ANODIC PROCESS OF ELECTROREFINING SPENT NUCLEAR FUEL IN MOLTEN LICl-KCl-UCl_3

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ABSTRACT

The need for the treatment of spent nuclear fuel has increased substantially over the last decade. Several methods of treatment are being investigated throughout the nuclear industry [1-3]. Argonne National Laboratory (ANL) has developed and demonstrated an electrometallurgical process for the Department of Energy (DOE) to treat sodium bonded spent nuclear fuels [4]. This process has been performed on irradiated fuel elements from the Experimental Breeder Reactor (EBR-II). One of the key steps in the demonstration was electrorefining of spent driver fuel in a molten LiCl-KCl-UCl_3/liquid cadmium system using a pilot scale electrorefiner (Mark-IV ER). The Mark-IV ER is located in the Fuel Conditioning Facility at the ANL-West site in Idaho.

The spent driver fuel consists of uranium, zirconium, bond sodium, and fission products. The primary purpose of the electrorefining was to separate the uranium from the other fuel components. For this purpose, the spent fuel elements with cladding were chopped into segments of 0.635 cm in length and placed into fuel dissolution baskets (FDBs). During the electrorefining, the FDBs filled with chopped fuel segments were the anode and a steel mandrel was the cathode. The bond sodium and fission products, which are thermodynamically more active than the uranium metal, chemically reacted with and displaced the UCl_3 from the molten salt. Uranium was then electrochemically dissolved from the fuel segments and deposited onto the cathode. Zirconium and fission products, which are thermodynamically nobler than uranium, were ideally retained within the FDBs with cladding hulls. Eventually, the cladding hulls, zirconium, and noble metal fission products will be solidified into metal waste for long term disposition. Therefore, the process goals for the anodic process were to maximize the dissolution of uranium and to retain most of the noble metals.

This article summarizes the experimental results and engineering aspects of the anodic process for electrorefining of 100 spent driver assemblies (the demonstration project). The focus is on the anode due to its unique geometry (FDBs loaded with chopped irradiated fuel segments), complex chemical compositions, high demanding process goals, and their significance to the entire spent fuel treatment process. Chemical analysis results of cladding hull samples were used as the key criteria to evaluate the effectiveness of the uranium dissolution and noble metal retention. Parametric study indicated that the diffusion of U^{3+} in the porous fuel matrix was the controlling step to the uranium dissolution from the chopped fuel segments. The chopping length of the fuel segments and anode rotating rate affected the uranium dissolution rate. Anode resistance was the most effective parameter indicating completeness of uranium dissolution and noble metal retention. The experience and data gained from the demonstration project are important since the Department of Energy (DOE) has decided to apply this technology to treat the remaining EBR-II and other sodium bonded spent fuels [5].

REFERENCE

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