previous experience with an engineering-scale version of the electrorefiner shows that the effective bulk density of a solid cathode deposit is approximately 1.0 kg/l which corresponds to an effective actinide volume fraction of 5%. The NOE value for the effective actinide volume fraction in the solid cathode deposit was set at 40% to allow for uncertainties in the process and for experiments to increase the solid cathode bulk density. The unlikely and extremely unlikely density events for a solid cathode were defined as achieving effective actinide volume fractions of 60% and 100% respectively in the solid cathode deposit. The densities assumed for the NOE, the unlikely density event, and the extremely unlikely density event far exceed any solid cathode density that has been achieved by this process. The design of the liquid cathode precludes density events in a liquid cathode.

The electrorefiner is designed to meet specific design criteria with regard to tipping, sliding, or other motions due to the design basis earthquake. The overhead cranes are designed to meet

similar criteria. These criteria limit the potential for changes in reflection around the electrorefiner. The unlikely and extremely unlikely reflection events shown in Table 2 are defined very conservatively to bound any credible change in reflection due to equipment motion, equipment failure, or the design basis earthquake.

The reference geometry for a liquid cathode is determined by the liquid cathode crucible. The reference geometry for a solid cathode is determined by the solid cathode mandrel (the bar on which the actinides deposit) and the horizontal and vertical scrapers in the electrorefiner. The unlikely and extremely unlikely geometry events for both types of cathodes were assumed to be formation of a spherical actinide deposit.

The assumed geometry events bound any credible cathode shape for several reasons. First, the liquid cathode crucible constrains the shape of the liquid cathode, and there is no physical mechanism which would cause the actinides to form such a shape inside this crucible. Second, the scrapers produce a cylindrical solid cathode as long as the solid cathode rotates. Failure of the rotation mechanism is detectable. Third, the actinides form a long, thin deposit along the length of the mandrel. Fourth, a short circuit would develop as soon as the solid cathode deposit contacts either the vessel wall or the cadmium which is readily observable.

The calculational model for the criticality analysis consisted of the electrorefiner with two solid cathodes and two liquid cathodes positioned in the electrolyte. This model is conservative because the normal nuber cathodes of either variety is two. The unlikely and extremely unlikely events were assumed to occur in both solid cathodes and both liquid cathodes simultaneously. This assumption increases the conservatism of the analysis and also reduces the required number of calculations.

Table 3 shows the computed results for the criticality analysis of the cathode deposition operation. In principle, the five unlikely events and the five extremely unlikely events lead to 25 event pairs. Elimination of event pairs in the same category reduces the total to 20. The total number of cases is further reduced because some events are not defined, e.g., the unlikely material

event. Finally, the number of cases was reduced by imposing the extremely unlikely reflection event for all cases. Using combinations of more than two unlikely and extremely unlikely events whenever possible increases the conservatism of the analysis and decreases the required number of calculations. The event combinations in Table 3 cover all possible combinations of events from Table 2.

Acknowledgements

Argonne National Laboratory is operated by the University of Chicago for the United States Department of Energy under Contract W-31-109-Eng-38.

7.0 References

- 1. J.P. Ackerman, "Chemical Basis for Pyrochemical Reprocessing of Nuclear Fuel," *I&EC Research*, 1991, 29, 141.
- 2. Z. Tomczuk, et al., "Uranium Transport to Solid Electrodes in Pyrochemical Reprocessing of Nuclear Fuel," *J. Electrochem. Soc.*, 1992, 139, 3523.
- 3. L. M. Petrie and N. F. Landers, "KENO V.a-An Improved Monte Carlo Criticality Program with Supergrouping," Oak Ridge National Laboratory Report NUREG/CR-0200, Volume 2, Section F11, ORNL/NUREG/CSD-2/V1/R2, December 1984.
- J. A. Bucholz et. al., "Standard Composition Library," Oak Ridge National Laboratory Report NUREG/CR-0200, Volume 3, Section M8, ORNL/NUREG/CSD-2/V3/R2, December 1984.
- 5. R. N. Blomquist, "VIM." Proceedings of Advances in Mathematics, Computation, and Reactor Physics, Pittsburgh, PA, April 1991.
- 6. H. C. Paxton, "Los Alamos Critical-Mass Data," Los Alamos National Laboratory Report LA-3067-MS, Rev. 1, December 1975.

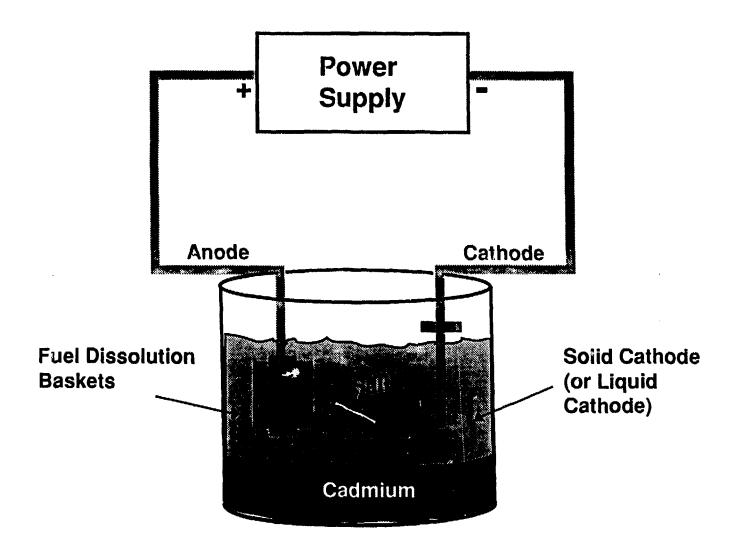


Figure 1. Schematic of the Fuel Cycle Facility Electrorefiner

Table 1. Reference States and Normal Operations Envelopes

for Routine Operation of the Electrorefiner.

			Normal
		Reference	Operations
Description	Units	State	Envelope
Salt Phase			
HM Concentration	weight%	7	9.5
HM Content	kg	34	46
Pu:U Ratio		2 to 1	6 to 1
Cadmium Phase			
Soluble HM	kg	20	40.5
Hold-up HM	kg	7	12
ER HM Inventory			
HM Content	kg	61	82
Anode Baskets			
HM Content/batch	kg	20	30
HM Content/anode	kg	10	15
HM Content/basket	kg	2.5	3.75
/oid fraction		0.43	0.39
		0.43	
Solid Cathode			0.39
Solid Cathode IM Content/batch	kg	20	27.8
Solid Cathode HM Content/batch HM Content/cathode	kg kg	20	0.39 27.8 15
M Content/batch M Content/cathode		20 10 1 to 100	27.8 15 1 to 6
M Content/batch IM Content/cathode		20	0.39 27.8 15
Solid Cathode HM Content/batch HM Content/cathode		20 10 1 to 100	27.8 15 1 to 6
M Content/batch M Content/cathode Pu:U ratio		20 10 1 to 100	27.8 15 1 to 6

Table 2 Definition of Events for the Contingency Matrix for Solid and liquid Cathodes in the Electrorefiner in CHC Zone P4

NOE/Reference	Unlikely	Extremely Unlikely
1 - Mass LC - 7.0 kg/ HM- 6.0 kg Pu, 1.0 kg U	10.5 kg HM- 9.0 Kg Pu, 1.5 kg U	14.0 kg HM- 12.0 kg Pu, 2.0 kg U
SC - 15.0 kg HM- 2.14 kg Pu, 12.86 kg U	20.0 kg HM- 2.86 kg Pu, 17.14 kg U	30.0 kg HM- 4.28 kg Pu, 25.72 kg U
2 - Material LC - Pu/U = 6:1	-	All Pu in LC
SC - Pu/U = 1:6	-	All Pu in SC
3 - Density ER at 500 °C VF - 40% in SC	ER at 20 °C, VF = 60% in SC	ER at 20 °C, VF - 100% in SC
4 - Reflection	Concrete base, 76 mm SS304 on two sides	Concrete on three sides
5 - Geometry LC - ternary crucible(s) in electrolyte	EAs form spheres	EAs form spheres
SC - cathode in electrolyte - cylindrical deposit-H/D = 1	Spherical deposit(s)	Spherical deposit(s)

1) LC - liquid cathode

SC - solid cathode

ER - electrorefiner

HM - actinides (heavy metal)

VF - volume fraction or packing fraction

SS304 - Type 304 stainless steel

2) Plutonium isotopics: 94.0 w/o Pu-239

5.5 w/o Pu-240

0.4 w/o Pu-241

Uranium isotopics: 78.0 w/o U-235 22.0 w/o U-238

0.1 w/o Pu-242

Table 3 Criticality Analysis for the Cathode Deposition Operation				
Case	Event Combination	k		
01	R1 x R2 x R3 x E4 x R5	0.49926 +/- 0.00138		
02	E1 x R2 x U3 x E4 x R5	0.75382 +/- 0.00383		
03	E1 x R2 x R3 x E4 x U5	0.63238 +/- 0.00174		
()4	U1 x E2 x R3 x E4 x R5	0.85636 +/- 0.00426		
05	R1 x E2 x U3 x E4 x R5	0.95120 +/- 0.00237		
06	R1 x E2 x R3 x E4 x U5	0.77901 +/- 0.00236		
07	U1 x R2 x E3 x E4 x R5	0.84417 +/- 0.00231		
08	R1 x R2 x E3 x E4 x U5	0.77040 +/- 0.00185		
()9	U1 x R2 x R3 x E4 x E5	0.55649 +/- 0.00162		
10	R1 x R2 x U3 x E4 x E5	0.60845 +/- 0.00182		

- 1) R Reference or NOE
 - U Unlikely Event
 - E Extremely Unlikely Event
 - 1 Mass Event
 - 2 Material Event
 - 3 Density Event
 - 4 Reflection Event
 - 5 Geometry Event
- 2) Each event is described by the combination of the letter R, U, or E with the integer 1, 2, 3, 4, or 5. For example, U1 describes the unlikely mass event from Table 1, and E2 describes the extremely—unlikely material event from Table 2.