DEMONSTRATION OF ANODE SUPPORTED CELL TECHNOLOGY IN kW CLASS STACK

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ABSTRACT

CFCL has developed anode supported cell technology for operation in the temperature range 700°C – 800°C. The cells have been scaled-up to 110x90mm in size and consist of a 3-layer laminate (support plus anode 500 - 700μm laminated with 20μm 8mol% yttria-zirconia electrolyte) and a screen-printed LSM cathode. The laminate structure is produced by tapecasting support anode and electrolyte tapes followed by roll lamination. Significant pore structure optimisation was required to allow operation with high steam content in the fuel. All production processes are being carried out on pilot plant production equipment (current capacity is 20 cells per week). The kW size stack consists of 22 layers of a 2x2 array design. Current collection and gas flow distribution has been developed in single cell and multicell monostack and a 2 layer 2x2 array prototype.

INTRODUCTION

CFCL has been developing SOFC technology for lower operating temperatures for several years. The major drivers included a larger choice in the selection of materials for interconnects and other stack and BoP components, lower costs of such materials, lower degradation, and potentially higher thermodynamic efficiency. After evaluating a number of options: scandia-zirconia electrolytes, magnetron sputtered 8YSZ layers on anode support structures (1) and laminated anode supported cells (2), CFCL focussed on the laminate cell technology fulfilling both performance targets and allowing ready up-scale of fabrication. With advanced cathodes, CFCL achieved up to 1W/cm² for 50x50mm cells. The technology has been scaled to 110x90 in size and the current production rate of 20 cell/week. With standard LSM cathodes, the larger cells achieve about 200mW/cm² at 750°C, but this output was sufficient for this stack demonstration. With a higher performing cathode significantly higher performance is achievable (over 1W/cm² at low Uf). This paper describes the recent PEN development work and the stacking technology and construction and testing of a kW class stack with anode supported cells.
RESULTS AND DISCUSSION

Cell development and production:

CFCL anode substrate cells are layered composites (2) consisting of:
- a porous, 700 μm thick nickel-zirconia substrate anode layer,
- a dense, 20 μm 8mol% yttria-zirconia electrolyte layer,
- and a thin perovskite cathode layer.

The methods used to produce the laminate cells are similar to those used in the production of electronic ceramics. Substrate and electrolyte sheets are tape cast, cut to size and laminated using a two-roll mill. The resulting structure is fired to a temperature sufficient to densify the electrolyte layer to greater than 96% of theoretical. The perovskite cathode is deposited on the electrolyte by screen printing and firing below 1200°C. Typically cells are produced in 2.0 cm² discs, and 25 cm² and 100 cm² plates.

Recent process developments have increased the permeability of the substrate and improved the reliability of the processing methods. 100 cm² cells are now the standard, and this size is seen to be sufficient for a commercial fuel cell product. The process methods have been scaled up to where the daily production of the cells has been moved out of the laboratory and into a pilot-plant environment.

Anode side Development

The use of hydrocarbon fuels in fuel cell stacks will necessitate moving large amounts of water in and out of the anode layer. In order to avoid losses due to concentration polarisation, development work was carried out to increase the permeability of the nickel-zirconia substrate. To accomplish this, pore-former of different particle size and in increased quantities has been used. This process change has increased the size and amount of porosity in these layers.

To ensure good electrochemical performance at the anode/electrolyte interface, a separate thin anode layer with a finer porosity and nickel particle size has been introduced. This tape is laminated between the nickel-zirconia substrate and the electrolyte. The resulting anode structure is approximately 30 μm thick.

In order to achieve flat PENs it was necessary to match shrinkages of the three layers. The shrinkage behaviour was controlled through particle size of the zirconia and green density of the tape cast sheet (2). Testing of this three layer structure has shown no significant evidence of increased losses due to concentration polarisation. Figure 1 shows the V-I curves of a cell at 2.5, 40 and 60% water content in hydrogen.
Laminate Cell Testing

Long-term (>8000 hour) testing of a two-layer laminate PEN was conducted in a single cell test stand. A laminate PEN with a single anode substrate layer was tested over a period of 4000 hours at 740°C and the temperature was then raised to 750°C and the cell was run for another 4000 hours. The open circuit voltage was around 1.1 V (theoretical OCV). The stability of the cell was good, first the performance improved during the first 4000 hours, and then a very slow degradation was observed during the final 4000 hours. Post test analysis showed that the cell was connected only over about half of the cathode area so that some of the cell had not been active. The time dependent overpotential and ohmic losses determined by in-situ Galvanostatic Current Interrupt (GCI) experiments are shown in Table 1 below:

| Time of operation hours | Temp C | Ohmic losses $\Omega$ cm$^2$ | Overpotential losses MV (at 330 mA/cm$^2$) |
|-------------------------|--------|-------------------------------|---------------------------------------------|
| 139                     | 740    | 0.37                          | 248                                         |
| 3355                    | 740    | 0.35                          | 241                                         |
| 4004                    | 750    | 0.32                          | 239                                         |
| 8035                    | 750    | 0.34                          | 264                                         |

1 Data is normalised to a smaller area since post-test inspection of the cell showed that less 60% of the cell was active due to poor connection.

Development of Stacking Technology

Good connection on both cathode and anode side proved vital in order to achieve long term stability in stacks. Chemical bonds between current collectors and electrodes performed the best. A Nickel mesh structure was used as current collector on the anode.
side and a silver mesh on the cathode side. Chemical bonding was achieved with special contact layers between the electrodes and the meshes. Table 2 shows cell ohmic resistance losses with different mesh structures and with and without bonding layers. The arrangement with the lowest loss was used for stack testing.

Table 2: Ohmic cell losses as function of connection technology:

| Anode side                | Cathode side                          | Ohmic resistance Ωcm² |
|---------------------------|---------------------------------------|-----------------------|
| Ni mesh                   | Ag mesh 1 - Pt wire connection         | 1.09                  |
| Ni mesh                   | Ag mesh 2 - Pt wire connection; cell and current collector heat treated at 925°C prior to assembly | 0.58                  |
| Ni mesh + contact layer   | Ag mesh 2 - Pt wire connection + contact layer | 0.58                  |
| Ni mesh + contact layer   | Ag mesh 2 + contact layer with interconnect through connection | 0.30                  |

Cell Performance Testing

Figure 2 shows the performance of a 110x90mm cell over a period of 600 hours at temperatures of 750 and 775°C and current densities of 200 and 250mA/cm² with 2% H₂O – 98% H₂ as fuel and air.

![Figure 2. Cell performance at 750 and 775°C tested in alumina assembly](image)

This cell was tested in an alumina housing to obtain a reference performance without metal interconnect components. The contact technology giving the lowest ohmic loss (see Table 2) was used. At 750°C and 200mA/cm² the performance first improved, then degraded to about 150 hours, and then slowly improved again. In situ GCI tests measured constant cell ohmic losses of 0.31 Ωcm². The cell performance continued to improve at
current densities of 250 mA/cm² – overpotentials decreased (0.3 Ωcm²; 270 mV). At 775°C, 790 mV were obtained at 250 mA/cm².

**Stack Testing**

The effectiveness (output and stability) of the chosen stacking technology has been assessed in single and five cell stack configurations. A 2 layer 2x2 array stack is currently under evaluation and a 20 layer 2x2 array stack (kW class stack) is under construction for testing in mid May. A number of single cell stacks have been assembled and tested earlier to optimise stacking technology and decrease losses (see Table 2). Figure 3 shows a 1000 hour stability test at 750°C and 200mA/cm². The cell sealed well as evident from the open circuit voltage of 1.11 V for 2%H₂O - H₂ versus air. GCI experiments measured an ohmic resistance of 0.34 Ωcm² and overpotential losses of 220 mV. Cell performance was stable over the test period.

![Figure 3. Stability test in single cell stack](image)

A 5-cell stack was assembled using the same connection technology as the single cell stack above. Figure 4 shows the performance of the stack at 750°C and 200 mA/cm² over the test period of 500 hours. The stack sealed well and all 5 cells reached theoretical OCV. Under load cell performance initially decreased over the first 250 hours then stabilised, except for cell 4. Stability and performance of individual cells was to some extent fuel and air flow dependent. For example, lowering of the fuel flow after 250 hours of operation narrowed the performance spread of the cells. This stacking arrangement is characterised by low pressure drops across cells resulting in uneven flow distributions between layers due to dynamic pressure variations in the manifold.
Future Work

In the area of cell improvement, work in the near future will focus on PEN cost reduction, electrolyte flaw reduction and development of high performance cathodes technology. The focus in stacking technology development is on assessment and improvement of current collection technology for thermal cycling development A 20 layer 2x2 array stack is being constructed and testing will start in mid May.

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