

Title: MOLECULAR MODELING OF SOLID OXIDE FUEL CELL ANODES IN THE PRESENCE OF H₂S
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Objective

Solid Oxide Fuel Cells (SOFCs) are fuel cells which operate at high temperatures ranging from 700°C – 1000°C. This makes them a prime candidate for converting effluent gas (syngas) from coal gasification to useful energy. Recent research has focused on increasing efficiency of an SOFC to perform under syngas conditions, specifically by reducing the deterrent effect of hydrogen sulfide (H₂S). This component of syngas deteriorates the performance of the anode materials in SOFCs. A typical SOFC has yttria-stabilized zirconia (YSZ) and nickel oxide yttria-stabilized zirconia (Ni-YSZ) as the electrolyte and anode respectively. The purpose of this project is to develop a theoretical model to understand the interaction of the Ni-YSZ anode in the presence of the H₂S contaminant, and validate this model using an experimental setup.

Accomplishments to date

Computational analysis has been carried out using Gaussian 03, Cerius² (v. 4.8) and CRYSTAL06 software programs. In Gaussian, binding energies and frequency calculations were performed using DFT methods. Cerius² was used to study the interactions between individual gas molecules and the anode surface. These interactions were used to calculate the interfacial diffusivity of the gases. The results from these calculations were reported in the published paper, the binding energies for Ni-YSZ+H₂, Ni-YSZ+H₂S and Ni-YSZ+H₂+H₂S systems were -80.1 kcal/mol, -21.4 kcal/mol and -21.6 kcal/mol respectively. The trend suggests that hydrogen oxidation at the anode is more thermodynamically favorable and the presence of H₂S decreases this H₂ activity. Also reported in the paper is the change in diffusivity of hydrogen with the presence of H₂S.

Due to the discrepancies of Gaussian-calculated Raman frequency modes with experimentally-determined values, the CRYSTAL program was used for the spectroscopy analysis. The calculated Raman frequency modes obtained from CRYSTAL showed better agreement with experimental values. These data from both CRYSTAL and experiments will be analyzed both statistically and visually. The quantum chemistry calculations for the monoclinic structure of zirconium oxide will also be used as a basis for calculation of the cubic structure of YSZ (commercial electrolyte), and then extend this calculation to the anode structure with nickel.

In the experimental design, the planar solid oxide fuel cell setup have 30mm diameter and 270 μm thickness cells with anode (Ni-YSZ) and cathode (LSM-YSZ) diameters of 10mm. Nickel and silver meshes are being used as the anode and cathode current collectors respectively. This setup is still in the final development stages and will be used to demonstrate the ability of the design to generate power as well as validate the theoretical model.

Future Work

In the remaining time left on this grant,

- Perform frequency calculations (Raman) on cubic YSZ to validate theoretical structure for electrolyte.
- Extend the frequency calculation analysis to NiYSZ anode.
- Perform V-I scan on planar SOFCs in H_2 gas to test the experimental setup and develop baseline calculations to be used with H_2S containing gas streams.

List of paper published

A. Marquez, Y. DeAbreu, and G. G. Botte, *Theoretical Investigation of NiYSZ in the Presence of H_2S* , *Electrochemical and Solid-State Letters*, 9 (3) A163-A166 (2006).

U.S. patent/patent applications

None at this time.

Conference Presentations

- 212th Electrochemical Society Meeting, Washington DC (October 2007): *Computational and Experimental Analysis of Solid Oxide Fuel Cell Anodes in the presence of H_2S* . Oral Presentation.
- 211th Electrochemical Society Meeting, Chicago, Illinois (May 2007): *Theoretical Investigations of Solid Oxide Fuel Cells Anode Materials in the presence of H_2S* (Poster)
- Eastern Regional Chemical & Materials Engineering Graduate Symposium (October 2006): *Theoretical Investigations of Solid Oxide Fuel Cells Anode Materials* (Poster)
- 2005 AIChE Annual Meeting: A. Marquez, Y. De Abreu, and G. G. Botte, “*Theoretical Investigations of Solid Oxide Fuel Cell anode Materials*”, October 30th – November 4th, 2005, Cincinnati, OH.
- 30th International Coal Conference, April 2005: A. Marquez, Y. De Abreu, and G. G. Botte, “*Theoretical Investigations of Solid Oxide Fuel Cell anode Materials*”, The 30th International Technical Conference on Coal Utilization and Fuel Systems, April 2005, Clearwater, FL.
- 206th Electrochemical Society (ECS) Meeting, Fall 2004: A. Marquez, and G. G. Botte, “*Theoretical Investigation of the Electro-oxidation of Ammonia*”, October 2004, Hawaii.

Awards received as a result of supported research

None at this time.

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