Proton conducting dense ceramic membranes for hydrogen separation and Membrane reactor applications

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Recently we have prepared a thulium doped strontium cerate (SCT) material with a significantly higher hydrogen permeability. The membranes will offer much higher hydrogen permeance if the thickness of the membranes is further reduced. The present project is aimed at preparation of thin (1-50 μm) membranes of the proton-conducting SCT material with improved hydrogen permeation flux, and investigation of the electric and hydrogen permeation properties of this new membrane. Currently we are investigating the use of three simple methods to prepare the SCT films on porous supports. We started synthesis of thin SCT films on porous alumina substrates by a polymeric sol-gel dipcoating method. Perovskite type thin films of SCT could be formed by multiple dip-coating process. However, the SCT films formed were not gas-tight to helium. More study is underway to improve the gas-tightness of the deposited films. The second method that we have been working on is the RF sputter deposition of the thin SCT films. Direct deposition of SCT films using SCT ceramic disks prepared by the solid state method as the target exhibited extremely low deposition rate. On the other hand, gas-tight palladium and copper alloy films were obtained with alternative sputtering method followed by annealing process. These results demonstrated an effective way to prepare the SCT films: sputter deposition of the individual metals (Tm, Ce, Sr) film followed by annealing to obtain the pervoskite-type STC ceramic oxide films. The third method we are working on is to prepare the supported SCT films by dry-pressing a mixture of alpha alumina (as support) and STC powder. Promising results were obtained when the thermal expansion coefficient discrepancy effect was alleviated by composition gradient. The carbon-powder prepared porous substrate totally avoided the problem of physical property difference between the substrate and the SCT top layer.

Theoretical study was focused on modeling of hydrogen permeation through the dense proton-conducting ceramic membranes based on the ambipolar approach

considering transport of all three charged species (proton, electron and electron-hole). A model for hydrogen permeation through mixed proton-electron conducting ceramic membranes was developed based on the ambipolar diffusion theory by considering proton, electron and electronic hole transport in the membranes. A general equation was derived which correlates the hydrogen permeation flux to the concentrations and diffusion coefficients of the three charged species. The equation was further simplified into analytic expressions relating the hydrogen permeation flux to the hydrogen partial pressures at upstream and downstream sides for special cases. The results of the theoretical model were compared with experimental hydrogen permeation data of SCT membranes. Good agreement between the model and experimental data was obtained.

List of Publications/Presentations (DE-FG26-00NT40818)

Papers Accepted or Submitted:

X.Qi., S. Cheng, Y.S. Lin, "Modeling and experimental study of hydrogen permeation through proton conducting ceramic membranes", Eds: A. Ramanarayanan, W.L. Worrell, and M. Mogensen, Proc. 4th Internal. Symp. on Ionic and Mixed-Conducting Ceramics, The Electrochemical Society, Pennington, NJ, in press (2001)

X. Qi and Y. S. Lin, "Modeling of hydrogen permeation through proton conducting ceramic membranes", *Solid State Ionics*, Submitted (2001)

Papers Presented in Conferences:

X. Qi, S. Cheng and Y.S. Lin, "Modeling and Experimental Study of Hydrogen Permeation through Proton Conducting Ceramic Membranes", The Electrochemical Society Annual Meeting, September 2-7, 2001, San Francisco, California

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