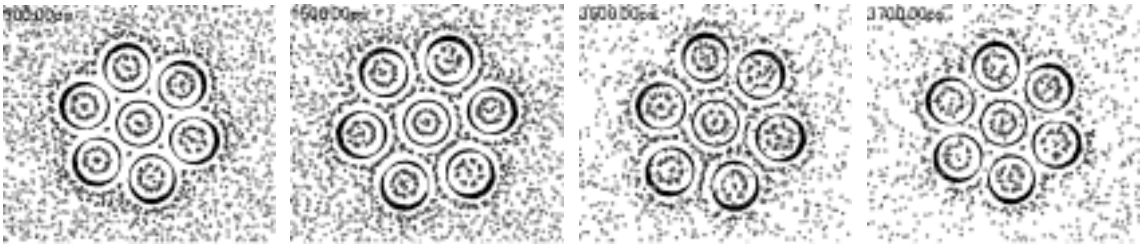
Molecular Dynamics Simulation of Hydrogen Absorption and Phase Transformation of Bundle of SWNTs

S. Maruyama[1,2], Y. Yoshino[2] and T. Kimura[2]

[1]Engineering Research Institute, The University of Tokyo, Japan
[2]Department of Mechanical Engineering, The University of Tokyo, Japan

The mechanism of efficient hydrogen storage with SWNTs was studied through classical molecular dynamics simulations. Assuming the physical adsorption of hydrogen to the surface of SWNTs, potential forms between H_2-H_2 and C-H_2 were both expressed by Lennard-Jones (12-6) functions. Each SWNT was regarded as rigid molecule and van der Waals potential between SWNTs was derived as a Lennard-Jones (8-4) function by integrating potential between carbon atoms. As shown in Fig. 1(a), 7 SWNTs were placed in a fully periodic system but each SWNT was shorter than the calculation domain so that hydrogen atoms could freely reach inside tubes and between tubes from longitudinal direction. During 500 ps simulation for 12 MPa and 77 K, the adsorption was observed only inside of tubes and outside of bundles as shown in Fig. 1(a). Here, the gravimetric storage density reached 5.7 wt%. We needed to once reduce the van der Waals potential between SWNTs to observe the phase transformation from the close packed bundle to the structure accommodating a hydrogen layer between tubes as in Fig. 1(b). Here, the gain of storage density was 1.6 wt %. Then, the phase transformation in the opposite direction was simulated by reducing the hydrogen pressure step wisely down to 6 MPa as shown in Fig. 1(c) and Fig. 1(d). By this phase transformation back to the close packed tubes, the storage density of 6.0 wt % in Fig. 1(c) reduced to 4.0 wt % in Fig. 1(d). Here, the considerable reduction of hydrogen atoms ‘inside’ tubes (0.7 wt %) was observed; suggesting the importance of the van der Waals force through surface of SWNTs. Through similar simulation for a bundle of (16,16) SWNTs, the effect of diameter of SWNTs was also examined.



(a) 12 MPa (b) Transformed (c) 6 MPa (d) Tranformed

Fig. 1 Phase transformation of a small bundle of (10,10) SWNTs with hydrogen absorption. Note that carbon atoms are not shown and SWNT are shown as bonds.