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MATERIAL ACCOUNTANCY IN AN ELECTROMETALLURGICAL FUEL CONDITIONING FACILITY

D. Vaden, R. W. Benedict, K. M. Goff,
R. W. Keyes, and R. D. Mariani

Argonne National Laboratory - West
P. O. Box 2528
Idaho Falls, ID 83403

R. G. Bucher and A. M. Yacout

Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

ABSTRACT

The Fuel Conditioning Facility* (FCF) treats spent nuclear fuel using an electrometallurgical process that separates the uranium from the fission products, sodium thermal bond and cladding materials. Material accountancy is necessary at FCF for two reasons: first, it provides a mechanism for detecting a potential loss of nuclear material for safeguards and security; second, it provides a periodic check of inventories to ensure that processes and material are under control. By weighing material entering and leaving a process, and using sampling results to determine composition, an inventory difference (ID) results when the measured inventory is compared to the predicted inventory. The ID and its uncertainty, based on error propagation, determines the degree of assurance that an operation proceeded according to expectations. FCF uses the ID calculation in two ways: closeout, which is the ID and uncertainty for a particular operational step, and material accountancy, which determines an ID and its associated uncertainty for a material balance area through several operational steps. Material accountancy over the whole facility for a specified time period assists in detecting diversion of nuclear material. Data from depleted uranium operations are presented to illustrate the method used in FCF.

I. INTRODUCTION

The Fuel Conditioning Facility (FCF), located at Argonne National Laboratory in Idaho, has two different hot cells which house the remotely operated equipment to electrometallurgically treat spent metallic nuclear fuel. Figure 1 shows the process steps. The fuel assemblies are transferred into the air-filled hot cell where the intact assemblies are stored until the fuel elements are separated from the fuel assembly hardware using the vertical assembly dismantler (VAD). The intact fuel elements, which consist of the nuclear materials inside welded stainless steel cladding, are stored individually in element storage magazines. The treatment operations begin after the elements are loaded into element chopper magazines, which can hold 44 elements (approximately 3.0 kg heavy metal).

The element chopper magazines are transferred to the argon-filled hot cell where operations with exposed actinide metals may be performed. The element chopper shears the fuel-containing portion of each element into fuel segments. The non-fuel portion, which is called the plenum, and samples are collected in separate containers. During the shearing process, some of the fission gases are released to the cell atmosphere and the fuel segments are collected in fuel dissolution baskets.

The loaded baskets are transferred to the electrorefiner (ER) which uses an electrochemical process to separate the uranium from the fission products. The ER consists of a molten lithium chloride-potassium chloride salt phase which contains a certain quantity of dissolved uranium chloride. Also, a liquid cadmium phase is underneath the salt phase. The loaded baskets are attached to an electrode assembly and become the anode of an electrical circuit. When current is passed the fuel is dissolved and the uranium is transported to either the cadmium pool or a steel mandrel which is the cathode. The transuranics, alkali, alkaline earth, rare earth, and halide fission products accumulate in the salt phases as metal chlorides. Transition metal fission products end up as insolubles in the cadmium phase or retained in the fuel dissolution baskets. The uranium that is collected on the cathode is removed and is transferred to the cathode processor.

The cathode processor distills off salt adhering to a cathode and consolidates the remaining uranium metal into ingots. The salt is returned to the electrorefiner and the uranium ingots are

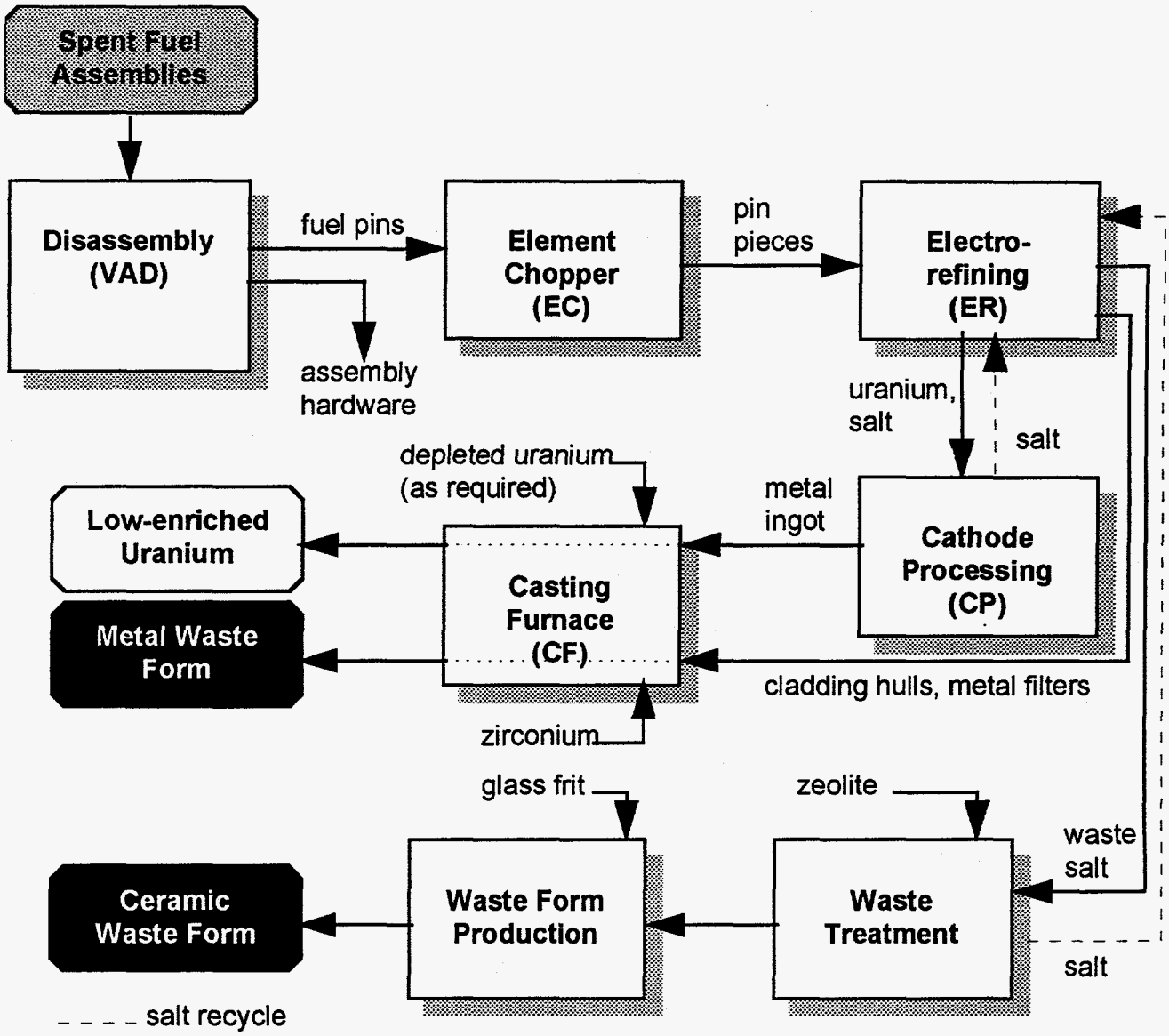

Figure 1: Simplified process flow diagram for Fuel Conditioning Facility

transferred to the casting furnace. Depleted uranium is mixed with the uranium ingots (with approximately 63% U-235) from the cathode processor to produce a final product containing less than 20% U-235. A sample is taken to establish the final composition.

The casting furnace is also used to produce a stainless steel -zirconium metal waste form. The undissolved cladding hulls from the electrorefiner and insolubles are contained in this waste form.

The other fission products and transuranics in the electrorefiner salt phase are placed in a ceramic waste form. The equipment for this process will be added to FCF in the future.

FCF fuel treatment differs significantly from traditional PUREX process facilities in both process technology and safeguards implications. For example, FCF processes the fissile material only in batches and transfers solid, discrete items within the facility. No liquid streams containing fissile material are transferred within the facility, nor enter or leave the facility. Nonetheless, material control and accountability is necessary to comply with DOE Order 5633.3A, "Control and Accountability of Nuclear Materials." The following gives some examples of applying material control and accountability to the FCF process equipment.

II. MATERIAL CONTROL AND ACCOUNTABILITY

Because nuclear material processed in FCF is contained in many types and forms, material accountability requires that the nuclear material content of all flows entering and exiting a material balance area and the quantities of nuclear material in the ending inventory be known. The ID is defined as the difference between the measured inventory and what is expected to be in the inventory based on the previous inventory and measured flows into and out of the process. The ID is calculated via the following equation[1]

$$ID = BI - EI + TI - TO \quad (1)$$

where BI and EI are the beginning and ending inventories and TI and TO are the transfers of nuclear material into and out of the material balance area, respectively. Because measurement errors will occur, the actual amount of material measured will differ somewhat from the expected quantity, creating a non-zero ID. The probability of detecting the loss of a given quantity of material (the loss detection capability) depends upon the uncertainty associated with the determination of the ID. FCF material control and accountability methods propagate all measurement and sampling uncertainties to give a standard error. The limit of error for the inventory difference is simply double the standard error, or two sigma. The limit of error means the measured ID has a 95% probability of being within two sigma of the true ID, which is zero if all materials have been measured and accounted for and all sources of error are used in determining the limit of error.

FCF process operations occur in two hot cells, the air cell and the argon cell, but for DOE reporting the cells are considered one material balance area. The air cell operations are based on individual item counts since the composition is not varied and items are only broken into other discrete items. Material accountancy in the air cell is similar to any storage area and is not described further in this paper.

In the argon cell, the nuclear material changes size, shape and chemical form, so a more sophisticated material accountancy method is required. Two types of material balance calculations are employed: batch closeout, which is the inventory difference for a single process, and material accountancy, which is an inventory difference over several criticality zones and a specified time interval. The element chopper, electrorefiner, cathode processor and casting furnace are considered together with the fuel element and uranium ingots being the primary input and output, respectively.

The closeouts have two different steps which are based on the available information. First a mass balance is performed based on the total weights of the materials that enter and leave a piece of equipment during a batch. This balance must meet a specified accuracy or operations are halted to investigate possible sources of error. This check provides the assurance that operations proceeded as planned and the inventory difference from the measured weights lie within expected limits. After analytical chemistry results are received, a second batch closeout is performed which checks expected and measured compositions. For new items, the expected mass and compositions are based on operational models and prior experience. In this manner, the two step closeout provides the best data for every item in FCF. Since items will be changing their masses and compositions at different times, an on-line mass tracking computer system has been implemented that reads and stores the item weights and compositions. This system provides a model of discrete accountable items distributed in space and time and constitutes a complete historical record.[2]

With this database, material accountancy over all or part of the facility can be calculated for any specified time interval using the item weights and compositions with their associated uncertainties. Material accountancy uses the best available information, which may include either measured or model weights. The computer code, Materials Accounting With Sequential Testing[3] (MAWST) is used to propagate the errors and establish the inventory difference and limit of error.

The following accountability examples focus on the element chopper, electrorefiner and cathode processor/casting furnace. Closeout data from initial depleted uranium operations are presented as examples.

A. Element Chopper

Figure 2 shows the process streams and the associated containers that enter and leave the element chopper.

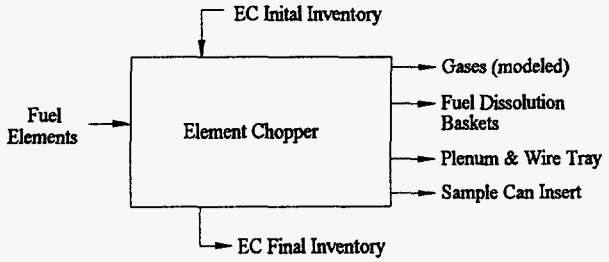

Figure 2: Element Chopper Process Streams

The fuel elements in the element chopper magazine are not individually weighed but their total weight is compared to their modeled weight as a method to verify no gross problems have occurred. The modeled weights and composition are based on fabrication data for the elements. The changes in composition that occurred during irradiation in the reactor are modeled through a physics database for EBR-II.[4] An individual fuel element's composition is contained in a one-dimensional data file, called an ISOZ file. An ISOZ file

contains polynomial functions of the axial distribution of nuclides in a irradiated fuel element.

$$m_{nt}(z) = \sum_{s=1}^{S_t} \sum_{p=0}^{P_{ts}} a_{tspn} z^p, \quad z_{nts}^B \le z \le z_{nts}^T \qquad (2)$$

$m_{nt}(z)$ axial mass distribution of nuclide n and nuclide type t.

a_{tspn} the mass distribution polynomial coefficient for nuclide type t, expansion section s, exponent p, and nuclide n.

z^B_{nts} the lower boundary for nuclide n, nuclide type t in ISOZ expansion section s.

z^T_{nts} the upper boundary for nuclide n, nuclide type t in ISOZ expansion section s.

During irradiated operations, a sample will be taken to determine the burnup at a specified position so the model can be checked. This method has been verified previously with samples from irradiated fuels, but the FCF element chopper has only used depleted uranium so the following data does not illustrate this feature.

The element chopper, using a user-specified chopping recipe, chops the fuel-containing portion of each element into 6 mm to 19 mm long fuel segments. Depending on the chopping parameters, material goes to either a fuel dissolution basket (FDB), the plenum and wire tray (PWT), a sample can insert (SCI), or EC holdup. A specified percentage of any fission gases found goes to the argon cell. While chopping fuel elements in the EC, the mass distributions are integrated between lengths specified in the chopping recipe. Integrating the axial mass distributions generates mass data for the container's zero-dimensional ISOZ file. A zero-dimensional ISOZ file contains the mass of each isotope present in a container or location. Summing all isotopic masses determines the total mass in each location. The FDB ISOZ files represents the process material entering the ER for fuel processing.

An element chopper closeout requires weighing all EC output streams, using the EC inventory (EC holdup) from the previous closeout, and estimating the final inventory to determine the element chopper's ID via Equation 1. Once a container's net weight is known, the isotopic masses in the ISOZ files are scaled via Equation 3 so the ISOZ file's total mass equals the container's net weight.

$$I_M = I_P \times \frac{X_M}{X_P} \qquad (3)$$

I_A the adjusted isotopic mass.

I_P the predicted isotopic mass.

X_M container X's measured net weight.

X_P container X's predicted net weight.

Table 1 shows an EC closeout for chopping 58 sodium bonded depleted uranium-zirconium fuel elements. The input uranium mass was based on the fabrication data for the elements. The output uranium mass was first estimated from a model for the chopping operation. This model assumes that the stainless steel cladding and fuel had a specified axial distribution. The depleted uranium fuel slugs contained no plenum gases, so no gases escaped during the chopping operation. After the chopping was completed, the fuel dissolution baskets were weighed and the uranium mass was adjusted via Equation 3. The final inventory mass was modeled based on two chopping parameters: the element length loss per chop for the sodium bond and the element length loss per chop for the non-sodium components, which includes cladding and fuel. The negative ID on uranium indicates that more uranium exited than entered the process; however, the positive total mass ID indicates more process materials entered than exited. Since the element chopper does not change the form of the material, this closeout and the limited error are based on individual weights of items and the uncertainties are dominated by weighing errors. Since the ID is less than the calculated limit of error the result is acceptable for this batch. The results from multiple batch operations will be tracked so that possible bias in the composition assignment method or the weighing methods can be identified.

Table 1 - Element Chopper Closeout

Container	Total mass (kg.)	Uranium mass (kg.)
BI - EC Initial Inventory (from previous closeout)	0.0000	0.0000
TI - Input mass from ISOZ files	7.4372	4.2063
TO - FDB013 (weighed)	2.8860	1.9891
TO - FDB016 (weighed)	3.2156	2.2224
TO - PWT001 (weighed)	0.6742	0.0000
TO - PWT001 (weighed)	0.6308	0.0000
EI - EC Final Inventory (modeled)	0.0008	0.0006
ID = BI - EI + TI - TO	0.0298	-0.0058
Limit of Error	0.1560	0.0660

B. Electrorefiner

Figure 3 shows the process streams entering and exiting the electrorefiner. The ER inventories require knowing the mass and composition of the salt and cadmium phases,

neither of which can be weighed on a balance. The initial inventory uses the ending inventory from the previous batch. The final inventory uses model values based on the operations that have taken place. At the end of a batch, level measurements and salt and cadmium samples are taken. Using level measurements and the ER volume calibration[5], and additive volumes to calculate the density, the salt mass and cadmium mass are calculated. When analytical chemistry analysis of the samples are available, the uranium mass for each phase can be determined and compared with the expected masses. The total mass holdup reflects the difference between the cadmium, salt and uranium mass as detemined by level measurements and density and the calculated mass from the streams entering and exiting the ER. The uranium holdup inventory is estimated by process models and includes the insolubles that form from impurities in the system.

The input to the electrorefiner uses the weights and compositions that were established during the loading of the baskets. The output in the fuel dissolution baskets is based on the net weight of the baskets at the end of the operations. The composition of the material remaining in the fuel dissolution baskets will initially be based on process models, but the material will be sampled and refinements made to the composition when analytical data becomes available. The solid cathode output is initially based on the weight of the cathode with the composition based on a process model. The principal variable in this model is the amount of uranium metal and adhering salt. Data from the cathode processor operations provide an initial refinement of the composition and further refinement is made when analytical chemistry data of the metal product is available from the casting furnace operation.

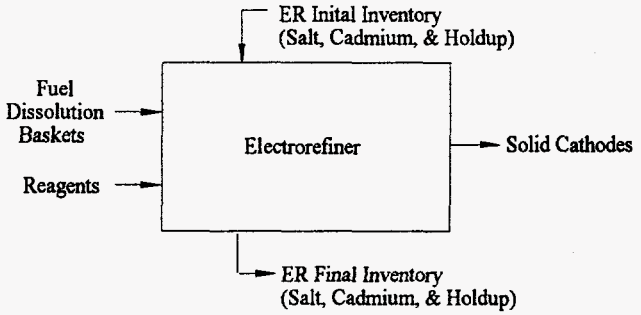

Figure 3: Electrorefiner Process Streams

Table 2 shows the ER closeout of batch ERDU02, which dissolved 14 kilograms of depleted uranium plates, added 8.8 kg cadmium chloride and produced a 3.8 kg solid cathode. The limit of error includes the uncertainties of the level measurements, the sampling and analysis, weight measurements and holdup models.

Table 2 - Batch ERDU02 Closeout

Container	Total Mass (kg.)	Uranium Mass (kg.)
BI - Salt Inventory	450.497	19.618
BI - Cadmium Inventory	536.450	0.000
BI - Holdup Inventory	8.645	0.304
TI - Fuel Dissolution Baskets	14.000	14.000
TI - Cadmium Chloride	8.776	0.000
TO - Fuel Dissolution Baskets	0.000	0.000
TO - Solid Cathode	3.836	3.075
EI - Salt Inventory	470.977	27.270
EI - Cadmium Inventory	539.073	2.839
EI - Holdup Inventory (Model)	0.685	0.304
ID = BI + TI - TO - EI	3.796	0.434
Limit of Error	9.593	0.806

Because the ID is smaller than the limit of error, the closeout is acceptable. With each closeout, the measured data gives the modeling code more information to predict with increased confidence the holdup inventory for subsequent batches.

C. Cathode Processor/Casting Furnace

The cathode processor (CP) distills off the salt adhering to a solid cathode and consolidates the remaining uranium metal into ingots.[6] The material accountancy is complicated because a process crucible (PC) coating material can potentially react with some of the uranium and create a minor waste dross stream. The casting furnace mixes depleted uranium with the CP uranium ingots to produce a low-enriched uranium product. A homogeneous sample is taken of this product by casting a small sample into glass molds. Figure 4 shows the process streams entering and leaving the cathode processor and casting furnace. Since these operations are closely related, the batch closeouts are combined together and shown in Table 3 for processing the 3.8 kg cathode from the ER.

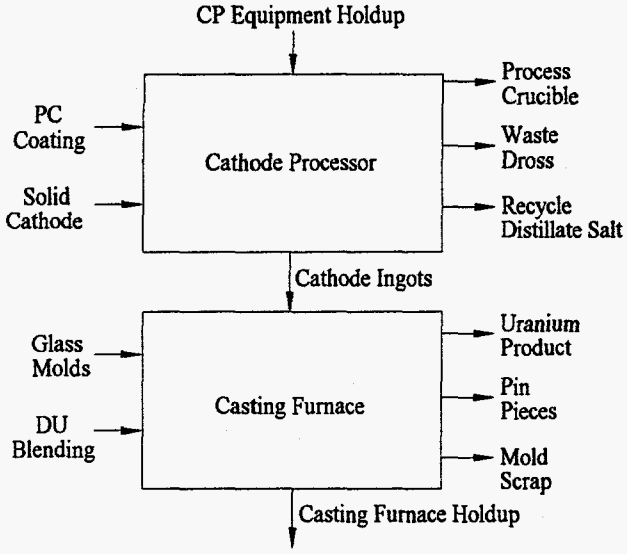

Figure 4: CP and CF Process Streams

Table 3 - Batch CPDU02 Closeout

Container	Total Mass (grams)	Uranium Mass (grams)
BI - CP Holdup	75.00	3.20
BI - PC Coating	74.00	0.00
TI - Solid Cathode	3820.00	3061.83
TI - CF DU Feedstock	3000.20	2986.10
TI - CF Glass Mold	55.00	0.00
TO - CP Waste Dross	434.00	355.19
TO - CP Distillate Salt	761.00	32.51
TO - CF Mold Scrap	64.00	8.94
TO - CF Heel Wt.	5256.00	5223.76
TO - CF Pin Pieces	332.00	329.96
EI - PC Holdup	44.00	36.01
EI - CP Holdup	106.00	1.32
EI - CF Holdup	5.00	4.97
ID = BI + TI - TO - EI	22.20	58.46
Limit of Error	Not Done	99.05

The inventories which are called equipment holdup are based on the differences in the measured total weights going in and out of the pieces of equipment. For the cathode processor, the holdup composition is assumed to be the ER salt composition when the solid cathode was produced. For the CP process crucible holdup, the composition is based on the uranium composition. For the casting furnace, the holdup composition is assumed to be the uranium metal unless a process upset occurs which justifies a change.

The uranium ID is higher than the total mass ID because the uranium content in the waste dross stream sample may not represent the uranium content in all the waste dross and process crucible holdup. The uranium ID has an uncertainty dominated by trying to obtain a representative sample from heterogenous material, whereas the total mass ID has an uncertainty based on the FCF balances.

The input streams, except the solid cathode, are well characterized, and their weights and compositions are used. The solid cathode has a composition established when received from the electrorefiner, and this composition is refined as described in the ER section.

The composition of the principal output stream, the uranium product, is determined from chemical analysis of the samples. The recycle distillate salt is assigned the ER salt composition, and since it is returned to the ER the masses will drop out of the overall material accountancy. The waste dross and mold scrap streams are assigned a composition based on either process models or sample analysis and contain minor amounts of uranium.

D. Material Accountancy

In general, the computation of an ID for material accountancy differs from that for the batch closeout of a process operation. For accountancy, a material balance area is defined as a set of one or more criticality zones, whereas, for closeouts, the area is composed of the equipment and containers that support the operation; these equipment and containers may constitute only a portion of the criticality zone(s) they occupy. Also, the material balance period for accountancy is nominally set at assigned intervals specified in DOE Orders, such as, one or six months, while the interval for closeouts is the time during which material in the specified batch is being processed.

Material accountancy requires quantitative knowledge of material, 1) present in the material balance area at the beginning and ending of the accountancy period and, 2) transferred into and out of the area during the period. Material accountancy performed over multiple zones, and therefore over multiple process operations, does not require knowledge of the material that is transferred between processes. The compositions of these inter-process materials are determined by process models, which at least for startup have large estimated uncertainties. Inventory and transferred materials for a properly designed material balance are well characterized by weighing

and analytical analysis of samples; this yields better known and significantly smaller uncertainties than similar closeout uncertainties.

Material accountancy can, in many instances, be configured to approximate a batch closeout. By assigning the area to be the appropriate criticality zone(s) and restricting the interval to processing times, the computed ID and uncertainty can closely approximately the closeout analysis. These methods have been employed to confirm many of the above batch closeout inventory difference calculations.

At present, chopped segments have not yet been processed through the ER and CP/CF, which prevents any attempt to conduct material accountancy on a single batch from the element chopper through the casting furnace. Starting from the ER through the CF, a temporary output stream is holdup on the harvested cathode mandrel. In the examples presented, this accounts for 12.68 grams of uranium and 16 grams total mass. However, as this material is also returned to the ER, it has no impact on overal accoutancy.

CONCLUSIONS

Material accountancy, based on data from depleted uranium operations involving the element chopper, electrorefiner, cathode processor and casting furnace, produced inventory differences less than their respective limits of error. Subsequent batch processing will provide additional data and information to help refine process models and improve the ability to predict the holdup inventory of subsequent batches.

NOMENCATURE

BI	beginning process or equipment inventory
EI	ending process or equipment inventory
TI	transfers into a process
TO	transfers from a process
$m_{nt}(z)$	axial mass distribution of nuclide n and nuclide type t.
a_{tspn}	the mass distribution polynomial coefficient for nuclide type t, expansion section s, exponent p, and nuclide n.
z^{B}_{nts}	the lower boundary for nuclide n, nuclide type t in ISOZ expansion section s.
z^{T}_{nts}	the upper boundary for nuclide n, nuclide type t in ISOZ expansion section s.
I_A	the adjusted isotopic mass.
I_P	the predicted isotopic mass.
X_A	container X's measured net weight.
X_P	container X's predicted net weight.

REFERENCES

1. "Guide for Implementation of DOE 5633.3B: Control and Accountability of Nuclear Materials", U.S. Department of Energy Office of Security Affairs, Office of Safeguards and Security, April 1995.

2. C. H. Adams, J. C. Beitel, G. Birgersson, R. G. Bucher, K. L. Derstine, R. W. Goin, R. W. Keyes, B. J. Toppel, and M. A. Vollmer, "The Mass Tracking System - Computerized Support for MC&A and Operations at FCF", this proceedings

3. R. R. Picard and J. F. Hafer, "MAWST: Materials Accounting With Sequential Testing", Los Alamos National Laboratory Report, N-4/91-633 (June 1991)

4. R. D. McKnight, J. A. Stillman, B. J. Toppel, and H. S. Khalil, "Validation and Application of a Physics Database for Fast Reactor Fuel Cycle Analysis", ANS 1994 Topical Meeting on Advances in Reactor Physics, Knoxville, Tennessee, April 11-15, 1994.

5. R. G. Bucher and Y. Orechwa, "Fuel Conditioning Facility Electrorefiner Volume Calibration", 36th Annual Meeting of the Institute of Nuclear Materials Management (INMM), Palm Desert, Calif., July 9-12, 1995.

6. B. R. Westphal, D. V. Laug, A. R. Brunsvold, and P. D. Roach, "Initial Cathode Processing Experiences and Results for the Treatment of Spent Fuel", this proceedings

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