2P19 Temperature dependence of the hydrogen adsorption sites on Zeolite-Templated carbon

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Carbon materials of large surface area, such as activated carbons, graphene-sheets, carbon nanotubes, and zeolite-templated carbon (ZTC: Figure 1), have attracted attention as hydrogen storage materials by physisorption. Among them, ZTC using Zeolite Y as a template has some structural features. It has a larger surface area than other materials, uniform micropores, and its hydrogen storage up to 2.2 wt% at 34 MPa at 303 K[1,2]. This hydrogen storage amount has been highest among any pure carbon materials. However, it has to increase the hydrogen storage capacity for practical use. To improve hydrogen storage capacity, it is indispensable to understand for the hydrogen diffusion process and its adsorption site on carbon. In this study, we have performed path integral molecular dynamics simulation at various temperatures to evaluate the temperature dependence of the hydrogen adsorption site on ZTC. In this simulation method, nuclei are treated as quantum mechanical particles, and can take account of quantum and thermal fluctuations with respect to molecular vibrational and rotational degree of freedom.

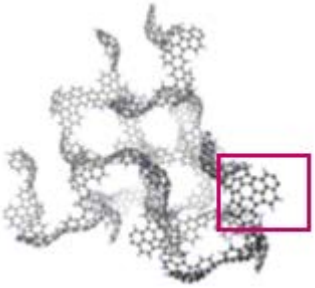

Figure 1 Crystal structure of ZTC

The minimum unit ($C_{36}H_{12}$: Figure 2) of ZTC which has a buckybowl structure including three pentagonal carbons was employed as a model structure [3-5]. Five carbons inside of buckybowl were selected from innermost to edge as the adsorption site for additional hydrogen atom. Path integral molecular dynamics simulations have been performed at 100, 300, 600, and 900 K using PM3 potential. The imaginary time of slices at 100, 300, 600, and 900 K are 64, 24, 12, and 8, respectively. Hydrogen atom at 100 K adsorbed on each carbon, while stable hydrogen adsorption sites are gradually decreasing as temperature becomes higher. This is because the additional hydrogen atom is easy to go over the potential barrier (Figure 3) between one to other carbon by thermal and nuclear quantum effects as temperature becomes higher.

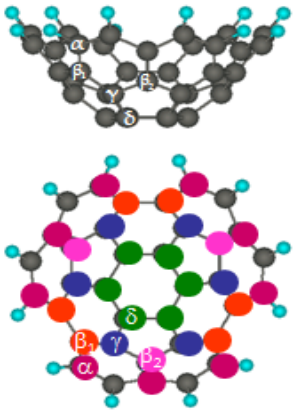

Figure 2 Minumum unit of ZTC ($C_{36}H_{12}$)

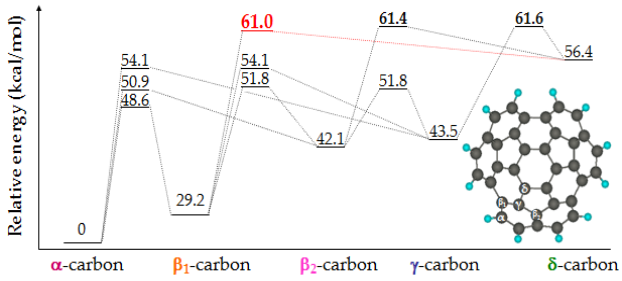

Figure 3 Relative energy of each carbon sites and transition states

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[1] H. Nishihara, et al., J. Phys. Chem. A, 113, 3189 (2009). [2] H. Nishihara, et al., Carbon, 47, 1220 (2009). [3] K. Suzuki, et al., J. Alloys. Compd., **509S**, S868 (2011). [4] K. Suzuki, et al., Compt. Theoret. Chem., **975**, 128 (2011). [5] K. Suzuki, et al., Theor. Chem. Acc., **130**, 1039 (2011).