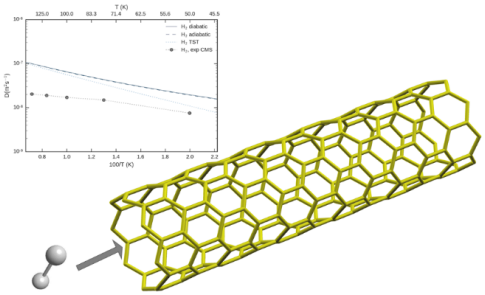
Importance of the Tunneling Effect on the Diffusion of H_2 along a Carbon Nanotube

Manel MONDELO-MARTELL, Fermín HUARTE-LARRAÑAGA

Department of Material Science and Physical Chemistry & Institute for Theoretical and Computational Chemistry, Universitat de Barcelona

Quantum confinement effects, arising from the interaction of small molecules embedded in nanostructured materials with nanometric, have fostered the discovery and tailoring of new physical-chemical phenomena with large relevance both at fundamental and technological level[1,2]. Quantum sieving, i.e. the possibility to separate a mixture of isotopologues of light molecules (e.g. H_2 from D_2) by either different mobility in the nanostructure (*kinetic sieving*) or different affinity to the pore (*selective adsorption* or *thermodynamic sieving*), is a paradigmatic example of such new processes only possible due to the quantum confinement effects[3].

In this communication we present our work on the calculation of the diffusion rates of H_2 and D_2 along a narrow SWCNT using rigorous quantum dynamics. The low diffusion barrier of the process implies that very long times are needed (20 ps) in order to obtain converged results, so a time-scale separation scheme was used in order to reach this time span. The diffusion rates obtained show a very noticeable contribution from the tunneling effect at low temperatures which affects previously reported results on the same system: the existence of low-energy resonances increases the diffusion rates compared with semiclassical results at temperatures below 125 K. This difference is much more noticeable in the case of hydrogen, preventing the kinetic sieving predicted with Transition State Theory.



1. T. Lu, E. E. M. Goldfield, and S. S. K. Gray, *J. Phys. Chem. B* **110**, 1742–1751 (2006).
2. Y. Inokuma, M. Kawano, and M. Fujita, *Nat. Chem.* **3**, 349–358 (2011).
3. H. Oh and M. Hirscher, *Eur. J. Inorg. Chem.* **2016**, 4278–4289 (2016).
4. M. Mondelo-Martell and F. Huarte-Larrañaga, *J. Phys. Chem. A* **120**, 6501– 6512 (2016).