Monolithic and flat plate solid oxide fuel cells (SOFCs) are currently being developed for a variety of electric power generation applications. The attractiveness of these SOFCs includes improved performance, high power density, and potential low-cost manufacturing. Monolithic SOFCs of various sizes have been fabricated by the tape calendering process. Methods are being developed for cofiring lanthanum chromite interconnects. Electrochemical performance of monolithic single cells and multicell stacks has been verified at 1000°C. Flat plate SOFCs, with thin (1-10 μm) electrolytes that allow reduced-temperature operation, have also been fabricated by tape calendering. Thin-electrolyte cells have been successfully operated at 600-800°C and for 1000 hours at 800°C without significant performance degradation.

1. INTRODUCTION

Solid oxide fuel cells (SOFCs) are presently under development for a variety of electric power generation applications. Current SOFC technology uses stabilized zirconia as the solid electrolyte and requires an operating temperature of about 1000°C. The attractiveness of the SOFC relates primarily to its solid state nature, its internal reforming capability, and its high-quality by-product heat, which can be used for cogeneration or other purposes. At present, two common configurations have been proposed and fabricated for SOFCs: tubular (the seal-less tubular design and the segmented-cells-in-series design) and planar (the monolithic design and the flat plate design) (1). The planar configuration has received much attention recently because SOFCs based on this configuration offer the advantages of improved performance, high power density, and potential low-cost manufacturing.

The tape calendering process has been developed for the fabrication of the monolithic solid oxide fuel cell (MSOFC). Tape calendering (for processing ceramic materials)
involves squeezing a softened thermoplastic polymer/ceramic powder mixture between two rollers to produce a continuous sheet of material (2). Individual sheets can be calendred to form the multilayer tapes required for the MSOFC. Recently, the feasibility of using tape calendring to produce thin (1-10 μm) zirconia electrolyte layers has been demonstrated. Flat plate SOFCs having thin electrolytes have been successfully operated at reduced temperatures.

2. MONOLITHIC SOLID OXIDE FUEL CELLS

Recent development efforts on the MSOFC have focused on three areas: fabrication of multicell stacks, cofiring of lanthanum chromite interconnect, and testing of cell and stack electrochemical performance.

MSOFC multicell stacks of up to 12-cell height and 25-cm² footprint area have been fabricated using the tape calendring process. Fabricated stacks have shown improved structural integrity and reduced structural defects. Optimizing cell designs and modifying materials and fabrication processes are key to these improvements. Optimizing cell designs involves mainly the development of a finite element model to perform stress analysis on the MSOFC. Analysis results are then used to recommend design changes for minimizing stresses and reducing structural defects. Modifying materials and fabrication processes includes the following: tailoring the characteristics of starting ceramic powders, optimizing batch compositions, performing defect analysis, changing forming and firing procedures, applying materials and process specifications, and implementing process control.

Cofiring work has concentrated on lanthanum chromite interconnects. Doped LaCrO₃ can densify to full density if fired alone in air at 1400°C. However, the material does not densify when fired in laminated form (laminated to electrode layers) under similar firing conditions (3). The main reason for the poor densification is the migration or wicking of liquid phases (responsible for assisting densification) from the LaCrO₃ interconnect into electrode pores at the interfacial regions. Recent experimental results show that LaCrO₃, laminated to thin anode and cathode layers (< 15 μm), can be fired to high density at 1400°C in air (4). In this case, the electrode pore volume is believed to fill with liquid phases from the interconnect during cofiring and densify, thus allowing the interconnect to also densify.

Fabricated MSOFC single cells and multicell stacks have been tested to demonstrate electrochemical performance. Single cell discs (about 5 cm²) are tested with electrical connections through platinum screen current collectors. The current collectors are attached to the cell by platinum paste. Attaching the current collectors in this manner uniformly distributes the current across the electrode surface without interfering with the electrochemical performance of the cell. The cell is cemented on an alumina tube to establish and maintain a seal between fuel and oxidant. Fuel flows inside the alumina
tube. This alumina tube is placed inside an outer alumina tube which contains oxidant gas. The single cell test setup is shown in Figure 1. The performance of single cells is evaluated in terms of open-circuit voltage (OCV), cell voltage under load (polarization), and area-specific resistance (ASR). Typically, single cells exhibit OCVs in good agreement with the theoretical value, indicating no cracks in the electrolyte. At 1000°C with a 97% H₂/3% H₂O fuel and air oxidant, cell ASR is in the range of 0.6 to 0.8 ohm cm². An example of single cell polarization curves obtained at different temperatures is given in Figure 2.

Multicell crossflow stacks are tested using a gas manifold schematically shown in Figure 3. In this setup, the stack is sandwiched between two flat zirconia plates. Zirconia bars are oriented and cemented in place so that the four edges of the stack are sealed to the bars to prevent gas cross leakage; this forms inlet and outlet passages for both fuel and oxidant gases. The zirconia bars provide support for the top plate. Figure 4 shows an example of the polarization curve of a two-cell stack obtained at 1000°C with hydrogen as the fuel and air as the oxidant. The OCV of the stack is lower than the theoretical voltage (2.1 V), indicating some gas cross leakage, probably through the seal. The stack shows excellent performance, with an ASR of about 1.0 ohm cm².

3. FLAT PLATE SOLID OXIDE FUEL CELLS

Flat plate SOFCs, with thin-film electrolytes that allow reduced-temperature (600-800°C) operation, are being investigated. Due to the nature of their fabrication and assembly, flat plate SOFCs can incorporate modifications resulting from lower operating temperatures more readily than the other designs. The benefits of reduced-temperature operation for SOFCs include wider choice of cell and ancillary materials, longer cell life, reduced thermal stress, improved reliability, and reduced fuel cell cost.

Tape calendering has been evaluated as a cost effective method for fabricating reduced-temperature flat plate SOFCs. The feasibility of using the tape calendering process to produce thin-film electrolytes required for this type of fuel cell has been demonstrated. Thin (1-10 μm) and dense ZrO₂ layers have been successfully fabricated on porous NiO/ZrO₂ support by tape calendering. This is the first time such a thin film has been produced by a conventional ceramic processing technique. The fabrication process involves progressive rolling of green (unfired) electrolyte-anode bilayer and cofiring of that bilayer at elevated temperatures. Figure 5 shows a SEM micrograph of a fracture surface of an electrolyte-anode bilayer. As can be seen from the figure, the electrolyte layer is fully dense, uniform, and well bonded to the porous anode. The nickel mapping of the bilayer clearly demonstrates the capability of the tape calendering process to produce micron-thick layers of uniform thickness (Figure 6).

Single cells based on thin zirconia electrolytes have been operated at reduced temperatures. Figure 7 shows polarization curves obtained at 600-800°C for a thin-
electrolyte single cell with a platinum paint cathode. The cell exhibits OCVs close to the theoretical voltages, indicating no significant gas leakage through the thin electrolyte. A power density of about 0.3 W/cm² has been achieved at 800°C operating temperature. A thin-electrolyte cell has operated for 1000 hours at 800°C without significant performance degradation. Post-test analyses have shown no cracking in the fully dense electrolyte and excellent bonding between the electrolyte and anode layers (Figure 8).

4. SUMMARY

Monolithic and flat plate SOFCs have been fabricated by the tape calendering process. Significant progress has been made on the fabrication of MSOFc multicell stacks and cofiring of lanthanum chromite interconnects. Fabricated MSOFc cells and stacks have been tested to confirm performance at 1000°C. The feasibility of using tape calendering to produce thin (1-10 μm) dense zirconia films (on nickel oxide/zirconia anode support) has been demonstrated. Flat plate SOFCs with thin-film electrolytes have been successfully operated at reduced temperatures of 600-800°C.

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Figure 1 Single Cell Test Apparatus

Figure 2 Polarization Curves of Single Cell at Different Temperatures
Fuel: 97%H₂/3%H₂O, Oxidant: Air
Figure 3  Gas Manifolding for MSOFC Stack Testing

Figure 4  Polarization Curve of MSOFC Two-cell Stack
Fuel: 97%H₂/3%H₂O, Oxidant: Air, Temperature: 1000°C
Figure 5 SEM Micrograph of Fracture Surface of Thin Zirconia Electrolyte on Anode Support

Figure 6 Nickel Mapping of ZrO₂ Electrolyte-NiO/ZrO₂ Anode Bilayer (Thickness of Electrolyte Layer: about 2 μm)
Figure 7  Polarization Curves of Thin-Electrolyte Cell at 600-800°C
Fuel: 97%H₂/3%H₂O, Oxidant: Air

Figure 8  Micrograph of Thin-Electrolyte Cell after 1000-Hour Testing