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Isotopic Characterization of HALEU from EBR-II Driver Fuel Processing

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TECHNICAL EVALUATION

Title: Isotopic Characterization of HALEU from EBR-II Driver Fuel Processing

TEV No.: 3537 Rev. No.: 0 Project No.: 31059 Date: 10/17/2018

1. Does this TEV involve a Safety SSC?	No	**Professional Engineer's Stamp** N/A
2 Safety SSC Determination Document ID	N/A	
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4. SSC ID	N/A	
5. Building	765	
6. Site Area	MFC	

7. Introduction:

The purpose of this technical evaluation is to provide the isotopic characteristics of the High Assay Low Enriched Uranium (HALEU) product recovered during the used nuclear fuel treatment operations occurring at Idaho National Laboratory.

8. N/A

9. Conclusions/Recommendations:

To provide sufficient information to perform an isotopic characterization of HALEU produced at Idaho National Laboratory:

- The analytical results of casting furnace pin samples provide sufficient information of the measured nuclides.
- Isotopic distributions based on ORIGEN calculations coupled with the analytical results provide the characterization for the non-measured nuclides.

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PROJECT ROLES AND RESPONSIBILITIES

Project Role	Name (Typed)	Organization	Pages covered (if applicable)
Performer	Dee Earl Vaden	C420	
Checker[a]	Brian R. Westphal	C420	
Independent Reviewer[b]	JC Price	U710	
CUI Reviewer[c]	N/A		
Manager[d]	Doug Crawford	U000	
Requestor[e]	Michael N. Patterson	U600	
Nuclear Safety[e]	N/A		
Document Owner[e]	Michael N. Patterson	U600	

Responsibilities

a. Confirmation of completeness, mathematical accuracy, and correctness of data and appropriateness of assumptions.

b. Concurrence of method or approach. See definition, LWP-10106.

c. Concurrence with the document's markings in accordance with LWP-11202.

d. Concurrence of procedure compliance. Concurrence with method/approach and conclusion.

e. Concurrence with the document's assumptions and input information. See definition of Acceptance, LWP-10300.

NOTE: *Delete or mark "N/A" for project roles not engaged. Include ALL personnel and their roles listed above in the eCR system. The list of the roles above is not all inclusive. If needed, the list can be extended or reduced.*

INTRODUCTION

Proposed Issue

To assess the feasibility of reusing High Assay Low Enriched Uranium (HALEU) recovered from the treatment of irradiated EBR-II fuel, an environmental assessment requires an isotopic characterization of the uranium product from the process deployed in the Fuel Conditioning Facility (FCF) at Idaho National Laboratory (INL). A best-available characterization is derived through a combination of measured values obtained through chemical and isotopic analysis and calculated values determined through the use of process modeling software and related programs.

ACRONYMS

CFBF	Casting Furnace Binary Fuel
EBR-II	Experimental Breeder Reactor II
FCF	Fuel Conditioning Facility
HALEU	High Assay Low Enriched Uranium (HALEU)
INL	Idaho National Laboratory
MTG	Mass Tracking System
ORIGEN	Oak Ridge Isotope Generator
PADB	Physics Analysis Database

Background

As part of past and ongoing operations, fuel elements irradiated in EBR-II undergo processing via the Electrometallurgical Treatment (EMT) system which resides in FCF. Under this process, the metallic uranium used in the original construction of the element is separated from the bond sodium and the majority of the fission products along with the associated minor actinides. The recovered uranium metal undergoes successive heating steps to remove process media (electrorefiner salt) and is simultaneously down-blended through the addition of natural or depleted uranium to achieve a final U-235 enrichment of less than 20%. During the heating process, the molten uranium is sampled via the production of a casting pin which is subsequently transferred to the Analytical Laboratory for analyses that provides the measured concentrations of some isotopes of interest (e.g., ^{234}U, ^{235}U, ^{236}U, and ^{238}U) thus characterizing the recovered uranium metal. This approach for characterization has been conducted on all of the nearly 4 metric tons of < 20% U metal recovered to date and is anticipated to continue for the remaining EBR-II spent fuel identified for future treatment. While this method provides for an accurate assessment of the inventory on the basis of the analyzed isotopes, complete characterization has required development of a method to determine the concentration of the non-measured isotopes (e.g., ^{232}U) as well. Based on the data accumulated from this inventory, a bounding approximation can be prepared that characterizes the HALEU material to be prepared from this inventory.

Evaluation

Uranium Isotope Determination

FCF utilizes a Mass Tracking System (MTG) that employs process models to estimate and track the mass and composition of process material moving through the facility [REF. 1]. Files representing the mass and composition of the incoming spent fuel were generated [REF. 2, 3, 4] from reactor burnup and isotope generation codes; the code employed for this purpose is ORIGEN, or Oak Ridge Isotope Generation [REF. 5], which is commonly used for such purposes. Because isotopes of an element possess the same chemical properties of that element, one can use the isotopic distributions from the MTG process models [REF. 6] along with the measured concentrations from sample analysis to determine the concentration of the non-measured nuclides. See Appendix A for an example of this calculated approach. The combination of isotopic values collected via sample analysis and those calculated from modeled values based on ORIGEN codes can be used to provide a representative approximation of the isotopic inventory of the HALEU to be produced from uranium metal recovered as a result of processing the irradiated EBR-II elements.

Appendix B provides a table containing average values for the four directly measured uranium isotopes ^{234}U, ^{235}U, ^{236}U, and ^{238}U, as well as maximum values for the calculated isotopes ^{232}U, ^{233}U, & ^{237}U for HALEU material intended to be produced from the FCF uranium product. These uranium isotope compositions values are judged to be bounding for the existing FCF uranium metal inventory, as well as for any future uranium recovered from ongoing treatment activities due to the incorporation of planned process enhancements intended to produce uranium metal with fewer impurities. It is recommended that the calculated compositions of U-232, U-233, and U-237 be used along with measured compositions of other uranium isotopes for assessment of environmental impact from HALEU fuel fabrication from FCF HALEU feedstock. Because the contribution of small amounts of these isotopes will have negligible impact on calculations of environmental impact of HALEU fuel fabrication, it is judged that the calculated U-232-, U-233, and U-237 values are suitable for the purpose.

Impurity and Isotopic Composition

Chemical and isotopic compositions determined using analytical chemistry methods have been obtained from a selected set of six casting batches [REF. 7], comprising roughly 202 kg of uranium. The six casting batches were selected to be conservatively representative of the impurity contents of FCF uranium product prepared for use as HALEU feedstock. An effort is underway to identify a process of blending of prior FCF uranium product ingots with newly produced FCF uranium product to produce HALEU feedstock ingots with further reduced impurities and more consistent U-235 enrichment; i.e., a process to improve prior and forthcoming FCF uranium product to material suitable for use as HALEU feedstock in HALEU fuel fabrication in gloveboxes. Early results, yet incomplete, indicate that further developments in the process scheme have the potential to result in further decontamination than that achieved in those six casting batches. Therefore, the average of the measured and chemical isotopic analyses from those six batches (Appendix C) is judged to be representative and conservatively bound the impurity contents of future casting batches of HALEU feedstock to be used in HALEU fuel fabrication. Because the objective in that previous six-batch study was to assess decontamination potential, the content of non-detectable uranium isotopes (U-232, U-233, and U-237) was not calculated. However, the U-232, U-233, and U-237 contents calculated for all prior casting batches, as described in the previous section, are the best-available estimate of contents in the HALEU feedstock going forward; the varying degrees of decontamination does not change the relative uranium isotopic composition.

CONCLUSION / RECOMMENDATION

The existing analytical reports in combination with the methodology used to calculate non-measured isotopes of interest provide an accurate and bounding estimate of the isotopic composition of the HALEU feedstock anticipated to be produced from the inventory of uranium metal recovered from the treatment of EBR-II fuel. This applies to both the existing inventory generated over the course of the past 25 years, as well as any future quantities to be generated as the result of ongoing treatment activities. The isotopic and impurity compositions as-measured in Appendix C, and supplemented with the calculated compositions for U-232, U-233, and U-237 as listed in Appendix B are recommended as a bounding composition (i.e., conservatively bounding uncertainties) for calculations of environmental impacts associated with HALEU fuel fabrication using HALEU feedstock from FCF.

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2. R. D. McKnight, "*ANL Calculational Methodologies for Determining Spent Nuclear Fuel Source Term*", International Conference on the Physics of Nuclear Science and Technology, Long Island, NY (US), July 1998.

3. D. Vaden, "*GENERATING ISOTOPE-Z (ISOZ) FILES FROM ORIGEN OUTPUT FAST FLUX TEST FACILITY FUEL ASSEMBLIES*", TEV-516, September 2010.

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6. D. Vaden, "*Fuel Conditioning Facility Electrorefiner Process Model*", Separation Science and Technology, Volume 41, Number 10, Pages: 1985 – 2001 (2006).

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Appendix A

Engineering Inputs

Tables A-1 and A-2 provide results of measured and calculated uranium isotopic composition derived for a single furnace casting batch (batch CFBF18A), as an example of those similarly derived from all FCF uranium product batches.

In Table A-1:

- The second column contains the measured weight fractions of uranium isotopes for casting furnace batch CFBF18A.
  - Note: the weight fractions of elemental and isotopic uranium are based on the entire sample, which is why the sum of the uranium isotopes is less than 100%. Table A-2 performs the calculations for the isotopic fractions of uranium and does sum to 100%.
- The third column contains the estimated weight fractions of uranium isotopes from the FCF MTG. The MTG process models, starting with the initial uranium isotopic distribution from ORIGEN calculations, estimated the isotopic distribution of uranium isotopes at the casting furnace.
- In the fourth column, the term "best available" means: if the measured weight fraction is available, use it; if not available, then use the estimated weight fraction. Note in the last row of the fourth column the best available weight fractions sum to a value not equal to the measured total uranium. The best available weight fractions are divided by their sum and multiplied by the measured uranium to produce the final uranium weight fractions shown in the fifth column.

Table A-1. Uranium Isotopic Distribution in Casting Furnace Batch CFBF18A (weight fraction)

Nuclide X	Weight Fraction Measured	Weight Fraction Estimated	Weight Fraction Best Available	Weight Fraction Final
^{232}U		8.700E-10	8.700E-10	8.725E-10
^{233}U		1.276E-07	1.276E-07	1.280E-07
^{234}U	2.104E-03	2.102E-03	2.104E-03	2.110E-03
^{235}U	1.903E-01	1.902E-01	1.903E-01	1.909E-01
^{236}U	6.314E-03	6.310E-03	6.314E-03	6.333E-03
^{237}U		1.158E-14	1.158E-14	1.161E-14
^{238}U	7.983E-01	7.978E-01	7.983E-01	8.007E-01
U	9.970E-01	9.970E+00	9.970E-01 + 4.350E-05	9.970E-01

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In Table A-1:

- The second column contains the measured isotopic fractions of uranium isotopes for casting furnace batch CFBF18A.
- The third column contains the estimated isotopic fractions of uranium isotopes from the FCF MTG. The MTG process models, starting with the initial uranium isotopic distribution from ORIGEN calculations, estimated the isotopic fractions of uranium isotopes at the casting furnace.
- In the fourth column, the term "best available" means: if the measured isotopic fraction is available, use it; if not available, then use the estimated isotopic fraction. Note in the last row of the fourth column the best available isotopic fractions sum to a value greater than one. The best available isotopic fractions are divided by their sum to produce the final uranium isotopic fractions shown in the fifth column.

Table A-2. Uranium Isotopic Distribution in Casting Furnace Batch CFBF18A (isotopic fraction)

Nuclide X	g X / g U Measured	g X / g U Estimated	g X / g U Best Available	g X / g U Final
^{232}U		8.927E-10	8.927E-10	8.926E-10
^{233}U		1.310E-07	1.310E-07	1.310E-07
^{234}U	2.110E-03	2.164E-03	2.110E-03	2.110E-03
^{235}U	1.909E-01	1.959E-01	1.909E-01	1.909E-01
^{236}U	6.333E-03	6.484E-03	6.333E-03	6.333E-03
^{237}U		1.190E-14	1.190E-14	1.190E-14
^{238}U	8.007E-01	7.954E-01	8.007E-01	8.007E-01
Total	1.000E+00	1.000E+00	1.000E+00 + 4.363E-05	1.000E+00

Appendix B

HALEU Assay

Analyte	Units	Average	Minimum	Maximum
Total U	wt. %	99.95	99.89	99.97
U232^	ppbU	0.66	0.07	5.04
U233^	ppbU	49.12	4.88	318.43
U234*	iso % U	0.17	0.16	0.21
U235*	iso % U	19.39	18.97	19.99
U236*	iso % U	0.58	0.50	1.22
U237^	pptU	0.06	0.00	0.22
U238*	iso % U	79.86	79.05	80.36

^ Isotope content beyond detection limits, so calculated using method described in Appendix A

* Actinide isotopes measured analytically

ppbU = g per billion grams of uranium

iso. % U = isotope wt.% of total U

pptU = g per trillion grams of uranium

Appendix C

Measured Chemical and Isotopic Composition of Six FCF Uranium Product Casting Batches (202 kg)

Analyte	Units	Weighted Average	Minimum	Maximum
Total U	wt. %	99.67	98.06	99.99
Zr	ppm	101.45	59.80	146.50
Si	ppm	77.60	40.00	130.00
Y	ppm	6.05	5.00	10.00
Fe	ppm	133.13	39.90	574.00
Cr	ppm	28.15	15.00	115.00
Ni	ppm	43.14	30.00	68.80
Mo	ppm	40.21	10.00	75.00
Mn	ppm	79.47	13.80	190.00
Ru	ppm	77.23	40.00	130.00
Cd	ppm	12.17	5.00	20.00
Al	ppm	101.24	20.00	285.00
Tc	ppm	75.00	65.00	85.00
Li	ppm	15.60	5.00	40.00
K	wt. %	0.05	0.01	0.15
Na	ppm	82.29	35.00	200.00
Ba	ppm	5.00	5.00	5.00
Sr	ppm	5.00	5.00	5.00
Sr90	ppb	15.77	0.31	62.50
Nd	ppm	95.92	25.00	200.00
Sm	ppm	56.82	20.00	160.00
Tc99	ppm	0.15	0.06	0.28

Analyte	Units	Weighted Average	Minimum	Maximum
Cs135	ppm	2.67	2.00	3.00
Mn54	ppt	3.04	0.04	12.52
Co60	ppt	27.81	0.27	176.99
Nb95	ND	ND	ND	ND
Zr95	ND	ND	ND	ND
Rh106	ND	ND	ND	ND
Ru106	ND	ND	ND	ND
Sb125	ppt	102.51	2.88	480.77
Cs134	ppt	24.99	0.23	153.85
Cs137	ppb	8.00	0.55	22.38
Ce144	ppt	67.11	1.88	313.48
Eu154	ppb	0.22	0.00	1.11
Eu155	ppb	0.22	0.00	1.04
Am241	ppb	61.23	2.92	291.55
U234	iso. %U	0.16	0.16	0.18
U235	iso. %U	19.28	18.92	19.42
U236	iso. %U	0.52	0.49	0.56
U238	iso. %U	79.76	79.03	80.03
Np237	ppm	17.11	13.90	21.40
Pu239	ppm	83.57	58.73	103.00
Pu240	ppm	2.24	1.46	2.71
Total Pu	ppm	NM	NM	NM

ppm = parts per million, by mass
ppb = parts per billion, by mass
ppt = parts per trillion, by mass
iso. % U = isotope wt.% of total U
ND = Not detected
NM = Not measured; total Pu was not measured. Np237, Pu239, and Pu240 isotopes are shown as ppm of material mass.
NOTE:

- The elemental impurities consist of isotopes in their natural isotopic abundance based on cooling time and the original source of the elements.
- Elemental Sr and Ba were below the minimum detection limit (MDL) of 5 ppm.
- Measurements below the MDL are listed at the MDL for conservatism.