INL/EXT-18-51906-Revision-1

Isotopic Characterization of HALEU from EBR-II Driver Fuel Processing

June 2021

DeeEarl Vaden

Changing the World's Energy Future

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/EXT-18-51906-Revision-1

Isotopic Characterization of HALEU from EBR-II Driver Fuel Processing

DeeEarl Vaden

June 2021

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Isotopic Characterization of HALEU from EBR-II Driver Fuel Processing

1. Effective Date	04/23/20	**Professional Engineer's Stamp**
2. Does this TEV involve a Safety SSC?	No	**N/A**
3. Safety SSC Determination Document ID	N/A	
4. SSC ID	N/A	
5. Project No.	----	
6. Engineering Job (EJ) No.	N/A	
7. Building	765	
8. Site Area	MFC	

9. Objective / Purpose

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

10. If revision, please state the reason and list sections and/or page being affected.

Modified Appendices B and C to reflect the results of the nine driver reguli analyzed to date in place of six selected FCF U-5Fs ingots.

11. Conclusion / Recommendations

Provide enough information to perform an isotopic characterization of HALEU produced at Idaho National Laboratory:

- The analytical results of driver reguli samples provide enough information of the measured nuclides.
- Isotopic distributions based on ORIGEN calculations coupled with the analytical results provide the characterization for the non-measured nuclides.

CONTENTS

APPENDIXES

PROJECT ROLES AND RESPONSIBILITIES

Project Role	Name	Organization	Pages Covered (if applicable)
Performer	Dee Earl Vaden	C420	See eCR 677014
Checker[a]	Beau Ballard	U410	See eCR 677014
Independent Reviewer[b]	Brian Preussner	C420	See eCR 677014
CUI Reviewer[c]	Travis Killian	U940	See eCR 677014
Manager[d]	Michael N. Patterson	U600	See eCR 677014
Requestor[e]	Michael N. Patterson	U600	See eCR 677014
Nuclear Safety[f]	N/A	N/A	N/A
Document Owner	Michael N. Patterson	U600	See eCR 677014
Reviewer	Michael N. Patterson	U600	See eCR 677014

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. Authorizes the commencement of work of the engineering deliverable. See Appendix A.
f. Concurrence with the document's assumptions and input information. See definition of Acceptance, LWP-10200.

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.*

SCOPE AND BRIEF DESCRIPTION

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 via process modeling software and related programs.

ADDITIONAL INFORMATION

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 most of the fission products along with their 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 ^{235}U enrichment of less than 20%. Using a specially designed “double-stack” crucible in a high temperature furnace produced several ingots between 3 and 4 kg with some under 2 kg. Drill fines sent to the Analytical Laboratory for analyses provided the measured concentrations of some isotopes of interest (e.g., ^{234}U, ^{235}U, ^{236}U, and ^{238}U) which characterizes the recovered uranium metal in these reguli. This approach for characterization has been conducted on seven reguli batches analyzed to date and is anticipated to continue for the remaining EBR-II spent fuel identified for future treatment. While this method provides an accurate assessment of the inventory based on the analyzed isotopes, complete characterization necessitates the development of a method to determine the concentration of the non-measured isotopes (e.g., ^{232}U) as well. The data accumulated from this inventory provides the basis for a bounding approximation that characterizes the HALEU material generated from this inventory.

ACRONYMS

AL	Analytical Laboratory
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

ASSUMPTIONS

- The sample results include the measured uranium isotopes ^{234}U, ^{235}U, ^{236}U, and ^{238}U.
- The ORIGEN output and FCF process models provide the unmeasured uranium isotopes.

REFERENCES

1. C. H. Adams, et. al., "*The Mass Tracking System - Computerized Support for MC&A and Operations at FCF*" Proceedings of the Embedded Topical Meeting on DOE Spent Nuclear Fuel & Fissile Material Management, Reno, NV, June 1996, p. 369.

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.

4. G. G. Galbreth, "*GENERATING ISOTOPE-Z (ISOZ) FILES FOR MK-IA AND MK-II GENERATION FUEL ASSEMBLIES*", ECAR-1747, January 2012.

5. M. J. Bell, "*ORIGEN — The ORNL Isotope Generation and Depletion Code*," ORNL 4628, Oak Ridge National Laboratory (May 1973).

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

Appendix A

Engineering Inputs

Table A-1 lists the AL log numbers used to generate the data tables in the appendices.

Table A-1. Analytical log numbers used for each Regulus batch

Batch	AL Log number(s)
CPDC003	103567, 103568, 103569
CPDC004	103550, 103551, 103552
CPDC005	103808, 103809
CPDC006	103852, 103853, 103854
CPDC007	103913, 103914, 103915
CPDC008	104009, 104010, 104011
CPDC009	104007, 104008

Because AL did not report the isotopes ^{232}U, ^{233}U, and ^{237}U in the sample analyses, their concentrations were estimated by multiplying the predicted uranium isotope distribution in the reguli by the ratio [measured ^{238}U / estimated ^{238}U] as illustrated in Table A-2 for reguli batch CPDC009, the last batch to date with AL sample results. The relative deviation is calculated from the two grey cells in each row.

Table A-2. Uranium Isotopic Distribution in Regulus from Batch CPDC009

Nuclide X	Mass Fraction Measured gX / gSample	Mass Fraction Estimated gX / gRegulus	Ratio	Mass Fraction Final gX / gRegulus	Relative Deviation	Final Isotope Distribution gX / gU
^{234}U	1.700E-03	1.717E-03		1.700E-03	0.000E+00	1.700E-03
^{235}U	1.969E-01	1.975E-01		1.969E-01	0.000E+00	1.969E-01
^{236}U	5.300E-03	5.360E-03		5.300E-03	0.000E+00	5.301E-03
^{238}U	7.960E-01	7.955E-01		7.960E-01	0.000E+00	7.961E-01
^{232}U		3.154E-10	1.001E+00	3.156E-10	6.166E-04	3.157E-10
^{233}U		8.281E-08	1.001E+00	8.286E-08	6.166E-04	8.288E-08
^{237}U		2.856E-13	1.001E+00	2.858E-13	6.166E-04	2.859E-13

Appendix B

HALEU Uranium Assay Seven Batches of U-5Fs Reguli

Analyte	Units	Average	Standard Deviation	Minimum	Maximum
Total U*	wt. %	99.572	0.980	98.043	100.371
^{234}U*	iso % U	0.169	0.009	0.158	0.179
^{235}U*	iso % U	19.655	0.194	19.346	19.892
^{236}U*	iso % U	0.518	0.016	0.493	0.538
^{238}U*	iso % U	79.658	0.209	79.390	79.983
^{232}U^	ng / gU	0.287	0.172	0.150	0.641
^{233}U^	ng / gU	75.771	12.170	53.175	84.067
^{237}U^	pg / gU	0.316	0.056	0.236	0.386

* Actinide isotopes measured analytically
^ Not in AL reports, so estimated using the method described in Appendix A
iso. % U = isotope wt.% of total uranium
ng / gU = g per billion grams of uranium
pg / gU = g per trillion grams of uranium

Appendix C

Measured Composition of Seven Batches of U-5Fs Reguli

The subsequent table contains the concentration of the non-uranium nuclides measured in the seven reguli batches. See Appendix B for the concentration of uranium isotopes.

Analyte	Units	Average	Standard Deviation	Minimum	Maximum
Zr	ppm	79.47	26.05	45.30	110.00
Si	ND	ND	ND	ND	ND
Y	ppm	28.31	22.67	3.91	80.80
Fe	ppm	181.10	184.32	38.80	785.00
Cr	ppm	14.25	2.47	12.50	16.00
Ni	ND	ND	ND	ND	ND
Mo	ND	ND	ND	ND	ND
Mn	ppm	42.84	24.79	7.28	94.30
Ru	ND	ND	ND	ND	ND
Cd	ND	ND	ND	ND	ND
Al	ppm	74.25	24.44	46.60	103.00
Tc	ND	ND	ND	ND	ND
Li	ND	ND	ND	ND	ND
K	wt. %	ND	ND	ND	ND
Na	ND	ND	ND	ND	ND
Ba	ND	ND	ND	ND	ND
Sr	ND	ND	ND	ND	ND
Sr90	ppb	4.49	4.37	0.32	12.54
Nd	ND	ND	ND	ND	ND
Sm	ND	ND	ND	ND	ND

Analyte	Units	Average	Standard Deviation	Minimum	Maximum
Tc99	ppm	0.29	0.08	0.22	0.38
Cs135	ppm	0.03	0.03	0.01	0.07
Mn54	ND	ND	ND	ND	ND
Co60	ND	ND	ND	ND	ND
Nb95	ND	ND	ND	ND	ND
Zr95	ND	ND	ND	ND	ND
Rh106	ND	ND	ND	ND	ND
Ru106	ND	ND	ND	ND	ND
Sb125	ND	ND	ND	ND	ND
Cs134	ND	ND	ND	ND	ND
Cs137	ppb	6.30	9.45	0.78	34.68
Ce144	ND	ND	ND	ND	ND
Eu154	ND	ND	ND	ND	ND
Eu155	ND	ND	ND	ND	ND
Am241	ppb	6.32	2.26	4.37	10.50
Np237	ppm	16.32	4.41	5.20	21.34
Pu239	ppm	81.52	39.42	8.63	128.00
Pu240	ppm	2.30	1.11	0.33	3.61
Total Pu	NM	NM	NM	NM	NM

ppm = parts per million, by mass
ppb = parts per billion, by mass
ppt = parts per trillion, by mass
ND = Not detected or below the minimum detection limit
NM = Not measured; total Pu was not measured. Np237, Pu239, and Pu240 isotopes are shown as ppm of material mass.