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Conversion of EBR-II Fuel to HALEU Oxide Pellets

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Idaho Falls, Idaho 83415

http://www.inl.gov

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Leah Squires,* Cynthia Adkins,* Colt Heathman,* Beau Ballard*

* *Idaho National Laboratory, P.O. Box 1625 Idaho Falls, ID 83415.* leah.squires@inl.gov

INTRODUCTION

High-assay low-enriched uranium (HALEU) is an important component of fuel for many next generation advanced nuclear reactor concepts that are being designed. Presently, the United States does not have the infrastructure or enrichment capacity to keep up with HALEU demand for these reactors. Downblending high-enriched uranium (HEU) is one method whereby some of the needed HALEU can be produced domestically. Here we describe a process for repurposing HEU recovered from Experimental Breeder Reactor II (EBR-II) driver fuel at Idaho National Laboratory (INL) to HALEU product suitable for fuel fabrication. The objective of this study is to better describe the thermochemical process chemistry for the conversion of uranyl peroxide hydrate ($UO_4 \cdot 4\ H_2O$) into uranium dioxide (UO_2), which involves heating the $UO_4 \cdot 4\ H_2O$ precursor in a 6% H_2/Ar atmosphere as shown in the solid/gas reaction in Eq. 1.

(1)

RESULTS

A laboratory-scale HALEU polishing process consisting of dissolution, solvent extraction, and precipitation was developed in cooperation with Pacific Northwest National Laboratory (1,2). This process removes fission products, plutonium, neptunium, and metallic impurities from material obtained from the drip cast process used to prepare metal uranium for EBR-II fuel fabrication Fig.1. The metal material was first converted to liquid uranyl nitrate then purified via a modified UREX solvent extraction process developed for this work. Upon the addition of hydrogen peroxide the material was precipitated as uranyl peroxide tetrahydrate ($UO_4 \bullet 4H_2O$).

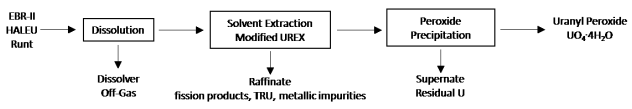

Fig. 1. Polishing process for $UO_4 \bullet 4H_2O$ production.

This material was then converted to UO_2 by heating under reducing atmosphere. Differential thermal analysis (DTA) along with thermogravimetry (TG) was performed under 6% H_2/Ar atmosphere to monitor the oxygen content of the material while identical heating runs were performed simultaneously in a larger scale furnace located in an inert atmosphere glovebox. After conversion to UO_2 the material was pressed into pellets and sintered. Fig. 2 shows the progression of the material from $UO_4 \bullet 4H_2O$ powder to UO_2 powder to UO_2 pressed pellets to UO_2 sintered pellets.

Fig. 2. Progression of material from $UO_4 \bullet 4H_2O$ to pressed and sintered UO_2 pellets.

DTA/TG data in Fig. 3 shows the loss of water molecules followed by oxygen loss during the process as decreasing steps in the TG curve along with associated peaks in the DTA data.

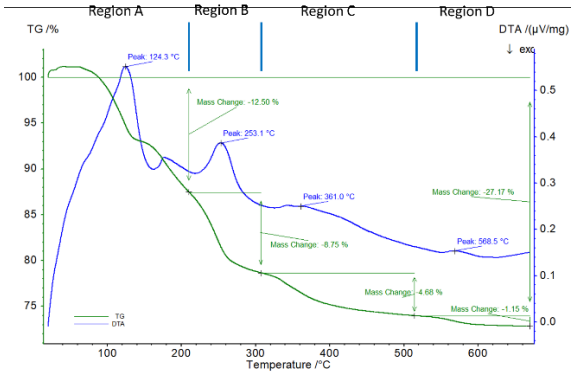

Fig. 3. TG and DTA signals (uV/mg) of uranyl peroxide during calcination from 25-670°C. Heating was performed at a rate of 10°C/min. The regions represent temperature ranges for loss of water and oxygen.

TG/DTA data has been used to monitor similar reactions (3). The TG/DTA data shows waters of hydration lost in Region A, conversion of UO_4 to UO_3 in region B,

conversion of UO_3 to U_3O_8 in region C and conversion of U_3O_8 to UO_2 in region D. Identification of these conversions was supported by the associated mass reduction at each step. Step 1 was associated with a 10.06% mass reduction, step 2 was associated with a 10.91% mass reduction, step 3 was associated with a 3.23% mass reduction and step 4 was associated with a 1.07% mass reduction. A second heat performed on post test material from the first heating cycle showed a -0.09% mass reduction indicated that the material was fully reduced during the first heating sequence. Identical heating runs were performed on the bulk material to obtain enough UO_2 to press into pellets.

DISCUSSION

The process described produced HALEU oxide pellets from EBR-II fuel. TG/DTA data was used to monitor the reduction and ensure the product was taken to UO_2. Further development of this process includes the production of HALEU metal from these pellets using a direct oxide reduction with calcium metal as a reducing agent. Analysis of the HALEU metal produced in this process is pending and results should be forthcoming. Due to the high demand for HALEU and its limited availability, methods of retrieving usable forms HALEU from existing stockpile materials are of great importance. This demonstration shows a path forward for producing HALEU material from existing uranium bearing fuel.

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