

Solid Oxide Fuel Cell Commercialization, Research, and Challenges

by Eric D. Wachsman and Subhash C. Singhal

World wide interest in solid oxide fuel cells (SOFCs) has increased dramatically over the last ~20 years as indicated in part by the tremendous growth in attendance at the International Symposium on Solid Oxide Fuel Cells (see sidebar on page 43).

In the United States, the U.S. Department of Energy's (DOE) Office of Fossil Energy's (FE) National Energy Technology Laboratory (NETL), in partnership with private industry, educational institutions, and national laboratories, is leading the research, development, and demonstration of high efficiency, fuel flexible SOFCs and coal-based SOFC power generation systems for stationary market large central power plants.¹ This Fuel Cell Program has three focus areas under the Solid State Energy Conversion Alliance (SECA): cost reduction, coal-based systems, and research and development. The SECA cost reduction goal is to have SOFC stacks capable of being manufactured at \$170 per kilowatt (2007 basis) by 2010. Concurrently, the scale-up, aggregation, and integration of the technology will progress in parallel leading to prototype validation of megawatt (MW)-class fuel flexible products by 2015. The SECA coal-based systems goal is the development of large (greater than 100 MW) integrated gasification fuel cell power systems. Under this program, in February 2009, two fuel cell stacks (around 10 kW each), developed by FuelCell Energy Inc. in partnership with Versa Power Systems achieved 5,000 hours of service. The stacks also exhibited an overall degradation of only 1.7 percent and 2.6 percent per 1,000 hours. Siemens' stack also surpassed 5,000 hours in April 2009. Another SECA Industry Team, Delphi, and the U.S. Navy, have made noteworthy development progress in early markets for truck auxiliary power units (APUs) and proof-of-concept for unmanned undersea vehicles (UUVs), respectively.

Using tubular (cylindrical) SOFCs, Siemens fabricated a 100 kW system for distributed power generation.^{2,3} This system has now operated for over four years in USA, The Netherlands, Germany, and Italy without any detectable performance degradation. It has provided up to 108 kW of ac electricity at an efficiency of 46%. Siemens tubular cells have also been used to fabricate and field test over a dozen 5 kW size CHP units, each about the size of a refrigerator. These

units gave excellent performance and performance stability on a variety of hydrocarbon fuels. However, at present, their cost is high; future such units are expected to use higher power density alternate tubular geometry cells to drive down the cost.

Several hundred 1 kW size combined heat and power (CHP) units for residential applications were field tested by Sulzer Hexis of Switzerland; however, their cost and performance degradation was high and stack lifetime too short. With improved sealing materials and sealing concepts, planar SOFC prototype systems in the 1 to 5 kW sizes have recently been developed. Significant progress has now been made in producing and field testing about 1 kW size SOFC-based CHP (combined heat and power) systems for residential applications. In Japan, over 50 such prototype systems have been installed in homes to provide electricity and hot water and collect performance data for eventual commercialization within about a year. A photograph of one such unit is shown in Fig. 1. These systems are being built by Tokyo Gas, Osaka Gas, and Eneos in Japan using SOFCs produced by Kyocera Inc. The systems operate on natural gas and provide an electric conversion efficiency of about 45-50%.

Similar CHP systems are also being produced and field tested in Australia, New Zealand, and Europe by Ceramic Fuel Cells, Ltd. of Australia. Both the Japanese and the Australian CHP systems use SOFCs based on the conventional yttria-stabilized zirconia electrolyte and operate at about 750-800°C. On the other hand, Ceres Power Ltd. of UK is developing SOFCs based on a ceria-based electrolyte for operation at 550-600°C for use in wall-mountable residential CHP systems, also of about 1 kW size.

Another application of SOFC systems is in the transportation sector. The polymer electrolyte membrane (PEM) fuel cell is generally regarded as the fuel cell of choice for transportation applications. PEM fuel cells require pure H₂, with no CO, as the fuel to operate successfully. However, presently no H₂ infrastructure exists, and on-board reformer systems to produce H₂ from existing fuel base (gasoline, diesel) are technically challenging, complex, and expensive. Furthermore, it is difficult to eliminate the CO entirely from the reformatre stream. In contrast, SOFCs can use CO along with H₂ as fuel, and their higher operating temperature and availability of water on the anode side makes on-cell or in-stack reformation of hydrocarbon fuels feasible. Also,



FIG. 1. SOFC-based residential CHP system showing the SOFC unit (left) and the hot water tank (right).

no noble metal catalysts are used in SOFCs reducing cost of the cells. The initial application of SOFCs in the transportation sector will be for on-board auxiliary power units (APUs). Such APUs, operating on existing fuel base, will supply the ever increasing electrical power demands of luxury automobiles, recreational vehicles, and heavy-duty trucks. Delphi Corporation has developed a 5 kW APU using anode-supported planar SOFCs.⁴ This unit is intended to operate on gasoline or diesel, which is reformed through catalytic partial oxidation. The building blocks of such an APU consist of an SOFC stack, fuel reformation system, waste energy recovery system, thermal management system, process air supply system, control system, and power electronics and energy storage (battery) system. One of the most opportunistic applications for the SOFC is in APUs for transport trucks. This allows the drivers to turn the truck engines off while they're sleeping and use the much more efficient fuel cell APU to deliver power for environmental control of the driver's cab. There is an estimated 200,000 trucks per year that are produced that would need these APUs, which provides an attractive commercialization route for these fuel cells. By limiting idling time and running a SOFC instead of the main engine, emissions are reduced, noise is nearly eliminated, and operators realize significant fuel savings. The SOFC also has the capability of using a variety of fuels, including natural gas, diesel, bio-diesel, propane, gasoline, coal-derived fuel, and military logistics fuel. In 2008, Delphi Corporation and Peterbilt Motors Company successfully demonstrated a Delphi SOFC APU powering a Peterbilt Model 386 truck's "hotel" loads on diesel. During testing at Peterbilt's Texas headquarters, the Delphi SOFC provided power for the Model 386's electrical system and air conditioning and maintained the truck's batteries—all while the Model 386's diesel engine was turned off.

Residential CHP systems will, in all likelihood, be the first commercial products based on solid oxide fuel cells. Technological spinoffs of SOFCs into a variety of other applications areas, especially APUs, will add to market penetration, increase manufacturing production volume, and lower SOFC cost.

SOFC Research Challenges

The challenges for the fuel cell community are to reduce cost and increase reliability. These challenges extend from the cell itself, to the stack interconnect and seals, to the balance of plant.

There has been a tremendous effort to lower the operating temperature of SOFCs from ~1000°C to ≤ 800°C, for cost and reliability considerations.

Simultaneously there has been an even larger effort to increase the operating temperature of PEM fuel cells above 100°C, for performance and fuel poisoning considerations. Somewhere in between is the optimum operating temperature for a fuel cell, depending on fuel choice and degree of external fuel processing (vs. relying exclusively on internal reforming).

While there has been some success at developing high-temperature PEM fuel cells operating at temperatures around 140°C, the power densities and fuel flexibility of these systems are limited. Moreover, there is significant concern that the hydrogen infrastructure necessary for PEMFCs will make this a future technology with limited market penetration.

In contrast, SOFCs can operate on both current conventional fuels (*e.g.*, natural gas, gasoline, and diesel) and biofuels (biogas, ethanol, and biodiesel). As such, SOFCs offer great promise as a clean and efficient process for directly converting chemical energy to electricity while providing significant environmental benefits (they produce negligible CO, HC, or NO_x and, as a result of their high efficiency, produce about one-third less CO₂ per kWh than internal combustion engines). Moreover, SOFC operation on biofuels is the most energy efficient means to utilize home grown carbon neutral fuels.

Unfortunately, current SOFC technology must operate in the region of ~800°C to avoid unacceptably high polarization losses. These high temperatures demand specialized (expensive) materials for the fuel cell interconnects and insulation; and significant time and energy to heat up to the operating temperature. Therefore, development of SOFCs

to provide reasonable power output at lower temperatures would make SOFCs both more cost competitive with conventional technology, and significantly reduce start-up times which is critical to transportation and portable power applications.

Development of Anode Supported Cells

One of the biggest breakthroughs in lowering the SOFC operating temperature, while maintaining high power densities, was the development of anode supported cells. Techniques such as tape calendering⁵ and colloidal deposition⁶ allowed for the fabrication of anode supported thin (~10 µm) electrolytes. An SEM cross section of a typical anode supported SOFC is shown in Fig. 2. With the advent of this technology the major polarization loss transitioned from the electrolyte to the cathode. This can be seen in Fig. 3 which shows the relative polarization losses in a typical anode supported SOFC.

Deconvolution of Cathode Polarization

Because of the dominance of cathode polarization, the recent major SOFC research emphasis has been on developing higher performance cathodes. Over the last several years, cathodes have progressed from p-type electronically conducting lanthanum manganese based (*e.g.*, La_{1-x}Sr_xMnO_{3-δ} - LSM) to composites of LSM with the electrolyte (yttria stabilized zirconia - YSZ) to both add ionic conduction and increase the triple phase boundary region (where the oxygen reduction

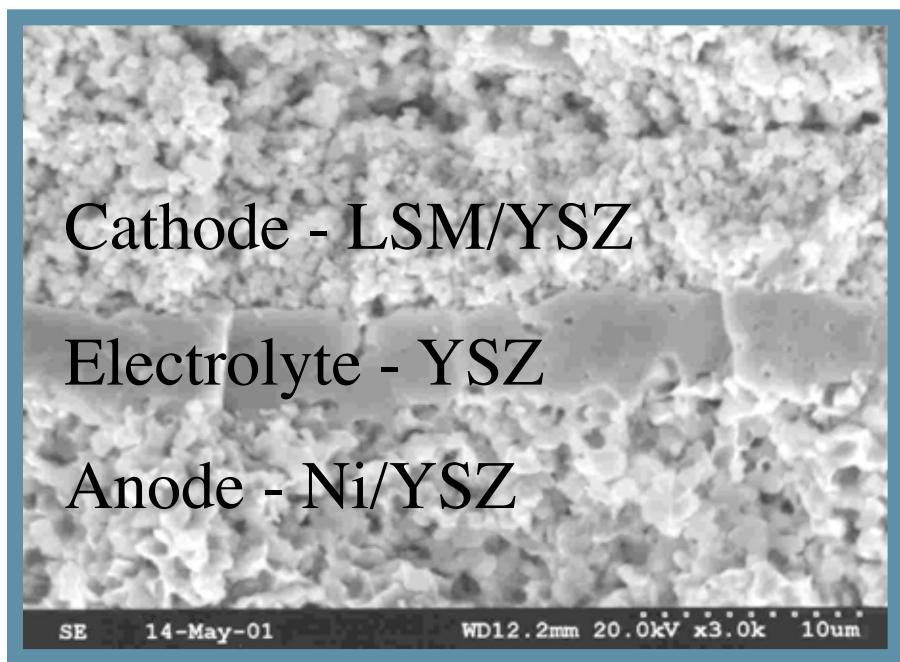


FIG. 2. Cross sectional SEM image of typical SOFC (Ref. 7).

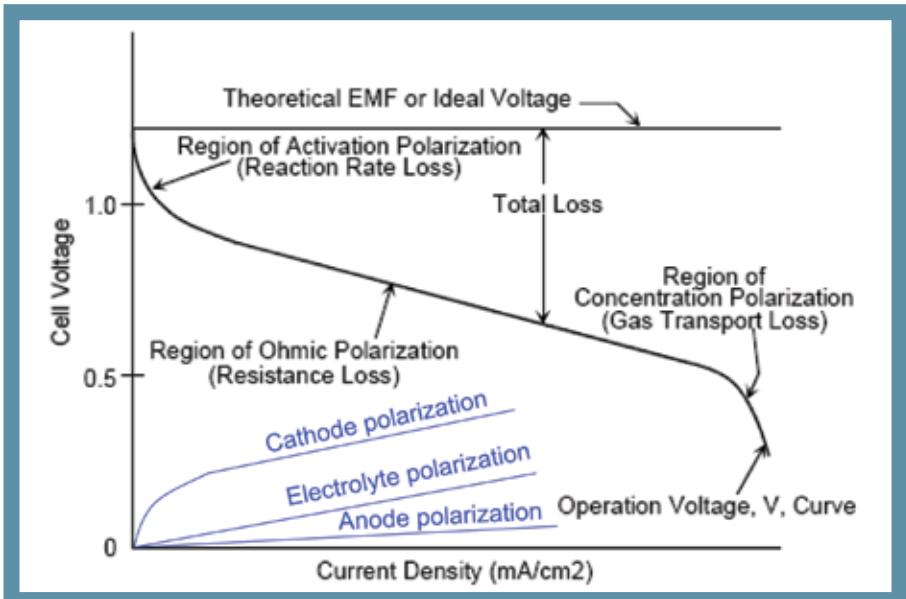


FIG. 3. SOFC current-voltage behavior indicating relative polarization losses.

reaction occurs); to the use of mixed ionic-electronic conducting oxide such as $\text{La}_{1-x}\text{Sr}_x\text{Co}_{1-y}\text{Fe}_y\text{O}_{3-\delta}$ (LSCF). More recently, impregnation techniques were employed to reduce the activation over-potential by depositing nano-dimensional catalyst in the cathode structure (see figure 4).⁸ The combination of these high performance cathodes with anode supported cell technology has resulted in power densities on the order of $\sim 1\text{W/cm}^2$ at 800°C .^{6,7}

These increases in cell performance, while impressive, have been to a large part Edisonian in nature. The fact is that the cathode process is extremely complex with multiple potential series

and parallel mechanistic steps. An example of this is shown in Fig. 5 for the simple case of O_2 reduction at a Pt/YSZ interface.⁹ In order to rationally design higher performance cathodes, a more fundamental understanding is necessary, and this requires the deconvolution and quantification of the various contributions to cathode polarization: electrocatalytic reduction (activation polarization), ionic and electronic conduction (ohmic polarization), and gas diffusion (concentration polarization).

The first important step is to quantify the effect of structure. Several groups

have approached this by measuring the impedance of well-defined (circular or square) micro-electrodes.¹⁰ However, more recently analytic techniques have been developed with the ability to quantify the structure of cathodes with actual/random structures. Using a focused ion beam (FIB)/SEM the 3D salient cathode features were quantified and directly related to electrochemical performance.¹¹⁻¹³

Figure 6 shows a FIB/SEM 3D reconstruction of a Siemens SOFC cathode, obtained from a series of sequential SEM images. Phase contrasting allows for identification of each phase: cathode (blue), electrolyte (orange), and pore (transparent). This allows for quantification of the critical microstructural parameters necessary to determine the sources of cathode polarization.

By quantifying phase boundaries we can calculate the triple phase boundary length (L_{TPB}) and surface area (S_v). These are the features where the oxygen reduction reaction occurs, and thus are the critical parameters for understanding activation polarization. Figure 7 shows the direct relationship between L_{TPB} and the charge transfer polarization; and S_v and the adsorption polarization, from impedance spectroscopy of LSM on YSZ.

With a quantified microstructure we can start to determine the fundamental reactions that occur at each feature of the microstructure. Independent measurement of the reaction rates of cathode materials by catalytic techniques, e.g., O-isotope exchange,^{14,15} provide the necessary mechanistic information. By integrating measurements of fundamental rate constants, with quantified microstructures of the measured materials, and comparing to electrochemical measurements, a fundamental and rational based approach to cathode development can provide the framework for further reductions in cathode polarization and thus SOFC operating temperature.

Higher Conductivity Electrolytes

If cathode polarization is no longer the major loss in SOFC performance, then the ohmic contribution of the electrolyte will once again dominate. The problem is, the conductivity of the conventional YSZ electrolyte is insufficient at lower temperatures, even for a thin ($\sim 10\text{ }\mu\text{m}$) electrolyte. Higher conductivity, alternative electrolytes will become necessary. Figure 8 compares the conductivity of YSZ with alternatives such as ceria and bismuth oxide based electrolytes.

Gadolinia doped ceria (GDC) has probably received the most attention as a lower temperature electrolyte due to its high conductivity,¹⁶ and recently an even more conductive ceria based

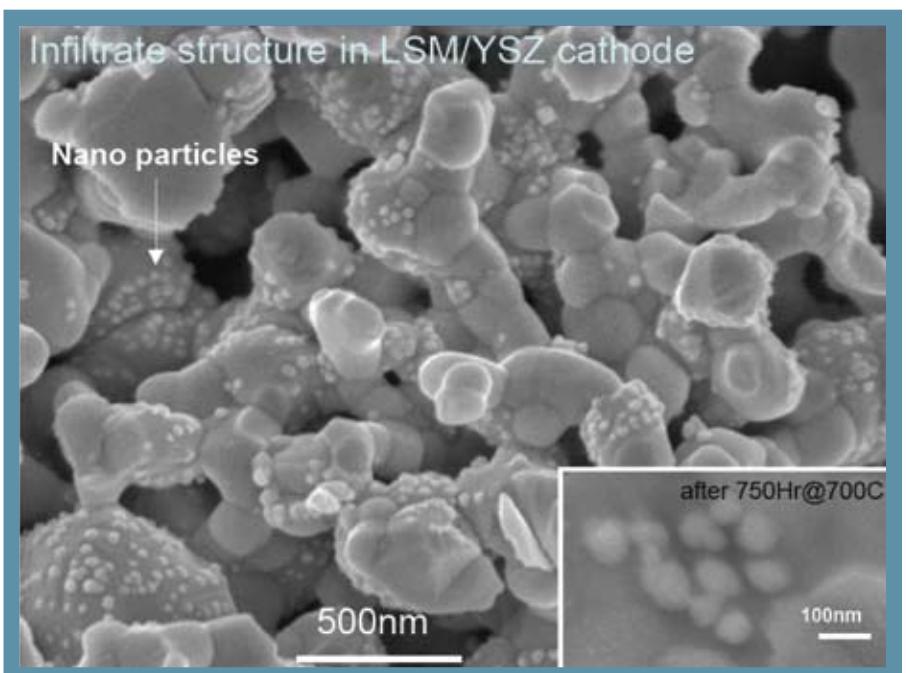


FIG. 4. SEM image of infiltrated cathode (Ref. 8).

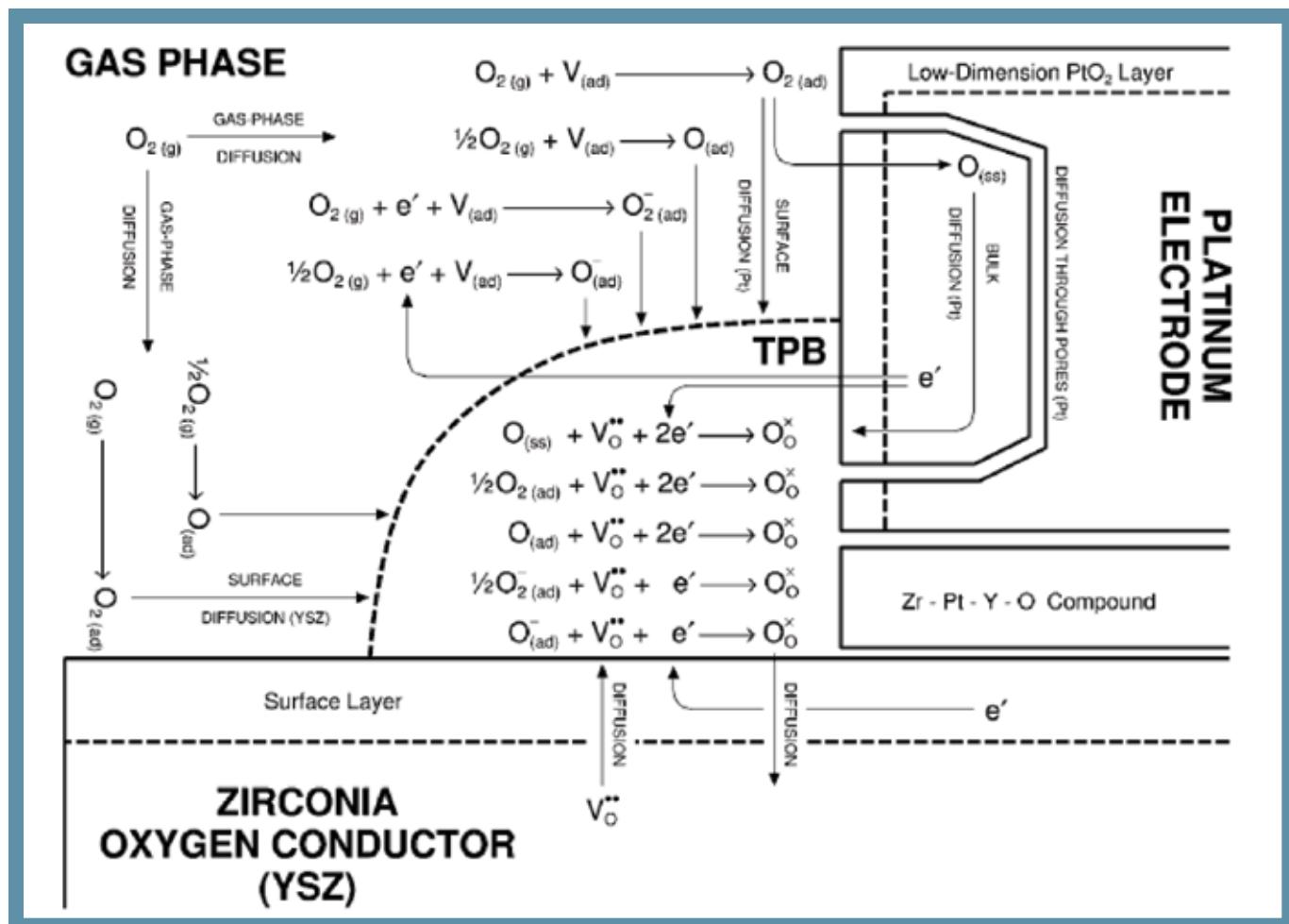


FIG. 5. Mechanistic oxygen reduction reaction at simple air/Pt/YSZ triple phase boundary (TPB) (Ref. 9).

electrolyte, samarium-neodymium doped ceria (SNDC) was developed.^{17,18} However, the highest conductivities have been obtained with bismuth oxide based electrolytes, such as erbia stabilized bismuth oxide (ESB),¹⁹ and recently a dysprosium-tungsten stabilized bismuth oxide (DWSB) was developed with even higher conductivity.^{20,21} At 500°C, the conductivity of SNDC is 20X that of YSZ, and the conductivity of DWSB is 100X that of YSZ; thus creating the opportunity for high power density low temperature SOFCs.

Unfortunately, high oxygen mobility is a result of weak metal-oxygen bonds, and thus these materials have lower stability under the low P_{O₂} at the anode (fuel side) resulting in mixed electronic-ionic conduction in CeO₂ electrolytes and decomposition to metallic Bi for Bi₂O₃ electrolytes. Addressing this issue is critical to future acceptance of these alternative electrolytes in practical SOFCs. Moreover, the use of a new electrolyte creates the need, and provides the opportunity, for a new compatible material set (cathode, anode, interconnect, etc.). ■

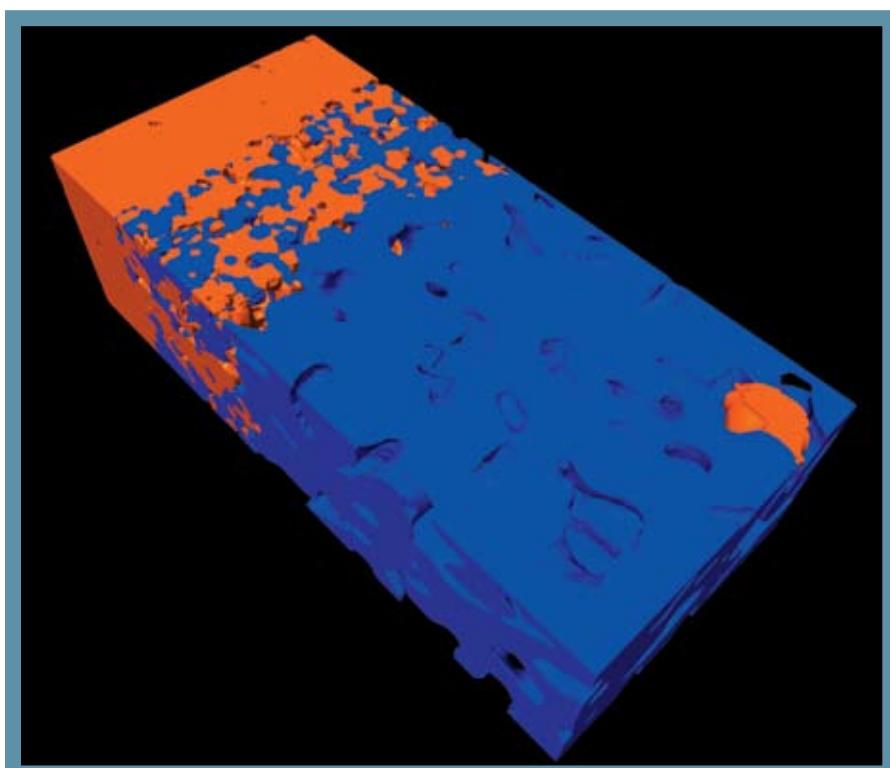


FIG. 6. FIB/SEM three dimensional reconstruction of SOFC cathode (Ref. 12).

About the Authors

ERIC D. WACHSMAN is the past Chair of the High Temperature Materials Division, Director of the University of Maryland Energy Research Center, and the William L. Crentz Centennial Chair in Energy Research at the University of Maryland (UMD). Prior to his recent move to UMD, Dr. Wachsman was the Director of the Florida Institute for Sustainable Energy at the University

of Florida. Dr. Wachsman's research interests are in ionic and electronic conducting ceramics, from fundamental investigations of their transport properties and heterogeneous electrocatalytic activity, to the development of moderate temperature solid oxide fuel cells (SOFC), gas separation membranes, and solid state sensors. He is an ECS Fellow and Editor-in-Chief of *Ionics*. He may be reached at ewach@umd.edu.

SUBHASH C. SINGHAL is a Battelle Fellow and Director of Fuel Cells at the Pacific Northwest National Laboratory (operated by Battelle for the U.S. Department of Energy), Richland, Washington. Prior to joining his present position in April 2000, he was manager of Fuel Cell Technology at the Siemens Westinghouse Power Corporation in Pittsburgh, Pennsylvania, for over 28 years. His research interests are in high temperature materials and advanced energy conversion systems, particularly solid oxide fuel cells. He is a member of the National Academy of Engineering and a Fellow of ECS, the American Ceramics Society, ASM International, and the American Association for the Advancement of Science. He has won several awards related to fuel cell technology including the Grove Medal, the Christian Friedrich Schoenbein Medal, and the Fuel Cell Seminar Award. He may be reached at singhal@pnl.gov.

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FIG. 7. Effect of LSM microstructure on cathode polarization; dissociative adsorption as a function of pore surface area, and charge transfer polarization as a function of triple phase boundary length; at 800°C in air (Ref. 13).

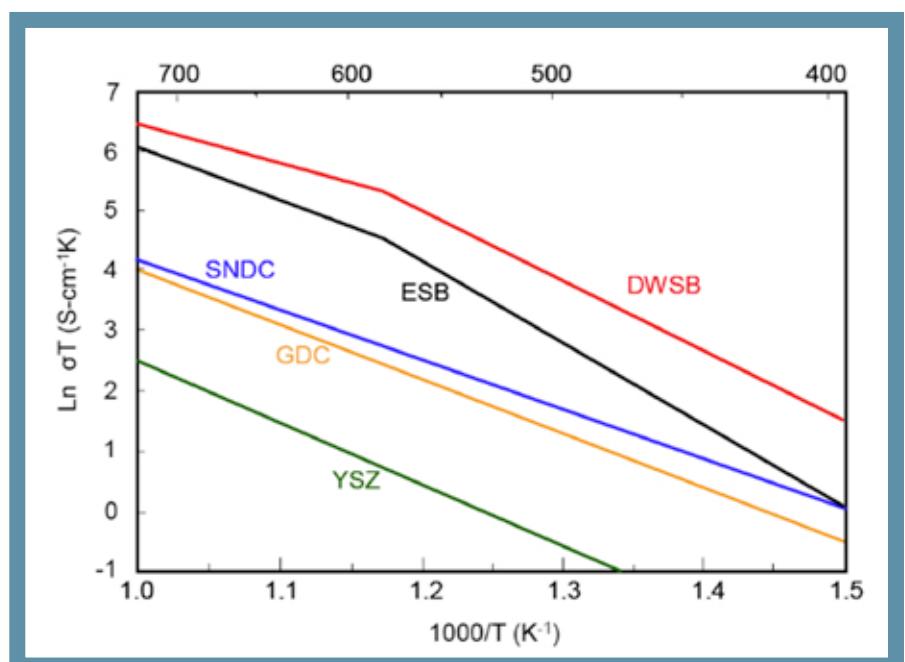


FIG. 8. Conductivity of conventional, YSZ, and alternative oxide electrolytes.

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International Symposium on Solid Oxide Fuel Cells

This continuing symposium series provides an international forum for the presentation and discussion of developments related to solid oxide fuel cells based on zirconia or another oxide electrolyte. Topics addressed include materials for cell components. (e.g., electrolyte, electrodes, and interconnections); fabrication methods for complete cells and components; cell design, electrochemical performance and modeling; stacks and systems for residential and automotive applications; and field tests of SOFC demonstration systems.

This symposium series started in 1989 at the ECS fall meeting in Hollywood, Florida, and has since become the leading symposium in the field of solid oxide fuel cells. The symposium is held every two years, rotated among the U.S., Europe, and Japan, and is co-sponsored by the SOFC Society of Japan. S. C. Singhal has been the lead symposium organizer since its inception.

The eleventh symposium in this series (SOFC XI) is being held at the **216th ECS meeting in Vienna, Austria, October 4-9, 2009**. At this symposium, 375 papers, a record number, covering all aspects of solid oxide fuel cells will be presented.

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