

# Direct Electrochemical Oxidation of CH<sub>4</sub> in a Solid Oxide Fuel Cell

Rahul Singh, Felipe Guzman and Steven Chuang\*

Department of Chemical and Biomolecular Engineering, The University of Akron, OH, 44325-3906 (USA)

\*schuang@uakron.edu

## Introduction

The direct electrochemical oxidation of hydrocarbons in a solid oxide fuel cell (SOFC) is an attractive energy conversion approach attributed to (i) higher energy conversion efficiency, (ii) simplicity in the overall system architecture and balance of plant and (iii) availability of constant concentration of fuel in the anode compartment providing a stability factor<sup>1</sup>. CH<sub>4</sub> is the most common hydrocarbon fuel of interest because of its abundance and availability in the form of natural gas. The extreme operational temperature of SOFC at 700–1000 °C provides a thermodynamically favorable pathway to deposit carbon on the most commonly used Ni-YSZ anode from CH<sub>4</sub>, thus deteriorating the cell performance, durability and stability<sup>2</sup>.

DFT (Density Functional Theory) calculation on the basis of thermodynamic and kinetic energy path for carbon formation on Ni surface suggested those stepped Ni surfaces are more active towards C-C bond formation than planar Ni surfaces<sup>3</sup>. The poor catalytic activity of Cu to catalyze C-C bond formation makes it an ideal candidate for blocking the step sites on Ni to avoid carbon deposition<sup>4</sup>. This paper will present a detailed investigation of direct electrochemical oxidation of CH<sub>4</sub> on a Cu-Ni bimetallic anode, focusing on the microstructure and performance characteristics (voltage-current curve and impedance) of Ni and Cu-Ni anode before and after long term exposure to CH<sub>4</sub>.

## Materials and Methods

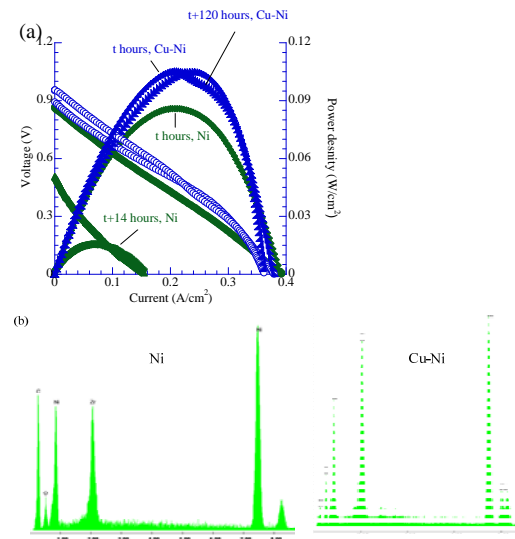
The SOFC used for this study consisted of Ni and Cu-Ni anode (700 µm), YSZ (Yttria stabilized Zirconia, TZ-8Y) electrolyte (18 µm) and LSM-YSZ (Lanthanum Strontium Manganese oxide, Heraeus) cathode (40 µm) fabricated by tape casting technique. The anode supported cells were obtained by firing the green discs at 1400 °C for 4 hr. The resulting cell was screen printed with 50 wt% LSM-YSZ cathode layer and a pure LSM cathode current collector layer.

The fuel cells were tested in 50 vol% of fuel (H<sub>2</sub> and CH<sub>4</sub>) and N<sub>2</sub> balance (total flow rate = 200 cm<sup>3</sup>/min) at 750 °C. The current voltage performance was measured by a Solartron Multichannel Cell Test system and post-experimental study, was characterized by SEM-EDAX (FEI Quanta 200) and XRD (Philips Analytical X-ray, Cu tube anode @ 1.54 Å). The characterization and performance analysis was performed for a fresh Ni and Cu-Ni on H<sub>2</sub> and CH<sub>4</sub> and results were compared.

## Results and Discussion

Figure 1(a) shows the change in current-voltage performance with time for Ni and Cu-Ni anode exposed to CH<sub>4</sub> at 750 °C. The maximum power density for CH<sub>4</sub> exposed to Ni anode was 0.085 W/cm<sup>2</sup> initially. The fuel cell was further exposed to CH<sub>4</sub> for 13 h resulting in 85 % decrease in the power indicating that power generation ability of the Ni-SOFC was severely affected after prolong exposure to CH<sub>4</sub>. Blockage of pores at anode for CH<sub>4</sub> fuel,

Ohmic resistances and diffusion limitation phenomena can significantly reduce the performance of the cell.



**Figure 1.** (a) Current-voltage performance variation with time for CH<sub>4</sub> on Ni and Cu-Ni anode at 750 °C, and (b) EDAX spectra of Ni and Cu-Ni anode exposed to CH<sub>4</sub> at 750 °C.

The exposure of CH<sub>4</sub> over a fresh Cu-Ni anode produced 0.1 W/cm<sup>2</sup> at 750 °C and after 120 h of exposure, the power performance remained constant. This result suggests that addition of Cu to the Ni cermet anode indeed limits the deactivation phenomena of anode from exposure to CH<sub>4</sub> fuel. Analysis of EDAX results shown in figure 1(b) further confirms the ability of Cu-Ni anode to reduce the carbon deposition observed by a lower carbon peak intensity.

## Significance

The direct reforming of CH<sub>4</sub> in a SOFC has a potential of commercialization in the area of auxiliary and portable power units and battery chargers. The microstructural and performance characteristics information would guide the development of an effective anode for the direct CH<sub>4</sub> solid oxide fuel cell.

## References

1. Kawano, M., Matsui, T., Kikuchi, R., Yoshida, H., Inagaki, T., Eguchi, K., *J. Electrochem. Soc.*, 154, B460 (2007)
2. Liu, J., Barnett, S.A., *Solid State Ionics*, 158, 11 (2003)
3. Galea, N.M., Kadantsev, E.S., Ziegler, T., *J. Phys. Chem. C*, 111, 14457 (2007)
4. Gorte, R.J., Vohs, J.M., McIntosh, S., *Solid State Ionics*, 175, 1 (2004)