QUARTERLY PROGRESS REPORT

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Project Title: Fuel Cells R&D

Covering Period: July 1, 2010 through September 30, 2010

Date of Report: October 8, 2010

Recipient: Sandia National Laboratories

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TABLE OF CONTENTS

Project Title: Task 1 Water Transport Exploratory Studies, B&R # EB4209, EEW112798.

Covering Period: April 1, 2010 through September 30, 2010

Date of Report: October 8, 2010

Principle Investigator: Ken S. Chen, 505-844-5783, kschen@sandia.gov
Other Key National Lab Researchers:

Subcontractors Funded through AOP Task: Chao-Yang Wang, Penn State Univ.
Yun Wang, Univ. of California, Irvine

Industrial Partners: none

DOE Managers: Jason Marcinkoski

Project Objective: To develop analytical and multi-dimensional numerical models for simulating (1) water transport and removal in a PEM (proton exchange membrane or polymer electrolyte membrane) fuel cell under normal operation conditions, and (2) ice formation and thawing during the start-up of a PEM fuel cell under subzero temperatures or freezing conditions.

Background: Particularly during peak power conditions, water generated in the oxygen reduction reaction and transported from anode to cathode via electro-osmotic drag must be removed efficiently in order to achieve and maintain high PEM fuel cell performance. Under freezing conditions, water within a PEM fuel cell can freeze. The formation of ice within the MEA (membrane electrode assembly), in particular in the cathode catalyst layer, can hinder reactant transport and degrade cell voltage so as to limit the cold-start performance of a PEM fuel cell.

In this sub-project, we propose to develop analytical and multi-dimensional models for simulating 1) water transport and removal under normal operation conditions; and 2) ice formation and thawing during the start-up of a PEM fuel cell under subzero temperatures or freezing conditions. Specifically, we propose to develop sub-models for liquid water removal via droplet detachment and evaporation. Furthermore, we propose to develop simplified and multi-dimensional models for simulating water transport and removal under normal operating conditions, ice formation and thawing during start-up under subzero temperatures or freezing conditions.

Status: Accomplishments and work done since the last quarterly report are as follows: 1) Completed the Q3 milestone, "Develop a model for elucidating ice thawing or melting during PEM fuel cell start-up from a subzero temperature"; 2) Completed the Q4 milestone, "Combine models developed previously to arrive at an integrated model that can simulate both condensation/evaporation and freezing/melting"; 3) Published in the *Journal of The Electrochemical Society* the work on developing a current-ramping strategy for rapid startup of a PEM fuel cell from subzero temperatures; 4) Developed a 1-D model for analyzing three-phase transient behavior during the post-cold start of PEM fuel cells and reported the work in a presentation at the 8th International Conference on Fuel Cell Science, Engineering and Technology held in Brooklyn, NY, June 14 − 16, 2010; 5) Made the first attempt on predicting through-plane water distribution and comparing computed water profiles with neutron data and documented the work in a proceeding paper and a presentation.

Plans for Next Quarter and Key Issues: Improve the model fidelity for predicting through-plane water distribution and compare improved model prediction with neutron imaging data.

Patents: none.

Publications/Presentations

1. F. M. Jiang, C. -Y. Wang, and **K. S. Chen**, "Current ramping − a strategy for rapid startup of PEM fuel cells from subfreezing environment", *Journal of The Electrochemical Society,* **157** B342−B347 (2010).
2. A. Nandy, F. Jiang, S. Ge, C.-Y. Wang, and **K. S. Chen**, "Effect of cathode pore volume on PEM fuel cell cold start", *Journal of The Electrochemical Society*, **157** B726-B736 (2010).
3. Y. Wang and **K. S. Chen**, "Predicting through-plane water distribution in a polymer electrolyte fuel cell", in ASME Proceedings of FUELCELL2010, paper #33029 (2010).
4. A. Nandy, F. Jiang, C.-Y. Wang, and **K. S. Chen**, "Modeling Three-phase Transients during Post-Cold Start of Polymer Electrolyte Fuel Cells", presentation at the 8th Int. Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33110.
5. Y. Wang and **K. S. Chen**, "Predicting through-plane water distribution in a polymer electrolyte fuel cell", presentation at the 8th Int. Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33029.

Project Title: Task 2 Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells

Covering Period: April 1, 2010 through September 30, 2010

Date of Report: October 8, 2010

Principle Investigator: Ken S. Chen, 505-844-5783, kschen@sandia.gov
Other Key National Lab Researchers: Brian Carnes (SNL)

Subcontractors Funded through AOP Task: Penn State Univ., Ballard Power Systems

Industrial Partners: none

DOE Managers: Jason Marcinkoski

Project Objective: The objectives of this project are twofold: 1) to develop and validate a two-phase, three-dimensional transport model for simulating PEM (polymer electrolyte membrane) fuel cell performance under a wide range of operating conditions; and 2) to apply the validated PEM fuel cell model to identify performance-limiting phenomena or processes and develop recommendations for improvements.

Background: Despite tremendous research efforts and a large number of models published in the literature, a comprehensive, multi-physics computer model suitable for practical use by PEM fuel cell engineers and designers, particularly in transportation and stationary applications, remains absent. Modeling PEM fuel cell operation is challenging because it involves multiple components and phases, multi-dimensionality, and complex physics with highly coupled transport phenomena and electrochemical reactions. The significantly disparate length scales in transport and the presence of liquid water within the PEM fuel cell when operating under practical current loads, relatively high inlet relative humidity or moderate cell temperature, further add to the modeling challenges.

In this project, we shall employ a combined computational and experimental approach. First, we'll develop a two-phase, three-dimensional, transport model for simulating PEM fuel cell performance under a wide range of operating conditions. Next, we'll validate our transport model in a staged approach using experimental data available from the literature and those generated by team members. Lastly, we'll apply the validated model to identify performance-limiting phenomena or processes and develop recommendations for improvements. The major task in this proposal is the development of the PEM fuel cell transport model, which requires input parameters and validation. Once the model is validated, we shall apply it to identify performance-limiting phenomena or processes and develop recommendations for improvements. As required by DOE, we shall also publically disseminate and document the model, and compile data generated. Consequently, at a high level there are five tasks in this project: 1) develop a two-phase, three-dimensional, PEM fuel cell transport model; 2) measure model-input parameters and generate model-validation data; 3) validate the two-phase, three-dimensional PEM fuel cell transport model; 4) identify performance-limiting phenomena or processes and develop recommendations for improvements; and 5) disseminate/document the models developed and compile data generated.

Status: Accomplishments and work done since the last quarterly report are as follows: 1) Completed the Q3 milestone, "Develop a first-generation, 3-D, partially two-phase, single-cell PEM fuel cell model"; 2) Met the Q4 milestone, "Complete the development of a 3-D, partially two-phase, single-cell PEM fuel cell model"; 3) Completed and documented in a proceeding paper the preliminary coupling of the PEM fuel cell model with DAKOTA (a toolkit for design, optimization, and uncertainty quantification developed by SNL); 4) Completed and reported in a conference presentation the work on implementing a preliminary sub-model for simulating cathode MPL (microporous layer) effect; 5) Simulated performance of a PEM fuel cell with zigzag flowfield and reported the work in a conference presentation; 6) Investigated water transport through cathode MPL-GDL using a pore-network model and reported/documented the effort in a conference presentation and 2 journal publications; 7) Measured along-channel current and temperature profiles in a 12 kW PEM fuel cell stack; 8) Visited industrial partner, Ford Motor Company, and held in-depth technical discussions; 9) Made a project review presentation at the 2010 DOE Vehicle Technology and Hydrogen Program Annual Merit Review and Peer Evaluation Meeting, Washington DC, June 7-11, 2010; 10) Made a project review presentation at the DOE Fuel Cell Tech Team July 14 Meeting, USCAR Office, Southfield, MI on July 14 2010; and 11) Presented an invited paper at the 2010 LANL(US)/AIST-NEDO(Japan) Fuel Cell Workshop, Hyatt Regency Waikiki Beach Resort and Spa, Honolulu, HI, August 9 – 11, 2010.

In addition, the Project PI, Ken Chen, served as the chair for the track titled "Modeling, Design, and Optimization for Low Temperature Fuel Cells − I", which had 9 sessions with a total of 44 papers (including 32 proceeding papers) at the 8th International Conference on Fuel Cell Science, Engineering and Technology held in Brooklyn, NY, June 14 − 16, 2010. Ken also served as session chair and co-chairs at the above-mentioned conference.

Plans for Next Quarter and Key Issues: Initiate efforts on i) developing a first-generation, 3-D, fully two-phase, single-cell PEM fuel cell model and ii) performing validation of the 3-D, partially two-phase, single-cell PEM fuel cell model developed previously in Year 1.

Patents: none.

Publications/Presentations

1. K. S. Chen, B. Carnes, F. Jiang, G. Luo, and C.-Y. Wang, "Toward developing a computational capability for PEM fuel cell design and optimization", in the ASME Proceedings of FuelCell2010, paper # 33037 (2010).
2. B. Carnes, K. S. Chen, F. Jiang, G. Luo, and C.-Y. Wang, "Systematic parameter estimation and sensitivity analysis using a multidimensional PEMFC model coupled with DAKOTA", in the ASME Proceedings of FuelCell2010, paper # 33038 (2010).
3. G. Luo, Y. Ji, C.Y. Wang, and P. K. Sinha. "Modeling liquid water transport in gas diffusion layers by topologically equivalent pore network", *Electrochimica Acta,* Electrochimica Acta 55 (2010) 5332–5341.

4. Y. Ji, G. Luo, and C.Y. Wang, “Pore-Level Liquid Water Transport Through Composite Diffusion Media of PEMFC”, *Journal of The Electrochemical Society,* 157 (2010) B1753-B1761.
5. K. S. Chen, “Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells”, in *DOE Hydrogen Program FY 2010 Annual Progress report* (2010).
6. K. S. Chen, B. Carnes, F. Jiang, G. Luo, and C.-Y. Wang, “Toward developing a computational capability for PEM fuel cell design and optimization”, presentation at the 8th Int. Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33037.
7. B. Carnes, K. S. Chen, F. Jiang, G. Luo, and C.-Y. Wang, “Systematic parameter estimation and sensitivity analysis using a multidimensional PEMFC model coupled with DAKOTA”, presentation at the 8th Int. Fuel Cell Sci., Eng. & Tech. Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33038.
8. F. Jiang, G. Luo, C.-Y. Wang, and K. S. Chen, “Simulation of a PEMFC with Zigzag Flow Field”, presentation at the ASME Eighth International Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33108.
9. G. Luo, F. Jiang, C.-Y. Wang, and K. S. Chen, “Numerical Modeling of Micro-porous Layers in a Two-Phase Multidimensional PEM Fuel Cell Model”, presentation at the 8th Int. Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper # FuelCell2010-33164.
10. K. S. Chen, “Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells”, presentation at the 2010 DOE Vehicle Technology and Hydrogen Program Annual Merit Review and Peer Evaluation Meeting, Washington DC, June 7-11, 2010.
11. K. S. Chen, “Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells”, presentation at the DOE Fuel Cell Tech Team July 14 Meeting, USCAR Office, Southfield, MI (2010).
12. K. S. Chen, “Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells”, invited presentation at the 2010 LANL(US)/AIST-NEDO(Japan) Fuel Cell Workshop, Hyatt Regency Waikiki Beach Resort and Spa, Honolulu, HI, August 9 – 11, 2010.

Project Title: Task 3 Anion Exchange Membrane Fuel Cell

Covering Period: May 1, 2010 to September 30, 2010

Date of Report: September 30, 2010

Recipient: Sandia National Laboratories

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Subcontractors: None

DOE Managers: Jason Marcinkoski

Project Objective: The objectives of this task are twofold: (1) synthesize both random and ordered anion exchange membranes and (2) characterize the polymers' physical properties such as IEC, water uptake and conductivity.

PROJECT STATUS:

To confirm our discovery in the previous quarter that the properties of anion exchange membranes are strongly dependent on the solvent used during the formation of quaternary ammonium groups, a series of membrane samples was prepared and sent to LANL for IR analysis. The results clearly showed a peak due to the O-H bending vibration in the samples that had been prepared in aqueous trimethylamine solutions whereas this peak was absent in the spectra of the samples that had been prepared in solutions of trimethylamine in ethanol. This served to confirm our conclusion that the presence of water during the amination step leads to the formation of hydroxide ions which can compete with the trimethylamine as a nucleophile. The result is that benzyl alcohol groups are formed instead of the desired quaternary ammonium groups and the membranes have ion exchange capacities that are lower than expected. These results have been included in a draft of a publication which describes our discovery.

We have prepared and delivered two sets of membrane samples to our collaborators at LANL for fabrication into MEAs and fuel cell testing. These membranes were all made with the Deils-Alder poly(phenylene) backbone with benzyltrimethyl ammonium cation groups attached and they were all prepared by soaking in ethanolic rather than aqueous solutions of trimethylamine. The membranes could also be dissolved in methanol to be used as the ionomer in theelectrodes. The best polarization curve obtained is shown in figure 1. The cell used in figure 1 was operating at 60 ^{o}C with H_2/O_2 and Pt black (4 mg/cm^2) on each electrode. The cell performance is similar to the current state-of-the-art and represents the benchmark for all future cells that will be prepared as part of this project utilizing next generation membranes, ionomers, and catalysts. In addition to the introduction of new materials, new MEA fabrication techniques are expected to play an important role in improving cell performance.

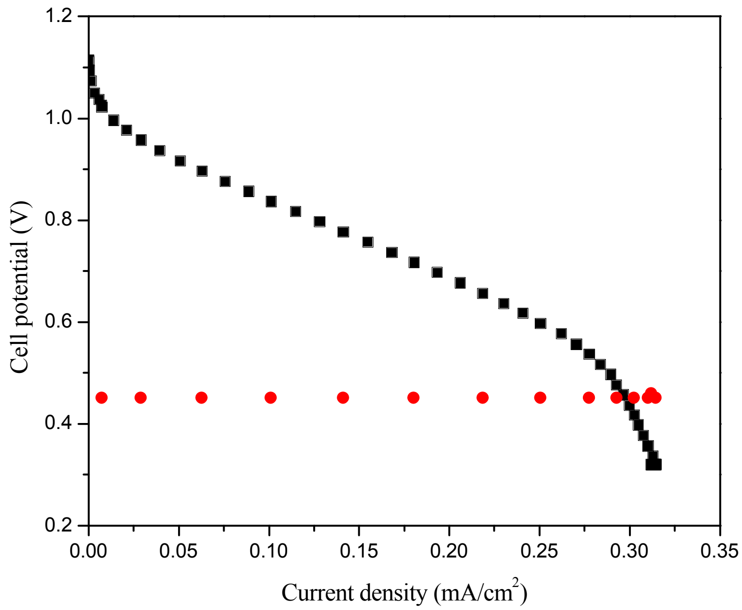

Figure 1. Polarization curve from alkaline fuel cell made with Sandia membrane and ionomer.

Several of the membranes delivered to LANL became brittle when dried out and were difficult to fabricate into MEAs. In order to improve the mechanical properties of these membranes when dry, a second set of membranes which contain a plasticizer have been sent to LANL for evaluation. Results from these membranes are expected in the next quarter.

Plans for next quarter:

In the upcoming quarter we plan to continue improving the mechanical properties of the membranes by blending in other plasticizers or elastomers to reduce brittleess. We also plan to continue work on the synthesis of anion exchange membranes with resonance-stabilized cationic groups, particularly quanidinium-type cations.

Patents: none

Publications/presentations:

M. R. Hibbs, "Recent Advances in Anion Exchange Membranes for Alkaline Fuel Cells", invited presentation at the 2010 NEDO-LANL Fuel Cell Workshop, Hyatt Regency Waikiki Beach Resort and Spa, Honolulu, HI, August 9 – 11, 2010.

Project Title: IEA Stationary Fuel Cell Annex

Covering Period: June 1st through September 31st, 2010

Date of Report: September 31st, 2010

Recipient: Sandia National Laboratories

Subcontractors: None

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Project Objective: The goal of this project is to support the DOE Hydrogen, Fuel Cells, and Infrastructure Technologies Program in collaborating internationally with the International Energy Agency (IEA) on research related to stationary fuel cell systems.

PROJECT STATUS

Subtask 1.1— Analyzing strategies for stationary fuel cell systems in the context of diverse, international markets.

We prepared research results on stationary fuel cell systems relevant to international researchers, developers, and markets. We prepared research results for an upcoming semi-annual meeting of the International Energy Agency's (IEA) Advanced Fuel Cell (AFC) Annex XIX Stationary Fuel Cell Systems working group in Copenhagen, Denmark on Oct. 13th-14th, 2010. The presentation will be entitled "Combined Cooling, Heating, Hydrogen, and Electric Power Fuel Cell Systems." Based on feedback from the previous IEA expert working group meeting on from April 27th-28th 2010, we updated related stationary fuel cell system models.