Conductivity of Oriented Samaria-Doped Ceria Thin Films Grown by Oxygen-Plasma-Assisted Molecular Beam Epitaxy

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Due to global demands for increased energy resources and sustainable and renewable alternative energy, technologies such as solid-oxide fuel cells (SOFCs) are becoming the significant focus of researchers across the globe. In an effort to make this technology economically and practically viable for everyday use, it is of paramount importance for researchers to understand the fundamental aspects that control the performance of an SOFC. The heart of the SOFC is the electrolyte material that transports oxygen ions from cathode to anode. High-quality, epitaxial thin films of doped cerium oxide (a potential material for SOFC electrolytes at low and intermediate temperatures) are an excellent resource that allows researchers to explore the fundamental mechanisms that control the ionic conduction in SOFC electrolytes.

With the increasing demand for energy efficiency and the realization that greenhouse gas emissions must be reduced, technologies such as solid-oxide fuel cells (SOFC) are gaining significant attention. The fundamental understanding of the actual mechanisms that govern the ionic conduction in SOFC electrolytes is still elusive. A major hurdle in overcoming this bottleneck stems from the lack of studies based on high-quality materials. Use of conventional powder-processed pellets with porosity and grain boundaries makes analysis more complicated. Oxygen-plasma-assisted molecular beam epitaxy capabilities housed at EMSL have allowed researchers to synthesize high-quality epitaxial thin films with controlled dopant levels and thickness. Among the various materials used for low- and intermediate-temperature SOFC electrolyte application, samaria (Sm_2O_3, samarium oxide) doped ceria (CeO_2, cerium oxide) (SDC) is expected to be the best possible material. Hence, high-quality epitaxial SDC films were grown on sapphire (0001) substrates.

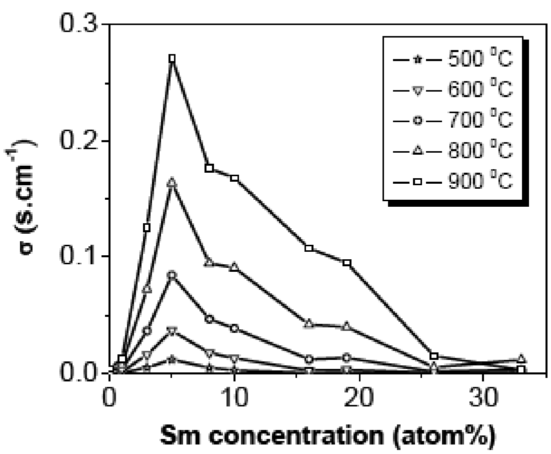

Figure 1. *The conductivity of Ce1-xSMxO2*Δ films as a function of samaria concentration for the temperature range of 500-900°C.*

The grown films were characterized using various *in-situ* and *ex-situ* techniques to understand their structure, crystalline quality, elemental distribution, and chemical composition (oxidation states of the cations). After establishing the optimum growth conditions, films with various amounts of dopant concentrations were grown (1 – 33 atom % samaria). Electrochemical impedance measurements were carried on these films using a four-probe van der Pauw method. The conductivity data obtained from these measurements were analyzed as a function of temperature and dopant concentration (Figure 1). Some of the salient features from this work are highlighted below:

- Ceria films with more than 10 atom% samaria were found to show polycrystalline features, while the films with less than 10 atom% samaria were found to be epitaxial, highly oriented films.
- Rutherford backscattering spectrometry and x-ray and electron diffraction measurements confirmed the high-quality, epitaxial nature of the films.
- 5 atom % samaria doping was found as an optimum concentration to obtain maximum conductance among various compositions.
- The reduction in the conductivity in films with high dopant concentration is attributed to the polycrystalline nature of the films. In the polycrystalline films, grain boundaries may act as scattering centers for the oxygen ions, hence the observed decrease in the overall conductivity.
- The higher ionic conduction at lower temperatures in the highly oriented SDC films is attributed to the alignment of oxygen vacancies [generated to retain the electrical neutrality of the ceria ($Ce^{4+}O^{2-}$) crystal when doped with Sm^{3+}]. The aligned oxygen vacancies help efficiently transport oxygen ions across the electrolyte from cathode to anode.

Various ongoing user research programs at EMSL are focused on exploring the finer details behind these observations and are aimed at developing a comprehensive understanding of the fundamental mechanisms that control the performance of nanoscale doped-oxide materials as potential SOFC electrolyte materials. This research was published in *Electrochemical and Solid State Letters* (Yu et al. 2008).

Citation:

Yu ZQ, SV Kuchibhatla, LV Saraf, OA Marina, CM Wang, MH Engelhard, V Shutthanandan, P Nachimuthu, and S Thevuthasan. 2008. “Conductivity of Oriented Samaria-Doped Ceria Thin Films Grown by Oxygen-Plasma-Assisted Molecular Beam Epitaxy.” *Electrochemical and Solid State Letters* 11(5):B76-B78.