Structure and dynamics of amorphous ice including carbon dioxide

Y. Naoshima and T. Ikeda-Fukazawa

Department of Applied Chemistry, Meiji University, Japan

In interstellar molecular clouds, various elements such as hydrogen, oxygen, carbon, and nitro gen are condensed onto dust grains, and formed H_2O amorphous ice and various molecules (e. g., CO_2, NH_3, CH_4, H_2CO, and so on). The various gas molecules, which are included in amor phous ice as impurities, undergo chemical evolutions to organic molecules through various pr ocesses [1]. Thus, the structure and properties of amorphous ice including gas molecules are i mportant to understand the molecular evolutions of organic molecules [2]. To investigate the effects of included gas molecules on the structure and properties of amorphous ice, the molec ular dynamics (MD) calculations of amorphous ice including CO_2 were performed.

We used an interatomic potential model (KAWAMURA potential model) for the MD simulati ons [3]. The amorphous ice was prepared by quenching of a liquid phase consisting of 368 wa ter molecules and n CO_2 molecules (n = 0–64) from 360 K to 200 K with 1.0 K/fs in cooling r ate. After annealing at 200 K, the system was cooled to 60 K with 1.0 K/fs. The density of the system at 60 K depends on the time period of the annealing at 200 K, and was controlled to b e 0.960 g/cm^3 in the present study. For the system with n > 4, CO_2 molecules formed a cluster before the quenching at 360 K. To prepare a system with isolated CO_2 molecules, pure amorp hous ice was formed by quenching with the same procedure. Then, m H_2O molecules were rep laced with m CO_2 molecules (m=1–61) at 60 K.

The results show that the density of H_2O ice with a CO_2 cluster is larger than that of pure ice a nd the density increases with increase in n. On the other hand, the density of H_2O ice with isol ated CO_2 molecules decrease with increase in m. To investigate the mechanisms of the density changes in H_2O ice, the vibrational densities of states of H_2O were calculated. The results sho w that the frequency of O–H stretching mode with both of isolated CO_2 and CO_2 cluster is hig her than that of pure amorphous H_2O ice. This indicates that the included CO_2 has effect to we aken the strength of hydrogen bonds of surrounding H_2O ice. The mean coordination number of H_2O in amorphous ice including isolated CO_2 is smaller than that of pure amorphous H_2O i ce. On the other hand, the mean coordination number of H_2O ice with CO_2 cluster is close to t hat of pure amorphous H_2O ice except for the interface with CO_2 cluster. These results sugges t that the isolated CO_2 has effects to decrease the density of H_2O ice due to decreasing of hydr ogen bonding number of H_2O molecules, whereas the cluster CO_2 causes an increase in densit y by distortion of local structure of ice in the interface. From the results, we discuss the effects of carbon dioxide on structure and dynamics of amorphous ice.

References

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