Project Title:

First principles calculation of Earth and Planetary Science

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1. Background and purpose of the project, relationship of the project with other projects

Intensive searches for alternative energy resources have been motivated due to the increasing fossil energy consumption and its related global environmental concerns. Hydrates of natural gases such as methane hydrate, CH4-H2O (the most abundant natural form of clathrate hydrate) prevailing in deep-sea sediments and permafrost have received active attention and been proposed as one of these energy resources. The global reserve of natural gas in the hydrate form is estimated to be significantly larger than that from traditional fossil fuels and will become a valuable future energy resource. More recently, hydrogen hydrate is of great interest because it facilitates environmentally clean (water is the only by-product) and highly efficient energy conversion. From astronomical point of view, water is known to be a major constituent of Uranus and Neptune. On the other side, hydrogen is the most abundant element in the universe, making up 75% of normal matter by mass and over 90% by number of atoms. Water and hydrogen under high pressure may form hydrogen hydrate. Therefore, study of water and hydrogen at high pressure has wide implications in astrophysics and knowing their physical properties is also important for understanding the structure and the formation history of these planets. The behavior of hydrates under pressure can also provide valuable information on water–water interactions and interactions of water with a wide range of guest molecules. Furthermore, studying H2O and H2 mixtures may provide insight into the nature of hydrogen-rich atmosphere in the large-body interstellar ice embryos postulated to exist during planet formation.

2. Specific usage status of the system and calculation method, Result, Conclusion

We investigated structural changes, phase diagram, and vibrational properties of hydrogen hydrate in filled-ice phase C2 by using the first principles molecular dynamics simulation. It was found that the experimentally reported ‘cubic’ structure is unstable at low temperature and/or high pressure: The ‘cubic’ structure reflects the symmetry at high (room) temperature where the hydrogen bond network is disordered and the hydrogen molecules are orientationally disordered by thermal rotation. In this sense, the ‘cubic’ symmetry would definitely be lowered at low temperature where the hydrogen bond network and the hydrogen molecules are expected to be ordered. At room temperature and below 30 GPa, it is the thermal effects that play an essential role in stabilizing the structure in ‘cubic’ symmetry. Above 60 GPa, the hydrogen bonds in the framework would be symmetrized and the order-disorder transition would disappear. These results also suggest the phase behavior of other filled-ice hydrates. In the case of rare gas hydrate, there would be no rotation-nonrotation transition since the guest molecule keeps its spherical symmetry at any temperature.

3. Schedule and prospect for the future

We continue to study interesting properties of various gas hydrates.

Fiscal Year 2012 List of Publications Resulting from the Use of RICC

[Publication]

Jingyun Zhang, Jer-Lai Kuo, Toshiaki Iitaka, "First principles molecular dynamics study of filled ice hydrogen hydrate", J. Chem. Phys. 137, 084505 (2012).

http://link.aip.org/link/?JCP/137/084505

[Oral presentation at an international symposium]

T.Iitaka, Ice and Gas Hydrate Under High Pressure,

Workshop on Structure and Dynamics of Water in Gas, Liquid and Solid Phases,November 28 - 30, 2012 Dr. Poe Lecture Hall, Institute of Atomic and Molecular Sciences, Academia Sinica

http://www.iams.sinica.edu.tw/ASWaterWorkshop2012/speakers

Toshiaki I IITAKA, J. Zhang, and J.L. Kuo、

First principles molecular dynamics study of filled ice hydrogen hydrate,

Physics and Control of Clustering Solids, Awaji Yumebutai, Awaji Island Japan.

5-7, Nov, 2012.

http://www.cmp.sanken.osaka-u.ac.jp/confs/jfsem12/Program.pdf