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Advancing Neutron Scattering Techniques for the Development of Atomic-Scale Modeling of Nuclear Materials

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ABSTRACT:

The recent multinational declaration to triple nuclear energy capacity by 2050 emphasizes the urgent need for advances in nuclear technologies to reduce greenhouse gas emissions. A key component of this bold effort is the extended utilization of the existing nuclear fleet, which demands a deeper understanding of how critical nuclear materials in current-generation technologies incorporate defects and structural disorder under prolonged operating conditions. For example, at high burnup and under accident conditions, UO_2, the most common nuclear fuel, can accommodate excess oxygen, leading to off-stoichiometry and phase transitions that significantly impact fuel performance. The actual complex atomic-scale structural evolution of UO_{2+x} under extreme conditions, which includes defect clustering, oxygen diffusion, and incorporation of soluble fission products, remains very poorly understood.

To address these challenges, this project will leverage recent advances in state-of-the-art neutron scattering techniques at Oak Ridge National Laboratory to probe sub-nanoscale structures in nuclear materials such as UO_{2+x}. Neutron scattering is an ideal probe for these systems as it offers enhanced sensitivity to light elements such as oxygen (unlike more widely used X-ray scattering techniques) and can provide insights into atomic-scale structural information like aperiodic defect clusters and short-range order. In fact, multi-scale modeling of our preliminary neutron scattering data has already indicated that the formation and interactions of defects in $UO_{2.07}$ are more complex than previously understood, with the overall interpretation of the data significantly influenced by the operative length scale of the experimental probe. Therefore, using uranium oxides synthesized with variable oxygen content and lanthanide cation doping levels as a model system, we will employ total scattering experiments with pair distribution function analysis to develop atomic-scale models of these important nuclear materials. Importantly, the output from reverse Monte Carlo modeling of multi-scale neutron scattering data will not only be compared with computational simulations (*e.g.,* molecular dynamics) but can also be used as the input initial configurations for extended computational modeling. Thus, this research project will provide essential atomic-scale data on defect behavior and interaction mechanisms enabling improved modeling of physiochemical nuclear fuel properties such as diffusivity and thermal conductivity.

In alignment with the strategic vision of the Department of Energy's Office of Nuclear Energy, this project aims to provide critical insights into the behavior of nuclear materials under extreme conditions and the performance of nuclear fuels during prolonged operation. The findings of this study will aid in refining computational models that predict material performance, ultimately driving innovation and sustainability within the nuclear fuel cycle. Finally, although neutron scattering is well established in other energy-related fields, it remains historically underutilized in nuclear materials research. Therefore, this project seeks to cultivate a new generation of nuclear materials scientists skilled in advanced characterization techniques like neutron scattering, offering students hands-on experience at large user facilities through collaborations with national laboratories.