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ABSTRACT

Surface Charge Redistribution in SOFC Materials

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Using polarization dependent soft X-ray absorption spectroscopy (XAS), the electronic structure of transition metal elements at the near surface region of relevant SOFC materials can be studied in an element resolved manner with high sensitivity. Employing a test facility that uses a symmetric cell (two back-to-back identical half cells) through which an oxygen ion current is driven not by a chemical potential as in a complete cell, but by a bias voltage, allows for superior control of the electrochemical environment occurring within the cells.

One strength of soft X-ray XAS is the ability to determine elemental valence and electronic structure information for the determination of substitutional doping sites. We have studied the electronic structure of undoped and Y-doped SrTiO₃ (0-10 % atomic doping) by polarization dependent X-ray absorption spectroscopy, determining that the Y is substitutional into the Sr A-site as Y³⁺ and that charge neutrality is maintained by the introduction of oxygen vacancies (the Sr remaining as Sr²⁺ and the Ti as Ti⁴⁺). We have also identified the substitution site for V-doped, and Mo-doped STO. Of additional utility is that by placing these doped STO materials in reducing and oxidizing environments and under electrical potentials, we can thermally quench the materials and attempt to capture the electronic structure of the oxidized and reduced states. For these systems, we find that the electronic structure of the elements used for doping remain unchanged, although the Ti remains as Ti⁴⁺, the intensity of the Ti L₂ and L₃ X-ray absorption features (branching ratio) varies in response to the electrochemical environment. By using the appropriate X-ray selection rules for linear and circular polarized light, we can identify which electronic orbits are filled and emptied in response to the electrochemical environment and quantify the oxygen vacancy concentration.