Complex Defect Physics in Oxides for Nuclear Energy Applications

Christopher R. Stanek
Los Alamos National Laboratory

Atomic scale defects (e.g. vacancies, interstitials) are very common in materials used in nuclear reactors, such as in the fuel, cladding and reactor vessel. The most studied of these defects originate from damage imparted to the crystalline lattice from fission events. However, defects that are not due to radiation damage are also present in a wide range of materials used in nuclear energy applications. In this talk, three examples will be used to show how a combination of atomic scale modeling, combined with experiments can yield deep insight in to defect physics, which ultimately allows for optimization of material performance. The three examples to be discussed are (1) nuclear fuel, (2) crystalline waste forms and (3) scintillation radiation detectors. (1) For nuclear fuel, interesting aspects of uranium dioxide (UO_2) thermal conductivity (such as surprising anisotropy) were recently found to be due to complex interaction between phonons and magnetic spins, the latter of which acts as a native defect and are the source of the fundamentally low thermal conductivity of UO_2. (2) Removing medium-lived isotopes $^{137}Cs^{1+}$ and $^{90}Sr^{2+}$ from the nuclear waste stream would result in a significant volume increase for a geologic repository. When designing robust crystalline waste forms for encapsulation of these isotopes, the impact of formation of their chemically disparate daughter products ($^{137}Ba^{2+}$ and $^{90}Zr^{4+}$, respectively) on waste form stability must be known. Initial calculations suggested that daughter products produced via transmutation are a unique type of defect worthy of experimental study. To overcome the practical difficulty associated with the 30-year half-life of these isotopes, we have devised an accelerated chemical aging approach, which relies on the use of very short-lived isotopes to assess the impact of daughter product formation on crystalline stability. (3) The resolution of radiation detectors based on scintillators is dependent on the light output of the scintillator, and an ideal scintillator converts all of the incident energy to light. However, atomic scale defects interfere with this conversion process. The talk will conclude with a recent example of how the light yield of a well-studied scintillator compound ($RE_3Al_5O_{12}$ garnet) was doubled via defect and band gap engineering.

Chris Stanek is a staff scientist in the Materials Science and Technology Division of Los Alamos National Laboratory. He received his B.S. in Materials Science and Engineering at Cornell University where he was a McMullen Scholar and his Ph.D. in Materials from Imperial College London (under the supervision of Prof. Robin Grimes). His research interests focus on the interaction between multidimensional defects in ceramics, primarily via atomistic simulation techniques. Stanek has a particular interest in materials for nuclear energy, including transmutation fuels, crystalline waste forms and scintillator radiation detectors. Currently, Stanek is the Fuels, Materials and Chemistry (FMC) focus area lead of DOE Energy Innovation Hub for the Advanced Modeling and Simulation of Nuclear Reactors, where FMC aims to integrate microscale materials science in to constitutive and behavioral models to predictively address nuclear fuel-related challenge problems. Stanek has published over ninety papers related to defect behavior in ceramics.