In Situ Raman Spectroscopy for Determining Actinide Speciation and Concentration

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Actinide / Fission Product
Separations

Collaborators: N/A

ABSTRACT:

The electrochemical reduction of uranium oxide (UO_2) based nuclear fuel to a metallic form, a critical step in incorporating used nuclear fuel (UNF) into a pyroprocessing based fuel cycle, has been developed and demonstrated on an engineering scale at various institutions. However, there are two issues that need to be addressed prior to commercialization of the process: low process efficiency and materials accounting for non-proliferation. Both these issues require accurate knowledge of the composition (speciation and concentration) of the actinides in the electrolyte. There is currently no analytical technique that can provide this information accurately. *Thus, there is a knowledge gap in the ability to determine the speciation and concentration of actinides in the electrolyte.* This proposal aims to develop an in situ analytical method consisting of Raman spectroscopic analysis during electrochemical polarization.

Objectives: Specialized laboratory molten salt reactors with automated control and a suite of spectroscopic analytical techniques available in the PI's laboratory uniquely qualify the team to execute this study. Recently, the PI has developed the capability to conduct in situ Raman spectroscopy of molten salts. Lanthanides will be used as surrogates to represent the spectroscopic and electrochemical properties of actinide chlorides. The proposed research will measure the Raman spectrum of various lanthanides, functioning as surrogates for actinides, in molten LiCl-KCl at 500°C while varying the concentration of UCl_3 in the melt. The objectives include: (i) Development of a methodology for in situ spectroscopy of the LiCl-KCl-UCl_3 system, (ii) In Situ Raman Spectroscopy of lanthanide chloride in molten LiCl-KCl-UCl_3, (iii) Quantification of the composition of multicomponent molten systems, and (iv) Demonstrating the ability of the above technique to quantify the concentration of multiple chloride in molten LiCl-KCl-UCl_3 during electrochemical simulation of the refiner.