Title: Understanding the Role of Defects in the Radiation Response of Nanoceria

Research in last decade has shown that oxide and mixed oxide systems with pyrochlore and fluorite have better structural stability under high energy radiation. Nanoceria has fluorite structure and its properties can be tuned by controlling the defects within the lattice. This work aims at understanding the phenomena occurring on irradiation of nanoceria through experiments and atomistic simulation.

At first, research was conducted to show the ability to control defects in nanoceria lattice and understand the effect in tailoring its properties. The defect state of nanoceria lattice was controlled through doping; with a lower valence state rare earth element europium. Extensive characterization was done using high resolution transmission electron microscopy (HRTEM), UV-Visible spectroscopy (UV-Vis), X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy to understand the effect of dopant in modifying the chemical state of nanoceria. The defects originating in the lattice and redox state were quantified with increasing dopant concentration. The photoluminescence property of the control and doped nanoceria were evaluated with respect to its defect state. It was observed that defect plays an important role in modifying the photoluminescence property and that it can be tailored in a wide range to control the optical properties of nanoceria.

Having seen the importance of defects in controlling the properties of nanoceria lattice, experiments were conducted to see the effect of radiation in ceria oxide thin films of different crystallinity. The cerium oxide thin films were synthesized using oxygen plasma assisted molecular beam epitaxy (OPA-MBE) growth. The thin films were exposed to high energy radiation over a wide range of fluence (10^{13} to 10^{17} He+ ions/cm^3). The chemical state of the thin film was accessed by carrying out in-situ XPS for each dose of radiation. It was found that radiation induced defects within both the ceria thin films and the valence state deviated further towards non-stoichiometry cerium oxide (increase in Ce^{3+} state) with radiation.

The experimental results from ceria oxide thin film irradiation were studied in the light of simulation. Classical molecular dynamics and monte-carlo simulation were used for designing the model ceria nanoparticle and studying the interaction of the lattice model with radiation. Electronic and nuclear stopping at the end of the range were modeled in ceria lattice using classical molecular dynamics to simulate the effect of radiation. It was seen that displacement damage was the controlling factor in defect production. The simulation suggests that nanosized cerium oxide has structural stability under radiation and encounters radiation damage due to the dual valence state of 4+ and 3+. A portion of the study will focus on observing the lattice stability of cerium with increasing concentration of the lower valence (Ce^{3+}) within the lattice. With current theoretical understanding of the role of redox state and defects during irradiation, the surfaces and bulk of nanoceria can be tailored for radiation stable structural applications.

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Approved for distribution by Sudipta Seal, Committee Chair, on June 1, 2012.

The public is welcome to attend.