

**SECTION A. Project Title:** Electrochemical Manipulation and Radiolytic Evaluation of Organic Phase Neptunium

**SECTION B. Project Description and Purpose:**

The goal of this project is to establish the feasibility of electrochemical manipulation of neptunium (Np) oxidation states in organic solutions representative of those envisioned for advanced used nuclear fuel (UNF) reprocessing solvent systems. Recovery of the actinide elements from UNF by large scale reprocessing technologies has the potential to reduce US dependence on front-end nuclear fuel-cycle activities and provide a means of significantly reducing the long-term radiotoxic burden of nuclear waste on geological repositories. In summary, the proof of concept to employ novel electrochemical techniques to precisely control the oxidation state distribution of Np in the organic phase of envisioned UNF reprocessing solvent system formulations will provide much needed fundamental thermodynamic and kinetic data, and potentially provide support for the development of a simplified electrochemical process for actinide recovery with favorable efficiency and economy.

The goal is to establish the feasibility of electrochemical manipulation of Np oxidation states in organic solutions representative of those envisioned for advanced UNF reprocessing solvent systems. To achieve this goal two research objectives will be initiated and executed as outlined.

1. Evaluation of Novel Functionalized Electrodes on Complexed Neptunium Redox Distribution in Organic Media. The proposed hypothesis is that application of electrochemical techniques will facilitate the generation of singularly valent complexed Np species in organic media that can be subsequently manipulated using traditional UNF reprocessing solvent extraction technologies. To evaluate this hypothesis, novel functionalized electrodes – developed by the Florida International University (FIU) Co-PIs to quantitatively generate atypical actinide oxidation states will be employed in conjunction with proposed advanced reprocessing solvent system formulations, specifically concentrated HNO<sub>3</sub> contacted with DEHBA/DEHiBA in organic solvents, to promote the formation of extractable Np complexes, which will be subsequently characterized by electrochemical and optical methods, e.g., UV-visible spectroscopy. Characterization will: (i) facilitate optimization of the proposed electrochemical system conditions for Np oxidation state manipulation; (ii) identify the accessible electrochemical window for Np complexes in the organic phase; (iii) report characteristic optical spectra for each accessible complexed Np oxidation state; and (iv) determine the lifetime and partitioning of a given atypical Np oxidation state in the organic phase post electrochemical generation. Together these data will determine the feasibility of leveraging electrochemical techniques for Np oxidation state manipulation under envisioned reprocessing conditions. The proposed experiments will be initially performed using cerium (Ce) as a surrogate before advancing to U as an actinide surrogate before work with Np. Cerium surrogate experiments may be performed at the INL Energy Innovation Laboratory (EIL), at which all of the necessary analytical equipment is available, in addition to direct access to the INL CR2's Foss Therapy Model 812 gamma irradiator instrument, see Research Objective 2. U and Np experiments will be conducted at INL's Materials and Fuels Complex (MFC) Radiochemistry Laboratory (RCL), a state-of-the-art radiological facility, wherein on-hand stocks of U and Np are available for this work. Further, the necessary electrochemical and optical equipment are also available at the RCL.

2. Radiolytic Evaluation of Novel Electrodes and Singularly Valent Complexed Neptunium Solvent Systems. The proposed hypothesis is twofold:

(i) novel electrode functionality will decrease with increasing absorbed dose – owing to radiolytic cleavage and chemical transformation of surface bound functional groups – however, their functional lifetime will be sufficient for a typical reprocessing dose regime study ( $\leq 500$  kGy); and

(ii) radiolytic processes, i.e., reactions involving radiolytic transients, will dictate the practical lifetime of electrochemically generated complexed Np oxidation states in the organic phase. To evaluate these hypotheses, three irradiation subtasks will be executed:

2.1. The suite of novel electrodes investigated by Research Objective 1 will be subject to an independent gamma irradiation study, wherein each electrode will be irradiated in the presence and absence of organic media, and then used to attempt generation of the aforementioned singularly-valent complexed Ce/U/Np species. Electrode efficiency vs. absorbed dose (0-500 kGy) will be evaluated using the same electrochemical and optical techniques as in Research Objective 1.

2.2. The gamma radiolytic lifetime of singularly valent complexed Ce/U/Np species will be evaluated in the same manner as previous aqueous phase Np and Am work published by the INL CR2. This will involve monitoring the concentration of a given Ce/U/Np oxidation state as a function of dose, and then comparing with their inherent lifetimes, as evaluated in Research Objective 1.

2.3. Radiation-induced reaction kinetics for the reaction singularly valent complexed Ce/U/Np species with radiolytic transients (e.g., the organic solvent radical cation) will be measured by pulsed electron radiolysis techniques using the Brookhaven National Laboratory (BNL) Laser Electron Accelerator Facility (LEAF). These experiments will require the design of sealed optical cuvettes with the ability to perform electrochemical manipulations at the LEAF beam line so as to generate fresh singularly valent complexed cerium/neptunium species as necessary. These types of time-resolved experiments have been successfully performed by the INL CR2 for various actinides and solvent systems.

Gamma irradiations (Research Objective 2.1 and 2.2) will be performed using the INL CR2's Foss Therapy Model 812 located in Energy Innovation Laboratory (EIL) for cerium experiments, and/or a Nordion Gammacell located in the INL MFC Fuels and Applied Science Building (FASB) for neptunium experiments. The work completed in EIL and CR2 involves non-radioactive material. Novel electrode irradiations can be performed using either irradiator. The other irradiator that will be used for the project will be located in Materials Fuels Complex (MFC) Radiochemistry Laboratory (RCL). Possible generation loss of approximately 10's of grams worth of radioactive and hazardous waste will be due to pipettes, wipes, etc., and will be properly disposed of by Waste Generator Services (WGS). All radioactive waste will be generated in RCL which contains all the necessary documentation for radionuclide waste generation.

**SECTION C. Environmental Aspects or Potential Sources of Impact:**

**Air Emissions**

N/A

# DOE-ID NEPA CX DETERMINATION

## Idaho National Laboratory

### Discharging to Surface-, Storm-, or Ground Water

N/A

### Disturbing Cultural or Biological Resources

N/A

### Generating and Managing Waste

Generation of waste includes approximately a gram of radioactive and hazardous waste via pipettes, wipes, etc. The waste will be classified and disposed of in accordance with INL procedures and DOE regulations/requirements.

### Releasing Contaminants

Any time that chemicals are used there is a potential for spills.

### 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.

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

Final Environmental Impact Statement for the Waste Isolation Pilot Plant (DOE/EIS-0026, October 1980) and Final Supplement Environmental Impact Statement for the Waste Isolation Pilot Plant (SEIS-I) (DOE/EIS-0026-FS, January 1990)

**Justification:** The proposed R&D activities are consistent with CX 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); 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 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."

NEPA coverage for the transportation and disposal of waste to WIPP are found in the Final Waste Management Programmatic Environmental Impact Statement [WM PEIS] (DOE/EIS-0200-F, May 1997) and Waste Isolation Plant Disposal Phase Supplemental EIS (SEIS-II) (DOE/EIS-0026-S-2, Sept. 1997), respectively. The 1990 ROD also stated that a more detailed analysis of the impacts of processing and handling TRU waste at the generator-storage facilities would be conducted. The Department has analyzed TRU waste management activities in the Final Waste Management Programmatic Environmental Impact Statement (WM PEIS) (DOE /EIS-200-F, May 1997). The WM PEIS analyzes environmental impacts at the potential locations of treatment and storage sites for TRU waste; SEIS-II addresses impacts associated with alternative treatment methods, the disposal of TRU waste at WIPP and alternatives to that disposal, and the transportation to WIPP.

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CX Posting No.: DOE-ID-INL-21-181

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

Approved by Jason L. Anderson, DOE-ID NEPA Compliance Officer on: 2/07/2022