
Monitoring of Actinide Concentrations in Molten LiCl-KCl Salt using Alpha Spectroscopy

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ABSTRACT:

The vulnerability of a closed fuel cycle comes from the risk of diversion of SNM for ill-purposed use. The ability to monitor actinides to prevent and detect the misuse of SNM throughout the flowsheet of pyroprocessing is of paramount concern to both national and global security. Success in this area is critical to enhancing the control of nuclear materials and to better safeguarding the current and the future US fuel cycle. Electrorefining is a key step used in the electrochemical processing flowsheet to separate the actinides from the fission products in the used nuclear fuel. There is a critical need to monitor the concentration of actinides in the salt after the drawdown process and during the pyroprocessing, and preferably this monitoring should involve an on-line monitoring method. Promising voltammetry-based molten salt actinide monitoring technologies and spectroscopic techniques, such as laser induced breakdown spectroscopy, are currently being developed by DOE. There are also research activities to develop a potentiometric sensor that would be selective for PuCl₃ concentration in the salt. While all of these approaches are promising, none have yet been proven to be both practically implementable and sufficiently accurate for safeguards. All viable alternatives need to be considered and investigated to the extent that resources allow, because this is arguably the single most challenging issue to be resolved for commercial-scale implementation of pyroprocessing.

The objective of this research is to fabricate sensor prototypes for measurement of actinide concentrations in molten salt and test it in a real salt environment up to 500°C. The novel approach using in-salt electro-deposition to pre-concentrate salt onto sensor before measurement will greatly increase the accuracy in actinides quantification for use in material accounting. We will deposit a layer of boron-doped diamond (BDD) thin film on the surface of a wide band gap semiconductor sensor that will act as a conductive electrode when doped with boron to over 4000 ppm. Using BDD as an electrode will enable the collection of actinide isotopes in solution directly on sensor through electrodeposition. After the sensor is removed from the salt, alpha spectroscopy can be used to determine the amounts of actinide isotopes in the deposit. This can then be correlated back to the concentration of these actinides in the salt. The benefit over voltammetry using alpha spectroscopy is improved ability to quantify specific actinides that might have similar electrochemical reduction potentials and even isotopes that are electrochemically indistinguishable. Notably, the sensor is reusable, because the actinide film on the detector can be reversibly removed through application of an oxidizing potential after the sensor is placed back into the salt.