NEUP Nuclear Energy University Program
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Project Title

Investigation into the processing parameters of phosphate-based dehalogenation for chloride-based waste salt

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Program: Fuel Cycle

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ABSTRACT:

In a recent DOE roadmap report on the development of the iron phosphate waste forms for salt waste stream treatment, several areas were identified that need to be developed to advance this approach towards a higher Technology Readiness Level (TRL).[3] The proposed research will focus on the following **objectives** to address the most critical needs, as described below.

Dechlorinate electrorefiner (ER) and molten salt reactor (MSR) salt simulants with various phosphate precursors (Task 1). The salt waste stream, the material used to immobilize portions of the partitioned salt waste, and the processing method to retain the radionuclides directly influence waste form consistency and properties. Dechlorination reaction rates and process flow sheets need to be developed for different waste streams and different phosphate precursors.

Collect glass property-composition data and develop models based on the glass-forming regions (Task 2). A glass-forming region for the iron phosphate compositions with suitable chemical durability has been identified; however, phase separation and crystallization are often observed upon canister centerline cooling. Data is needed on the thermal stability of these glasses and the effects of secondary phase development on durability. Glass forming ability-composition models need to be developed for different waste loadings and additives (e.g., Fe_2O_3, Al_2O_3), which are specific to each different salt composition; we plan to target 2-3 separate salt compositions for this work.

Determine how various crucible materials react with the phosphate products and byproducts under static and dynamic conditions (Task 3). The corrosive and fluid nature of phosphate melts leads to significant interactions with the crucibles during dehalogenation and vitrification into the final iron phosphate waste form. Additionally, unwanted crystal formation can occur, which can lead to a reduction in the waste form performance and processing issues. Evaluation of crucibles materials is needed to provide information for potential process equipment.

Evaluate the effect of processing variables on dechlorination efficacy and chlorine-bearing species in the off-gas (Task 4). The use of different phosphate precursors and processing variables (e.g., temperature, time, and gas flow) changes the properties of the intermediate phosphate glass (viz. before the addition of the iron) as complete dechlorination might not be achieved. Additionally, off-gas compositions are unknown, and potentially problematic, for some of the potential phosphate precursors. Process ranges need to be determined to understand key processing parameters.

Develop a process for reacting the recovered NH_4Cl with metals that need to be fed into the system (U, Na, Li, K, etc.) (Task 5). The secondary off-gas waste produced during processing must be managed and ideally recycled. Recycling will avoid the generation of a secondary waste stream containing Cl and provide a means by which to recharge the systems with UCl_3. This process is particularly needed for an MSR that may be enriched with costly Cl-37 or be contaminated with long-lived radioactive Cl-36 ($t_{1/2} = 3\times10^5$ y).