

SEMINAR

Friday, February 16, 2007

1:30 PM

MSC 100

First-Principles Elucidation and Design of Catalytic Sites and their Environments

Dr. K-D. Kreuer

Max-Planck-Institut für Festkörperforschung

Heisenbergstr. 1, D-70569 Stuttgart, Germany

The electrolyte of PEM fuel cells is usually a sulfonated polymer such as DuPont's Nafion®. Such ionomers naturally combine, in one macromolecule, the high hydrophobicity of their backbone with the high hydrophilicity of the sulfonic acid functional group, which gives rise to a constrained hydrophobic/hydrophilic nano-separation^[1]. The sulfonic acid functional groups aggregate to form a hydrophilic domain, which is formed by connected „channels“ or „pores“ and this is hydrated in the presence of water. It is within this continuous domain that protonic conductivity occurs: protons dissociate from their anionic counter ion ($-\text{SO}_3^-$) and become solvated and mobilized by the hydration water. Apart from the coupled transport of water and protonic charge carriers there is also significant transport of water as such. The nature of this transport (diffusional, collective flow) and the transport parameters are essentially determined by the microstructure, the ion exchange capacity and the water uptake^[2].

This presentation provides a quantitative model for the water uptake with the elastic constants and the heat of hydration being the only parameters. It is shown that significant pressures may build up in such membranes and that pressure gradients have to be introduced as separate driving forces (apart from electrochemical potential gradients) into the transport equations to fully describe the behavior under fuel cell operating conditions.

The model not only provides a rationale for the understanding of the behavior of the different types of membrane (hydrocarbon versus perfluorinated, effect of composite formation), it may also be useful in exploring the potentials (limits) of sulfonic acid based proton conducting membranes in general.

In a first step towards these limits, sulfonated electron-poor polysulfones of extremely high IEC have been prepared and characterized not only with respect to transport but also to stability. The results are very encouraging and suggest that an increase of the operation temperature of conventional PEM fuel cells by about 20° C is feasible^[3].

Finally, a brief introduction will be given into radically different approaches aiming at water-free proton conductors relying on the amphoteric properties of heterocycles (e.g. imidazole) and phosphonic acid^[4,5].

[1] K. D. Kreuer *J. Membrane Science* **2001**, 185, 29

[2] K. D. Kreuer, S. J. Paddison, E. Spohr, M. Schuster *Chem. Rev.* **2004**, 104, 4637

[3] M. Schuster, K.D.Kreuer, H. Anderson, J. Maier; *Macromolecules* **2006**, electronically available

[4] M. Schuster, T. Rager, A. Noda, K. D. Kreuer, J. Maier, *Fuel Cells* **2005**, 5, 355

[5] S. J. Paddison, K. D. Kreuer, J. Maier, *Physical Chemistry Chemical Physics* **2006**, 8, 4530