

Fabrication and Electrochemical Performance of Unit Anode Supported Intermediate Temperature Solid Oxide Fuel Cells by Single Step Process

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In this work micron sized particles of NiO, Gd_{0.1}Ce_{0.9}O_{1.9} (GDC10), and La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O₃ (LSCF48) were used in the preparation unit anode supported planar Intermediate Temperature Solid Oxide Fuel Cell (IT-SOFC). The laboratories of ICB in Dijon and FC-LAB in Belfort France were aimed at preparation of large active area planar cells by low cost effective and efficient fabrication process with high performance and characterizing them in real time stack for use in APU application. The cells were prepared by novel single step and in expensive, process. These cells were characterized electrochemically to have I-V and I-P curves. The study of microstructure was carried on the cells. The initial results obtained were reported in this paper with still large possible room for optimization to further improve the performances of the cells. The maximum power density obtained was 466mW.cm⁻² at 648°C. At 500°C 1.002V were obtained.

Introduction

Solid Oxide fuel cells (SOFC) are extensively studied in the recent years due to its greatest advantages like high electrochemical conversion efficiency, with low toxic emission and flexibility to use variety of fuels (1-2). During initial stages, in conventional SOFCs yttria stabilized zirconia (YSZ) were used as an electrolyte which was best suited for operating temperatures of 800°C to 1000°C. These high operating temperature utilization leads to variety of problems, right from difficulties in materials selection, fabrication, low performance and reliability of this system which in turn affected the mass commercialization (3). The material selection and the fabrication play a crucial role in the performance of cells with main criteria being reduction of operating temperature down to less than 700°C. These low temperature cells are known as Intermediate Temperature Solid Oxide Fuel Cells (IT-SOFC). So far effective commercialization of SOFCs, all factors must be carefully tailored to achieve low possible working temperature with high performance design, low material degradation, longer life time and a cost effective manufacturing process. Anode supported planar cells with thin electrolyte film are widely preferred than tubular design, electrolyte and cathode supported cells for their low fabrication cost, low ohmic resistance and high performance (4-6). The previously reported preparation method of anode supported planar cells from literature is done in two steps: first step being the preparation of half cell with thin electrolyte film on anode support and sintering at high temperature to get dense electrolyte followed by application of cathode on half cell and sintering to achieve a unit working cell (7).

The application of thin electrolyte film and cathode is generally done by spin coating (8) chemical vapor deposition CVD (9), dip coating (10), screen printing (11). These methods involve high initial capital costs and many steps of sintering at high temperature increases the cost of fabrication and thus lead to costly processes. In this paper, the preparation of the large active area (50 cm^2) planar cells prepared by simple, facile tape casting process is explained and initial results obtained from 10 cm^2 active area cells are presented.

Experiment

NiO from Sigma Aldrich and GDC 10 powders from Neyco and in house produced LSCF48 were used as raw active materials for the preparation of tape casting slurry to obtain unit cell. The slurries for anode support, anode functional layer, electrolyte and cathode layers were prepared in two steps process. First active materials were ball mixed with solvent and dispersant for 24 hours in second step binders and plasticizers were added and balled for another 24 hours to obtain a homogenous slurry. For anode support slurry carbon black pore former is used and studied electrochemically.

The cells preparation is explained in our previous work (12, 13). These unit cells were electrochemically characterized in open flange Fiaxell device. 10 cm^2 active area cells are tested in this device with humidified hydrogen containing 3% of water as fuel and air as oxidant in a fixed flow rate. Microstructure study was carried out in Scanning electron microscope (SEM) device with the model number SU1510.

Results and Discussion

Figure 1 Shows the voltage and power density of 10 cm^2 planar cells measured as a function of current density in the temperature range of 500°C to 650°C . The initial OCV values at 500°C , 550°C and 600°C and 650°C were 0.920V , 0.862V , 0.785V , and 0.667V respectively. The maximum power density of $47 \text{ mW}\cdot\text{cm}^{-2}$ at 600°C was obtained.

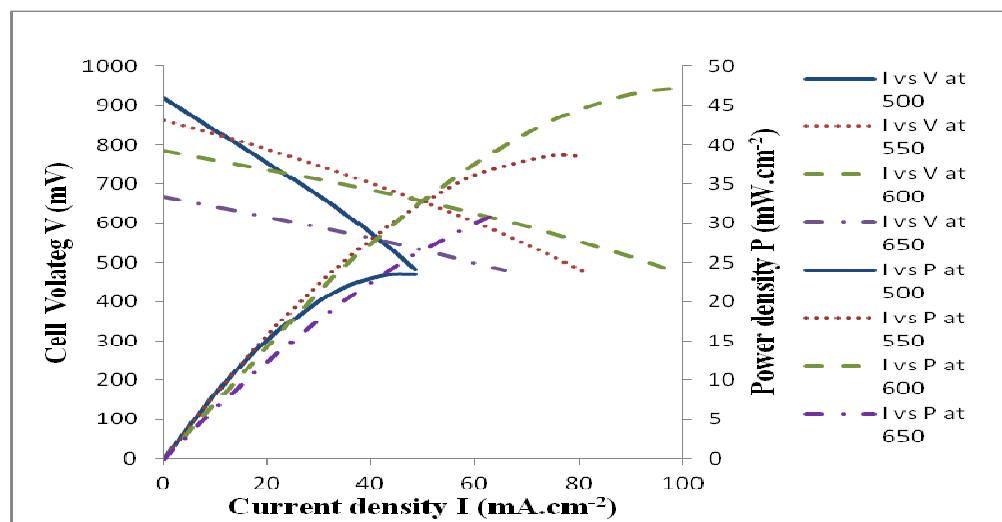


Figure 1. Voltage and Power density of unit planar cell as a function of current density

Figure 2a. Shows the Back Scattered SEM image of cross section of unit cell. The cathode on the left side of the image was made from composite of LSCF48-GDC10. The white electrolyte layer in the cross section image was prepared from GDC10 slurry, and anode functional layer (afl) was made of NiO-GDC10 without carbon black pore former. The anode support layer was the same as an anode active layer but carbon black was used as a pore former. From the figure 2a, it can be seen that the thickness of cathode was around 10 μm , electrolyte thickness was maintained around 15 μm and afl thickness was 15 μm and the remaining being anode support layer. The mixing of carbon black was even and it shows homogenous pore distribution. Since the thickness of cathode layer was less than 10 μm , small bubbles were formed during fabrication. This were observed by SEM on the cathode surface and it is shown in figure 2b.

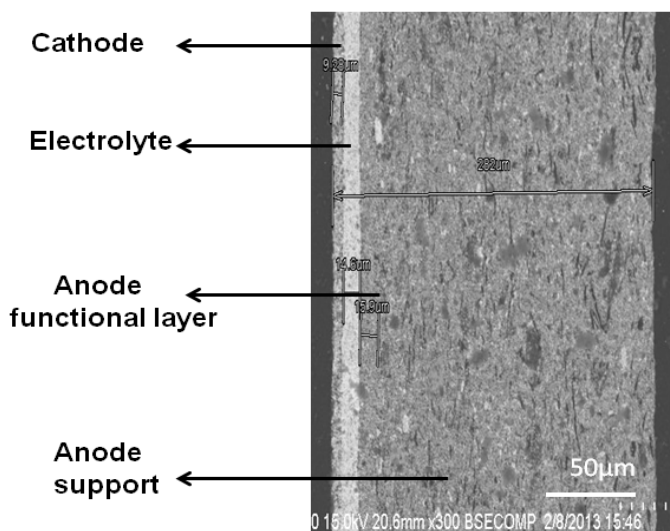


Figure 2a. Back scattered SEM image of Cross section of unit planar cell.

The probable reason for low power density can be due to water logging on the bubbles from electrochemical reaction, which might rise internal resistance and as well as cause possible delamination of cathode. There can also be some other possible unfavorable reactions which are yet to be explored.

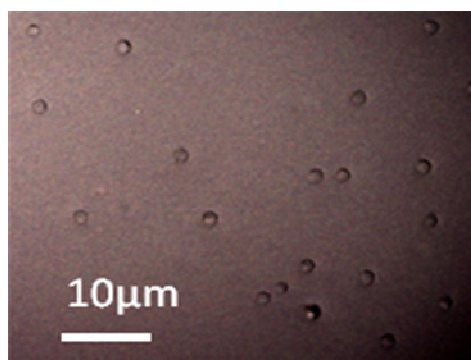


Figure 2b. Cathode surface with bubbles.

The microstructure and the process of fabrication were partly optimized in each layer of cell to obtain the maximum of 1.002 V at 500°C and maximum power density of 466mW.cm⁻² at 648°C as reported in (13). These initial results obtained by this new process show promising cell performances. More engineering and optimization of electrodes, electrolyte and microstructure are needed to realize this process at a commercial scale.

Conclusion

Large area flawless planar cells are effectively fabricated by the single step process. The initial power density results from first cells at 600°C were 47mW.cm⁻². Small optimization lead us to obtain maximum of 1.002 V at 500°C and maximum power density of 466mW.cm⁻² at 648 °C, and confirms that this is an effective process for mass fabrication at low cost compared to the present process of preparation of cell.

Acknowledgments

Authors would like to thank region of Bourgogne and region of Franche-comte for the financial support, and also express word of gratitude to Raphel Ihringer from Fiaxell Sarl (EPFL Scientific Park A) for helping in electrochemical measurements of large cells.

References

1. B.C.H. Steele, *Nature*, **400**, 619, (1999).
2. K. Joon, *J. Power Sources*, **71**, 12, (1998).
3. F. Tietz, H-P Buchkremer, D. Stover, *Solid State Ionics*, **152-153**, 373, (2002).
4. Y. J. Leng, S. H. Chan, K. A. Khor, S. P. Jiang and P. Cheang, *J. Power Sources*, **117**, 26, (2003).
5. L. Blum, W.A. Meulenber, H. Nabielek, *J. Appl. Ceram. Tech*, **2**, 482, (2005).
6. Y.J. Leng, H. Chan, S.P. Jiang, K.A. Khor, *Solid State Ionics*, **170**, 9, (2004).
7. H.Y. Yung, S-H. Choi, H. Kim, J-W. Son, J. Kim, H-W. Lee, J-H. Lee *J. Power Sources*, **159**, 478, (2006).
8. X. Xu, C. Xia, S. Huang, D. Peng, *Ceram. Intern*, **31**, 1061, (2005).
9. Z. Xu, J. Sankar, S. Yarmolenko, *Surf. Coat. Tec*, **52**, 177, (2004).
10. C.R. Xia, S.W. Zha, W.G. Yang, R.R. Peng G.Y. Meng, *Solid State Ionics*, **133**, 287, (2000).
11. P.V. Dollen, S. Barnett, *J. Amer. Ceram. Soc*, **88**, 3361, (2005).
12. V. Sivasankaran, L. Combemale, G. Caboche, *French patent*, FR12/59643.
13. V. Sivasankaran, L. Combemale, M.C. Pera, G. Caboche, *Fuel cells - from Fundamentals to Systems*, (accepted, 2013).