Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

PPS24-P01 Room:Poster Time:May 1 18:15-19:30

Importance of deuterium fractionation of ethanol by grain surface reactions: experiment of H-D tunneling substitution

OSAKA, Kazuya[1*] ; OBA, Yasuhiro[1] ; KOUCHI, Akira[1] ; WATANABE, Naoki[1]

[1]Inst Low Temp Sci, Hokkaido University

Since we have demonstrated the importance of tunneling grain surface reactions in deuterium fractionation of molecules, many works have targeted this process. To date, we have shown that the grain surface reactions play a crucial role in deuterium enrichments of water, formaldehyde, methanol, and methylamine. In this talk, we present the results of experiment on H-D substitution tunneling reactions of ethanol on cryogenic surfaces. Although C_2H_5OH was observed toward interstellar clouds, its deuterated species have not been detected. However, it was found that its homologous, CH_3OH can be highly deuterated by H-D substitution reactions on grain surfaces and thus it should be reasonable to focus on the potential importance of this process for ethanol. We demonstrated that deuterated methanol is efficiently produced by tunneling reaction of H atoms at very low temperatures relevant to grain surfaces in clouds. H-D reactions predominantly occur in CH_3-CH_2- groups but were hardly observed in an ?OH group which is consistent with the methanol case.

Keywords: deuterium enrichment, ethanol, grain surface reaction

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Ion-induce nucleation experiment II: free energy of the water-cluster ion

HIDAKA, Hiroshi[1*] ; NAKAI, Yoichi[2] ; KOJIMA M., Takao[3] ; WATANABE, Naoki[1]

[1]Inst. Low Temp. Sci., Hokkaido Univ., [2]RIKEN Nishina Center, [3]RIKEN Atomic Physics Laboratory

Ion-induce nucleation in gas phase is an important mechanism for grain formation in various circumstances. However, the number of works regarding this formation mechanism is very limited. To investigate the elementally processes of ion-nucleation mechanism, we recently developed a new apparatus (See, the presentation by N. Watanabe in this session). Using this apparatus, the cluster ion formation with an ion core mass-selected, which is the first stage of nucleation, can be observed quantitatively. In this presentation, we show the results of experiment on water-cluster ion formation in which free energies with the size of cluster have been determined. The experiment was performed at temperatures in range of 230-400 K with the supersaturation ratio of about 10^{-3}-10^{-2}.

Keywords: interstellar grain, cluster ion, nucleation

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Detection of levitation dust around the asteroid by Hayabusa-2 LIDAR

OSHIGAMI, Shoko[1*] ; SENSHU, Hiroki[2] ; WADA, Koji[2] ; KOBAYASHI, Masanori[2] ; NAMIKI, Noriyuki[2] ; MIZUNO, Takahide[3]

[1]National Astronomical Observatory of Japan, [2]Planetary Exploration Research Center, Chiba Institute of Technology, [3]Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency

The micron-size particles are continuously produced at the surface of airless bodies like the Moon and asteroids by innumerable micro impacts and thermal stress related to large temperature difference between daytime and nighttime. Previous asteroid missions have revealed smooth appearance of topography on 951 Gaspra, 243 Ida, and 433 Eros suggesting that these asteroids are covered with particles smaller than resolution of camera images. Particularly, the exploration of Eros by NEAR Shoemaker has revealed as smooth surface as a liquid water at the base of craters whose diameter is between 20 and 300 m. This "pond" is consistent with stagnant dusts of diameter smaller than 50 microns. Based on this observation, dust levitation hypothesis was proposed. According to this hypothesis, a photoelectric effect of solar UV positively charges both dust and the surface. Then a balance between electric repulsion and gravity causes 0.5-microns dusts to oscillate vertically over the surface of Eros long period of time. When a dust has a horizontal velocity, it transfers laterally until it reaches to a shadow of topography where electrostatic field is weaker than surroundings. Thus topographic depression such as a crater becomes a sink of levitating dusts.

LIDAR is one of four remote-sensing instruments onboard Hayabusa-2, and is used to measure altitudes of the spacecraft from a surface of the asteroid, 1999 JU3, for not only secure navigation but also scientific investigation of a C-type asteroid. Hayabusa-2 LIDAR has been improved from that onboard Hayabusa which explored and returned samples from asteroid 25143 Itokawa. A new function called dust count mode is implemented to Hayabusa-2 LIDAR to observe spatial distribution of dust number density in 8 levels with resolution of 20 m in bore sight direction. LIDAR can hardly observe lateral distribution of dusts, but distinguish a weak reflection of thin dust cloud from that of the surface. To plan an operation of the dust count mode observation is difficult because the number density of asteroid dust is not known at all. Instead, we evaluate the lower bound of number density that is geologically important for morphology of asteroid surface. For a given number density of dusts and under an assumption that a characteristic time of levitation is the rotation period of 1999 JU3, the rate of embayment of craters is calculated. If this rate of embayment is greater than that of crater production, we need to take into account a modification process for the study of crater morphology and crater counts of 1999 JU3. This lower bound is calculated to be 10^6 m^{-3} for a cloud of dusts whose radius is larger than a few microns. Then we set this value as a target of the dust count mode observation.

A detectability of dust count mode is dependent on sensitivity of Hayabusa-2 LIDAR and an altitude of the spacecraft. We calculate a reflection from dusts using Mie scattering model assuming that a diameter of dust particle is constant and is larger than the wavelength of laser, that is, 1064 nm. A characteristic distance between dusts is also assumed to be sufficiently larger than the wavelength so that interaction between dust particles is negligible. Using a lidar equation, we calculate a peak power of backscattering light from a dust cloud for various sets of the distance, the number density, and the dust radius. The peak power of reflection is generally stronger than noise level of the detector. The reflection from dust cloud is so weak that the targeted number density of 10^6 m^{-3} is hardly higher than the detection limit. Even at the lowest altitude, the reflection from a dust cloud of 10-microns radius for 10^6-m^{-3} number density is equivalent to the detection limit. If the dust radius is 5 microns, number density more than 10^7 m^{-3} is necessary to be detected. Therefore we plan to start the dust count operation from the HP and attempt to conduct as much operations as possible at low altitude.

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Condensation of forsterite under protoplanetary disk conditions

TACHIBANA, Shogo[1*] ; TAKIGAWA, Aki[2] ; MIYAKE, Akira[2] ; NAGAHARA, Hiroko[3] ; OZAWA, Kazuhito[3]

[1]Dept. Natural History Sci., Hokkaido Univ., [2]Dept. Geol. Mineral., Kyoto Univ., [3]Dept. Earth Planet. Sci., Univ. Tokyo

Meteoritic evidence indicates that dust condensation occurred in the early stage of solar system evolution. In this study, we succeeded in performing condensation experiments of forsterite under controlled protoplanetary-disk conditions, which will make significant contribution to understanding silicate formation and chemical fractionation in protoplanetary disks.

Condensation experiments were carried out in the system of Mg_2SiO_4-H_2-H_2O. A mixed gas of H_2 and H_2O was flowed into a continuously evacuated infrared vacuum furnace at a controlled rate to keep a pressure constant. Synthetic forsterite powder in an It crucible was heated as a gas source. A part of evaporated gases were condensed on a Pt mesh located at a cooler region in the chamber. The pressure and temperature conditions were close to those of protoplanetary disks. The total pressure of the system was 5.5 Pa, and the substrate temperature ranged from 1320 to 1160 K. The H_2O/H_2 ratio was set at 0.015, which was about 15 times larger than the solar ratio. The SiO/H_2 ratio was evaluated to be about 0.7-2 % of the solar ratio from the weight loss rate of the gas-source forsterite. Experimental duration ranged from 6 to 237 hours.

Sub-micron to micron-sized condensates covered with Pt substrates at 1160 and 1275 K, but no condensates were found at 1320 K. The typical size of condensates at 1160 K was less than 1 micron irrespective of experimental duration and no effective growth of each condensed grain was observed. Condensates at 1275 K for >40 hours partly had several micron-sized flat regions. EDS analyses showed that chemical compositions of condensates were consistent with the stoichiometry of forsterite, and their EBSD patterns were well fitted with the patterns from crystalline forsterite. Coincident EBSD patterns were obtained from the flat region of condensates at 1275 K, suggesting that the area was covered with a single crystal. TEM observation of condensates at 1160 K also found that the condensates were polycrystalline forsterite with a thickness of 30-150 nm, and infrared absorption spectra of condensates show clear 10-micron absorption features resembling those of crystalline forsterite. These evidence indicates that polycrystalline forsterite condensed at 1275 and 1160 K.

The mean free path of gas molecules under the present experimental conditions is less than 1 mm, and the evaporated forsteritic gas and the ambient H_2-H_2O gas are expected to be well mixed. Supersaturation ratios (S) for experiments at 1320, 1275, and 1160 K are thus estimated to be $<$1.2, $<$10, and $<$1000-2000. These supersaturation ratios correspond to the supercooling of $<$5, $<$60 and $<$170 K, respectively.

No condensates were found at 1320 K because the degree of supersaturation was too small for nucleation of forsterite or even the vapor was not saturated with forsterite ($S<1$). The condensates at the supercooling of $<$170 K (1160 K) imply that heterogeneous nucleation of new grains occurred successively on preexisting grains. On the other hand, with the supercooling of $<$60 K (1275 K), some grains seem to have grown up to several microns, and some seem to have newly nucleated on preexisting grains, suggesting that both nucleation and growth of each condensate occurred.

These differences would result in a structural difference in forsterite dust condensed in protoplanetary disks. Fluffy aggregates of sub-micron sized fine particle would form with a supersaturation of $>$1000, while aggregates of micron-sized grains would form with S of 10 that could be an analogue of amoeboid olivine aggregates in chondrites.

Keywords: forsterite, condensation, protoplanetay disk

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A New Experiment for Organic Molecule Formation by Catalytic Reactions on the Surface at Low Temperature and Pressure

KIMURA, Yuki[1*] ; TSUCHIYAMA, Akira[2] ; NAGAHARA, Hiroko[3]

[1]Tohoku University, [2]Division of Earth and Planetary Sciences, Graduate School of Sciece, Kyoto University, [3]Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo

Abundant H_2, CO and N_2 gases react to be more complex molecules mainly on the cooled surface of cosmic dust particles in the molecular cloud and/or primitive solar nebula [1]. The production of organic molecules and subsequent evolution to organic materials in the solar nebula may contribute to the primordial organic system of the Earth. Catalytic chemical reactions are possible production pathway of organic materials in the solar nebula after the formation of simple molecules on nanometer sized cosmic dust particles in the molecular clouds. Experimentally, organic molecules ranging from methane (CH_4), ethane (C_2H_6), benzene (C_6H_6) and toluene (C_7H_8), to more complex species such as acetone (C_3H_6O), methyl amine (CH_3NH_2), acetonitrile (CH_3CN) and N-methyl methylene imine (H_3CNCH_2) have been produced using such as the Fischer-Tropsch type (FTT) and Haber-Bosch type (HBT) reactions on analogs of naturally occurring grain surfaces [2]. Previous studies were performed at higher-temperature ($>$573 K) and pressure ($\sim$1 atm) than the expected conditions in the solar nebula [3-6]. However, since the actual environment is at lower temperature and pressure, it is not clear whether the previous experimental results can be extrapolated to the solar nebula. Our group seeks to elucidate the reaction rates of chemical reactions including isotopic fractionation at lower temperature (100-500 K) and pressure (10^{-3}-10^{0}) and their contribution to the primordial organic system of the Earth.

We are constructing a vacuum chamber based on a new concept to conduct the experiments mentioned above. The chamber with a differential pumping system has a temperature-controlled substrate, a Fourier transform infrared spectrometer (FT-IR), and two quadrupole mass spectrometers (Q-MSs). The substrate has an iron or silicate thin film for FTT and HBT reactions and the FT-IR measures the vibration modes of adsorbed and produced molecules on the surface and the Q-MSs detect volatile and nonvolatile molecules, respectively. As a result, reaction rates of molecules such as H_2, CO, N_2 and NH_3 on iron or silicate substrate will be obtained as a function of temperature and pressure.

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Keywords: Organic molecules, Catalytic reactions, Protoplanetary system

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Dust movement and chemical evolution of proto-solar disk

NAGAHARA, Hiroko[1*] ; NAKATA, Mamoru[1] ; OZAWA, Kazuhito[1]

[1]Hiroko Nagahara

Origin and evolution of the protoplanetary system have been developed mostly from the dynamic point of view, which includes two competing theories, and whichhas been improved by astrophysical observation of exoplanets and extrasolar planetary systems. On the other hand, examination of meteorites and samples by planetary explorations such as lunar samples, cometary particles, and regolith particles of the asteroid Itokawa enable us to gain insight into the evolution of the Solar System. Although those primitive materials give various information, they are not linked to the physical processes in the primary Solar System.

The purpose of this study is to demonstrate how chemical composition distribution evolves over time in the early stage of the proto-solar disk. In order to combine physical processes and chemistry, we have developed a new model consisting of chemical equilibrium calculation and particle tracking equations. At first, we calculate the chemical composition of starting particles at each position in the protoplanetary disk, to track their each motion in the evolving disk, and to analyze the bulk composition of particles that came from various positions in particular time and space. Then, the dynamic evolution of individual particles is calculated in one-dimensional steady-state disk model. In an early stage, particle located in the inner region of the disk have a composition rich in refractory components and those outside have unfractionated CI-like composition. Particles in average move inward by the angular momentum conservation, but a little fraction of them move outward by the turbulent diffusion. Therefore, mixing of refractory particles from inside and CI-like materials from outside takes place, and the mixing ratio vary with time and space.

Because of inward movement of many particles, the relative fraction of particles from outside increases with time for one particular region in the disk, that is, the bulk chemical composition of particles is getting more CI-like. Similarly, the bulk chemical composition of particles at particular place is getting more CI-like with time. Calculations with model parameters of higher temperature of the disk suggest that longer time is needed to replace refractory-rich compositions by a CI-like composition. It is because the radial distance between fractionated particles with refractory-rich composition and unfractionated CI-like materials is longer in a high temperature disk.

Comparing these results and the composition of CM, CO, CV chondrites, it is concluded that CV composition can be reproduced at the most inner region, CO in the next, and CM most outer region in the disk. The present work shows that the composition of carbonaceous chondrites were formed at the asteroid belt region at the early stage of disk evolution with the wide spread of high temperature region.

Keywords: protoplanetary disk, chemical evolution, dust movement, chemical equilibrium