ABSTRACT

ECN is carrying out a materials development program for ceramic SOFC components together with two major system and stack developers in Europe. The development program has resulted in optimized, upscaled cells with an internal resistance of 0.3 Ohm.cm². Maximum sintered cell dimensions are in the order of 300–400 cm² for respectively circular and rectangular cells. Manufacturing techniques are tape casting for the electrolyte and screen printing or tape casting for the electrodes. Between the metallic separator plate and the cells a ceramic, intermediate layer is applied, in order to improve electrical contact and to stabilize the interface between the stack components. Sealing of the cells is carried out with a glass gasket which is also made by tape casting. In addition, a component development program for low temperature (800°C) SOFC operation is carried out. Experimental data for cells and stacks are presented.

INTRODUCTION

SOFC development at ECN (1,2) is carried in collaborative research projects with Siemens AG (Germany) and Sulzer Innotec (Switzerland). In these projects ECN has concentrated on the materials aspects of cell and stack components with the aim to obtain cells with high performance and on manufacturing techniques for upscaled components. The optimized and upscaled cells will be applied in the flat plat or "multiple array" reactor concept of Siemens (3) or in the flat plate HEXIS concept of Sulzer Innotec (4). In both cases a metal separator plate will be used. For reasons of reduction of costs of the total system, a development of low temperature (800°C) SOFC components is also carried out in collaboration with the partners.

UPSCALED STATE-OF-THE-ART CELLS

For optimization of the cell performance different cathode and anode powder
morphologies have been investigated. The objectives of the optimization process were: 1) a large amount of three-phase boundaries at the electrolyte/electrode interface and 2) optimum adhesion of the electrodes to the electrolyte. The results of these investigations have been described in detail in several papers (5,6,7). From the electrode powders electrode/electrolyte sandwiches were made by tape casting, which were characterized with impedance spectroscopy. With this method a semi-quantitative measure could be derived for the three-phase boundary length, which proved to be a valuable method for screening optimized electrode microstructures for high performance cells. Under ideal testing conditions, i.e. low utilization of the fuel gas and Pt-current collectors, the internal resistance of standard (10x10 cm²) ECN cells is in the order of .3 Ohm.cm² (Fig. 1). Under specific gas compositions which simulate the local conditions along the electrodes of an upscaled cell, endurance tests over 3000 hours have been carried out (Fig. 2).

LOW TEMPERATURE CELLS

In order to be cost-competitive with conventional power plants (50-100 MW) the operation temperature of SOFC needs to be lowered to temperatures around 800°C. At lower temperatures, the use of state-of-the-art balance of plant (BOP) components can be envisaged. In addition, longer life-time is assured, due to reduced corrosion effects of metal parts in the SOFC stack. In collaboration with the partners, the work at ECN is focusing on three lines: 1/ optimization of the microstructure of state-of-the-art electrode materials through improved ceramic processing, 2/ development of alternative manufacturing techniques for thin ceramic components and 3/ development of alternative electrolyte/electrode materials with optimized microstructures.

Manufacturing techniques have been developed for 5x5 cm², self-supporting, thin electrolytes of TZ–3Y (60 µm) and 20 mol% gadolinia-doped ceria (130 µm). Electrolyte structures with a density of >97% can be formed by sintering at 1500°C. In addition, a calendaring and cofiring technique is developed for manufacturing electrode supported, thin (40 µm) electrolytes.

The performance of a cell with a thin (60 µm) TZ–3Y electrolyte and state-of-the-art electrodes with optimized microstructures is shown in Fig. 3 as a function of operation temperature. More recently, cells with a 10 mol% Gd-doped Ce–oxide electrolyte of 130 µm, a state-of-the-art Ni/zirconia cermet anode and a cathode with the composition La₆Sr₄Fe₈Co₂O₃ were tested. At 650°C a power output of 220 mW/cm² was observed (8). Further improvement for these cells is to be expected from optimization of the cathode microstructure with the above described impedance spectroscopy approach.
STACK TESTING

For the metal separator plate, ECN has focussed on the development of a simplified design with respect to manufacturing costs and gas distribution. For optimization of the stack characteristics a small scale design of HA230 or the Plansee alloy of Siemens is used for cells with an active electrode area of 15 cm². More recently, a revised design has been manufactured for upscaled (10x10 cm²) cells. The design is characterized by internal manifolding in the separator plate and the electrolyte. Sealing of the cells is carried out with a glass gasket along the outer edges of the cell housing and around the gas-passages.

In order to avoid Cr-evaporation from the separator plate and to optimize the electrical contact between the separator plate and the cell, an intermediate layer is applied between the two components. The characteristics of this intermediate layer have been optimized by evaluating different powder morphologies/compositions and deposition techniques. The intermediate layer may consist of a coating on the separator plate and/or a separate, metallic or ceramic cushioning element between the separator plate and the cell.

Stack tests with metal current collectors using optimized ECN components of 5x5 and 10x10 cm² have been carried out by ECN and partners. The results of the different tests are reported in Table 1. Most important for an optimum performance is the character of the ceramic intermediate layer between the metal separator plate and the cell.

Table I. Stack testing experiments with ECN cells and metal current collectors.

| TEST BY ACT. ELECT. | OXIDANT/ TEMP (°C) | TIME (HRS) | I at .7V (mA/CM²) |
|---------------------|--------------------|------------|-------------------|
| ECN 15 O₂/920       | 48                 | 500        |
| SIEMENS 81 O₂/950   | 10                 | 550        |
|                    | AIR/950            | 11         | 250               |
|                    | O₂/850             | 1000       | 225               |
| SULTZER 2.4 AIR/1000| 350                | 250        |
|                    | AIR/920            | 800        | 170               |

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CONCLUSIONS

The SOFC program at ECN is carried out in direct collaboration with two major system/stack developers and manufacturers in Europe, i.e. Siemens and Sulzer Innotec. The program concentrates on the development and upscaling of optimized, high performance stack components, which are tailored to the needs of system partners. By ceramic processing and electrochemical characterization with impedance spectroscopy of electrode/electrolyte half-cells, high performance cells with low internal resistance at 930°C could be manufactured. In addition, low temperature cells for a second generation of SOFC's have been developed with optimized state-of-the-art materials and with alternative materials, which potentially offer the possibility to lower operation temperatures to 700–800°C. Future work will concentrate on durability, use of natural gas, cost reduction of the stack components and optimization of low temperature cells.

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Figure 1. Performance of upscaled (10x10 cm² electrolyte) with standard electrodes (81 cm²) and platinum current collectors with oxygen or air on the cathode side and low utilization on the anode side.
Figure 2. Performance of small cell (2x2 cm² electrolyte) with standard electrodes (3 cm²) and platinum current collectors as a function of time at 40% utilization of the anode gas (humidified hydrogen) and air on the cathode side.

Figure 3. Performance as a function of temperature of a cell (10 cm²) with a thin (60 μm) electrolyte of TZ-3Y and state-of-the-art electrode materials, which were optimized for low temperature operation. Tests were carried out with platinum current collectors.