Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

We put science to work.™

Savannah River National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

A U.S. DEPARTMENT OF ENERGY NATIONAL LABORATORY • SAVANNAH RIVER SITE • AIKEN, SC

Feasibility of Processing the Experimental Breeder Reactor-II Driver Fuel from the Idaho National Laboratory through Savannah River Site's H-Canyon Facility

V. E. Magoulas

July 2017

SRNL-STI-2017-00420, Revision 0

SRNL.DOE.GOV

DISCLAIMER

Printed in the United States of America

Prepared for
U.S. Department of Energy

Keywords: *EBR-II*

Retention: *Permanent*

Feasibility of Processing the Experimental Breeder Reactor-II Driver Fuel from the Idaho National Laboratory through Savannah River Site's H-Canyon Facility

V. E. Magoulas

July 2017

Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

Savannah River National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

Acknowledgements

This author would like to acknowledge Bill Clifton, H-Area Technical Support; Glynn Dyer, Mission Development/Engineering Support; Dan Leduc, Packaging Technology; and SRNL's Separations and Actinide Science group for their support in this study.

LIST OF ABBREVIATIONS

DU	depleted uranium
EBR-II	Experimental Breeder Reactor II
HEU	highly enriched uranium
INL	Idaho National Laboratory
LEU	low enriched uranium
NCSA	nuclear criticality safety assessment
NCSE	nuclear criticality safety evaluation
SNF	spent nuclear fuel
SRNL	Savannah River National Laboratory
U	uranium
U-5Fs	uranium with 5% "fissium"
USQ	unreviewed safety question
Zr	zirconium

1.0 Introduction

Savannah River National Laboratory (SRNL) was requested to evaluate the potential to receive and process the Idaho National Laboratory (INL) uranium (U) recovered from the Experimental Breeder Reactor II (EBR-II) driver fuel through the Savannah River Site's (SRS) H-Canyon as a way to disposition the material. INL recovers the uranium from the sodium bonded metallic fuel irradiated in the EBR-II reactor using an electrorefining process. There were two compositions of EBR-II driver fuel. The early generation fuel was U-5Fs, which consisted of 95% U metal alloyed with 5% noble metal elements "fissium" (2.5% molybdenum, 2.0% ruthenium, 0.3% rhodium, 0.1% palladium, and 0.1% zirconium), while the later generation was U-10Zr which was 90% U metal alloyed with 10% zirconium. A potential concern during the H-Canyon nitric acid dissolution process of the U metal containing zirconium (Zr) is the explosive behavior that has been reported for alloys of these materials. For this reason, this evaluation was focused on the ability to process the lower Zr content materials, the U-5Fs material.

The uranium is recovered from the irradiated sodium bonded metallic EBR-II fuel using an electrorefining process for separating the sodium from the uranium. The uranium is then converted from a highly enriched uranium (HEU) to a low enriched uranium (LEU) using a furnace to mix the molten HEU with other depleted uranium (DU) metals to result in a LEU ingot. Samples can be taken of the material during its molten state.

2.0 Results and Discussion

The scope of this study was to provide a detailed flowsheet that investigates the compatibility of the low enriched metallic uranium recovered from the treatment of irradiated sodium bonded fuel, with the chemical separation process employed in the SRS H-Canyon facility. A concern was raised over the presence of Zr in the recovered uranium material from the EBR-II fuel in that it may be problematic in the current configuration of the H-Canyon process. The flowsheet development focused on the EBR-II U-5Fs material. In addition the study was to improve the understanding of the physical form required for addition to the H-Canyon process, as well as packaging configuration and handling of the uranium from U-5Fs.

The SRNL evaluation consisted of two parts; one part was to evaluate the feasibility of a chemical processing flowsheet for H-Canyon to accept the uranium composition in conjunction with current H-Canyon spent nuclear fuel (SNF) disposition mission, and the second part centered on how the material would need to be packaged for receipt and handling at SRS for processing. For the flowsheet development evaluation, SRNL received four 25 gram samples representative of the highest potential Zr content in the U-5Fs material and performed dissolution, chemical analysis, and characterization of the material. H-Canyon Technical Support and SRNL developed a strategy for integrating the dissolving of the INL LEU in H-Canyon along with the SNF. The strategy would be to dissolve multiple spent fuel charges first, then add the EBR II material in the final charge. SRNL performed experiments to demonstrate this flowsheet. The results of the H-Canyon flowsheet compatibility are documented in *Dissolution of Low Enriched Uranium from the Experimental Breeder Reactor II Fuel Stored at the Idaho National Laboratory,* SRNL-STI-2017-00263, Rev. 0, Savannah River National Laboratory, Aiken, SC (April 2017) reference 1. There will be MC&A issues to resolve with this strategy.

Part two of the study dealt with packaging and handling of the packaged material. Based on initial radiation dose rates, the thought was to receive in leak tight containers as a cask shipment to L basin, where the material would then be loaded into L bundles for transport to H-Canyon for dissolution. Based on the SRNL dose rate measurements on the initial samples, the beta component was higher than the gamma, and therefore would be shielded if packaged in a shipping container. Given this information, the approach for packaging and receipt of the material changed to using carbon steel cans within a Type B shipping container. INL would package the material in a “can-bag-can” configuration, using only carbon steel cans and nylon bagging material in an approved Ttype B shipping container. INL packaged ~2.7 kg of LEU in the form of small diameter rods into a Gavin can to collect additional rad dose measurements. The values recorded by INL were around 250 mrem/hr on contact. The containers could be received into H Area in the same manner that the plutonium metal was previously handled for the HB-Line plutonium oxide production mission. The cans would be removed from the shipping container in the Crane Maintenance vestibule, and then loaded into reusable charging bundles in the H-Canyon Crane Maintenance Area for charging to the dissolver. This LEU material would be the final charge in the SNF dissolver batch.

A material packaging evaluation was performed by SRNL. The evaluation included the 9977, 9975, and the ES-3100 containers. For the 9977 package, the concentrations of each radionuclide listed in the sample results are within the limits of content envelope AC.5 as stated in Appendix A. Although up to 18 kg of uranium can be shipped under envelope AC.5 of the 9977 Addendum 2, the actual amount to be shipped will depend on ensuring the dose rate at the exterior of the 9977 package remains at or below exclusive-use limits. The 9975 package has the same limits as the 9977 package content envelope AC.5, but will only allow 13.5 kg contents even though it has additional shielding. The ES-3100 content envelopes do not address all the isotopes found in the EBR II samples examined, therefore a new content envelope would have to be established in a content amendment in order to use this package.

3.0 Conclusions

It is feasible for the INL U-5Fs uranium material to be processed through SRS as a potential disposition path[1]. Although Zr content in the material was an original concern, the processing showed that the U-5Fs EBRII material is bounded by a Zr content of 1000 µg Zr/g U. Another concern is the presence of chloride salts from the INL electrorefiner which are associated with the LEU ingots as a potential H-Canyon corrosion concern. The samples showed less than the detection limit for chloride in solution, but the samples may not be a good representation due to the casting process. An acceptable flowsheet and dissolution strategy were developed for processing the INL U-5Fs uranium material from the EBR-II in conjunction with the SNF processing at H-Canyon. A packaging and handling approach was also established on condition that the radiation dose rates are ~250 mrem/hr on contact after packaging as provided by INL.

4.0 Recommendations, Path Forward or Future Work

If the decision is made for the EBR-II U-5Fs material to be dispositioned through H-Canyon, the following work, at a minimum, would be required prior to any potential receipt of this material to SRS:

- Establish a Shipper/Receiver Agreement to include MC&A confirmation and verification requirements
- Review of the Shipping Package Qualification in the Storage Surveillance program for potential storage in K-Area. A review of the safety basis evaluation (USQ, NCSE, and NCSA) would be required with potential changes.

Also there is a potential for an additional study to try to include the U-10Zr EBR II material in this possible disposition path. Based on the test results for the Zr content in the U-5Fs samples, there may be a potential to receive and process the U-10Zr material too, but additional testing would be required.

5.0 Reference

1. W. E. Daniel, T. S. Rudisill, P. M. Almond, and P. E. O'Rourke, *Dissolution of Low Enriched Uranium from the Experimental Breeder Reactor II Fuel Stored at the Idaho National Laboratory,* SRNL-STI-2017-00263, Rev. 0, Savannah River National Laboratory, Aiken, SC (April 2017).

Appendix A.

ES3100 and 9977 Content evaluation for EBRII materials

3.0 PACKAGING OPTIONS

3.1 Model 9977 Package

The 9977 package is licensed in the U.S. for the shipment of uranium metal and small concentrations of various fission products. The model 9977 has a single containment vessel with a 6.065 inch diameter and a usable length of 20.25. EBR II ingots can be cast to fit into the 9977 PCV and to ensure that the dose rate at the surface of the package is less than 200 mrem/hr. The concentrations of the Isotopes of interest for shipping in sample EXP 103-Idaho-LEU-S28-4A-U-253 are listed in Table 1. The concentrations of the Isotopes of interest for shipping in sample EXP 104-Idaho-LEU-S28-4A-U-253 are listed in Table 2.

Table 1 EBR II SIGNIFICANT ISOTOPES in EXP 103.

Isotope	Weight [g]	Wt %	ppm
Co-60	1.00E-12	2.77E-11	2.76875E-07
Sb-125	2.69E-11	7.44E-10	7.44E-06
Cs-137	4.87E-08	1.34E-06	1.34E-02
Tl-208	9.12E-17	2.52E-17	2.52E-13
Eu-155	-	-	-
Bi-212	5.68E-15	1.57E-13	1.57E-09
Pb-212	5.99E-14	1.65E-12	1.65E-08
Ra-224	4.35E-13	1.20E-11	1.20E-07
Pa-233	2.23E-12	6.15E-11	6.15E-07
Pa-234m	1.28E-15	3.52E-14	3.52E-10
Th-234	2.57E-11	7.09E-10	7.09E-06
U-235	5.55E-01	1.53E+01	1.53E+05
Np-237	6.57E-05	1.81E-03	1.81E+01
Am-241	9.40E-09	2.59E-07	2.59E-03
U-238	3.62	-	-

Table 2 EBR II SIGNIFICANT ISOTOPES in EXP 104.

Isotope	Weight [g]	Wt %	ppm
Co-60	1.36E-12	3.07E-11	3.07E-07

Sb-125	3.20E-11	7.20E-10	7.20E-06
Cs-137	3.55E-08	8.00E-07	8.00E-03
Tl-208	1.33E-16	2.99E-15	2.99E-11
Eu-155	1.75E-10	3.94E-09	3.94E-05
Bi-212	8.44E-15	1.90E-13	1.90E-09
Pb-212	8.99E-14	2.03E-12	2.03E-08
Ra-224	8.13E-13	1.83E-11	1.83E-07
Pa-233	3.26E-12	7.34E-11	7.34E-07
Pa-234m	1.46E-15	3.30E-14	3.30E-10
Th-234	3.12E-11	7.04E-10	7.04E-06
U-235	6.57E-01	1.48E+01	1.48E+05
Np-237	9.62E-05	2.17E-03	2.17E+01
Am-241	1.10E-08	2.49E-07	2.49E-03
U-238	4.44	-	-

Assuming the g/g-U column in the ingot information file (x 100) is a true weight % of the material, the part per million (ppm) concentrations of the isotopes are shown in Tables 1 and 2. These concentrations are well below the 1000 ppm limit for each isotope in both samples. This limit is specified in section 1.2.2 of the contents section of Addendum 2 of the 9977 SARP (The exception is U-235, which is considered part of the Uranium metal as opposed to an impurity). Content envelope AC.5 of Table A.1.1 in Addendum 2 for this SARP establishes a limit of 18 kg for Uranium metals consisting of up to 95% U-235.

A scoping shielding analysis indicates that moving the package with 18 kg of the EBR II samples would result in a contact dose rate of 5 to 10 mrem/hr at the 9977 package side. Therefore, amount of material that can be shipped in 9977 packages under the AC.5 Content envelope will be based on actual dose rates.

3.2 Model 9975 Package

The 9975 package is licensed in the U.S. for the shipment of plutonium and uranium metal and oxide. It has two containment vessels each with a greater than 0.25 inch thick wall and a lead shield with a minimum thickness of 0.423 inches. The inner containment vessel designated the Primary Containment Vessel (PCV) has a cavity that is approximately 5.03 inches in diameter with a minimum usable length of 15.1 inches. 9975 content envelope C.1 allows for 13.5 kg of uranium up to 100% enrichment with wt% limits for U-232, U-233 and U-236 of 1x10-5%, 0.5% and 40% respectively. All other isotopes are restricted to 1000ppm (same as 9977) so that both samples in Tables 1 and 2 are compliant with the C.1 content envelope. Since the 9975 has a ½ inch thick lead gamma shield whereas the 9977 has no shielding, dose rates on the exterior of the 9975 with 13.5 kg contents are expected to be less than the 9977.

3.3 Model ES-3100 Package

The ES-3100 is licensed in the U.S. for the shipment of uranium metal and oxide as well as plutonium oxide in 3013 containers under specific conditions. It has a single thin wall vessel with an internal cavity which is 5.06 inches in diameter and 31 inches long and has no gamma shielding. Current ES-3100 content envelopes do not address the radionuclides included in the EBR II samples. Therefore, in order to ship the EBR II material in the ES-3100 a new content envelope would have to be established in a content amendment.