Advanced Raman Spectroscopy for Characterization of *f*-Element Coordination Chemistry and Multiphasic Nuclear Waste Forms

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ABSTRACT:

This project will enhance capabilities at Clemson University (CU) to probe the chemistry and structure of radioactive materials relevant to nuclear separations and waste stabilization. Specifically, this project will fund the purchase of a new confocal Raman microscope to support student and faculty research within the CU Nuclear Environmental Engineering Sciences and Radioactive Waste Management Center (NEESRWM). This added spectroscopic capability will expand NEESRWM's current suite of analytical and radioanalytical instrumentation and provide a dedicated Raman microscope for analysis of *f*-element (particularly actinide) coordination chemistry in extreme environments (high pH, temperature, and ionic strength), analysis of actinide oxide structure, and analysis of multiphasic nuclear waste forms and other radioactive materials.

Raman spectroscopy provides a direct probe of the electronic structure of polarizable chemical bonds. Given that structure and bonding directly influence macroscopic chemical reactivity, Raman spectroscopy provides an invaluable tool for understanding radionuclide behavior in complex and extreme systems, ranging from contaminated environments to nuclear separations processes to the complex chemical environments of various waste form technologies. Four primary research-use objectives directly relevant to nuclear science and engineering are envisioned for this project. These include the examination of (1) the coordination chemistry of actinide complexes and compounds, including uranyl phosphates; (2) the chemistry and structure of actinide and technetium oxides, before, during, and after solubility studies in aqueous solutions; (3) the structure and polydispersity of nuclear waste forms, ranging from multiphasic ceramics to cementitious materials; and (4) the identification of alteration of radioactive materials after long-term environmental exposure. Procurement of a confocal Raman microscope, equipped with two laser excitation sources (532 nm and 785 nm), filters for polarized Raman spectroscopy, a temperature-controlled sample stage, accessories that will facilitate routine analysis of liquid samples and microfluidic cells, and focus-tracking software that will allow semi-automatic Raman spectral mapping of solids with textured or uneven surfaces, will enable these research objectives and provide CU, DOE-NE, and the NSUF a unique resource for the characterization of radioactive materials.