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ABSTRACT

Spent fuel from the Experimental Breeder Reactor-II (EBR-II) is treated at the Fuel Conditioning Facility (FCF) located at Argonne National Laboratory-West (ANL-W). A number of sensitive nuclear material streams are present in the facility, which mainly include material in the input spent fuel, the element chopper, the electrorefiner, the cathode processor, and the casting furnace. Different types of measurements and calculations are performed in order to estimate the amount of material in those streams. There are three basic types of measurements: weight, elemental and isotopic fractions, and liquid level measurements. Materials in some streams are determined using validated models, which include the mass and composition of the input spent fuel. This paper describes the different streams of sensitive nuclear material for the facility, and the methods for their measurements or estimation as part of the MC&A system at FCF. It also describes the analytical and sampling uncertainties associated with those measurements and estimates, which are required for the facility variance propagation calculations.

INTRODUCTION

The application of electrometallurgical treatment processes to spent fuel from the Experimental Breeder Reactor-II (EBR-II) at Argonne National Laboratory-W Fuel Conditioning Facility (FCF) is approaching the end of its demonstration phase.[1] The treatment is based on electrorefining the fuel in molten-salt electrolyte. The following summarizes the different treatment processes.

The spent fuel assemblies are transferred from the reactor to FCF where they are processed in a vertical assembly dismantler (VAD) in order to separate the irradiated fuel elements from the assembly hardware. The fuel elements are chopped into 0.6 cm segments at an element chopper (EC). The fuel-containing segments are collected in fuel dissolution baskets (FDB's). The loaded baskets are transferred to the Mk-IV electrorefiner (ER) where the electrochemical process is used to separate uranium from fission products. The ER contains both salt and cadmium phases. The salt phase, which rests on top of a liquid cadmium phase, is a molten LiCl-KCl phase that contains a quantity of dissolved UCl_3. During the electrorefining, the

uranium in the chopped segments in the FDB's (anode) is oxidized, and uranium is collected by reduction onto a steel mandrel cathode. Most of the fission products and plutonium in the fuel remain in the salt phase. When the cathode is removed from the ER, salt and possibly cadmium adheres to it. The uranium collected on the cathode is transferred to a cathode processor (CP) where the adhering salt is distilled off and the remaining uranium metal is consolidated into ingots. To remain within the MC&A classification for the facility, depleted uranium is added at the CP to produce an ingot that does not exceed 50% ^{235}U enrichment. The distilled salt is returned back to the ER while the uranium ingots are transferred to a casting furnace (CF) in order to generate low-enriched uranium products and determine their composition. In the casting furnace, appropriate quantities of depleted uranium are added to blend down the enrichment of the final product to less than 20% ^{235}U. The casting batch constituents are melted together to form a homogeneous alloy, and a portion of the molten metal is cast into quartz molds to form sample pins that are chemically analyzed for composition.

As discussed above, various material streams exist in the facility, and some of those streams contain significant amounts of sensitive nuclear material (SNM). Consequently, accurate estimation of SNM in those streams is important, not only for operations but also for material control and accountability (MC&A). One of the success criteria [(2)] for the project is the demonstration of a MC&A system that shows the historical inventory difference for uranium and plutonium are within control limits based on variance propagation of measurement and sampling error, as specified in DOE order 5633.3B.[(3)]

The following sections identify the different streams of SNM, whether it is directly measured (ER salt and cadmium, cladding hulls, and cast furnace ingot), or estimated based on models (input uranium and ER holdup). The different types of uncertainties associated with SNM in these streams, which are needed for the MC&A variance propagation requirements, are also discussed. These types of uncertainties include measurement, model, sampling, and pseudo-instrument uncertainties. Activities related to the estimation of these uncertainties are also included.

MEASURED STREAMS

Weight, liquid level, and sample content measurements are systematically used around the facility to determine the amount of SNM associated with a number of material streams. Those streams include the electrorefiner salt and cadmium, the cladding hulls, and the low-enriched uranium product from the casting furnace.

The ER tank contains an upper fluid layer of a mixture of LiCl-KCl salt and a lower fluid layer of liquid cadmium. The mass of SNM in the ER is determined from the product of the mass of each fluid layer and the weight fraction of SNM in each layer. The salt and cadmium masses are determined from estimates of their volumes and densities. The weight fractions of components in the fluids are determined by chemical analysis of two salt samples and two cadmium samples taken from the ER. Both the weight fractions and isotopic fractions of uranium and plutonium are measured. The ER fluid volume is calibrated against the liquid level measurement at the ER operational temperature.[(4)] A correlation determines the volume for a given level measurement.

The densities of the salt and the cadmium are calculated by the method of additive volumes. This method assumes that the mixture volume is a weighted-average of the volumes of the constituent components. In this case, a mixture density is a function of the individual component densities and their weight fractions (the densities obtained by calculation and by measurement agreed within 0.1%). This leads to salt and cadmium densities that are determined as a function of the ER operating temperature, the individual densities of the pure components, and the weight fractions of each.[4] The weight fractions of components are determined by chemical analysis of the salt and cadmium samples. Multiplying the SNM fractions in the salt (or cadmium) by the salt (or cadmium) mass determines its content of SNM.

At the EC, irradiated fuel elements are chopped into segments, and placed in fuel dissolution baskets (FDB's). After electrorefining, cladding hulls segments remain in the baskets (one anode consists of four FDB's). After removal from the ER, the cladding hulls become part of the element hardware waste stream. However, this stream contains a relatively small amount of uranium that is attributed to two sources: incomplete dissolution of uranium in the fuel and UCl_3 in the adhered salt matrix. The amount of uranium in this stream is determined through weight measurement of the individual FDB's that contain the hulls, estimate of the mass of the adhering salt, and chemical analysis of cladding hulls and ER salt samples. A hull sample is taken from each FDB (after mixing the hulls), where each sample consists of 10 hull pieces.

A number of streams of in-process material, such as, the ER cathode deposit and cathode processor ingot, are weighed but not sampled for MC&A purposes. The compositions of those streams are determined when chemical analysis results for the casting furnace samples become available. Other streams that are determined from weight measurements are the CP holdup (assumed to be ER salt) and the CP dross (the dross is a product of the reaction between the cathode material and the processing crucible coating).

A casting furnace batch consists of the ingot produced at the cathode processor with added depleted uranium in order to reduce the enrichment of the final product to less than 20%. The casting furnace product is homogeneous and the pin samples produced at this stage are representative of the product. The sample analysis results combined with weight measurement of final product provide measurement-based estimates of SNM in this stream.

ESTIMATED STREAMS

There are no direct measurements of the mass and composition of the input irradiated fuel as it enters the facility. The estimates of the mass and composition of the fuel are determined from the fresh fuel specifications combined with burnup calculations. Validated computer codes and methods calculate the change in the as-fabricated composition due to reactor burnup.[5] The calculated values are verified by measurements performed on samples obtained for each batch (an EC batch consists of four subassemblies) at the element chopper. When the measured data become available, the measured and calculated data are compared to determine that both are consistent. Details of the comparison and correlation between the measured and calculated values have been described.[6]

Holdup in the ER is presently modeled as uranium in the form of oxide; this holdup is not measured by sampling. The increment in holdup is assumed to be proportional to the uranium input to the ER. Based on conservation of uranium mass in the initial conditioning of the salt with depleted uranium, the holdup coefficient is estimated. This coefficient is being evaluated using mass balances over the ER during subsequent depleted and irradiated uranium operations.

MEASUREMENT UNCERTAINTY

There are three basic types of measurements that are performed routinely in FCF. These include weighing process material in containers, measuring levels of the ER liquids, and analyzing samples for elemental and isotopic fractions.

Weighing measurement uncertainties are estimated from periodic characterization and daily linearity checks of each in-cell mass balance.[7] Random and systematic uncertainties range between 0.3 and 7.5 gm, and between 0.1 and 5.8 gm, respectively, depending on the maximum capacity and operating environment of the balance. During each calibration period, uncertainties are recorded for each balance.

Level measurements of the salt and cadmium in the ER determine the volumes of the two fluids. Combined with the density of each fluid, the salt and cadmium masses are determined. As discussed previously, the volume calibration of the ER vessel prior to operations established the relationship between liquid level and volume at the operational temperature and provided random and systematic volume uncertainties of 0.34 and 0.40 liters, respectively. The salt and cadmium densities are calculated using additive volumes, which assumes that the specific volumes of the individual components in a solution are constant; that is, the liquid density is a function of the weight fractions and densities of the individual components. Variance propagation of the weight fraction and density errors of the individual components yield estimated uncertainties of approximately 1% for the densities of both the salt and cadmium.

The analysis of samples for elemental and isotopic fractions of uranium and plutonium is performed with either isotope dilution mass spectrometry or alpha spectrometry. The majority of measurements for MC&A employ isotope dilution mass spectrometry; the analytical laboratory estimates the one standard deviation uncertainty to be 0.5%. If the plutonium is present only in trace amounts, it is measured using alpha spectrometry, which has an uncertainty of 25%. This is the case for estimating plutonium in the cadmium pool and the low-enriched uranium product.

MODEL UNCERTAINTY

The uncertainties for the components in the fuel are determined from the uncertainty in the mass and composition of the as-fabricated fuel before irradiation in EBR-II and from the accuracy of the neutronics code calculations that determine the change in composition with burnup. The mass error in the fresh fuel is less than several tenths of gram and the composition error is about 1%. The uranium uncertainties for fuel input to the process are dominated by the error in the fabricated fuel mass and composition. The estimated overall uncertainty for uranium in the spent fuel is 1.1%.[5] On the other hand, the error in the plutonium estimates is determined by the

errors in the neutronics codes calculations alone, since there is no plutonium in the fresh fuel. Comparisons of calculated and measured plutonium in irradiated fuel segments have indicated a random error of about 4.3%. In addition to the random error, the comparisons have established a calculations bias of +6.4%.[8] Differences between calculated and measured values for each fuel assembly may lead to adjustments of the calculated inventory files. However, no adjustment was necessary during the demonstration program due to the close agreement between these values.

As mentioned before, the ER holdup is estimated through a holdup model that is based on accumulating information about the electrorefiner mass balance during operations. The difference between the measurable ER inventory at the beginning of a batch and the end of a batch (measurable inventory difference) is related to the nonmeasurable holdup in the ER. Analysis of the ER inventory difference data provides preliminary estimate of the uncertainty associated with the ER holdup.

SAMPLING UNCERTAINTY

Errors arise because of inhomogeneities in sampled materials; the inability to obtain a representative sample is expressed as sampling uncertainty. Most of the significant streams in FCF have been demonstrated to be homogeneous and, therefore, require no sampling errors. However, two streams, the ER cadmium and the waste cladding hulls, are extremely inhomogeneous.

An experiment to quantify the inhomogeneity in the cadmium has established a random sampling uncertainty of 13.2%. Significant quantities of uranium or plutonium in the cadmium for an accountancy inventory period can cause this sampling error to be a dominant source of uncertainty in the inventory difference.

To ensure a representative sampling of cladding hulls, a procedure was implemented for mixing and sampling; the resulting sampling uncertainty was determined to be about 30%. If irradiated fuel in the anode baskets is not adequately dissolved (e.g., only 80% of the uranium is dissolved), this error will be an important contribution to inventory difference uncertainty. However, the average electrochemical dissolution of uranium is approximately 96%.

The impact of the large sampling uncertainties on the inventory difference uncertainty can be reduced in the future. For example, performing the cadmium sampling at the points where there are minimal amounts of SNM in the cadmium, will sharply reduce the impact of the cadmium sampling error on the inventory difference uncertainty. Also, in the future, when the cladding hulls are processed in the cathode processor into a metal waste ingot, the sampling errors will be eliminated since the complete batch will be processed into a homogeneous form.[9]

OTHER UNCERTAINTY

For propagation of variance, all quantities are represented as the product of a mass estimate and a composition estimate. In several streams, the mass estimate is not a single measurement. For example, the ER salt mass is determined by multiplying its volume, based on a measured height

and an experimental volume calibration equation, and density, derived from the additive volumes model using measured concentrations. This estimate is assumed to be determined by a single "pseudo-instrument" whose uncertainty is obtained by a detailed propagation of component variances for a representative situation. Pseudo-instruments are also required for the ER cadmium and holdup. Table I list the uncertainties associated with the different streams at FCF, and the pseudo-instrument errors associated with some of those streams.

Table I. Errors for Material Streams at FCF (include pseudo instruments errors).

Stream/Component	Mass (%)	Measurement/Model (%)	
		Uranium	Plutonium
Mk-IV ER Salt	1.17(*)	0.5	0.5
Mk-IV ER Cadmium	1.17(*)	0.5	25
Irradiated Driver Fuel	1.0	1.1	4.3
CP Ingot	(**)	0.67(*)	25
CP Holdup	(**)	8.4(*)	8.4(*)
CP Dross	(**)	2.9(*)	N/A
Cladding Hulls	(**)	0.5	25
Product CF Ingot	(**)	0.5	25

(*) Pseudo Instrument Error
(**) Mass is estimated by multiple balance measurements; mass uncertainty is dependent on balance and calibration period.

SUMMARY

The MC&A accounting system at FCF is dependent on accurate estimation of a number of sensitive nuclear material streams and variance propagation of errors associated with those streams. This paper summaries the methodology for estimating the SNM present in the facility, whether by using direct measurements or by validated models. The uncertainties needed for variance propagation are also discussed, and the most recent estimates of these uncertainties are included. These uncertainties included measurement, model, sampling, and pseudo-instrument uncertainties.

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