星間空間における宇宙風化を模擬した水素イオン照射実験
Space weathering in the interstellar medium by hydrogen ion irradiation

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宇宙空間にはダストと呼ばれる固体微粒子が普遍的に存在する。ダストは主に進化末期星の大気外層や、超新星爆発で放出されたガスから生成されると考えられている。また、結晶質ケイ酸塩ダストは星周に存在するが、星間空間にはほとんど存在しないことが観測から分かっている。星間空間に結晶質ケイ酸塩が存在しない理由の一つに、結晶質ケイ酸塩の構造破壊による非晶質化が考えられる[1]。構造破壊の原因としては、１．粒子同士の衝突、２．宇宙線の照射、３．超新星爆発の衝撃波によって加速された星間イオンの照射、４．初期太陽系での太陽風照射、が考えられる。本研究では、星間空間におけるこのような破壊現象を広義の宇宙風化と呼ぶ。

星間空間での宇宙風化を模擬して、これまでにolivine、enstatite、diopsideなどへの照射実験が行われているが、実験の容易さから最も豊富なガス種である水素ではなくヘリウムイオンを使っているものが多い[1-4]。しかし、結晶内部の構造変化やブリスターとよばれる水ぶくれ構造の特徴は照射ガス種によって異なる可能性がある [5]。そこで本研究では、主にH^+イオンの照射による構造変化の条件を定めることを目的とし、ダスト模擬物質へのイオン照射実験を行った。

星間ケイ酸塩ダストを模擬した物質として、olivine (San Carlos産)、enstatite(タンザニア産)、合成単結晶forsterite、serpentine($(Mg_{2.8} Fe_{0.2})_3Si_2O_5(OH)_4$)、$MgSiO_3$組成の急冷ガラスを用いた。また、IDP、始原隕石中に豊富に含まれるとされるFeSを模擬したpyrrhotite($Fe_{0.9}S$)と、鉄隕石も用いた。試料は大きさ3x5x0.5 mmに加工し、表面は1 μm以下まで研磨後、表面を化学研磨処理した。

実験は若狭湾エネルギー研究センターで行った。加速エネルギー40keVのH_2^+、10keVのH_2^+イオンを用い、それぞれ照射量10^{16}~10^{18} ions/cm^{2}および 10^{17} ions/cm^{2}を照射した。照射時間が60分以上になる試料に関しては温度上昇を防ぐため水冷ステージを利用した。照射後の試料は走査型電子顕微鏡(SEM)により表面構造の変化を観察した。

olivine、enstatite、forsterite、serpentine、pyrrhotite表面にブリスターの生成が認められた。40keV H_2^+イオンを照射した場合、ブリスターが生成する照射量は、olivine、enstatite、serpentine、pyrrhotiteに関しては10^{18} ions/cm^{2}以上で、forsteriteは10^{17} ions/cm^{2}以上であった。鉄隕石と$MgSiO_3$ガラスには表面変化は観察されなかった。10keV H_2^+イオンを照射した場合、すべての試料から表面変化は観察されなかった。olivine, forsterite, serpentine, pyrrhotiteのブリスターは楕円形状であった。同一試料上のブリスターサイズはほぼ均一であったが、forsterite上のブリスターが最も小さく100 nmほどの大きさで、olivineとpyrrhotiteのブリスターは約3 μm、serpentineはやや大きく4-5 μmほどであった。enstatiteには波状の形状をしたブリスターが数μmの間隔で並んでいるような構造がみられた。

Matsumoto (2014)は鉄含有量の多いolivine試料に対して10 keV H_2^+イオンを10^{17} ions/cm^{2}照射し、ブリスター生成を示した [4]が、本研究では同様条件でブリスターを確認できなかった。これは試料の鉄含有量の違いに起因すると考えられ、鉄を多く含むolivineの方が構造が破壊されやすいことを示唆する。enstatiteに関しては、40keV H_2^+イオンの照射でブリスター形成に必要な照射量は、先行研究で調べられた50keVのHe^+イオンによる非晶質化照射料より一桁多い[2]。Pyrrhotiteに関しては、過去の1MeV Kr^+イオン照射実験において結晶完全な非晶質化は認められなかったが[6]、本実験において40keV、H_2^+、10^{18}ions/cm^{2}でブリスターが生成することがわかった。これらの違いはイオン質量と大きさの違いによるものであると考えられる。

今後、照射試料の内部構造を集束イオンビーム加工および透過電子顕微鏡観察により行う予定であり、SEM観察の結果と合わせて報告する。

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キーワード：宇宙風化、ダスト、照射実験、星間空間
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エンスタタイト凝縮実験に向けた装置改良とSiO_2-MgO蒸発実験

Development of experimental setups for condensation experiments of enstatite and evaporation experiments of SiO_2-MgO

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Enstatite whiskers elongated to the [001] axis were identified in chondritic porous interplanetary dust particles (CP-IDPs), Antarctica Micrometeorite (AMM), and samples from a comet Wild 2, while those elongated to the [001] axis are common in minerals occurred in the Earth and meteorites [1-4]. The condensation conditions of enstatite whiskers, however, have not been shown quantitatively. Moreover, the difference in the morphology of enstatite whiskers may reflect their different condensation conditions in the early solar system. In order to constrain on the formation conditions of enstatite whiskers with different crystal habits, we aim to construct the experimental setup that can control the condensation temperature, supersaturation ratio, and partial pressure of a hydrogen gas. In this presentation, we report the current status of our experimental setups and preliminary results of SiO_2-MgO evaporation experiments.

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あかり衛星による小惑星起源ダストの観測
AKARI observations of asteroidal dust

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The zodiacal emission is the thermal emission from the interplanetary dust and the dominant diffuse radiation in the mid- to far-infrared wavelength region. From the results of the Infrared Astronomical Satellite (IRAS) observations, it was found that there are many small-scale structures in the zodiacal emission distribution, such as dust band pairs at the ecliptic latitudes of +/-1.4 and +/-2.1 degrees (the inner band) and +/-9.3 degreed (the outer band). It is suggested that recent disruption events among multikilometer bodies in the main asteroid belt within the last several million years are major supply sources of the dust particles in the dust bands, and they produce a edge-brightened toroidal distributions of dust. We conducted observations of the asteroidal dust bands with the Japanese infrared satellite AKARI in mid- and far-infrared wavelengths region. AKARI clearly detects the zodiacal dust-band structures. We derived the heliocentric distances of dust bands from the AKARI observations: 1.9 AU and 2.2 AU from the Sun for the inner and the outer bands, respectively. Based on the AKARI spectroscopic observations in mid-infrared, the dust particles in the outer band show a slight different spectral shape of the silicate feature from those of other regions which are thought to be cometary origins. It is highly probable that we obtain the mid-infrared spectra of micron-sized asterodial dust grains for the outer band regions. We will discuss the size distribution and silicate composition for the dust grains in outer bands.

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赤外分光法によるアモルファス氷の表面構造の解析
Analyses of surface structure of amorphous ice using infrared spectroscopy

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In interstellar molecular clouds, water molecules deposit on dust grains, and forms amorphous ice. The amorphous ice includes various molecules such as CO, CO_2, NH_3, CH_4, H_2CO, and so on. These molecules undergo chemical evolutions to organic molecules through various processes. Because the deposited amorphous ice has a large surface area due to its uneven structure, the surface of amorphous ice has an important role for chemical evolutions of included molecules in molecular clouds [1].

Structure of amorphous ice depends on formation condition [2], and has been mainly classified by density into two types: low-density amorphous (LDA) and high-density amorphous (HDA) ice. The densities of the LDA and HDA ice are 0.94 ±0.03 and 1.1 ±0.1 g cm^{-3}, respectively. Although there are various studies for structures and properties of amorphous ice in bulk states, only a few studies have been reported for surface. In the present study, the surface structure of amorphous ice was analyzed by measurements of infrared (IR) spectra for thin film of vapor-deposited amorphous ice.

Amorphous ice was prepared with vapor deposition of distilled and degassed water on a substrate of oxygen-free copper at 43 K. The deposition rate was controlled to be a value in range of 0.02–0.61 nm/min. The inner pressure of the vacuum chamber during the deposition was about 5.0 $\times10^{-5}$ Pa. The IR spectra were measured using Shimadzu IRPrestage-21.

The spectral features change with deposition. According to the assignments of IR spectra for amorphous ice [3], the O–H stretching vibration band observed in 2800–4000 cm^{-1} was decomposed into three vibration modes (i.e., in-phase and out-of-phase modes of symmetric stretching, and asymmetric stretching mode). From the variations of the wave number of the out-of-phase mode, the variations in water structure of amorphous ice were analyzed.

The result shows that the wave number of the out-of-phase mode increases as the thickness of the amorphous ice increases, and gradually approaches a constant value of 3480 cm^{-1}. From the observed variations, the thickness of the surface layer is estimated to be around 20 nm. Furthermore, the wave numbers of the surface layer and bulk state are estimated to be 3420 and 3480 cm^{-1}, respectively. Because these values are close to those for HDA and LDA ice [4], the internal part of the sample is confirmed to be HDA ice. For the surface layer, the density is smaller than that of the internal part. Therefore, it is supposed that the structure is a close representation of LDA ice. Furthermore, it was found that the surface structure of the amorphous ice depends on deposition rate. From the results, we discuss the structure and dynamics of amorphous ice surface.

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アモルファス氷表面構造の密度依存性
Effects of Density on Surface Structure of Amorphous Ice

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In interstellar molecular clouds, various molecules (for instance, H_2O, NH_3, CO, CO_2, and so on) are formed from elements such as H, C, O, and N by deposition on dust grains [1]. Most of H_2O exists as a thin shell of amorphous ice around dust grain. The deposited molecules undergo chemical evolutions to organic molecule through various processes [2]. The structure of amorphous ice surface is an important factor to understand the molecular evolution in molecular clouds.
Amorphous ice is formed by vapor deposition of H_2O gases [3] or compression of crystalline ice [4]. The structure of amorphous ice depends on formation processes and is classified into two types: high-density amorphous (HDA) and low-density amorphous (LDA) ices [4]. To investigate the effects of internal structure on surface structure, the molecular dynamics (MD) calculations of amorphous ice were performed.
The KAWAMURA potential model [5] was used for the MD calculations. The amorphous ice was prepared by quenching a liquid phase consisting of 2760 water molecules from 280 to 235 K with 2.5 K/fs in cooling rate. After annealing at 235 K, the system was cooled to 10 K. The density of amorphous was controlled with the time period of the annealing at 235 K. An infinite surface was simulated by replicating the cell in the directions parallel to the surface using periodic boundary conditions. The pressure was kept at 0.1 MPa. The layer with 5 Åin thickness from the outmost atom was analyzed as the surface layer.
The calculations show that the density of amorphous ice at 10 K depends on the temperature history before the cooling to 10 K. In the case that the time period of annealing at 235 K was 1400 ps, for instance, the formed amorphous has the minimum density of 0.962 g/cm^3 at 10 K. For the case that the system was directly quenched from 300 to 10K without annealing at 235 K, the density reaches its maximum value of 1.095 g/cm^3. Because a significant change of density was observed at 235 K, we controlled the density of internal part of amorphous ice with time period of the annealing at 235 K.
The result shows that the density of the surface layer is smaller than that of the internal part, and increases as the density of the internal part increases. The decrease in molecular density observed in the surface layer is attributed to the uneven structure and large amplitude of the thermal vibrations of water molecules. To investigate the contribution of the uneven structure, the radical distribution function (RDF) was calculated. The position of the second peak, which is observed at around 4.1 Å, of surface layer is about 0.3 Ålarge than that of internal part. This suggests that the surface layer has a lower density even if there is no unevenness. The amplitude of the thermal vibration is measured by the atomic displacement parameter (ADP). The calculation indicates that ADP of surface layer is larger than that of internal part, and depends on internal density. This result is consistent with the calculation of RDF. In the RDF result the broadening of peaks was observed for surface layer. It is concluded that the structure of surface layer is affected by density of internal part. This effect of internal structure on surface structure of amorphous ice might have important implications for molecular evolution in molecular clouds.
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低温真空下におけるヘルプガス含有二酸化炭素ハイドレートの形成メカニズム
Formation of Carbon-Dioxide Clathrate Hydrates including Help Gases under Vacuum Conditions

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In interstellar molecular clouds, water molecules condense on dust grains. The condensed water exists as amorphous ice in the cold clouds and is transformed into various structures depending on thermal conditions and compositions of including molecules. Blake et al. [1] proposed the presence of clathrate hydrates in cometary ice. From the results using transmission electron microscopy and Fourier transformed infrared spectroscopy, they showed the phase transition of vapor deposited amorphous ice including CH_3OH and CO_2 into type-II hydrate at around 120 K.
Clathrate hydrates are inclusion compounds consisting of water molecules and a variety of guest molecules. Most hydrates form one of two distinct crystallographic structures, type-I and -II, depending on the sizes and shapes of the guest molecules. The structure of CO_2 hydrate formed under a high-pressure condition is type-I [2]. For the hydrate from the vapor deposited amorphous ice by Blake et al. [1], the structure was type-II due to the help-gases effect of CH_3OH. For the CO_2 hydrate grown epitaxially on a hydrate under vacuum condition, the structure depends on the structure of the hydrate as the substrate [3]. In order to investigate the formation mechanisms of CO_2 hydrate including help-gases under vacuum conditions, we analyzed infrared spectra of vapor deposited amorphous ice including CO_2 and C_3H_6O during warming.
The CO_2/H_2O ratios of the prepared gas mixtures were 0.13–16.8. The gas mixtures were deposited onto a substrate of oxygen-free copper at 43 K. After the deposition, the substrate was warmed from 43 to 160 K. Infrared spectra were measured at approximately 1 min intervals during deposition at 43 K and at 2 K intervals during warming. Furthermore, CO_2 and C_3H_6O mixed hydrates were prepared from gas mixtures of H_2O, CO_2, and C_3H_6O with various compositions, and were measured spectra with the same procedures.
From the variation in spectral features of H_2O–CO_2 ice with warming, remarkable changes were found at 82 K. The wave numbers of the O–H stretching modes of H_2O and the C–O asymmetric stretching modes of CO_2 change significantly at this temperature. Furthermore, significant gas release and exothermic temperature rise were observed. These results suggest that the crystallization of amorphous ice begins at 82 K. From the wave numbers of C–O asymmetric stretching modes, the formed crystal is determined to be type-I hydrate. From the formation conditions of CO_2 hydrate, we propose the phase diagram of the H_2O–CO_2 system under vacuum conditions. In addition, the formation processes of mixed hydrate including CO_2 and C_3H_6O were analyzed. The result shows that the transition temperature of the mixed hydrate depends on the guest composition. From the results, we discuss the formation mechanisms of clathrate hydrates under vacuum conditions.

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アンモニアのモンモリロナイト及びサポナイトへの吸着・脱着に伴う窒素同位体分別
Nitrogen isotopic fractionation of ammonia accompanied with adsorption-desorption process on montmorillonite and saponite

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Solar system objects show wide variation in the nitrogen isotopic composition. The δ^{15}N value (‰, normalized as vs. Air) ranges from -400‰ in the solar wind [1] and Jovian atmosphere [2] to around +1500‰ of comets, chondrites, and interplanetary dust particles (IDPs)[3, 4]. These pristine solar system materials also show occasional extreme ^{15}N-enrichment up to +5000‰ [5, 6]. This δ^{15}N variation in the solar system objects is considered to have been inherited from the cold interstellar environments [7]. However, only a few models can explain the interstellar chemistry to enhance ^{15}N-enrichments, such as photodissociation of nitrogen molecules and subsequent formation of ammonia [8], and conceivable isotopic fractionation processes have not well understood.

In the study, we conducted simple adsorption-desorption experiments using ammonia gas and two clay minerals (montmorillonite and saponite). From the results, we propose the adsorption-desorption process of ammonia on grain surface of interstellar dusts as a potential mechanism for ^{15}N-enrichmentin interstellar environments. Ammonia is a simple nitrogen-containing molecule and one of major nitrogen carriers in the molecular clouds. It is a highly reactive chemical and regarded as a precursor for other complex nitrogen-containing molecules. Thus, adsorption of ammonia could be a first step for grain surface chemistry to form more complex organic molecules. The adsorption-desorption experiments were performed to examine the possibility of nitrogen isotopic fractionation of ammonia by adsorption and desorption on the clay minerals. Each clay mineral sample, with controlled amount of water (0, 5, 10, 20 wt.% vs. dry cry minerals), was sealed into the glass serum bottles with ammonia gas (+27‰, SI Science). The glass vial was rested for a week under room temperature (25℃) to reach a stable state before analysis. After the initial analysis was completed, the glass vial was vacuumed for 1 to 8 hours to examine the results under vacuumed environment. The nitrogen isotopic composition of the adsorbed ammonia at each step of the experiments was determined by nanoEA/IRMS technique [9].

The results showed that adsorbed ammonia generally had larger δ^{15}N value than the initial ammonia gas. The degree of isotopic fractionation from the initial ammonia were +2.4~+40.3‰ for montmorillonite and -1.6~44.4‰ for saponite as Δ^{15}N value. There was also a negative correlation observed between the adsorbed ratio (wt. %) and the δ^{15}N value. When the glass vial was vacuumed for only 1 hour, the δ^{15}N of the remaining ammonia increased (+64.1‰ for montmorillonite and +60.1‰ for saponite as Δ^{15}N value). However, when the glass vial was vacuumed more than 1 hour, selective removal of $^{15}NH_3$ was observed. The water content of the sample was not an influential factor throughout the experiments. The results suggest that adsorption and desorption of ammonia on clay minerals causes significant nitrogen isotopic fractionation (Δ^{15}N=−1.6~+64.1‰) and could be a potential mechanism for ^{15}N-enrichment in interstellar environments.

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Keywords: nitrogen isotopic fractionation, adsorption, ammonia, molecular clouds, grain surface chemistry

星間氷を模擬した光化学反応実験生成物のアミノ酸分析
Amino acid analysis of photochemically processed interstellar ice analogues

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Amino acids are one of major constituent of living organisms on the Earth, so they are essential molecules for the origins of life. Many scenarios for the endogenous production of amino acids on the early Earth have been proposed [e.g., 1, 2]. On the one hand, the extraterrestrial delivery of them is also considered as a likely candidate [e.g., 3, 4]. Carbonaceous chondrites are known to contain abundant amino acids up to 250 ppm [5] and more than 80 species were identified [6]. In addition, the simplest amino acid: glycine was also detected in the cometary grains returned by stardust mission [7]. The extraterrestrial amino acids are considered to have been formed by aqueous alteration on the meteoritic parent bodies or in the presolar interstellar environments. Although, the formation process, as well as the place to be taken, is still on a debate, the existence of amino acid in comets as wall as the isotopic signatures of them suggest that at least some amino acids and the precursors of them are interstellar origin [8].

In the study, we conducted laboratory experiments to synthesize interstellar ice analogues from typical interstellar gases and analyzed their amino acid composition. For the experiments that simulate the condition in the interstellar medium (ISM), we utilized an apparatus called PICACHU: Photochemistry in Interstellar Cloud for Astro-Chronicle in Hokkaido University. In this PICACHU apparatus, the typical ISM gases (H_2O: CH_3OH: NH_3 = 2:1:1) were introduced into the chamber and deposited onto the surface of a sappier glass there, which were refrigerated at ~12 K. During the deposition of the ice, UV photons were continuously irradiated under highly vacuumed condition for 71 hours, because photochemical reactions in the ice, which were processed by stellar ultraviolet (UV) photons and cosmic rays, are important for the formation of complex organic molecules in ISM. In addition, we made two ice samples and one of them was further irradiated by UV for 232.5 hours after heated to room temperature in order to simulate subsequent decomposition in warmer environments. Then, the organic residues of the two ice samples were extracted by methanol and analyzed by GC-MS and GC/NPD after the derivatization for GC analysis.

The results showed that 11 species of amino acids were formed in the interstellar ice analogues, which were irradiated only at ~12 K. The most abundant amino acid was glycine and the second most abundant one were β-alanine and alanine. Their abundances are about fourth of that of glycine. The abundance of each amino acid generally decreased with the increase of the carbon number of the amino acids. This trend was consistent with the previous study [9]. Then, the further irradiated sample showed the general decrease in the abundances of amino acids, but some of them were still remained. The survived ratio differed among amino acids and glycine was the most resistant to the irradiation (~80% remained.). These results indicate that amino acids can be formed by the UV irradiation to the interstellar ice and can survive subsequent irradiation in the warmer environments.

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