

## Copper Sintering in Cu-Ceria-Sm<sub>0.2</sub>Ce<sub>0.8</sub>O<sub>1.9</sub> Anodes

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### Introduction

Solid oxide fuel cells with Cu-ceria based anodes have been developed for the direct utilization of hydrocarbon fuels to potentially simplify system design and reduce system cost (1). Ceria is the catalyst for hydrocarbon oxidation in the anode, and Cu is the current collector that conducts electrons away from the anode-electrolyte interface (2, 3). Because of low melting points of Cu oxides and high processing temperatures to obtain dense electrolytes, new fabrication techniques have been successfully developed to prepare Cu-ceria based anodes (2, 4). Due to the uniqueness of these techniques, copper oxide in the anodes only covers the surface of the porous anode structure. During the reduction from Cu oxide to Cu in a H<sub>2</sub> stream, Cu sintering may happen and degrade cell performance.

In this work, the stability of cells with a Cu-ceria-Sm<sub>0.2</sub>Ce<sub>0.8</sub>O<sub>1.9</sub> (SDC) anode is studied in H<sub>2</sub> at 973K. We report the cell performance over 9 hours, and present a model involving Cu sintering in the anode to interpret cell-performance variations. A strategy to impede Cu sintering is discussed and explored.

### Experimental

Single cells in this study are fabricated using the pressing method developed previously (2), and the anode composition is Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC where the percentage is the constituent weight over the weight of SDC in the anode. Tantalum oxide is incorporated into some anodes using its isopropoxide to suppress Cu sintering. Platinum and gold current collectors are attached to the cathode and anode, respectively. The cells are then sealed onto an alumina-supporting tube using a high-temperature ceramic cement (Aremco<sup>TM</sup>). The cathode is exposed to air, its active area is ~1.2cm<sup>2</sup>. The Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC anode is exposed to Ar when temperature is raised from ambient temperature to 973K. Then H<sub>2</sub>, with a flow rate of 20 ml/min at 1 atm, replaces Ar in the anodes. Cell current-voltage (I-V) curves and impedance spectra are collected as a function of time using a Solartron 1287 interfaced with a Solartron 1250.

The four-probe method is used to measure the conductivity of a porous Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC sample during the reduction of copper oxide to Cu in H<sub>2</sub> at 973K. Porous Cu<sub>20%</sub>-ceria<sub>10%</sub>-YSZ anodes are also prepared using the tape casting techniques (4), and are reduced in H<sub>2</sub> using different temperature profiles. Their morphologies are then examined using a JEOL 6300 SEM equipped with an energy-dispersive X-ray spectroscopy (EDS). The accelerating voltage is 15keV and the typical magnification is 5000.

### Results and Discussion

Fig. 1 displays the variation of cell maximum power densities (MPD) and conductance with time in H<sub>2</sub> at 973K. The MPD of the cell with Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC anode drops from 129mW/cm<sup>2</sup> to 32mW/cm<sup>2</sup> as the time increases from 15 to 105 minutes. Then it increases slowly to 25mW/cm<sup>2</sup> after 540 min. The degrading trend of cell conductance matches the declining MPD, indicating that the MPD variation is mainly associated with Cu sintering in the anodes.

To impede Cu sintering in the anodes, tantalum oxide is included in a Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC anode. The resultant cell exhibits improved stability as shown in Fig. 1. The cell MPD in this case decreases much slowly. After 9-hr operation, its MPD is still 97mW/cm<sup>2</sup> approximately 4 times higher than that of the cell without Ta<sub>2</sub>O<sub>5</sub> in the anode. Cell conductance gradually decreases from 0.76S/cm<sup>2</sup> to 0.65S/cm<sup>2</sup>, but it does not display the slump behavior. These promising results suggest the possibility of suppressing Cu sintering by modifying the present anode design.

### Conclusions

This work focuses on the study of copper sintering in SOFCs with Cu-ceria-SDC anodes. Copper sintering happens at 973K, and it can lead to the loss of anode conductivity and the degradation of cell performance over 9-hr operation on H<sub>2</sub> fuel. However, it is possible to impede Cu sintering and improve anode stability with a modified anode design.

### Acknowledgements

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### References

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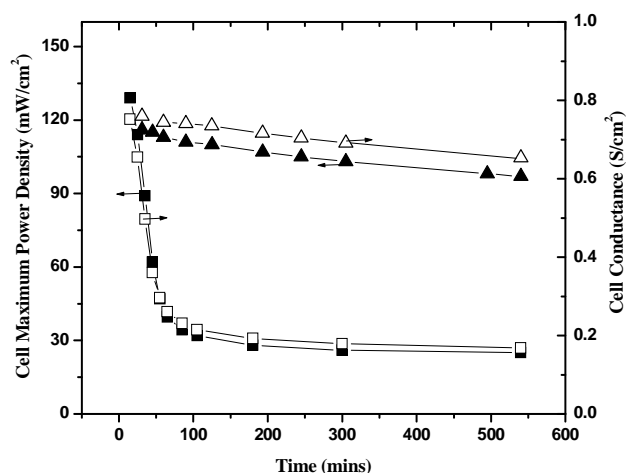


Fig. 1 The variation of cell maximum power densities and conductance with time in H<sub>2</sub> at 973K. Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC anode (square), and an improved Cu<sub>25%</sub>-ceria<sub>16%</sub>-SDC anode with the addition of Ta<sub>2</sub>O<sub>5</sub> (triangle).