Dynamical Behaviour of H_2 Molecules on Graphite Surface. A Molecular Dynamics Study.

Jean-Marc Simon[1], Ole-Erich Haas[2], Signe Kjelstrup[2], Astrid Lund Ramstad[2]

1. Institut Carnot de Bourgogne, UMR 5209 CNRS-Université de Bourgogne, France. jmsimon@u-bourgogne.fr

2. Institutt for kjemi, NTNU, 7491 Trondheim, Norway

1. Introduction

The background of this work is the prediction made by Meland et al. [1] that access to the three-phase contact line in porous gas electrodes is via the interfaces that surround the contact line. Electrodes of this type are central in the polymer electrolyte fuel cell. In this work we investigated the dynamical behaviour of hydrogen molecules on the graphite carbon surface by molecular dynamics (MD) simulations.

2. Model and simulation details

The system was composed of two infinite planes of graphite distant by about 160 Å in contact with different amounts of hydrogen molecules. The surface was in the plane of the hexagonal structure. A rectangular basic simulation cell of about 44 Å long and 34 Å large in that plane was extended to infinity by the use of periodic boundary conditions. Both the graphite and the hydrogen were simulated using an atomic model and they were allowed to interact through inter and intra-molecular potentials and to move by integrating the equation of motion.

Systems with 50, 100, 150, 200 and 300 molecules of hydrogen were simulated at different temperatures 70K, 100K, 130K and 300K. By an analysis of the molecular trajectories at the surface, we could reach different dynamical properties like the self-diffusion coefficients. We also found information on the thermodynamics properties of adsorption.
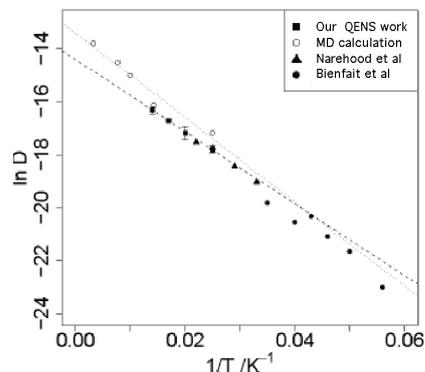

Figure 1: Arrhenius plot comparing our work (MD and QENS) with Narehood and Bienfait et al. [2-3]

3. Results and discussions

Our simulation conditions were such that the density on the surface was lower than that of a monolayer, it varied from 1.5×10^{-5} to 1.8×10^{-7} mol/m^2. The self-diffusion coefficients are presented in an Arrhenius plot in figure 1. They are compared with

results from Quasi Elastic Neutron Scattering (QENS) obtained by us and from the literature. Despite our simple model, the results show a good agreement both on the slope and on the absolute values.

The residence time t_r for a molecule on the surface slightly decreased at each temperature with increasing surface density. The mean value for each temperature is also plotted on figure 2 in an Arrhenius plot, it decreases as the temperature increases. From that curve we can deduce an activation energy of 3.0

Figure 2. Arrhenius plot of the residence time of hydrogen

kJ/mol in good agreement with the adsorption energy, but higher than the activation energy of the surface diffusion 2.2kJ/mol. At 300K, the residence time is about 5.7 ps, with an average self-diffusion coefficient of about $2\ 10^{-6}\ m^2.s^{-1}$, the average distance travelled by a molecule on the surface before desorption is then around 50 Å.

4 Conclusion and perspectives
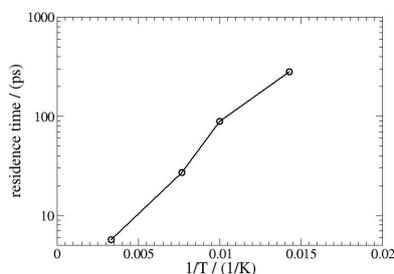
We have simulated hydrogen molecules adsorbed on graphite at different temperatures. Their dynamical properties were investigated showing that at 300K, despite the fact that the number of adsorbed molecules is low, the travelled distance on the surface is quite substantial, 50 Å. This result supports the proposal of Meland et al.[1] that hydrogen gas reaches the catalytic site (Pt) via the interfaces of the porous structure leading up to the site.

References

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