A Novel Integrated Stack Approach for Realizing Mechanically Robust Solid Oxide Fuel Cells

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Objectives

- Develop a suitable support material.
- Develop a suitable interconnect material and verify that dense, conductive layers are achieved.
- Develop techniques allowing accurate patterned deposition of the fuel cell components with pattern resolution of < 0.1 mm.
- Develop processes for depositing patterned fuel cell components, using a colloidal deposition process, e.g. screen printing.
- Demonstrate successful operation of integrated stack elements containing 5 to 20 cells and interconnects.

Key Milestones

- Develop and optimize processing of a suitable support material.
- Develop processing methodology based on screen printing for patterned deposition of fuel cell components.
- Develop a suitable interconnect material and processing techniques.
- Verify electrode layers with suitable conductivity, structure, and electrochemical performance.
- Verify formation of thin dense YSZ electrolyte layers.
- Successful demonstration of integrated SOFC stacks with 5 – 20 cells.

Approach

In this project, a new approach to stacking SOFCs is being developed. The geometry has all active SOFC components and the interconnect deposited as layers on a low-conductivity support. We term this an integrated SOFC (ISOFC) design because of the strong analogy with integrated circuits, where many devices and interconnects are on a single chip. The integration of SOFCs and interconnects on the same support has several advantages, including the reduction of electrical resistances associated with pressure contacts between the cells and interconnects, relaxation of fabrication tolerances required for pressure contacts, reduction of ohmic electrode losses, and reduction of interconnect conductivity requirements.

Processing methodologies are being developed for fabricating the ISOFC materials: electrolyte (Y-stabilized Zirconia, YSZ), anode (Ni-YSZ), cathode ($La_{0.8}Sr_{0.2}MnO_3$-YSZ), and interconnect. Partially-stabilized zirconia is being used for the support material. Ag, Pt, and $LaCrO_3$ are being tested as interconnect materials. Screen printing is being developed as the primary processing method. A centrifugal casting technique has also been developed for depositing electrolyte layers.

Results

PSZ supports were optimized by statistical design of experiments with sintering temperature, time, filler amount, and calcining temperature as the primary variables. Good results were achieved with a maximum strength of 300 MPa and a porosity of 30%. Electrical conductivity was measured and found to be low enough to not substantially shunt the ISOFC device.

The ISOFC design requires that electrodes have sufficient conductivity to transport current across each cell without significant loss. In the present design, the sheet resistance should be ≤ 2 Ω/square. Ni-YSZ anode sheet resistances easily met this criterion, with values < 1 Ω/square.

Given that the electrode thickness was ≈ 10 µm, the Ni-YSZ conductivity was ≈1 x 10^3 S/cm in the 500ºC-800ºC temperature range, in agreement with literature values [1].

For LSM-YSZ cathodes, the sheet resistance for a 10 mm layer was found to be quite high, > 100 Ω/square. Adding a second cathode layer of pure LSM decreased this value to < 20 Ω/square. Additional cathode layers, and/or a decrease in cell width, will thus be required to achieve the required cathode conductivity.

The screen printing process scheme was as follows. First, the anode was deposited, followed by the electrolyte layer and interconnect. Next, these layers and the support were co-sintered at 1400ºC. Then, the cathode layer was deposited. Then, the cathode was sintered. Figure 1 shows photographs taken of screen printed patterns showing the deposition sequence. Below the photographs are schematic drawings showing cross-sections of the patterned layers.

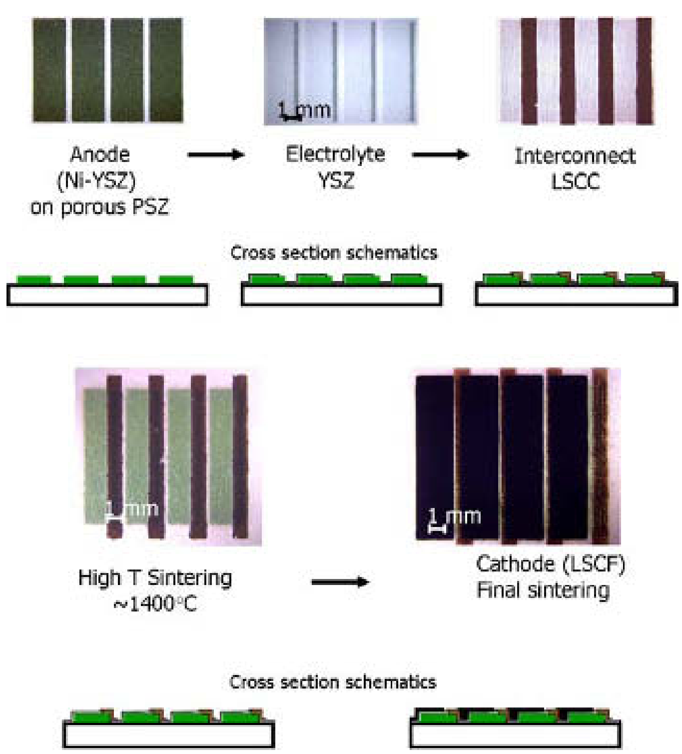

Figure 1. Screen printed ISOFC layer images with cross-sectional schematic drawings.

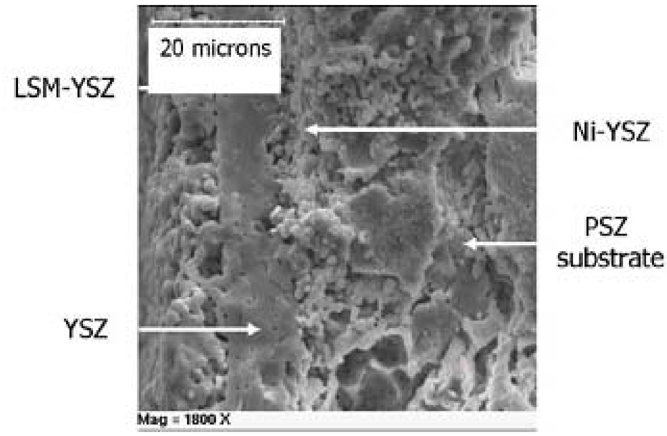

Figure 2. Cross-sectional SEM micrograph of screen printed ISOFC layer.

Figure 2 shows a cross-sectional SEM image of an ISOFC printed on a porous PSZ substrate. The fired layer thicknesses are about 10 µm and the cathode and anode are visibly porous. The YSZ electrolyte is fairly dense with only a small amount of closed porosity visible in the sample. Furthermore, the substrate shows an open pore network, which allows gas transport to the electrode.

At this time, the electrolyte layers obtained by screen printing are not sufficiently dense to produce good fuel cells. Thus, the initial demonstrations of ISOFC devices have been carried out using a centrifugal casting technique to produce dense electrolyte layers. All the other layers were produced by screen printing. Four-cell ISOFCs similar to those described above were prepared and tested with air as the oxidant and humidified hydrogen as the fuel. The maximum stack voltage of 3.2V, was lower than the expected value of 4.4V (1.1V per cell). It is not clear why the voltages were less than expected, but presumably improved fabrication will provide better results. Stack current and power increased with increasing temperature, as expected for solid oxide fuel cells. The maximum power was ≈ 25 mW at 700ºC. This relatively low value is presumably due to insufficient conductivity of the cathode layers.

Conclusions

We have addressed the key issues pertaining to the development of the ISOFC, i.e. materials and processing required to achieve a resistive porous substrate, electrically conductive and porous electrodes, accurate patterned deposition, dense electrolyte, and approximate substrate-layer shrinkage match. We have also carried out a preliminary demonstration of the device. Future work will include improved processing techniques, optimization of interconnect material, and demonstration of larger ISOFCs with better performance.

References

1. Minh N. Q., "Ceramic Fuel Cells," *J. Am. Ceram. Soc*., **76** [3], 563-88, (1993).