diffusion-fundamentals

The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Molecular Simulation of Gas Transport in Nanoporous Carbons

Qiong Cai, Alex Buts, Nigel Seaton, Mark Biggs

Institute for Materials and Processes, University of Edinburgh, Mayfield Road, Edinburgh EH9 3JL, UK, E-Mail: n.seaton@ed.ac.uk

1. Introduction

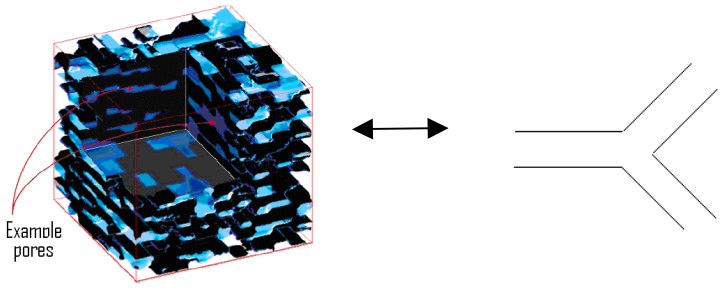
Nanoporous carbon materials are widely used in industry, medicine and elsewhere for separation processes, whilst they are also utilized in new applications such as catalyst supports, capacitors, gas storage and biomedical engineering applications [1-4].

Understanding the transport of species within the pore space is a key aspect in the design of nanoporous materials. A wide variety of factors, such as the pore and molecular sizes and shapes, the interactions between the molecules and the surface of the pore, and the connectivity of the pore network, contribute to the overall transport and separation properties [5]. Hence, detailed models are needed for taking into account the effect of such factors. However, despite the fact that considerable theoretical work, computer simulations and experimental studies have been devoted to this class of problems, our understanding of these phenomena remains incomplete.

We are carrying out a molecular simulation and pore network modelling study to investigate the effect of pore structure on transport in nanoporous carbons. We aim to develop a validated pore-level transport model which can be applied in the design of these materials.

2. Simulation method

In this work, a "pore network model" (PNM) is used to analyze a "virtual porous carbon" (VPC) [6], which is a highly realistic (but much less tractable) representation of a real carbon. These models are illustrated in Figure 1. Based on an analysis of adsorption, the pore size distribution (PSD) and the pore network connectivity (defined as the mean number of pores meeting at a junction) of the VPC are obtained and used to generate a PNM with the same effective structure (from the point of view of diffusion) as the VPC [7].



Virtual porous carbon

Pore network model

Figure 1 Illustration of the two models used.

Transport of three species (CH_4, CF_4 and SF_6) in each pore of the PNM is modelled by Molecular Dynamics (MD) simulation. There are several ways that one can measure the transport diffusivity of species in a MD simulation. The dual control-volume grand canonical molecular dynamics (DCV-GCMD), the external field non-equilibrium MD (EF-NEMD), and the equilibrium MD (EMD) methods have been successfully used by several authors for computing non-equilibrium transport coefficients of confined fluids [8-10]. In this work, EMD is used because of its simplicity and efficiency.

The effective transport coefficients for each species in the PNM as a whole is determined using an approximate solution method: the Monte Carlo renormalized effective medium approximation (MC-REMA) [11]. This method is based on the solution of a mass balance at each node of the network. The PNM parameters, i.e. the PSD and the network connectivity, and the transport coefficients in individual pores (calculated by EMD) are the inputs to the MC-REMA calculation. In this way, we can evaluate the performance of the PNM in describing diffusion in real carbons, represented here by the realistic VPC model.

3. Conclusion

In this work, transport of three species (CH_4, CF_4 and SF_6) in pores with a range of pore sizes is studied by EMD simulation. The transport coefficients in these single pores will be incorporated to a pore network (PNM) to calculate the effective transport coefficient for each species by MC-REMA approach.

References

[1] K. Takashi, Carbon 38 (2000) 269-286.
[2] A. W. Heinen, J. A. Peters, and H. Bekkum, Applied Catalysis A: General 193 (2000) 194-195.
[3] W. M. Qiao, Y. I. Korai, M. Y. Hori, T. Maeda, Carbon 39 (2001) 2355-2368.
[4] M. Minamisawa, H. Minamisawa, S.Yoshida, N. Takai, J. Agric. Food Chem.52 (2004) 5606-5611.
[5] D. D. Do, Adsorption analysis: equilibria and kinetics, Imperial College Press, London, 1998.
[6] M. J. Biggs, A. Buts, D. Williamson, Langmuir 20 (2004) 5786-5800.
[7] Q. Cai, A. Buts, N. A. Seaton, M. J. Biggs, Article in preparation.
[8] G. Arya, H. C. Chang, E. J. Maginn, J. Chem. Phys.115 (2001) 8113-8124.
[9] A. M. Vieira-Linhares, N. A. Seaton, Chem. Eng. Sci. 58 (2003) 5251-5256.
[10] D. J. Keffer, P. Adhangale, Chem. Eng. J. 100 (2004) 51-69.
[11] L. Zhang, N. A. Seaton, AIChE Journal 38 (1992) 1816-1824.