

# Hydrogen Permeation of Cermet [Ba(Zr<sub>0.8-x</sub>Ce<sub>x</sub>Y<sub>0.2</sub>)O<sub>3-α</sub>/Ni] Membranes

Chendong Zuo\*, T. H. Lee, S.E. Dorris, U.  
Balachandran, and Meilin Liu\*

Energy Technology Division, Argonne National  
Laboratory, Argonne, IL 60439

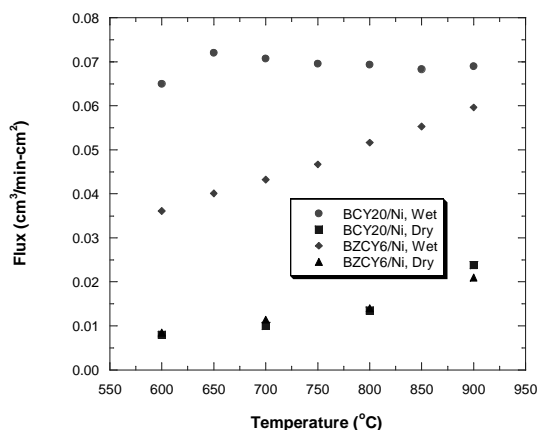
\* School of Materials Science and Engineering,  
Georgia Institute of Technology  
Atlanta, GA 30332

As the demand for hydrogen increases, a practical way of separating pure hydrogen from a gas mixture (e.g., synthesis gas) is becoming more important. Among the various separation techniques, dense membranes of mixed ionic-electronic conductors (MIECs) offer a simple way for obtaining hydrogen from gas streams at high temperatures.

Although various perovskite-type oxides have been reported to have high proton conductivity in a hydrogen-containing atmosphere, the chemical stability of these materials under practical conditions is still a concern.[1] For example, BaCeO<sub>3</sub>-based proton conductors display the highest conductivity among this class of materials; however, it is unstable in a H<sub>2</sub>O, CO<sub>2</sub>-containing atmosphere.[2] In contrast, BaZrO<sub>3</sub>-based proton conductors have excellent stability but relatively low proton conductivity.[3] These results suggest that the solid solution between cerate and zirconate may offer a good compromise between proton conductivity and chemical stability.[4,5]

Compositions in the BZCY [Ba(Zr<sub>0.8-x</sub>Ce<sub>x</sub>Y<sub>0.2</sub>)O<sub>3-α</sub>] system were prepared by solid-state reaction. The metal-ceramic BZCY/Ni mixture prepared by mechanically mixing 40 vol.% of metal with ceramic powder.

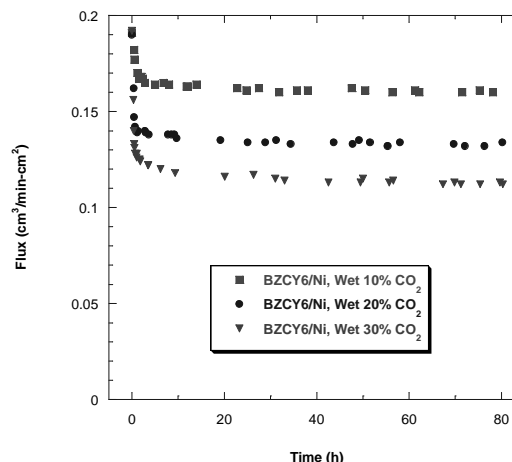
As shown in Fig. 1, the flux for BZCY6/Ni increased with temperature from 600 to 900°C, reaching 0.06 cm<sup>3</sup>/min-cm<sup>2</sup> at 900°C for a 0.75-mm-thick membrane using wet 4% H<sub>2</sub>/balance He as the feed gas. The flux was almost same in the dry condition and only slightly lower than that of BCY/Ni at high temperature in wet condition.



**Figure 1.** Hydrogen flux through 0.75-mm-thick Ba(Zr<sub>0.8-x</sub>Ce<sub>x</sub>Y<sub>0.2</sub>)O<sub>3</sub>/Ni (x=0.8, 0.6) membranes using wet 4% H<sub>2</sub> (balance He) as feed gas .

To determine whether Zr substitution improved the stability of BZCY relative to BCY, we monitored the

hydrogen flux through composite membranes during exposure to CO<sub>2</sub>-containing feed gas. Fig. 2 shows the hydrogen flux through the 0.75-mm-thick membrane made with BZCY6(x=0.6) and 40 vol.% Ni versus time using wet feed gas that contained 10, 20, or 30% CO<sub>2</sub> at 900°C, each of the feed gas also contained 40% H<sub>2</sub> with a balance of He. For measurements with a given CO<sub>2</sub> concentration, the initial reading (time = 0h) was taken using 40% H<sub>2</sub> balance He. After a small initial decrease during the first hour of exposure, the hydrogen flux through BZCY6/Ni cermet membrane was stable for 80 h in atmospheres containing up to 30% CO<sub>2</sub>, the initial drop increase with CO<sub>2</sub> content increase.



**Figure 2.** Time dependence of hydrogen flux through 0.75-mm-thick BZCY6/Ni membranes in feed gas of wet x% CO<sub>2</sub> (x=10, 20, 30, balance 40% H<sub>2</sub>/He) at 900 °C .

## Acknowledgements

This work was supported by U.S. Department of Energy, Office of Fossil Energy, National Energy Technology Laboratory's Gasification Technologies Program, under Contract W-31-109-Eng-38.

## Reference

1. K.D.Kreuer, Solid State Ionics 97, (1997) 1-15.
2. S. V. Bhide and A. V. Virkar, J. Electrochem. Soc., 146, (1999) 4386-4392.
3. K.D.Kreuer; Solid State Ionics 125, (1999) 285-302.
4. K. Katahira, Y. Kohchi, T. Shimura, H. Iwahara, Solid State Ionics 138, (2000) 91-98.
5. K. H. Ryu, S. M. Haile, Solid State Ionics 125, (1999) 355-367.