USAXS, HESAXS, and XMT Studies of Solid Oxide Fuel Cell Layers

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Introduction

In solid oxide fuel cell (SOFC) development, control of the microstructure and chemistry in the various component layers is of primary importance in determining SOFC performance and cost. Since microstructure is particularly affected by materials choice and processing, good void and phase microstructure characterization is required. The pertinent scale range extends from tens of micrometers down to nanometers. The properties of such functional gradient materials depend on a complex superposition of spatial gradients in void morphology and structural phase composition. Quantifying these gradients can lead to an improved understanding of the underlying phenomena that govern the material properties.

To fulfill this goal, a combination of x-ray synchrotron-based research tools at the APS is being developed to provide a complete 3-D quantitative characterization of the void and phase microstructures as a function of position throughout the SOFC system [1, 2]. Specifically, this involves a combination of ultrasmall-angle x-ray scattering (USAXS) [3], high-energy small-angle x-ray scattering (HESAXS) with associated wide-angle x-ray diffraction (WAXD) [4], and computed x-ray microtomography (XMT) [5].

Methods and Materials

The SOFC materials were composed of a porous lanthanum-strontium-manganate (LSM) cathode, a denser cathode interlayer, a fully dense yttria-stabilized zirconia (YSZ) electrolyte layer, and a porous Ni/YSZ cermet anode [3]. Ranges of electrolyte and cathode interlayer thicknesses and of anode porosities were explored.

Absolute volume-fraction size distributions over much of the scale range of interest were obtained by USAXS at UNI-CAT beamline 33-ID at the APS [4]. Characterization of the fine features present and of the phase composition as a function of position at 5-μm spatial resolution were obtained by HESAXS/WAXD (XOR beamline 1-ID at the APS) [5]. A 3-D visualization of the SOFC layer microstructure for features on the micrometer scale and above was achieved by XMT (XOR beamline 2-BM at the APS) [6].

Results

Figure 1 shows USAXS results for the spatial variation perpendicular to the SOFC layers of the total void surface area (from Porod scattering) and the total void volume fraction (derived from an entropy maximization size distribution routine, MaxEnt) for a SOFC with a 20-μm electrolyte layer and a 20-μm-thick cathode interlayer. These results were obtained with an x-ray energy of 16.9 keV and a nominal incident beam slit height of 10 μm, although the actual slit definition was finer than this. The surface area and volume fraction curves were derived from the absolute-calibrated scattering data within the different SOFC layers by using appropriate contrast terms. Figure 1 suggests that the measured volume fraction and surface area would decrease to near zero in the electrolyte with a fine enough beam size and ideal sample alignment. Increased porosity in the anode away from the interfaces is also indicated, as are strong variations close to the electrode/electrolyte interfaces.

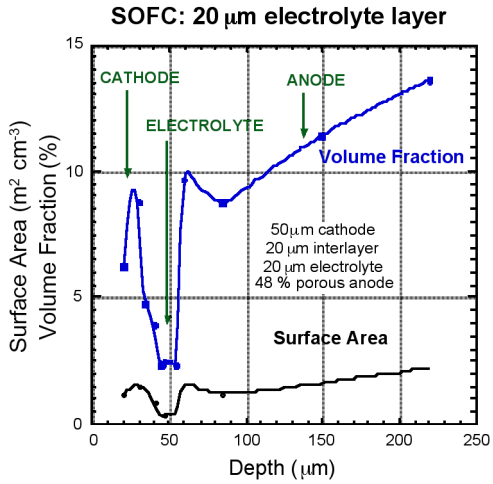

FIG. 1. Spatial variation of the void total surface area and volume fraction as a function of position perpendicular to the SOFC layers measured by USAXS.

Although HESAXS does not have the Q range necessary to determine void size distributions, it has provided similar surface area curves for a SOFC by using an x-ray energy of 80 keV. This high energy also enables WAXD diffraction patterns to be taken at the same locations as HESAXS. Figure 2 shows such a set of WAXD diffraction patterns at a series of positions through the layers of a SOFC with a 16-μm-thick

electrolyte layer and a 20-μm-thick cathode interlayer. Use of these two techniques together allows the spatial variation of the surface area (and hence the microstructure) and the phase composition to be followed through the SOFC layers and across the interfaces.

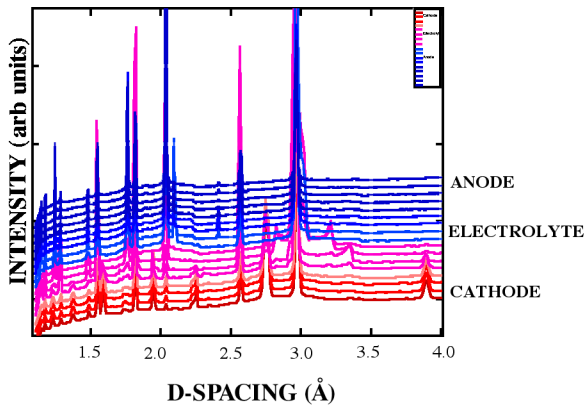

FIG. 2. WAXD data at successive positions through the SOFC layers with a 16-μm electrolyte and 20-μm cathode interlayer.

To characterize the SOFC layer microstructures at length scales significantly above a micrometer and to evaluate the integrity of the thin electrolyte layer, XMT studies were also made of the SOFC sections, which were reduced to 200 × 200 μm in cross section for these studies. Figure 3 shows a 3-D image reconstruction for a typical SOFC section. The porous anode and cathode regions separated by a dense electrolyte and cathode interlayer are clearly distinguishable.

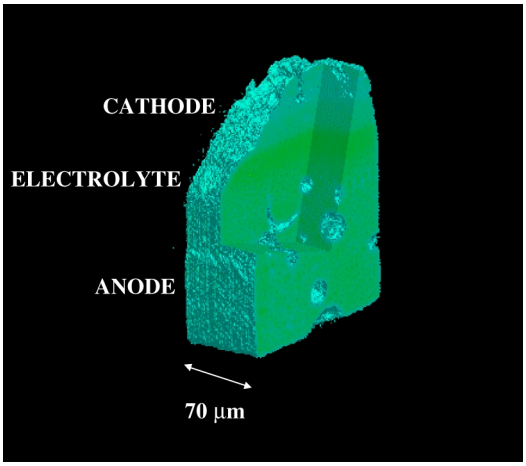

FIG. 3. A 3-D XMT reconstruction of an SOFC section having a 20-mm electrolyte layer and a 20-mm cathode interlayer.

Discussion

Representative void microstructures have been characterized and quantified through the anode, electrolyte, and cathode layers for a selected group of generic SOFC systems. The corresponding structural phase variations have also been determined by WAXD, with sample volumes closely corresponding to those used for HESAXS. Variations that occur with differences in processing have been explored and correlated with varying electrolyte and cathode interlayer thicknesses as well as with anode porosity. In particular, it has been demonstrated that the spatial resolution is sufficient to detect the abrupt microstructure and phase composition changes at the SOFC layer interfaces.

Correlations of microstructure and phase evolution in the cathode during service life, as a function of different compositions, morphologies, and doping, could be particularly valuable in optimizing cathode design. Similarly, the effects of corrosion in the anode can be explored, together with chemical effects at the electrode/electrolyte interfaces. Used together, the experiments discussed here can provide an improved quantitative understanding of how SOFC microstructures might be controlled through processing, and a similarly improved understanding may also be gained of how SOFC microstructures govern the performance properties as these evolve during service life.

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