Development of In-situ Spectroscopic Techniques to Study the High Temperature Chemistry in Plutonium Pyroprocessing Salts

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Molten salts, such as alkali metal halides, are used to processes actinide materials for both nuclear weapons and nuclear energy applications. At Los Alamos National Laboratory plutonium metal is purified by electrorefining (ER). The ER process is founded on the difference in stability of various metal impurity chlorides compared to $PuCl_3$. Voltage is applied to provide a means of mass transport through a NaCl-KCl electrolyte. Another pyrochemical process used in plutonium metal purification is Molten Salt Extraction (MSE). The MSE process is used to extract americium from molten plutonium metal. The chemical foundation of MSE is based on the metal chloride equilibria that is established when a molten plutonium-americium alloy is reacted with chlorine gas at temperatures around 800 °C. While these two processes have been in place at LANL for a number of years many fundamental questions still remain concerning the coordination chemistry and oxidation state of the species present in the molten salt at processing temperatures. In addition the MSE process currently has no in-line process monitoring capability to determine when all of the americium has been removed from the metal phase. This paper will discuss on going efforts at LANL to apply a suite of spectroscopic and electrochemical techniques designed to, first, provide a pathway to gaining much needed plutonium and americium speciation data and to, second, possibly provide a real-time process monitoring capability. Progress on the development of molten salt UV-vis and Raman instrumentation will be demonstrated first with rare-earth surrogate materials and then with relatively small-scale plutonium and americium materials.

The development of real-time high temperature spectroscopic techniques will impact a number of DOE programs including: Pit Manufacturing, Non-proliferation, and Basic Actinide Science by allowing us to assess potential process upsets, to clearly design the chemistry and engineering aspects of these processes (smart manufacturing) and more importantly, open new windows of discovery of what, at first glance, should be a mature area of science.