DOE-ID NEPA CX DETERMINATION
Idaho National Laboratory

CX Posting No.: DOE-ID-INL-20-222

SECTION A. Project Title: Radiation-Induced Late Actinide Redox Chemistry

SECTION B. Project Description and Purpose:

The proposed are experiments designed to understand radiation-induced actinide redox chemistry in a variety of different solutions (e.g., aqueous, ionic liquids, and organic media). The actinide series marks the point in the periodic table where nuclear instability and the effects of relativity on electronic structure define the basic characteristics of elements. The factors that determine fundamental electronic properties such as electron repulsion, spin-orbit coupling, and ligand-field effects are on similar energetic scales in actinides, and thus no single factor dominates; whereas in lighter elements electron repulsion and ligand-field splitting largely control electronics. Thus, there is significant interest in the physical properties and the chemical behavior of actinide complexes, particularly in uncovering the origin of chemical divergence from the elements that we are more familiar with such as transition metals. However, while there are research programs dedicated to synthesizing and characterizing actinide compounds as well as theory groups dedicated to improving the accuracy of electronic structure calculations on heavy elements, few programs are focused on understanding the radiolytic reactions of actinides even though these reactions cannot be prevented – they are as fundamental to the actinides as the large magnitude of spin-orbit coupling. This critical radiochemistry knowledge gap becomes further exacerbated as the actinide series traversed to the point where there is almost no published data beyond americium.

In order to address this hole in our basic knowledge of the radiation chemistry of actinides beyond plutonium, and ultimately be able to model and predict radiation-induced redox chemistry in a variety of chemical environments, a rigorous interdisciplinary research effort must be initiated. We propose to accomplish this by performing an extensive suite of both steady-state (gamma and alpha-self-radiolysis) and picosecond time-resolved (pulsed electron) irradiation experiments. This will involve leveraging a novel collaborative capability (Idaho National Laboratory Center for Radiation Chemistry Research, Florida State University, and Brookhaven National Laboratory) for the measurement of radionuclide reaction kinetics – experiments that currently are not and cannot be performed anywhere else in the world. These experiments will be complimented by state-of-the-art multi-scale modeling calculations to provide predictive, quantitative, mechanistic information into the radiolytic redox behavior of americium, curium, berkelium, californium, and einsteinium under a range of solution media. This work will provide important missing information that will augment previous aqueous phase efforts that have involved only individual actinides, where many kinetic parameters were estimated rather than measured, in addition to providing new data for less investigated solutions, e.g., ionic liquids and actinide complexes in organic media. The hypothesis driving this investigation is that measurements of thermodynamics and kinetics of radiolytic reactions of later actinides can be coupled with electrochemical data and modelling of general redox reactions to provide a deeper understanding of the complexities occurring for actinide complexes in solution, and that these data will yield greater predictive capabilities for actinide chemical behavior in a variety of applied scenarios (e.g., during used fuel recycling, implementation in energy technologies, and nuclear forensic analysis).

The purpose of this DOE BES project is to investigate the radiation-induced redox chemistry of the late actinides (americium onwards). Actinide materials will be supplied by Florida State University (FSU). The materials to be investigated are curium, californium, berkelium, and einsteinium. The latter two are currently being synthesized by the Oak Ridge National Laboratory (ORNL) isotope production campaign. Actinides materials received by INL – having been shipped from FSU - will be manipulated in a variety of solutions and solvent systems – similar to those currently used for reprocessing research at Radiochemistry Laboratory (MFC-1702). Initially the solutions to be used will be aqueous acidic solutions (e.g., nitric acid, perchloric acid, and hydrochloric acid) and complexes dissolved in organic media such as n-dodecane. Once in solution, the actinides will either be subject to external gamma fields delivered by the Fuel and Applied Sciences Building (MFC-787) gamma irradiator or self-radiolysis from their own radioactive decay. Changes in solution composition will be followed using currently available analytical capabilities at RCL, e.g., UV-Vis spectroscopy and NMR. Actinide solutions will also be prepared and shipped to Brookhaven National Laboratory (BNL) for pulsed electron irradiation to evaluate reaction kinetics. All actinide materials shipped to BNL will either be returned to INL or FSU. The irradiations performed as part of this work do not activate the samples.

SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

The use of organic solvents would result in minor air emissions. The emissions do not result from the irradiation process as the constituents are in a sealed system. The solvents identified, thus far, are not listed as hazardous air pollutants. Use of radioactive materials at MFC could result in radioactive emissions.

Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

FASB is a facility that is 50 years old. No structural or aesthetic changes will be made to the building.

Generating and Managing Waste

Project activities are likely to result in the generation of small quantities of industrial, low-level radioactive and mixed waste. Waste Generator Services would properly characterize and dispose of all waste generated.

Releasing Contaminants

When chemicals are used during the project there is the potential for spills that could impact the environment (air, water, soil).

Using, Reusing, and Conserving Natural Resources

All materials will be reused and recycled where economically practicable. All applicable waste will be diverted from disposal in the landfill where conditions allow.

SECTION D. Determine Recommended Level of Environmental Review, Identify Reference(s), and State Justification: Identify the applicable categorical exclusion from 10 Code of Federal Regulation (CFR) 1021, Appendix B, give the appropriate justification, and the approval date.

For Categorical Exclusions (CXs), the proposed action must not: (1) threaten a violation of applicable statutory, regulatory, or permit requirements for environmental, safety, and health, or similar requirements of Department of Energy (DOE) or Executive Orders; (2) require siting and construction or major expansion of waste storage, disposal, recovery, or treatment or facilities; (3) disturb hazardous substances, pollutants, contaminants, or Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)-excluded petroleum and natural gas products that pre-exist in the environment such that there would be uncontrolled or unpermitted releases; (4) have the potential to cause significant impacts on environmentally sensitive resources (see 10 CFR 1021). In addition, no extraordinary circumstances related to the proposal exist that would affect the significance of the action. In addition, the action is not "connected" to other action actions (40 CFR 1508.25(a)(1) and is not related to other actions with individually insignificant but cumulatively significant impacts (40 CFR 1608.27(b)(7)).

References:
10 CFR 1021, Appendix B to subpart D, items B3.6, "Small-scale research and development, laboratory operations, and pilot projects"

Justification:
Project activities are consistent with 10 CFR 1021, Appendix B, B3.6, "Siting, construction, modification, operation, and decommissioning of facilities for small-scale research and development projects; conventional laboratory operations (such as preparation of chemical standards and sample analysis); and small-scale pilot projects (generally less than 2 years) frequently conducted to verify a concept before demonstration actions provided that construction or modification would be within or contiguous to a previously disturbed or developed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration actions, meaning actions that are undertaken at a scale to show whether a technology would be viable on a larger scale and suitable for commercial deployment."

Is the project funded by the American Recovery and Reinvestment Act of 2009 (Recovery Act) ☐ Yes ☒ No

Approved by Jason Sturm, DOE-ID NEPA Compliance Officer on: 1/12/2021