LA-UR-20-26298
Approved for public release; distribution is unlimited.

Title: Current Needs in Radiochemistry

Author(s): Auxier, John David II

Intended for: Introduction for ACS Publication

Issued: 2020-08-14

Current Needs in Radiochemistry
J. D. Auxier II – Actinide Analytical Chemistry
Los Alamos National Laboratory

Introduction

There are a number of current needs in the development of the Pu and Am separations that underpin larger interest areas including nuclear waste issues, nuclear weapons, nuclear forensics, nuclear fuels and similarly related fields. In the large areas of interest, there are a number of sub-fields that include corrosion chemistry, pyrochemistry, solution-phase chemistry, rapid separations, environmental speciation, and other similar efforts. However, in all of the these fields it is important to realize that the underlying chemistry that underpins these efforts is similar in that Pu and Am are bound to ligands that can be in the solid, aqueous, or gas phase and separations occurs due to the preferential binding of the ligand to Pu or Am based on a number of factors that include pH, concentration, oxidation state, etc. In the past, most of the Pu and Am chemistry has occurred in limited scope within universities where micro-scale and bench-top scale chemistry are the dominate investigative theme. At US national laboratory and their international counterparts, larger scale reactions have been attempted such that industrial scale chemistry has been completed. Some of this technology has been harnessed in radiopharmaceuticals and nuclear fuel reprocessing. However, such bench-top research is expensive to due to the safety and security that is concerned when dealing with Pu, and scale-up of reactions is even more resource intensive. In an effort to offset this expensive, it would be of interest to explore the ability to predict selectivity based on the computational modeling of actinide chemistry. This has historically been limited by the abilities of large supercomputers, but with the emphasis on developing exascale computers within the US and similar efforts by other countries this predictive modeling has potential to produce significant results that would yield improved understanding of actinide chemistry.

There is a large push within DOE and NNSA to advance computing into the exascale regime, which allows for the movement from small molecule organometallic chemistry to modeling the chemistry of ions and complexes with extractants and the solution where they are contained. There is little precedence[1] for exploring the overlap of computational chemistry with solutions chemistry with regard to separations and selectivity[2]. This is in part because the software and basis sets for this type of work was underdeveloped or the computational power needed for the research was not yet ready and it is often easier to perform the separations chemistry in the laboratory to determine efficiency, selectivity, etc. In regard to actinide separations, performing the experiments in the laboratory is often more difficult due to the more regulated research environment, the difficulty in obtaining the starting materials, in addition to the radiological hazards with performing these research efforts.

Some specific examples of necessary improvements in the US would involve efforts to understand Pu in high salt environments for disposition at WIPP (waste isolation pilot plant) or

---

[1] Deborah A. Penchoff, Charles C. Peterson, Mark S. Quint, John D. Auxier, II, George K. Schweitzer, David M. Jenkins, Robert J. Harrison, and Howard L. Hall *ACS Omega* **2018** *3* (10), 14127-14143 DOI: 10.1021/acsomega.8b01800

[2] Deborah A. Penchoff, Charles C. Peterson, Jon P. Camden, James A. Bradshaw, John D. Auxier, II, George K. Schweitzer, David M. Jenkins, Robert J. Harrison, and Howard L. Hall, *ACS Omega* **2018** *3* (10), 13984-13993, DOI: 10.1021/acsomega.8b02068

other similar repositories. In this case, Pu and Am would be stored in drums and over time would be encapsulated in salt comprised of Mg and Ca chlorides. In other efforts, it would be to understand Pu and Am behavior in vitrified matrices where they elements are entrained in Na and Si glasses and left to be placed underground to be kept. The chemistry of these environments is complex and aging studies on these timeframes are often difficult if not impossible to model for the purposes of nuclear waste.

In a related vein, the separation of Pu and Am can help support nuclear waste in that segregated waste streams with lower activity provide the ability to store these compounds for longer time periods. This separation is made further important in that there is a need for a domestic supply for Am. Am production is necessary for oil and gas well-logging sources, smoke detectors and other important uses and the ability to meet nation demands for this element will require increased understanding of the chemical processes required to stand-up this capability. Likewise, the recent nuclear posture review indicates that there is need to increase the number of Pu pits to meet the US requirements. To meet this requirement, the Pu metal in the US stockpile has aged which involves the increased amount of Am in the metal. This chemistry must also be done in larger batches (e.g. greater than bench-top capacities) which will require improvements to the separations chemistry of Pu and Am.