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Potential Calculations and MD Simulations of *n*-Pentane in Silicalite-1

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1. Introduction

Recently, we have proposed *ab initio* fitted potentials representing the n-pentane/silicalite-1 interaction[1], but these potentials underestimate the heat of adsorption (q_{st}). In the next section, we propose new potential parameters which yield the correct heat of adsorption. Then, results of MD simulations applying these new potential parameters at various temperatures and concentrations are discussed in the last section.

2. Model

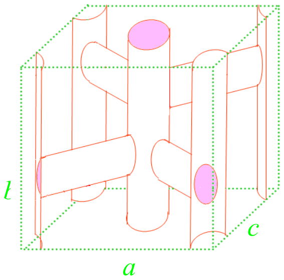

Fig.1 Schematic views of the silicalite-1 channel system

Silicalite-1, a silicious ZSM-5 has unit cell parameters of a = 20.07, b = 19.92 and c = 13.42 Å^2. The lattice consists of two distinct types of channels. Sinusoidal channels are directed along the [100] crystal axis while straight channels are directed along the [010] crystal axis. A three-dimensional network is given by the connectivity of the two types of channels (see Fig 1). The potentials representing the *n*-pentane/silicalite-1 interaction (obtained from quantum calculations) are fitted by a set of Lennard-Jones pair potentials. The united atom approximation is applied for *n*-pentane. The pentane/lattice interaction is represented by interactions of CH_n groups and the O atoms

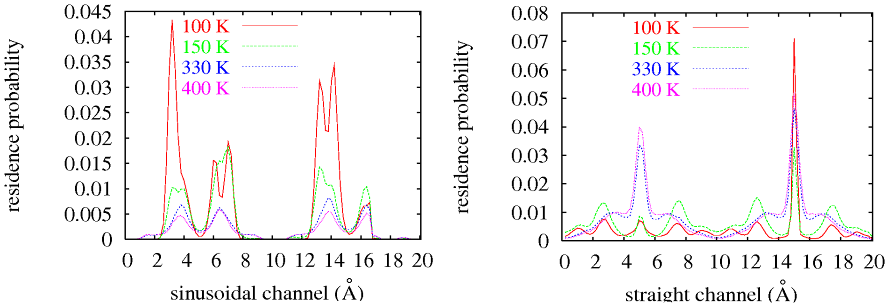

Fig.2 The distribution of sorbates in the channels at various temperatures. The loading of sorbates is 4 molecules per unit cell for all temperatures.

in which ε and σ for CH_3-O are 0.475 kJ/mol and 3.62 Å. The parameters of the CH_2-O interactions are ε =0.574 kJ/mol and σ= 3.71 Å. The interactions of CH_n groups with the Si atoms are implicitly taken into account in these effective parameters.

3. Results and Discussion

The temperature affects the distribution of sorbates in channels as shown in Fig. 2.
In Fig. 2, the intersections are taken into account as parts of straight channels. Dot lines show the centers of the intersections. It can be seen that the molecules prefer to stay in the channels at low temperatures and they migrate to reside more frequently in the intersections when the temperature increases. The reason is that whith increasing temperature entropic effects become more important. Sorbates have more kinetic energy and migrate to the intersection where they find more space to tumble, as well as for vibration and rotation. Increasing the entropy with a given loading of sorbates does not influence the distribution of guest molecules in the channels. This becomes evident by the curves shown in Fig. 3.

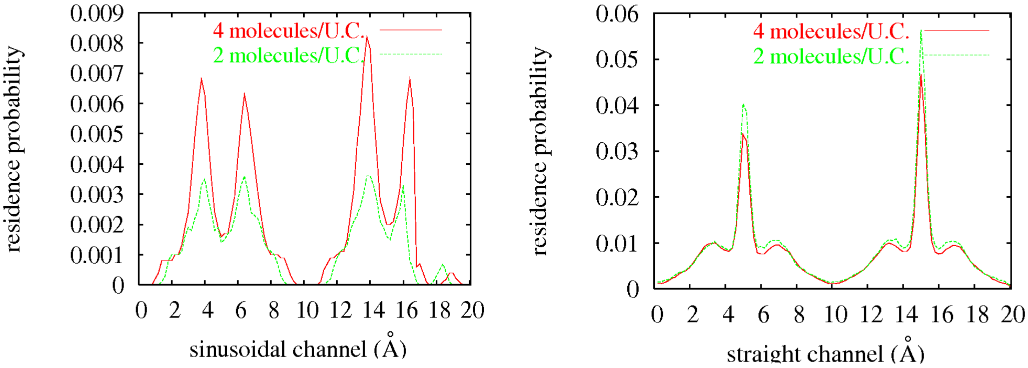

Fig.3 The distribution of sorbates in the channels at 330 K. The loading of sorbates is 2 and 4 molecules per unit cell.

For convenience in comparison of the peak positions, the integration of areas below curves for each loading is normalized equal to 1 for the entire distribution and not for each curve separately.

References

[1] A. Loisruangsin, S. Fritzsche, S. Hannongbua, Chem. Phys. Lett. 390 (2004) 485-490.

[2] C. Baerlocher, W. M. Meier, D. H. Olson, *Atlas of Zeolite Framework Types, fifth revised ed., Elsevier, Amsterdam,* 2000.