
A Novel and Flexible Approach for Converting LWR UNF Fuel into Forms that can be used to Fuel a Variety of Gen-IV reactors

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ABSTRACT:

The recycling of used nuclear fuel rods from light water reactors (LWRs) throughout the world has been identified as one of the priority research needs in the MRWFD campaign of the DOE.[1] The applicant/PI (Craig Barnes; University of Tennessee at Knoxville) and coPI G. D. (Bill) DelCul, (ORNL), describe a novel proposal to digest whole MOX-based fuel rods and obtain three product streams: 1) Pure decontaminated $ZrCl_4$; 2) pure UCl_4 and 3) a stream containing TRU/FPs as well as alloying metals (as chloride salts). The key elements of this strategy are first, the ability of thionyl chloride to convert metals and metal oxides into metal chloride salts and second, the propensity of many of these salts to cleanly crystallize from thionyl chloride. The objectives of this proposal are 1) to investigate and understand the reactions of thionyl chloride with depleted uranium dioxide and lanthanide surrogates for TRU and fission products (e.g. UO_2 , SrO , Sb_2O_3 , Cs_2O , La_2O_3 , Pr_2O_3). All reaction products will be identified and conditions by which reactions can be made to go to completion will be determined. The solubilities of the respective chloride salts in thionyl chloride will be determined. 2) The reaction kinetics will be studied to determine if different chlorination reactions proceed at different rates so that a kinetic resolution of products may be possible in our purification strategies. Finally, the third objective is to use the understanding gained in objectives (1) and (2) to develop a protocol and design a reactor to demonstrate the digestion of a fuel rod “simulant” and subsequent isolation of the three major product streams described above. The tasks of the proposed work parallel the objectives exactly. Task 1 will be to investigate and reactions of the metal oxides above and identify all products and yields. The solubilities of the chloride salts are also important to the success of this proposal and therefore will be investigated. Task 2 focuses on the kinetics of the reactions of thionyl chloride with the oxides above. Task 3 focuses on designing a purification protocol around the results from tasks (1) and (2) whereby the components of whole fuel rods may be transformed into a metal chlorides and separated from one another, essentially in one step. The successful achievement of the objectives of this project would provide a new, highly efficient protocol for the transformation of used nuclear fuel into 1) pure, decontaminated $ZrCl_4$ that may be reduced and reused as nuclear grade zirconium metal; 2) UCl_4 that could be used in some Gen-IV reactor designs (or converted to fluoride salts) and 3) a concentrated chlorinated TRU/FP stream that would be amenable to current electrochemical reconditioning schemes and thus also be useful as fuel components in Gen-IV reactors. Finally, the protocol we seek to develop effectively contains all radioactivity into a single reactor from which a concentrated stream of highly radioactive materials may be appropriately handled depending on composition and activity.

1. J. Law; T. Todd; Soelberg, N.; C. Pereira; Williamson, M.; Ebert, W.; Willit, J.; Jubin, R.; Vienna, J.; Lumetta, G.; Rudisill, T. *Prioritized List of Research Needs to support MRWFD Case Study Flowsheet Advancement*; ORNL: 2015, FCRD-MRWFD-2015-000571.