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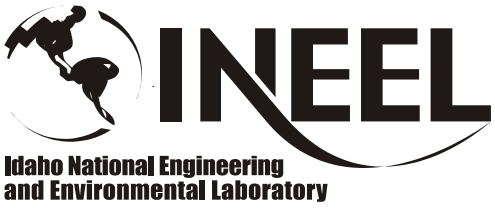

Radiological Characterization Methodology for INEEL-Stored Remote-Handled Transuranic (RH TRU) Waste from Argonne National Laboratory-East

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RADIOLOGICAL CHARACTERIZATION METHODOLOGY FOR INEEL-STORED REMOTE-HANDLED TRANSURANIC (RH TRU) WASTE FROM ARGONNE NATIONAL LABORATORY-EAST

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ABSTRACT

An Acceptable Knowledge (AK)-based radiological characterization methodology is being developed for RH TRU waste generated from ANL-E hot cell operations performed on fuel elements irradiated in the EBR-II reactor. The methodology relies on AK for composition of the fresh fuel elements, their irradiation history, and the waste generation and collection processes. Radiological characterization of the waste involves the estimates of the quantities of significant fission products and transuranic isotopes in the waste. Methods based on reactor and physics principles are used to achieve these estimates. Because of the availability of AK and the robustness of the calculation methods, the AK-based characterization methodology offers a superior alternative to traditional waste assay techniques. Using this methodology, it is shown that the radiological parameters of a test batch of ANL-E waste is well within the proposed WIPP Waste Acceptance Criteria limits.

INTRODUCTION

The Idaho National Engineering and Environmental Laboratory (INEEL) is proposing a radiological characterization methodology based on Acceptable Knowledge (AK) for RH-TRU waste generated at Argonne National Laboratory-East (ANL-E). The proposed methodology utilizes the Quality Assurance provisions of 40 CFR 194.22(b) to qualify the AK-based radioactive waste characterization by Peer Review.

The purpose of this paper is to show that the proposed methodology can be used to determine a technically defensible radionuclide inventory using AK, complemented by calculations using reactor physics principles without any additional confirmatory measurements to meet the Waste Isolation Pilot Plant (WIPP) transportation and disposal requirements. The advantages of the AK-based methodology include more reliable quantification of the radionuclides in the waste than any quantification through non-destructive assay or limited sampling and the minimization of worker exposure to radiation.

AK-BASED RADIONUCLIDE QUANTIFICATION

The radiological characterization approach the INEEL is developing, consists of the following steps:

(a) Collecting AK information which comprises of:

Identification of all fuel elements that contributed to specific batch of drums
Initial fuel composition
Irradiation history of the fuel

(b) Calculating radioisotope inventory from AK information using a combination of nuclear engineering and physics principles

(c) Determining the fraction of the fuel that went into the RH-TRU waste based on generator hot cell records

(d) Determining uncertainties in radioisotope contents from uncertainties in:

Initial fuel composition
Irradiation history of the fuel
Waste generation and collection processes

(e) Completing peer review of the INEEL approach per 40 CFR 194.22(b).

The above steps, except the last step on peer review, are discussed below using a test batch of RH-TRU waste as an example. The test batch consists of waste drums numbered 728-737 stored at the INEEL.

AK Collection

Based on official ANL-E records used for reporting Pu-239/U-235 present in waste drums 728-737, the RH-TRU waste was collected in the destructive examination of 14 fuel elements irradiated in the EBR-II reactor from 1987 to 1990. [Note: Hot cell data indicate there were more fuel elements examined than reported during this time period. This reporting discrepancy would contribute to the uncertainty in radionuclide content estimates.] Seven of these fuel elements were manufactured at ANL-W where the EBR-II reactor was located. These seven fuel elements had detailed initial composition data, including the quantities of the important plutonium isotopes (Pu-239, Pu-240, Pu-241, and Pu-242). The data were based on radiochemical analysis of sample fuel at the time the fuel elements were made. The other seven fuel elements that contributed to the RH-TRU waste in the batch were manufactured by contractors outside of the ANL-W. The AK for these seven fuel elements only provided the U-235, U-238, Pu-239, and Pu-240 quantities in the fresh fuel elements; the other uranium and plutonium isotopes had to be estimated based on the uranium enrichment and the Pu-240 to Pu-239 ratio.

Detailed records exist on the irradiation history of the fuel elements. The irradiation history includes the dates of irradiation, average power of the reactor during irradiation, and average burnup of the fuel subassemblies that contained the fuel elements. Most importantly, information is also available on the plutonium (Pu-239 and Pu-240) and uranium (U-235 and U-238) contents of the fuel elements after irradiation, which enables the calculation of the burnup of the individual fuel elements. Such information was based on reactor neutronic calculations. Such calculations for the reactor driver core had been validated by radiochemical analyses of irradiated driver core fuel elements.

Radionuclide Inventory Calculation

Most of the radioactive inventory in irradiated fuel comes from fission products. At the time of disposal of the RH-TRU waste at WIPP, the fission products would have aged more than 10 years since the time the fuel was last irradiated. For this time frame, the characterization of the radioactive inventory in the waste is greatly simplified because only a few radionuclides need to be quantified. Because of the combination of fission yields and decay half-lives, approximately 10 years after irradiation, the dominant fission products in irradiated fuel are limited to only Cs-137, Sr-90, and their decay daughters Ba-137M and Y-90, respectively. These four fission radionuclides would account for more than half the radioactivity in the waste. When plutonium was present in the fresh fuel, Pu-241 would account for approximately another 1/3 of the total activity in the irradiated fuel approximately 10 years after irradiation. Most of the remaining activity in the waste would be accounted for by Pu-239, Pu-240, and Am-241.

The inventory of the fission products and the actinides in RH-TRU waste can be calculated from the initial composition of the fuel, irradiation history of the fuel, and decay times. Such calculations are generally performed with an isotope generation and depletion code, such as ORIGEN2 (1). However, for the quantification of just a few nuclides, the use of a code is often not necessary, particularly for the quantification of the fission products, which can be calculated directly from simple formulas without much loss of accuracy compared to code calculations. For the actinides, because of the multiple generation and decay paths, code calculations are preferred when a high degree of accuracy is required. However, large uncertainties of radioactivity contents in RH-TRU waste exist due to factors other than inventory in the destructively examined fuel elements. Therefore, highly accurate calculations for inventories in fuel elements contribute little to the accurate quantification of the radionuclides in the waste. For this reason, simplified calculations instead of code calculations are formulated for the actinides also. These formulations have the advantages that the functional dependencies are transparent and only the burnup, not the detailed irradiation history, is needed for the calculations. The simple formulas for the calculations are presented below.

Fission Products

The amount of fission products generated in irradiated fuel is directly proportional to the heavy metal (uranium, plutonium, and higher actinides) mass in the fuel, the burnup of the heavy metal, and the fissionable-mass weighted yield of the fission products. Depletion of Cs-137 and Sr-90 due to neutron reactions is negligible, but decay of these radionuclides has to be accounted for because their relatively short half-lives. The mass of Cs-137 or Sr-90 can be calculated from the following formula:

$$M_{fp} = M_{hm}(BU)y(A_{fp}/A_{hm})e^{-\lambda t_D}(1-e^{-\lambda t_R})/(\lambda t_R) \qquad \text{(Eq. 1)}$$

where

M_{fp} = the mass of the fission product
M_{hm} = the mass of the heavy metal
BU = the fractional burnup of the heavy metal
y = the fissionable-mass weighted fission yield (atomic fraction)
A_{fp} = the atomic weight of the fission product
A_{hm} = the average atomic weight of the fissionable nuclides
λ = the decay constant of the fission product
t_D = the decay time after irradiation, and
t_R = the irradiation time.

Actinides

The estimates of the amounts of actinides in a fuel element due to irradiation can be greatly simplified when depletion due to burnup is ignored, when short-lived intermediate products are assumed to decay instantly, and when long-lived intermediate products are assumed stable. Such simplification introduces an error on the order of the burnup, which is generally less than 15% for fuel irradiated in the EBR-II reactor. For the generation of an actinide that requires n successive neutron reactions from an actinide initially present in the fuel, such an approximation leads to the following formula (2):

$$N_n \approx N_{00}\frac{\prod_{k=1}^{n}\sigma_k}{\sigma_f^n n!}(BU)^n \qquad \text{(Eq. 2)}$$

where

N_n = moles of actinide n produced
N_{00} = moles of initial actinide at top of the generation chain
σ_k = neutron reaction rate with actinide k in the generation chain of actinide n
σ_f = fissionable-mass weighted fission cross-section, and
BU = fractional burnup of heavy metal.

The above formula can be applied to all the actinides without much loss of accuracy for waste characterization purposes, but decay corrections must be made to account for decay of Pu-241, which has a short half-life of 14.4 years.

Waste Generation and Collection Information

Once the radioactive inventory in the fuel elements are known, the amount of that inventory going into the RH-TRU waste can be estimated by examining the operations on the fuel elements that produced the RH-TRU waste. These operations include cutting the fuel elements into examination samples, grinding the samples into suitable metallographic mounts, and polishing the samples. The large fuel particles created by these operations were picked up and stored as "high activity level" waste, while the fines and deposits on equipment that could not be easily picked up were cleaned with rags and tissue paper, which became the RH-TRU waste. Based on dimensions of the cutting tools and sample sizes, and the observed degree of cleanup of the fines, hot cell personnel estimated that, on average, 1.5% of the irradiated fuel in an examined fuel element went into the RH-TRU waste. When this percentage was multiplied into the radioactive inventory in the fuel element, the radioactive inventory in the RH-TRU waste was obtained.

Uncertainty Estimates

Three sources of uncertainty are considered for the uncertainty in the estimated amounts of radioactive inventory in the RH-TRU waste. First, the uncertainty in the inventory in that part of the fuel element that went to the waste is estimated based the burnup profile in the fuel element and the uncertainty in the calculated amount for a given burnup. The burnup profile is important in determining the uncertainty because it is generally not known which part of the fuel element went into the waste. Second, the uncertainty in the waste generation and collection process is estimated based on the variations in the number of samples taken and in the type operations performed on the samples. The uncertainty in the collection efficiency is estimated based on interviews with operations personnel and on observations on the setups in the hot cell. Third, uncertainties in bookkeeping are considered. These include examining the practice of waste reporting in batches, which often did not completely correspond to the actual waste generation and collection. These three types of uncertainty are then combined to arrive at the overall uncertainty in the estimated radioactivity in the waste.

In computing the uncertainties of combined quantities, such as the radioactive inventory from the several fuel elements that contributed to the batch of waste, correlation between the various parameters are considered. If two quantities are certain to be uncorrelated, such as between the inventory and collection efficiency, they are treated as such; if they are judged to be partially correlated, such as among those parameters derived from a common calculation method, they are treated as completely correlated. The algorithms of combining the uncertainties based these assumptions over-estimate the final uncertainties. This is a conservative approach to uncertainty determination.

RESULTS FOR A TEN-DRUM TEST BATCH

The AK-based radionuclide quantification methodology presented above was applied to a ten-drum test batch. As mentioned under *AK Collection*, the fuel elements that were reported to have contributed to this ten-drum (728-737) batch of waste were irradiated in the EBR-II reactor from 1987 to 1990 and destructively examined at ANL-E in 1990 and 1991. The burnup of the fuel elements that produced this batch of waste ranged from 0.7% to 12%. The plutonium to uranium ratios ranged from 0.15 to 0.25 and the enrichments of the uranium ranged from 0.25 to 0.8. From these very different ratios it is evident that the waste can be very inhomogeneous unless there was a deliberate attempt to homogenize the waste, which was not the case. For these reasons, the estimates based on AK and physics principles will be more reliable than assaying a limited number of waste samples. Preliminary results of the estimates of radiological parameters are shown in Table I along with comparisons with the proposed Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (3).

Table I. Comparison of calculated radiological parameters and WIPP Waste Acceptance Criteria.

Waste Acceptance Criterion	Results *	Limits	Factor of Safety
Pu-239 FGE (fissile gram equivalent)	18.6 FGE for all 10 drums	<32.5 FGE for 72B cask (1/10th the inventory limit. No error needs to be assigned)	43% below 1/10th of inventory limit of the cask
Pu-239 PE-C (equivalent Ci)	1.4 in all 10 drums	240	170 times below the limit
TRU alpha concentration	162 nCi/g lower limit 5,250 nCi/g upper limit	>100 nCi/g	62% higher than lower limit
Radionuclide Activity	26.0 Ci in all of the 10 drums (114 L per drum)	<23 Ci/L	Far below the limit
Decay Heat	0.080 W for all 10 drums	<50 W/RH-TRU 72B cask <0.14 W/drum and <0.43 W/cask as specified per content code	5 times below cask limit

* All values are at the 2-sigma (95%) confidence level.

CONCLUSION

The results of the radiological characterization of the batch of drums (728-737) using the AK-based calculation methodology indicate that for this particular waste stream, radiological characterization can be performed using AK. The results are robust enough to allow safe transportation and disposal without any additional confirmatory measurements. In addition there may not be any value added if measurements are performed because:

- Assignment of TRU isotopes based on gamma-spectroscopic measurements of fission products alone will not be reliable due to variation in fuel types (mixed oxide and metallic fuels), large variation in initial fuel composition (Pu/U ratios), and burnup.

- Representative samples for radiochemical analysis cannot be obtained because of the highly heterogeneous nature of the waste.

The 10-drum radiological AK Radioactive Waste Characterization report (4) for the test batch and the AK-based calculation methodology (5) will be submitted to Carlsbad Field Office for peer review. If the peer review panel and, subsequently, regulators accept the proposed methodology, the radioactive waste characterization of remaining batches will be performed using the approved methodology.

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