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Surface Self Diffusion of Hydrogen on Carbon Support by Quasielastic Neutron Scattering

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1. Introduction

The purpose of this work is to examine the theoretical hypothesis made by Meland et al. [1] that access to the three-phase contact line in porous gas electrodes is via the interfaces between the homogenous phases that surround the contact line. Electrodes of this type are central in the polymer electrolyte fuel cell; see Fuel Cell Systems Explained for the description [2], so the overall aim of our effort is thus to contribute to an improved understanding of these complicated heterogeneous catalytic structures. The hypothesis stems from an analysis of the platinum electrode surface as a hydrogen gas anode [1], using non-equilibrium thermodynamics for the heterogeneous system [3]. In this work we have used quasielastic neutron scattering (QENS) to investigate the surface self diffusion of molecular hydrogen on carbon black XC-72.

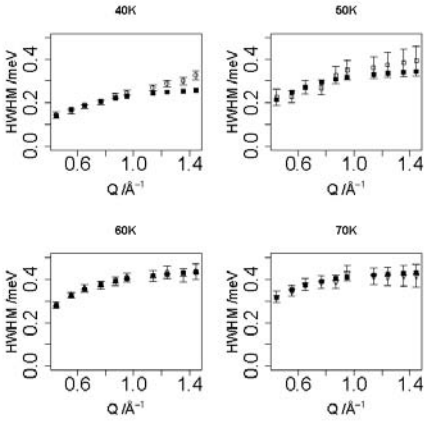

Figure 1: Fit of the model to the experimental data at each temperature.

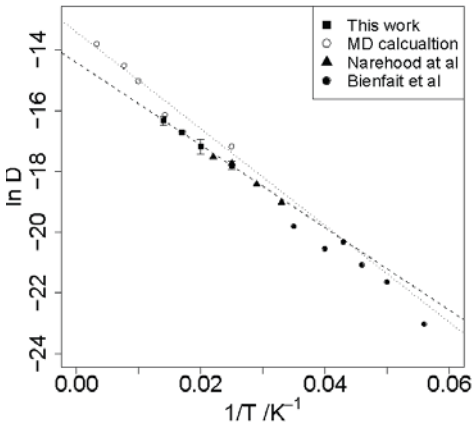

Figure 2: Arrhenius plot comparing our work with Narehood and Bienfait et al. [6-7] and NEMD simulations.

2. Results of Quasielastic Neutron Scattering Measurments

Our sample is a commercial carbon black, XC-72, from Cabot Corp. From BET measurements the surface area was found to be 204.3 m^2/g. The QENS experiments were performed at the direct geometry time of flight spectrometer IN5, ILL, Grenoble, France. Measurements to estimate the diffusion were conducted at 2, 10, 20, 30, 40, 50, 60 and 70K with varying hydrogen coverage, due to shifts of the equilibrium between adsorbent and gas phase. From the broadening of $S(Q,\omega)$ we found the surface self diffusion for molecular hydrogen. The Chudley & Elliott model was fitted to the experimental data. The Chudley & Elliott model describes the movement of a single molecule as it jumps between identical sites on a lattice [4-5], see Figure 1. The self diffusion coefficients calculated from the fit are presented in an Arrhenius plot in Figure 2. Previous results from Narehood and Bienfait et al. are also plotted for comparison. We have also done some non-equilibrium molecular dynamics with a similar system. The results from those simulations are in good agreement with the values we found from the experiment. See our other poster on the non-equilibrium molecular dynamics for more information [8].

3. Conclusion

From QENS we found the surface self diffusion of hydrogen and the Arrhenius coefficients for hydrogen self diffusion. We found that self diffusion on amorphous carbon is in very good agreement with that of previous experiments on carbon nano tubes and graphitic sheets.

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