ANL/RA/CP-95514

Conference paper to be submitted to The American Nuclear Society Third Topical Meeting DOE Spent Nuclear Fuel and Fissile Materials Management, September 8-11, 1998, Charleston, South Carolina.

FCF Metallic Waste Data Uncertainty Analysis

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*Work supported by the U.S. Department of Energy, Nuclear Energy Programs under Contract W-31-109-ENG-38.

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Metallic waste is a residual of the electrometallurgical treatment of the Experimental Breeder Reactor II (EBR-II) spent fuel. The treatment is based on electrorefining the fuel in molten salt, and currently it remains in the demonstration phase at the Fuel Conditioning Facility (FCF) at Argonne National Laboratory-West (ANL-W). A reference metallic waste form is produced by mixing 15% zirconium with the stainless steel cladding hulls, which remain in the fuel dissolution baskets (FDB's) after electrorefining, to form a metal alloy.[1,2] Estimates of uranium in this waste form are of importance to both operations and sensitive materials control and accountability (MC&A). Accurate estimates of the uranium in this product provide important information regarding the dissolution of uranium and the efficiency of the electrorefining process. Under certain operating conditions, non-negligible amounts of uranium were found in this stream, which made it an area of interest for MC&A. The estimates of uranium in this waste stream are currently provided through analysis of cladding hulls samples. The collected cladding hulls data and the errors associated with the data are discussed here, in addition to the effects of these errors on the overall facility ID variance.

CLADDING HULLS DATA

As part of the fuel treatment at FCF, irradiated fuel elements are chopped into 0.6 cm segments, and placed in anode baskets (FDB's). During the electrorefining process, uranium is transported from the FDB's into a solid cathode, and cladding hulls segments remain in the baskets (one anode consists of four baskets). In addition to UCl_3 in the salt matrix, part of the uranium remains in the basket undissolved.

The data considered here correspond to the first 10 batches of irradiated fuel operations at the FCF. Six samples were taken from the hulls associated with each of the first six anodes: one sample from the middle of each of three baskets and three samples from the bottom, middle, and top of the fourth basket.

Only four samples were taken from each of the last four anodes: one sample from the middle of each basket. Two types of errors are associated with estimates of uranium in those samples, which include the analytical errors and the sampling errors. Both errors are discussed next.

ANALYTICAL ERROR

The cladding hulls samples undergo both water wash and acid etch processes to separate the uranium contained in the hulls and the attached salt. The total amount of uranium in one basket can be estimated by

$$M^{U}_{Hulls} = (U_{water} + U_{acid}) \times \Delta W$$

where

U_{water} = fraction of uranium associated with the hulls sample water wash, mg/g

U_{acid} = fraction of uranium associated with the hulls sample acid wash, mg/g

ΔW = net weight of the FDB

Variance propagation of the above equation gives the random analytical error in the estimate of uranium in the cladding hulls. U_{water} and U_{acid} are estimated using mass spectroscopy and ICP analysis, which leads to 2 σ errors in the estimates of these quantities that range between 5 mg/g for the mass spectroscopy analysis and 50 mg/g or more for the ICP analysis. The weighing errors associated with the balances, σ_w, are about 3.5 gm. Using the irradiated operations data described in the previous section, the average error over all the data is 0.75% and the maximum error is about 3.5%. The errors associated with the top or bottom samples are not much different from the errors associated with the middle samples. Large values of error are associated with samples that contained small fractions of uranium.

SAMPLING ERROR

More dominant errors are introduced to the uranium estimates as a result of using hulls samples with uranium concentrations that are not representative of the bulk of uranium in the hulls. Two types of sampling errors are caused by the inhomogeneity of uranium in the hulls. The first type is a result of the within

basket inhomogeneity which leads to large axial variations in the concentrations of uranium in the baskets. The second type of sampling error is due to the inhomogeneity among the baskets (basket-to-basket variations).

Analysis of the data described before shows large sampling errors that exceed 50%, on average, due to the axial variations in the concentration of uranium within a basket. The basket-to-basket variations are relatively smaller (about 30% average error over the different anodes). Due to those large sampling errors a new sampling procedure is currently in place. The procedure involves mixing of the hulls before sampling and random sampling of the individual hulls within a basket (each hulls sample consists of 10 individual cladding pieces). This sampling is expected to reduce the axial sampling error to a large extent and can also lead to reduction in the basket-to-basket errors as well.

EFFECTS ON THE FCF INVENTORY DIFFERENCE VARIANCE

The overall sampling error associated with the uranium in the hulls is based on a combination of the two types of sampling errors discussed before which can lead to errors up to 60%. A previous sensitivity study described the MC&A models for the different processes within FCF.[3] The study evaluated the sensitivity of the FCF inventory difference (ID) variance to errors in the estimates of the different sensitive materials streams. Based on those MC&A models, the large sampling errors can cause the contribution of the uncertainty in the uranium remaining in the hulls to become important (unless the amounts of uranium in the hulls are small). For example, for errors over 60% and 800 grams of uranium remaining in the hulls, the hulls contribution to the facility ID variance was estimated to exceed 25% of the overall variance. The amount of uranium remaining in the hulls depends on the electrorefining operating conditions. Thus, in order to reduce the hulls contribution to the variance, better estimates of the uranium should be provided through reduction in the sampling errors. Improvements on those estimates are expected in the future through the improved sampling procedure described before in addition to the future use of NDA techniques to estimate the uranium in the metal waste.

References:

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