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Apatite and sodalite based glass-bonded waste forms for immobilization of ^{129}I and mixed halide radioactive wastes

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ABSTRACT:

Owing to its long half-life (~1.6×10^7 y) combined with its high solubility in water (in anionic state) and technical challenges associated with its capture in conventional radioactive waste forms (glasses or cements), the ^{129}I poses serious concerns for its geological disposal. The proposed research is aimed at developing sodalite, $Na_8(AlSiO_4)_6X_2$ and apatite, $Ca_5(PO_4)_3X$ (where X = Cl, I) based chemically durable glass-bonded ceramic waste forms for immobilization of ^{129}I and mixed halide wastes. The major challenge associated with the proposed research is to synthesize the waste forms with maximum waste loading but at low temperatures (<800˚C), considering the volatility of halides at higher temperatures. Accordingly, solution-based and hydrothermal routes will be explored to synthesize the sodalite and apatite based ceramic minerals which will be further bonded with glass binders by sintering. The major focus of the research effort will be on the ready practical applicability of these methods for ^{129}I immobilization.

The design of waste forms will be an iterative process where state-of-the-art materials science characterization techniques will be used to study the structural chemistry along with radiation, thermal and chemical stability of materials. The results obtained from these studies will be used further to optimize the synthesis routes, processing conditions and chemical compositions of glasses and ceramics in order to develop suitable glass-bonded ceramic waste forms for ^{129}I immobilization.

The current proposed technology in U.S. for the removal of iodine from reprocessing plant off-gas is to pass it through Ag-exchanged zeolite (AgZ) to form chemisorbed AgI. Since silver is a RCRA metal, iodine needs to be extracted from AgZ and further processed for immobilization in a durable waste form. On the other hand, radioactive iodine in U.K. is, currently, discharged to sea. However, considering the future changes to the regulatory practices, it will become necessary to review this strategy and develop durable waste forms for immobilization of ^{129}I. The results obtained from this project will provide much-needed baseline data for the development of advanced waste forms for immobilization of iodine (aqueous based reprocessing of fuel) and mixed halide wastes (molten-salt processing of fuel).

The team comprises of a principal investigator (PI) from Rutgers University, NJ with expertise in design and synthesis of glasses and ceramics (Prof. Ashutosh Goel), along with a Co-PI from Washington State University (WSU), WA with expertise in low-temperature synthesis and processing of sodalite and other ceramic minerals (Prof. John McCloy, WSU). Additional Pacific Northwest National Laboratory (PNNL) collaborators necessary for this project include those with expertise in solution based synthesis and chemical durability of ceramic waste forms (Mr. Brian Riley) and with expertise in the development of advanced ceramic waste forms for ^{129}I immobilization (Dr. Josef Matyáš). Rutgers will lead the tasks on synthesis and characterization of halide-containing calcium phosphate apatites and design of glass binder, WSU will lead the tasks on synthesis and characterization of halide-containing sodalite and investigation of alternative consolidation method including HIP and SPS. The PNNL collaborators will lead the tasks on investigating the chemical durability and radiation stability of the ceramic waste forms.