2O04

A computational study for the dynamics of water, methanol, and ammonia in periodic mordenite zeolite structure

Mohamed Elanany,[1] Katsumi Sasata,[1] Akira Endou,[1]
Momoji Kubo,[1] Parasuraman Selvam,[1] and Akira Miyamoto[1,2]

[1]*Department of Materials Chemistry, Graduate School of Engineering,*
[2]*New Industry Creation Hatchery Center (NICHe),*
Tohoku University, Aoba-yama 07, Sendai 980-8579, Japan

Introduction

In order to show clearly the interaction of water, methanol and ammonia in zeolites, we have chosen protonated mordenite (H-MOR) structure and used a novel tight-binding quantum chemical molecular dynamics program "Colors". The calculations were performed using the full topology of H-MOR employing the periodic boundary condition.

Method

The structure models of H-MOR, H_2O, CH_3OH, and NH_3 were geometrically optimized with the Vosko-Wilk-Nusair using local density approximation (LDA) whereas the energies were evaluated at the GGA level employing Perdew-Wang (PW91XC) exchange and correlation functionals. The molecular dynamics simulations were carried out for 2000 simulation steps with a time interval 0.1 fs.

Results and Discussion

Table 1 shows that the binding energy and the charge distribution calculated by "Colors", which are very close to the density functional method employing ADF program. Figure 1 depicts the optimized structure (unit cell) of H-MOR. The replacement of one of the tetravalent silicon by trivalent aluminum and a charge-compensating proton (Brønsted acid site) at the bridging oxygen creates a local distortion. Finally, using the optimized models we performed the dynamics by "Colors", and the simulation results will be presented.

Table 1. Results of "Colors" and ADF for $Si(OH)_4$, $Si_2O_7H_6$ and $Si_3O_9H_6$.

Molecule	BE (eV)	Hirshfeld charge			Code
		Si	O	H	
$Si(OH)_4$	-49.72	0.56	-0.28	0.15	ADF
	-47.98	0.41	-0.29	0.16	Colors
$Si_2O_7H_6$	-76.45	0.61	-0.32	0.16	ADF
	-77.47	0.54	-0.29	0.15	Colors

Fig. 1. The optimized structure of H-MOR.