Pyroprocessing: Bridging the Gap between Current Challenges and Future Innovations

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Pyroprocessing, as a critical field in actinide separations, plays a pivotal role in nuclear fuel cycles, waste management, and materials science. It encompasses the crucial steps of converting oxides to metals and the purification of metals. Pyroprocessing has been applied to used nuclear fuel (UNF) and plutonium processing flowsheets. In both applications, the core purification process is electrorefining. Due to significant resources required to perform electrorefining of actinide, innovation has been risky, costly, and limited. The innovation of electrorefining and other pyrochemical processes can be accelerated by reducing risk and cost in three ways: (1) development of electrochemical sensors and models, (2) maturing surrogate systems, and (3) scaling down operations.

Electrochemical Sensors and Models. Electrochemical sensors can improve our understanding of the behavior of actinides in molten salts, which can drive innovative approaches and designs for pyrochemical processes. However, due to the necessitated use of macro electrodes (i.e., high currents) and the prevalence of high concentrations in molten salts, the electrochemical behavior departs from classical electrochemistry models. For instance, the widely used Berzins-Delahay model is derived for the scenario of diffusion only, negligible ohmic resistance and pure metal deposits, which does not fully capture the intricacies of molten salt systems. To address these shortcomings, a physics-based model was developed specifically for electrodeposition in molten salts. The initial iteration of the model accounts for the combined effects of migration, ohmic resistance, deposition onto a foreign substrate, and activity of non-ideal solutions. Addition advancements in electrochemical measurements and analysis in molten salts are also discussed.

Maturing Surrogate Systems. The development of surrogate systems, while imperfect, offers a bridge to meaningful research experiences in low-risk testbeds for innovative approaches. Researchers can gain exposure to the complex phenomena occurring in electrorefining, explore molten salt chemistries, and design innovative processes without direct access to actinide materials. At BYU, we have successfully developed surrogates for electrorefining liquid metals in molten salts providing a low-risk, low-cost environment for initial testing of innovation in operation or process design. Some examples are highlighted including chlorine generation from molten chlorides, autonomous electrorefining, and dual-stage chloride volatility.

Scaling Down Operations. In addition to surrogates, BYU is exploring ways to reduce the scale of electrorefining and other electrochemical experiments involving actinides and molten salts. At smaller quantities, security, economic, and health risks are reduced, which enables more scientific studies of the electrochemical behavior and material properties of actinides. Using surrogates, the practical limit to scaling down electrorefining is being explored. Additionally, infrastructure is in place at BYU to perform pyrochemical experiments on sub-gram quantities of actinides.

The future of pyrochemistry hinges on our ability to deepen our understanding of chemistry of pyroprocessing operations while also inspiring and mentoring the next cohort of researchers. The development of surrogate systems, scaled-down studies, and physics-based models enable more opportunities for innovation in pyrochemical processing and greater engagement of students in the area.