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Sorbent regeneration, recycling, and transformation: A transformative approach to iodine capture and immobilization

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ABSTRACT:

Long-lived radioactive iodine (^{129}I) can be released into various off-gas streams during electrochemical or aqueous reprocessing of used nuclear fuel, as well as subsequent processes to immobilize salt waste from oxide reduction or actinide drawdown. Members of the Off-Gas Sigma Team under the Material Recovery and Waste Form Development Campaign under DOE-NE have identified areas where information is still needed based on known factors influencing the off-gas (e.g., type of facility, rate at which used fuel is processed, speciation and retention of volatile radionuclides in the process). One critical area still in need of research after decades of study is the advancement of the technology readiness level (TRL) of sorbents for iodine capture.

Whether a sorbent is required to physically trap (i.e., through physisorption) or chemically bind (i.e., through chemisorption) a radionuclide of interest, the complexity of the gas stream has a large impact on the performance (e.g., loading capacity, selectivity) and active life longevity of the sorbent. A solution to these performance and aging challenges is to examine the regeneration and recycling of sorbents. To understand the potential of this technology, the proposed research has two specific objectives:

Objective 1: Development of materials and processes for regeneration and recycling of sorbents

Based on the proposed flow sheets for pyroprocessing and aqueous reprocessing, several different off-gas streams will be generated. Therefore, having sorbents with tunable properties is critical for effective capture. Additionally, regeneration gas and temperatures could change depending on the sorbent. A combination of computational (electronic structure calculations, molecular simulation) and experimental studies will be conducted to understand (A) how the components in a primary off-gas stream interact with the getter (i.e., material that chemisorbs iodine) and the substrate (e.g., mordenite, binder) of a sorbent, and (B) how the initial capture off-gas stream (e.g., competing components, temperature) affects the regeneration lifetime of the sorbent. While zeolites will be the focus of the study, other materials may be included in the study if time permits and/or the performance of the selected zeolites is poor.

Objective 2: Transformation of iodine-loaded sorbents into stable waste forms

The formation of iodine-loaded sorbents into chemically and mechanically stable waste forms without the loss of semi-volatile radionuclides (e.g., I, Cs) is an enormous challenge that has been examined from a multitude of angles. Despite the extensive research in this area, there are still some unknowns regarding promising sorbents, especially for the secondary sorbents that would be used during regeneration (i.e., the release of iodine as HI(g) opens up the use for new materials previously unstudied). To feed empirical approaches with informed starting points, modeling approaches will be implemented. Studies will focus on formulating low-temperature binders and processing paths that could lead to chemically and mechanically durable phases. As immobilization of radioactive wastes requires a robust understanding of the phase stability in repository environments, waste forms will be irradiated to simulate ^{129}I decay.