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Immobilization of High-Level Waste Salt in Dechlorinated Zeolite Waste Forms

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ABSTRACT:

Glass-bonded sodalite (GBS) is the baseline waste form for the immobilization of salt waste generated from electrorefining EBR-II spent fuel. However, this waste form is only capable of holding up to 7.5 mass% salt. It has been estimated that disposal of INL's ER salt waste into GBS would take more than five years of full-time hot cell operation and result in an estimated 23 – 51 metric tons of waste from only 25 metric tons of original fuel waste. The resulting high waste processing and disposal costs for completion of the EBR-II spent fuel treatment project would discourage commercial use of electrochemical processing for closing the commercial nuclear fuel cycle. Since over half of the mass of this electrorefiner salt is chlorine in the form of non-radioactive chloride ions, it is intriguing to consider dechlorination to assist in waste volume reduction. The challenge is to devise a chemical process for achieving this dechlorination in a molten salt containing numerous chloride compounds of varying thermodynamic stability. A novel approach for achieving this dechlorination via exchange of the metal ions into ultrastable H-Y (USHY) zeolite has recently been reported in the literature. Using this process, we have estimated that a 3× reduction in the final waste volume compared to the GBS wasteform could be achieved. Furthermore, waste processing equipment can be simplified, designed for high throughput operation, and even used to separate and recover long-lived I-129 from the salt. In order to develop this H-zeolite ion exchange process to be both practical and economical, two main research objectives have been established for this project:

1) Optimize dechlorination of electrorefiner salt. The recently published findings regarding reaction of LiCl-KCl with USHY have demonstrated chlorination via measured off-gas of HCl. Full exchange was not achieved in the reported study (i.e., ~50%). Therefore, we endeavor to pursue optimized exchange of H^+ with metal ions in the zeolite via selection of zeolite drying conditions, salt/zeolite mixing efficiency, and the temperature cycle. In a later phase of the project, simulated fission products will be added to the salt to verify that the process works effectively for a wide range of salt compounds. Experiments will also be performed with iodide salts to determine if this process can also be used to separate and collect radioactive I-129 from ER salt.

2) Synthesize a sintered waste form to immobilize the metal-exchanged zeolite. The metal-exchanged (dechlorinated) zeolite will be in powder or granular form, which is unsuitable for permanent geologic disposal due to the potential for dispersion in the environment. We will, thus, examine various glass formulations and sintering processes in which the glass serves as a binder for the metal-exchanged zeolite. The key challenge is to select glasses with low softening temperatures that are chemically durable and compatible with the zeolite particles.

A diverse team, comprised of university and national laboratory collaborators, will combine expertise in the areas of nuclear waste disposal and glass chemistry to develop and optimize this process.