Ab Initio Molecular Dynamics Simulations of H_2 Formation inside POSS Compounds

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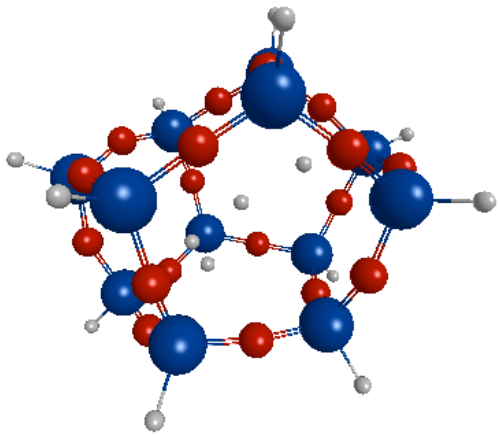
Polyhedral oligomeric silsesquioxanes (POSS), $[RSiO_{1.5}]_n$ (n = 4, 6, 8, 10, 12, …), referred to as T_n, have been the focus of considerable experimental and theoretical interest because of their wide variety of practical uses for many years. Especially, making use of the cage cavities for encapsulation of atoms and ions is one of the very exciting research areas of POSS compounds. The present study applies the ab initio molecular dynamics (AIMD) method to examine the mechanism and dynamics for H_2 formation process inside some POSS compounds in various situations.

Three types of H_2 formation were considered here : (I) $H + H@T_n \rightarrow H_2@T_n$[1], (II) $H + H_2@T_n \rightarrow (H + H_2)@T_n$, and (III) $H + (H + H_2) \rightarrow 2H_2@T_n$; n=8 and 12. The host molecules are T_8 with a cubic structure and the larger T_{12}. The various reactions, such as H_2 formation ($H + H \rightarrow H_2$), H-H exchange ($H + H_2 \rightarrow H_2 + H$) and escaping of a H or H_2 from the cage, take place depending on the initial condition.



The state of two hydrogen atoms and one hydrogen molecule moving inside T_{12} at a time of the AIMD trajectory for the reaction of $H + (H + H_2)@T_{12} \rightarrow 2H_2@T_{12}$

[1] Kudo, T., Taketsugu, T. and Gordon, M.S. *J. Phys. Chem. A*, **2011**, *115*, 2679.