

**WEDNESDAY, AUGUST 24, 2005, P.M.**

**SESSION 43A: INTERNATIONAL SYMPOSIUM ON FUEL CELL AND HYDROGEN TECHNOLOGIES**

MODELLING IV

Sponsor(s): Materials Science and Engineering Section, The Metallurgical Society of CIM

Room: Imperial Ballroom 7

Chairmen: K. KARAN, Fuel Cell Research Centre, Queen's University, Canada and

B. ZHOU, University of Windsor, Canada

PAPER 43A.1— 14:00

SIZE EFFECTS ON REACTIVITY OF SUPPORTED Pt NANOPARTICLES IN CO MONOLAYER OXIDATION: KINETIC MONTE CARLO SIMULATIONS.

B. ANDREAUS, NRC Institute for Fuel Cell Innovation, Canada

E. SAVINOVA, Boreskov Institute of Catalysis, Russia and

M. EIKERLING, Simon Fraser University, Canada

In spite of tremendous efforts in research and development of polymer electrolyte fuel cells (PEFCs), essential properties of the actual catalyst material, supported Pt or Pt-alloy nanoparticles, are not yet well understood. Fundamental understanding of the catalyst structure and the prevailing kinetic mechanisms is, however, of utmost importance for further progress.

In the present study, we investigate catalyst activity for CO oxidation from a theoretical point of view. Electrochemical CO oxidation serves as an important test reaction for the study of the fundamental properties of supported Pt nanoparticles. Recent experiments have revealed strong effects of catalyst particle size and supporting material on catalytic activity. It was found, that the activity towards CO oxidation decreases significantly when the particle diameter drops below 2.5 nm. We perform kinetic Monte Carlo simulations of CO oxidation on supported catalyst particles to elucidate this particle-size effect. The simulations are based on the active site concept, highlighting the role of active site formation and finite surface mobility of adsorbed electroactive species for catalyst activity. For comparison with experimental results, potential step experiments were simulated. The Monte Carlo simulations consistently reproduce effects of particle size and electrode potential found experimentally. In comparison with our simulations, analytical mean field models and nucleation and growth models exhibit inferior agreement with experimental data.

In summary, our findings support an oxidation mechanism via active site formation. The reason for the slow reaction kinetics on catalyst particles with diameters below 2.5 nm is a rather low CO surface diffusivity. The rates of active site formation, diffusion and CO oxidation via recombination with adsorbed OH can be determined from the simulations.

PAPER 43A.2 — 14:20

CATHODE CATALYST LAYER AS THE WATERSHED IN PEMFC.

M. EIKERLING and J-F. LIU, Simon Fraser University, Canada

The full competition between mass transport and reaction in PEMFC unfolds in cathode catalyst layers (CCLs). Recently developed models provide vital insights into effects of thickness and composition on effectiveness of catalyst utilization and performance. However, the CCL's key function in liquid water removal is rather unexplored. Studies of water balance in PEMFCs focus mostly on other cell components. Notwithstanding this prioritization, a simple calculation shows that the CCL is the PEMFC's favourite medium for conversion of liquid water into vapor. Porous structure and wetting properties steer the interplay of evaporation, transport and reaction. A model will be presented, that links pore space morphology, composition, distributed properties (concentrations, pressures, reaction rates, potential) with global performance (critical phenomena). It reveals how problems of flooding in the fuel cell could be mitigated, by properly adjusting catalyst layer structure and operation conditions and under which conditions all liquid water arriving in the CCL would leave it as water vapour through the CCL/GDL interface. Results for a simple variant of the model emphasize the role of the CCL as a watershed for the whole cell.

PAPER 43A.3 — 14:40

A METHOD FOR DETERMINING ANISOTROPIC TRANSPORT PROPERTIES IN FUEL CELL POROUS TRANSPORT LAYERS.

D. HAMILTON and J.G. PHAROAH, Queen's University, Canada

The electrode of a PEM fuel cell consists of two distinct sections - the catalyst layer and the porous transport layer (PTL). There are several kinds of PTL material, although all use carbon fibres as the solid fraction. The fibres can be

matted to form a paper, woven into a cloth, or chopped to form a felt. Despite the different morphologies of the media, they are all anisotropic in structure, that is the majority of continuous fibres lie perpendicular to the direction of bulk transport. Currently, no models account for anisotropy and all transport coefficients are calculated as volume weighted averages or from empirical correlations (not developed for fibrous porous media). The present work presents a new methodology for determining anisotropic transport coefficients in fibrous porous media. A numerical experimentation process is employed wherein random samples of the PTL are generated and used to solve a diffusion transport equation. Effective transport coefficients are determined by integration of flux across the domain boundaries once a steady state solution has been reached. Multiple random samples at each porosity are generated in order to predict statistical variance. The results are correlated for ease of implementation in full PEM models.

PAPER 43A.4 — 15:00

UNDERSTANDING THE ROLE OF ION TRANSPORT AND POROUS TRANSPORT LAYER (PTL) ON REACTION RATE DISTRIBUTION IN PEMFC CATHODE.

K. KARAN, W. SUN, D. HARVEY and J.G. PHAROAH, Fuel Cell Research Centre, Queen's University, Canada

Oxygen reduction reaction in PEM fuel cell cathodes involves participation of three reactants - oxygen, hydrogen ions, and electrons. Electrochemical reaction in the catalyst layer is, therefore, influenced by how these species are transported to the reaction site. The two-phase boundary locations at which percolating networks of ionically conducting electrolyte and electronically conducting Pt/C meet are feasible reaction sites. The electrolyte phase also provides pathway for the transport of dissolved oxygen. Most of the 3-D models describe the catalyst layer as an ultra-thin layer, thereby, losing useful information on the distribution of electrochemical reaction rate.

In this paper, we extend our previous work on 2-D cross-the-channel Modelling of PEMFC cathode based on agglomerate catalyst layer. In particular, we take a closer look at the influence of proton conductivity on the reaction rate distribution in the catalyst layer. As well, we re-examine the role of anisotropic transport coefficient of the porous transport layer (PTL), commonly known as the gas diffusion layer, on the current density distribution under the channel and the rib of the flow-field plates. Results from both a 2-D and a 3-D models will be presented.

PAPER 43A.5 — 15:20

MODELLING OF THE CONTAMINATION EFFECT INTRODUCED BY AMMONIA ON THE PERFORMANCE OF PEM FUEL CELLS.

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Contamination introduced by the impurities from the feed stream can impact the PEM fuel cell performance dramatically. The presence of these unwanted trace species such as CO, H<sub>2</sub>S, NH<sub>3</sub>, can adversely affect the fuel cell functioning. It has been reported that the major impact of CO and H<sub>2</sub>S contamination on the fuel cell performance is kinetic while the effect of NH<sub>3</sub> contamination is speculated as membrane conductivity reduction only. In this paper, a model regarding the NH<sub>3</sub> contamination is described and the mechanism of the NH<sub>3</sub> contamination is illuminated by the model calculations.

COFFEE BREAK — 15:40 – 16:00