

## 星間塵表面におけるエタノール重水素濃集の可能性：H-Dトンネル置換反応実験 Importance of deuterium fractionation of ethanol by grain surface reactions: experiment of H-D tunneling substitution

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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<sub>2</sub>H<sub>5</sub>OH was observed toward interstellar clouds, its deuterated species have not been detected. However, it was found that its homologous, CH<sub>3</sub>OH 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<sub>3</sub>-CH<sub>2</sub>- groups but were hardly observed in an -OH group which is consistent with the methanol case.

キーワード: 重水素濃集, エタノール, 星間塵表面反応

Keywords: deuterium enrichment, ethanol, grain surface reaction

## イオン誘起微粒子核生成 II: 水クラスターイオンの自由エネルギー Ion-Induce nucleation experiment II: free energy of the water-cluster ion

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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 elementary 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  $10^{-3}$ - $10^{-2}$ .

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Keywords: interstellar grain, cluster ion, nucleation

## はやぶさ2 レーザ高度計による小惑星周辺ダスト検出の試み Detection of levitation dust around the asteroid by Hayabusa-2 LIDAR

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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  $10^6 \text{ m}^{-3}$  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  $10^6 \text{ m}^{-3}$  is hardly higher than the detection limit. Even at the lowest altitude, the reflection from a dust cloud of 10-microns radius for  $10^6 \text{ m}^{-3}$  number density is equivalent to the detection limit. If the dust radius is 5 microns, number density more than  $10^7 \text{ m}^{-3}$  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.

## 原始惑星系円盤条件でのフォルステライト気相成長 Condensation of forsterite under protoplanetary disk conditions

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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  $\text{Mg}_2\text{SiO}_4\text{-H}_2\text{-H}_2\text{O}$ . A mixed gas of  $\text{H}_2$  and  $\text{H}_2\text{O}$  was flowed into a continuously evacuated infrared vacuum furnace at a controlled rate to keep a pressure constant. Synthetic forsterite powder in an Ir 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  $\text{H}_2\text{O}/\text{H}_2$  ratio was set at 0.015, which was about 15 times larger than the solar ratio. The  $\text{SiO}/\text{H}_2$  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  $\text{H}_2\text{-H}_2\text{O}$  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.

キーワード: フォルステライト, 気相成長, 原始惑星系円盤  
Keywords: forsterite, condensation, protoplanetary disk

## 低温低圧環境下における微粒子表面での触媒化学反応による有機分子生成実験に向けて

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

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Abundant H<sub>2</sub>, CO and N<sub>2</sub> 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<sub>4</sub>), ethane (C<sub>2</sub>H<sub>6</sub>), benzene (C<sub>6</sub>H<sub>6</sub>) and toluene (C<sub>7</sub>H<sub>8</sub>), to more complex species such as acetone (C<sub>3</sub>H<sub>6</sub>O), methyl amine (CH<sub>3</sub>NH<sub>2</sub>), acetonitrile (CH<sub>3</sub>CN) and N-methyl methylene imine (H<sub>3</sub>CNCH<sub>2</sub>) 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 (~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<sup>-3</sup>-10<sup>0</sup>) 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<sub>2</sub>, CO, N<sub>2</sub> and NH<sub>3</sub> 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



## 原始惑星系円盤進化にともなうダストの運動と組成分布 Dust movement and chemical evolution of proto-solar disk

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太陽系の力学的な進化を記述するために構築された惑星形成理論は、系外惑星や系外の円盤の観測によって改良が加えられ、近年まで発展してきた。他方、物質科学的な研究も、地球に飛来する隕石や探査によって得られた月試料、彗星の塵やいとかわの粒子などの分析によって発展してきた。太陽系の形成過程を解明する上で形成初期の情報を保持する物質科学的証拠は、円盤初期の物理過程に制約を与えるが、そのためには化学と物理モデルのカップリングが必要不可欠である。

原始惑星系円盤における物理と化学を統合して理解するため、本研究においては、原始惑星系円盤初期の粒子の化学組成を中心星からの関数として決定し、各粒子の円盤内の物理的移動を追跡することで、円盤内の粒子の総化学組成の時空間変化を調べることが目的とした。

モデルは、化学平衡計算とダスト粒子移流拡散方程式を基本とする。熱力学的平衡計算により、円盤内の各初期位置における凝縮相の組成を決定し、その組成は移動により変化しないものとした。各粒子の運動は一次元定常 $\alpha$ 円盤を仮定し、ラグランジアン法による移流として追跡した。初期に内側に存在する粒子は高温のため揮発性元素に枯渇した組成を持つ一方、外側の粒子は未分化な組成をもつ。粒子は時間とともに、全体としては太陽方向へ移動するが、乱流拡散の効果により円盤外向きの成分も存在する。内側の揮発性元素に枯渇した粒子と外側の揮発性元素を含む粒子との混合を解析した。

計算の結果、以下のことが明らかとなった。(1) 粒子は全体としては太陽方向へ移動するため、円盤内粒子の総化学組成は、各時間において円盤外側ほど未分化となる。(2) 特定の位置についてみると、時間経過とともに未分化な組成となる。(3) 円盤面密度、温度構造をパラメータとして検討した結果、高温の円盤ほど、円盤内側領域の組成が未分化なものに置き換えられるまでの時間が遅くなる。これは高温の円盤ほど分化した化学組成領域が広がるため、外側領域から未分化組成の粒子が移動するのに時間を要するためである。

これらの結果をC型コンドライトの組成と比較したところ、円盤内側からCV, CO, CMの順番に各コンドライトを説明する領域が存在するということがわかった。また本研究により、小惑星帯においてC型コンドライト組成を作り出すためには、初期に高温領域が広く広がった円盤、さらに早期に微惑星形成がおこることが必要であることが明らかとなった。

キーワード: 原始惑星系円盤, 化学組成, ダスト移動, 平衡計算

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