Analysis of Electrolytic Reduction Behavior by Oxide Fuel Characteristics

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Accumulation of spent nuclear fuel from nuclear power plants is a core challenge in nuclear energy technology. Pyroprocessing technology has been developed for recycling of the spent fuel into metal fuel for fast nuclear reactors by converting the spent oxide fuel into U/TRU(uranium/transuranium) metal ingots via electrochemical processes utilizing high-temperature molten salt. These electrochemical processes employ electrolytic reduction (also referred to as oxide reduction, OR) for the reduction of spent oxide fuel to metal, and electrorefining for the recovery of U/TRU [1–9].

Generally, molten LiCl containing Li_2O at 650°C is used as the electrolyte in the OR process. The spent oxide fuels, loaded in a permeable basket, and platinum (Pt) are used as the cathode and anode, respectively. The cathode reactions (1–3) can be summarized as follows:

$$Li^+ + e^- \rightarrow Li \quad (1)$$

$$MO_2 + 4Li \rightarrow M + 2Li_2O \ (\textit{salt phase}) \quad (2)$$

$$MO_2 + 4e^- \rightarrow M\ (\textit{actinide}) + 2O^{2-}\ (\textit{salt phase}) \quad (3)$$

Li_2O produced by reaction (2) in molten LiCl dissociates into Li^+ and O^{2-}:

$$Li_2O \rightarrow Li^+ + O^{2-}\ (\textit{salt phase}) \quad (4)$$

The anode reaction can be described as follows:

$$O^{2-}\ (\textit{salt phase}) \rightarrow O_2\ (\textit{gas}) + 4e^- \quad (5)$$

When an electrical potential is applied, the actinide metal oxide is reduced to metal and remains at the cathode. The oxygen ions (O^{2-}) produced at the cathode are transported through the salt and discharge at the anode to form O_2 gas [10–16]. Hence, the diffusion of O^{2-} ions from the inside of the oxide fuel to the bulk salt affects significantly the reduction rate and current efficiency during the OR process [17]. In this presentation, two previous reports [18,19] on analysis of electrolytic reduction behavior by oxide fuel characteristics will be summarized.

In the first study [18], we investigated complete OR of high-density UO_2 to metallic U without any remaining UO_2. The size and density of the used UO_2 pellets were ϕ 8.3 mm × 9.9 mm (H) and 10.67 g/cm^3, respectively, i.e., values that are similar to those of spent fuel pellets from pressurized water reactor. In the second study [19], We show that the reduction of REOs to rare earth metals in Li_2O–LiCl salt can be enhanced significantly by using lithium metal. Specifically, REOs in the fuel are reduced to a high extent in the electrolytic reduction of simulated oxide fuel in 1.0-wt% Li_2O–LiCl salt following the application of electrical charge in order to induce the production of Li metal.

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