Modeling High Fuel Utilization in Direct Oxidation SOFCs

Olga Costa-Nunes[1], John M. Vohs[1], Raymond J. Gorte[1]
[1]Department of Chemical and Biomolecular Engineering, University of Pennsylvania, PA 19104, USA.

Introduction

Work in our laboratory has recently demonstrated the direct, electrochemical oxidation of a variety of hydrocarbon fuels in solid-oxide fuel cells (SOFC) using Cu-based anodes [1,2]. Fuel cells that can produce electricity by direct electrochemical oxidation of hydrocarbons could have significant efficiency advantages over conventional systems. However, fuel dilution can also be a problem in direct-oxidation fuel cells because each fuel molecule produces a larger number of product molecules. Since water is produced at the anode by oxidation of the hydrocarbon fuel, there is the possibility of using this water to carry out reforming of unreacted hydrocarbons, thus increasing the concentration of fuel by the production of H_2, over a reforming catalyst within the stack.

In the work described in this paper, we examined the likely effect on cell performance that higher fuel utilizations would have by feeding a partially oxidized fuel to a model SOFC. To produce the products that would be obtained at higher conversions in direct oxidation of a hydrocarbon on an anode composed of Cu, ceria, and yttria-stabilized zirconia (YSZ), we pre-oxidized n-butane with varying amounts of O_2 on a ceria catalyst in a tubular reactor at 973 K before feeding the products to the SOFC. To determine the effect of having a reforming catalyst in the anode, we pre-oxidized the fuel over Pd/ceria catalyst. The results demonstrate that the addition of a reforming catalyst to the anode compartment significantly changes the products that are formed and should be effective for improving performance at high fuel utilizations.

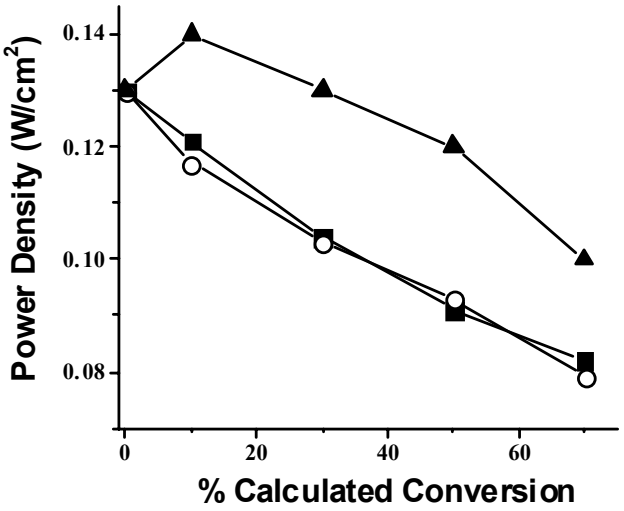
Results and Discussion

Figure 1 displays maximum power densities *versus* conversion trends obtained in different experiments. The fuel conversions were calculated as the amount of O_2 added to the oxidation reactor as a fraction of the amount of O_2 that would be required for complete combustion of the fuel. When ceria was used as a catalyst in the oxidation reactor, the maximum power density decreases continuously with fuel conversion, going from 0.13 W/cm^2 at 0% conversion to 0.08 W/cm^2 at 70% conversion, as Fig 1 indicates.

To simulate the effect of adding a reforming catalyst to the stack using the steam generated in the cell, we replaced the ceria catalyst with a Pd-ceria catalyst (1-wt% Pd) in the oxidation reactor. The performance levels using the Pd-ceria were remarkably higher as shown in Fig. 1, and the power densities were actually higher than we observe for pure n-butane at conversion less than 30%.

The reason for the improved performance of the cell when feeding the fuel oxidized by the Pd-ceria catalyst was elucidated by the composition of the fuel leaving the oxidation reactor, which was analyzed on a water-free basis using an on-line GC. The differences in the amounts of CO_2 and H_2 formed by the ceria and Pd-ceria catalysts provided evidences to support the explanation that remaining n-butane was reformed using H_2O generated by the oxidation over the Pd catalyst.

In the absence of reforming, it is important to determine whether CO_2 and H_2O simply dilute the fuel concentration or are chemically involved in the oxidation process at the three-phase boundary of the anode. To determine the effect of fuel dilution in the absence of chemistry, we tested a cell in n-butane diluted with He to a concentration that would be expected for various conversions, with the data also plotted in Fig. 1. The maximum power densities achieved following He dilution are almost identical to what we observe following oxidation of the fuel over the ceria catalyst. Therefore, the data in Fig. 1 indicate that the oxidation products, CO_2 and H_2O, affect cell performance primarily by diluting the fuel.



In summary, a drop of the SOFC performance with conversion was observed and associated to dilution in the products, CO_2 and H_2O. Using a good steam reforming catalyst overcomes the drop in performance and even improves it at high conversion conditions. The enhancement was attributed to electrochemical oxidation of H_2, which displays higher power densities than butane.

References

1. S. Park, J.M. Vohs, and, R. Gorte, *Nature*, 404 (2000) 265.
2. R. Gorte, S. Park, J.M. Vohs and C. Wang, *Adv. Mat*, 12, (2000) 1465.