Neutron Reflectometry Studies of Interfacial Phenomena in Actinide and Actinide-Related Thin Films

Izabela Kruk¹, Peng Wang², David D. Allred,³ and Kirk D. Rector¹, Jaroslaw Majewski,^{2,4*}

¹Los Alamos National Laboratory, Los Alamos, NM 87545, USA;

²Lujan Neutron Scattering Center, Los Alamos National Laboratory, Los Alamos, NM 87545, USA;

³Department of Physics and Astronomy, Brigham Young University Provo, UT 84602, USA,

⁴National Science Foundation, Div. of Molecular and Cellular Biosciences, Alexandria, VA 22314.

Studies of interfacial chemistry of actinides attract much attention due to interest in the power generation. In the power generation field, current research aims to reduce Pu and minor actinides in the spent light water reactor's fuel stockpiles. Therefore, considerable research efforts are underway to evaluate the suitability of Th as a nuclear fuel.

Herein, *in situ* neutron reflectivity technology measures changes in the scattering length density (SLD) and thickness of a thorium, uranium, and cerium metal films in controlled environmental conditions: time-dependent exposition to oxygen and water vapors in various temperature regimes. Among other phenomena, our research uncovered non-stoichiometric thorium oxides, ThO_x, preferentially generated between the metal and its thermodynamically favored dioxide layers. The near perfect stoichiometric lattice and relative low O solubility of ThO₂ film limits the availability as well as diffusivity of O species interacting with ThO, and hence prevents or slows the successive further oxidation. These observations suggest that ThO has many advantages over ThO₂ as a potential nuclear fuel such as good breeding performance, high thermal conductivity and density with good chemical and temperature stability. Our studies of interaction the Cerium thin films with water vapors revealed homogenous penetration of physisorbed water into the layers of various oxygen stoichiometry and its complete removal upon decreasing of the relative humidity.

Key Words: Actinides, Chemistry of Thin Metal Layers, Neutron Reflectometry, Nuclear Fuel.