

プレソーラーアルミナ粒子の内部構造分析による形成・変成履歴の推定 Crystal structure, morphology, and isotopic compositions of presolar alumina grains in unequibrated ordinary chondrites

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Corundum, the thermodynamically stable phase of Al_2O_3 , is predicted to be the most abundant refractory dust species condensed in envelopes around oxygen-rich asymptotic giant branch (AGB) stars. Many presolar Al_2O_3 grains, which are the survival circumstellar dust grains, have been identified from acid-residues of chondrites. The grain morphology and crystal structure of presolar grains may reflect condensation conditions in circumstellar envelopes of AGB stars and processing in the interstellar medium (ISM) and protosolar disk.

Using scanning electron microscopy (SEM) we obtained detailed secondary electron images, energy dispersive X-ray spectroscopy (EDS), electron backscattered diffraction (EBSD) patterns, and cathodoluminescence (CL) spectra of each Al_2O_3 grain prior to isotopic measurements. Focused ion beam (FIB) lift-out sections were made from the identified presolar grains and the interior structures were observed with a transmission electron microscope (TEM).

The Al_2O_3 grains were identified from acid residues of QUE97008 (LL3.05) by EDS and observed in detail by field emission (FE) SEM at the Carnegie Institution of Washington (CIW). Previously identified alumina grains from Semarkona (LL3.0), Roosevelt County 075 (H3.1), and Bishunpur (LL3.15) were also used in this study. CL spectra were obtained with a FE-SEM equipped with a Gatan Mono CL4 system at NIST. EBSD analysis was performed with an FEI Nova 600 FIB-SEM equipped with an HKL EBSD system at the Naval Research Laboratory (NRL). Isotope measurements were performed with the Cameca NanoSIMS 50L ion-microprobe at CIW. Oxygen isotopes of 163 grains were measured using ~ 100 nm Cs^+ beam rastered over each of the grains. An O^- beam was used to measure the Mg-Al isotopic compositions of the presolar and some solar Al_2O_3 grains. Ultra-thin sections of presolar grains QUE053, 060, and 067 were prepared with the NRL FIB-SEM. TEM studies were carried out at NRL with a JEOL 2200FS field-emission scanning transmission electron microscope (STEM).

Eight presolar grains from QUE97008 and one from RC 075 were newly found. Grain QUE060 is classified into Group 2 and has a subhedral shape with clear flat facets. The surface is smooth except for a face with a cavity. TEM diffraction patterns of the FIB section indicated that the grain consists of multiple corundum crystallites. Dark-field TEM image showed large (>100 nm) and small (<30 nm) scale orientation variation. The large-scale misorientation observed on the right side of the grain seems to relate to the cavity. Small-scale distortions occur uniformly within the grain. EDS spectra showed that the Mg/Al ratio of QUE060 is ~ 0.01 , and the NanoSIMS measurement revealed this high Mg content to be essentially pure radiogenic ^{26}Mg , with inferred initial $^{26}\text{Al}/^{27}\text{Al} \sim 0.01$, similar to other Group 2 grains.

QUE067 is a thin Group 4 grain with very irregular morphology. Its $^{27}\text{Al}/^{24}\text{Mg}$ ratio was three times lower than in QUE060, but its inferred $^{26}\text{Al}/^{27}\text{Al}$ ratio was similar. No EBSD patterns of crystals were obtained from the grain surface but TEM observation on the FIB section showed that the interior of QUE067 was corundum, not amorphous.

The subhedral shape and smooth surface of QUE060 suggest that this grain was likely single crystalline corundum when it condensed in a circumstellar envelope of a low-mass AGB star, and that the polycrystalline nature, voids and distorted crystal structure inside the grain are secondary features.

A possible process to form large-scale misorientation and the cavity is grain-grain collisions in a SN shock in the ISM. A high velocity collision creates a shockwave propagating inside the grain, finally forming a crater. Small-scaled distortions may have also formed by collisions with small particles in the ISM. Such collisions are less destructive than with larger grains, but their probability is high. Ion bombardment in the ISM may also contribute to the small-scale distortions.

キーワード: ダスト, 初期太陽系, コンドライト, プレソーラー粒子, 透過型電子顕微鏡, 晩期型巨星

Keywords: dust, early solar system, chondrite, presolar grain, transmission electron microscopy, evolved star

電子線トモグラフィ法による GEMS の 3次元構造の観察 3-D observation of GEMS by electron tomography

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彗星起源と考えられる無水惑星間塵 (CP-IDPs) には GEMS (Glass with Embedded Metal and Sulfides) と呼ばれる微小(数 100 nm) 粒子が多量に含まれている。GEMS は非晶質珪酸塩中に複数の Fe-Ni 金属、硫化物ナノ包有物 (10-50 nm) を有しており、太陽系の最も始原的な物質の一つと考えられている。また GEMS は、その赤外スペクトルの特徴が星間塵や星周塵のものとよく合致するサブミクロンサイズのダストであることから、いわゆる”astronomical silicate” の候補物質でもある [1]。このように惑星物質学上も天文学上も注目されている物質であるが、どこでどのように GEMS が生まれたのかは議論中である [2-5]。

Bradley らは、GEMS は原始太陽系円盤での様々な変成を免れた星間塵であり、結晶が宇宙線などの照射により非晶質化して形成したと主張している [2,4]。その根拠の一つとして透過型電子顕微鏡 (TEM) を用いた観察結果で、GEMS の中に 10nm 程度の relict grain と呼ばれる結晶が存在しその外形が GEMS そのものの外形と仮晶関係にある、というのがある [2]。しかし、そもそも relict grain がこの報告以外では観察されないこと [3] や、2次元の観察での外形評価は不定性が大きいことなどの疑問も残っている。

一方 Keller と Messenger は、GEMS は高温ガスの凝縮物であるというモデルを提案している [3,5]。ほとんどの GEMS に酸素同位体異常がみられない、元素存在度が粒子毎にばらつきが大きい、粒子表面にのみ硫化鉄が存在するという観察事実に基づき、GEMS は原始惑星円盤において高温ガスから金属鉄を含む非晶質珪酸塩が凝縮し、冷却するにつれて凝縮粒子表面の金属鉄が硫化したことにより形成した、と説明している。しかしこのグループの観察においても TEM による 2次元像観察を行なうため、真に硫化鉄が GEMS 表面のみに存在しているのか証明できていない。

GEMS の形成起源を明らかにするため、本研究では、これまで 2次元での観察に留まっていた GEMS の組織観察を、TEM を用いた電子線トモグラフィにより 3次元で行なうことを試みた。これにより、3次元構造が得られるだけでなく、従来の 2次元観察では試料の厚み (>50 nm) により制限を受けていた空間分解能が、透過像の画素サイズの数倍程度まで上げることが可能であり、nm オーダーでの不均一性をもつ GEMS のようなサンプルにとって極めて有効な観察手段である。観察には cluster CP-IDPs である、L2036AA5 cluster4, L2009O8 cluster13, W7262A2 の 3 試料を用いた。このうち W7262A2 はポリウレタンを用いて捕獲した IDP であり、従来の捕獲法で使用されるシリコンオイルによる汚染を受けないという特徴をもつ試料である [6]。

IDP 試料を樹脂に埋めミクロトームにより超薄切片を作成した。GEMS の平均粒径が約 180 nm であるため、切片厚みは 50-300 nm とした。電子線トモグラフィは京都大学の TEM (JEM-2100F, JEOL) を用いて行い、約 ± 65° 回転させながら 1° おきを取得した透過像を再構成し一連の断層像を得た。なお透過像は、明視野透過顕微鏡法 (BF-TEM)、高角散乱環状暗視野走査透過顕微鏡法 (HAADF-STEM) 法により取得した。

断層像中に含まれる鉱物相の推定を、STEM-EDS (エネルギー分散型 X 線分析法) で取得した 2次元元素分布像と比較することにより行なった結果、金属鉄は珪酸塩に含まれる事、硫化鉄は主に GEMS の表面に存在する事を確認した。このことは GEMS が凝縮物であることを示唆するモデル [3,5] と調和的な結果だが、今後サンプル数を増やして統計的に検討する必要がある。なお電子線トモグラフィを惑星間塵へ応用し断層像の再構成に成功したのは、今回の研究が世界初である。このことは今後の惑星物質科学の分析的研究上も意義深いといえる。

[1] Bradley et al. (1999) Science, 285, 1716

[2] Bradley and Dai (2004) ApJ, 617, 650

[3] Keller and Messenger (2011) GCA, 75, 5336

[4] Bradley (2013) GCA, 107, 336

[5] Keller and Messenger (2013) GCA, 107, 341

[6] Messenger et al. (2012) 43rd LPSC, 2696 (abstr.)?

キーワード: 惑星間塵, GEMS, 透過型電子顕微鏡, トモグラフィ

Keywords: IDP, GEMS, TEM, tomography

新たに開発した手法によるコンドリュールメルトへのシリカに富むガスの凝縮実験：
急速な低Ca輝石の析出
Condensation experiments of Si-rich gas into the chondrule melt for rapid low-Ca pyroxene formation using a new furnace

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はじめに

低Ca輝石(主としてエンスタタイト)とその多形は、太陽系岩石圏を構成する最も主要な相である。原始太陽系星雲における低Ca輝石の形成機構には不明な点が多い。低Ca輝石は、その凝縮過程でMg珪酸塩として先に晶出するフォルステライトとは反応関係にあり、フォルステライトとシリカに富むガスとの反応によりエンスタタイトが包晶反応として形成することが熱力学計算により予測されている(Grossman, 1972)。しかし、その反応は生成層(エンスタタイト)中の固体内拡散が律速するために遅く、最大分別凝縮で近似でき、また、ガスから直接するエンスタタイトは微量である(Imae et al., 1993)。Tissandier et al. (2002)によりコンドリュールメルトへシリカに富むガスが相互作用してピジョン輝石(CaO 5-7wt%)が析出することを実験的に示した。本実験的研究では、原始太陽系星雲環境を再現する新たな実験法を開発し、この装置を用いて短時間で多量の低Ca輝石(CaO 1wt%)を析出させることに成功したので報告する。

実験

全圧は水素ガス導入下で主として100Paに設定し、排気系にバタフライバルブを用い、隔膜真空系を用いて制御した。各実験の最高温度は輝石の安定な1200-1450°Cとし、冷却速度は主として約100°C/hとした。実験出発物質としてAllende隕石(CV3)の小片(1回の実験に~30-50mg)を用いた。上部に1mmの穴の開いたアルミナのるつぼ内には出発物質とシリカガス源としてシリカの粉末を試料と直接触れないように入れた。また対照実験としてシリカ粉末のない条件でも行った。

結果

シリカ粉末のない条件では微量の輝石が認められることがあるが、かんらん石が支配的に析出した。一方、シリカに富むガスのある条件では、顕著な変化が認められた。すなわち、熔融したAllende試料の1450°Cでは試料全体が低Ca輝石に変化しており、かんらん石をポイキリティックに取り囲んで成長していた。一方、1350°Cおよび1250°Cと最高到達温度がより低温になるにつれ晶出するかんらん石および輝石は鉄成分に富み、縁に主として低Ca輝石がより多く析出した。

議論

出発物質へのシリカに富むガスの衝突頻度は大規模なバルクの組成変化を引き起こすほど高くない。シリカに富むガス源が存在しない実験温度条件は、輝石の安定温度域であるので、準安定に存在したかんらん石がメルトと共存していたことを示唆する。シリカに富むガス源がある実験では、シリカに富むガスが輝石の析出を助けたと考えられる。

文献

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Tissandier L., et al. (2002) MAPS 37, 1377.

キーワード: 原始太陽系星雲, 凝縮, 低Ca輝石, シリカに富むガス, 低圧実験, コンドリュール

Keywords: primordial solar nebula, condensation, low-Ca pyroxene, Si-rich gas, low-pressure experiments, chondrule

原始惑星系円盤におけるアモルファスフォルステライト粒子表面での含水鉱物形成 Hydration of amorphous forsterite grains in protoplanetary disks

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Hydrous silicate dust can be thermodynamically stable at low temperatures ($\sim 200\text{K}$) in protoplanetary disks with the solar-system abundance of elements. Theoretical and experimental investigation (Fegley and Prinn, 1989; Imae et al., 1999) have shown that the hydration reaction between crystalline Mg-silicates and water vapor is too sluggish to occur during a lifetime of protoplanetary disks. However, infrared spectroscopic observation and investigation of extraterrestrial materials have shown that both crystalline and amorphous Mg-silicates may be present in protoplanetary disks and in the protosolar disk. Amorphous silicates are thermodynamically unstable, and thus could be hydrated more rapidly at higher temperatures than crystalline silicates (Nagahara and Ozawa, 2011). If hydration of amorphous silicates occurs within a disk lifetime, hydrated dust could be a source of water to terrestrial planets.

In this study, in order to investigate hydration of amorphous Mg-silicates, we conducted closed-system reaction experiments of amorphous forsterite and water vapor in sealed glass tubes at temperatures of 1023-423K and $P_{\text{H}_2\text{O}}$ of 0.05 -50 bar for 2-1344 hours. Run products were analyzed with FT-IR and XRD. We found that samples reacted at temperatures below 723K and $P_{\text{H}_2\text{O}}$ of 5-50 bar showed sharp $3\mu\text{m}$ absorption features, clearly suggesting the formation of hydrous phase(s). XRD analyses of some run products showed that the hydrous phase was serpentine. Because the stable temperature of hydrous phase(s) at $P_{\text{H}_2\text{O}}$ of 5-50 bar is $\sim 523\text{K}$, the present results indicate that hydrous Mg-silicates can be formed metastably from amorphous forsterite at higher temperatures than the thermodynamic prediction. Moreover, hydration of amorphous forsterite occurred after 2-hour heating at 623K and $P_{\text{H}_2\text{O}}$ of 5 and 50 bar, implying that the activation energy for hydration of amorphous forsterite is much smaller than that for crystalline forsterite.

Hydration reaction seems to have little $P_{\text{H}_2\text{O}}$ dependence at $P_{\text{H}_2\text{O}}$ of 5-50 bar, indicating that hydration is controlled by a reaction between amorphous forsterite with saturated adsorbing water molecules. No hydration occurred, however, at 523K and $P_{\text{H}_2\text{O}} < 1$ bar, which could be due to less effective adsorption of water molecules at low-pressure conditions.

キーワード: 原始惑星系円盤, マグネシウムケイ酸塩, 結晶質, 非晶質, フォルステライト, 含水鉱物
Keywords: protoplanetary disk, Mg-silicate, crystalline, amorphous, forsterite, hydrous mineral

鉄ダストの均質核生成時の付着確率 Sticking probability for homogeneous nucleation of iron dust

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Nucleation theories have been used to understand the condensation sequence, number density and size of cosmic dust in a gas outflow of dying stars or a gas plume after shock wave heating in the primitive solar nebula. However, it has been well known that nucleation rates obtained by nucleation theories and by experiments have a large difference. We believe that the reason is uncertainties of the physical parameters of nanometer sized particles. Therefore, it is still not successful to explain the characters of cosmic dust by a nucleation theory. To determine the physical parameters of nanoparticles and evaluate nucleation theories, we constructed an in-situ observation system of temperature and concentration during homogeneous nucleation in vapor phase using interferometry for both of ground based and microgravity experiments.

Nanoparticles are formed from a supercooled vapor after evaporation by electrical heating in a controlled gas atmosphere. Using the new system in lab, we succeeded to determine surface free energy and sticking probability of manganese nanoparticle from timescale for gas cooling and condensation temperature based on nucleation theories [1]. In this laboratory experiment, convection of gas atmosphere caused by thermal heating generates heterogeneity of nucleation environment, such as temperature and concentration profiles around evaporation source. If same kinds of experiments are performed in microgravity, evaporated vapor defuses uniformly and the temperature profile becomes concentric around the evaporation source. As the result, nucleation will occur at concentric position. Then, we can obtain physical properties with relatively smaller error bars and then we may be able to evaluate nucleation theories more precisely. Therefore, we also performed a microgravity experiments using an aircraft and the sounding rocket S-520-28 launched on December 17th, 2012.

We prepared specially designed Mach-Zehnder-type interferometers with an evaporation chamber and camera recording systems to fit the space and weight limitations of the rocket. Three systems, named DUST 1 to 3, with same configuration except evaporation source and gas pressure in the chamber were installed into the nosecone of the rocket. The evaporation source and gas atmosphere were tungsten and gas mixture of oxygen (4000 Pa) and argon (36000 Pa) for DUST 1, iron and argon (20000 Pa) for DUST 2, and iron and argon (40000 Pa) for DUST 3. The experiments were run sequentially and automatically started from 100 s after launch of the rocket. The evaporation source of iron was electrically heated under microgravity. Evaporated iron vapor was diffused, cooled and condensed in the gas atmosphere. The temperature and concentration at the nucleation site are determined from the movement of the fringe in the interferogram. Here, we will show the results of the homogeneous nucleation and determine the sticking probability of iron atoms into a nanoparticle based on nucleation theories. The results will be compared with that by ground based experiment.

[1] Y. Kimura, K. K. Tanaka, H. Miura, K. Tsukamoto, Direct observation of the homogeneous nucleation of manganese in the vapor phase and determination of surface free energy and sticking coefficient, *Crystal Growth & Design*, **12** (2012) 3278-3284.

キーワード: 核生成, 観測ロケット, 干渉計, その場観察, ダスト

Keywords: Nucleation, Sounding Rocket, Interferometer, In-situ Observation, Cosmic Dust

イオン誘起微粒子核生成I：装置開発とねらい Ion-induce nucleation experiment I: development of a new apparatus

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Mechanisms of grain nucleation have attracted researchers in various fields of science in connection with e.g. atmospheric aerosols and cosmic dust grains. Although there have been many theoretical and experimental works approaching this issue, the details of nucleation mechanism is still in debate. Most of works are performed assuming homogeneous nucleation in gas phase or heterogeneous nucleation on the bulk surfaces. The homogeneous nucleation often suffers from a "critical size" of particle and requires high supersaturation condition to gain the efficient formation rate, while the nucleation on the bulk surface may not be relevant to the first stage of grain formation in realistic environments. It is known that ion-induced heterogeneous nucleation would play an important role in the particle formation because in this mechanism ion-neutral interaction overcomes difficulties expected in neutral-gas-phase homogeneous nucleation. We recently developed a new experimental apparatus to investigate the elementary process of the ion-induced nucleation. We present the importance of ion-induced nucleation and advantages of the newly developed apparatus. Using this apparatus, we have been successful in obtaining an important physical parameter, free energy, of water cluster ions.

キーワード: 微粒子, 核生成, クラスターイオン

Keywords: cluster ion, ion-induced nucleation

すばる望遠鏡によるアイソン彗星の中間赤外線観測 Mid-infrared Observation of sungrazing comet C/2012 S1 (ISON) with Subaru+COMCIS

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Comets are the frozen reservoirs of the early solar nebula and are made of ice and dust. Dust grains in comets have been used to investigate the formation conditions of the solar system. A silicate feature is often observed in comet spectra in mid-infrared region, for example 11.3-micron, and may be used for probing early history of the solar system. In most cases the feature shows the existence of crystalline silicate together with amorphous silicate. Since the crystalline silicate grains are generally made through high-temperature annealing above 800K from amorphous ones, it is believed that the crystalline silicate grains produced at the inner part of the disk were transported to the outer cold regions where comet nuclei formed.

Comet C/2012 S1 (ISON) is a long-period Oort cloud comet, discovered in September 2012. In particular, comet ISON is a sungrazing comet, which is predicted to pass close by the Sun and Earth and becoming a bright object. Mid-infrared observations of this new comet and investigation of the 10-micron silicate feature help us to understand the formation of crystalline silicate grains in the early solar nebula.

We observed comet ISON in mid-infrared wavelength region using Cooled Mid-Infrared Camera and Spectrometer (COMICS) mounted on the Subaru Telescope on Mauna Kea, Hawaii. The observation of comet ISON was carried out on 2013 October 19th and 21st UT. Since the weather condition was not so good when we observed, we carried out N-band imaging observations (8.8 and 12.4 micron) and N-band low-resolution spectroscopy. The spectrum of C/ISON can be fit with 260–265 K blackbody spectrum when we use the 7.8–8.2 and 12.4–13.0 micron region as the continuum. The spectrum has only a weak silicate excess feature, which may be able to attribute to small amorphous olivine grains. We could not detect a clear crystalline silicate feature in the spectrum. We will compare the spectrum with other Oort cloud comets, such as comets C/2011 L4 (PanSTARRS) and C/2013 R1 (Lovejoy), and discuss the dust properties and the birthplace of the comet C/ISON.

Keywords: comet, dust, silicate, infrared

極低温原子間力顕微鏡によるアモルファス氷の表面構造観察 Observation of surface structure of amorphous solid water by atomic force microscope at low temperatures

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Amorphous solid water (ASW) is one of the important materials in space because it exists an abundant and effects to evolution of interstellar molecules. In molecular clouds, it is well known that chemical reactions on icy interstellar dust grains which consist of ASW mantle and mineral particle core are key processes in the formations of important organic molecules (ie. H₂CO, CH₃OH) and deuterium-enriched molecules. Although ASW surface play an important role as a field of chemical reactions, the surface structure of ASW has yet to be revealed.

Recently, we developed a low temperature atomic force microscope (AFM) for study of the surface structure of ASW. AFM is a powerful tool to study the surface structure of ASW because it can work even if the surface do not have a conductive property. In this presentation, we show AFM images of ASW surfaces which were formed at several conditions. ASW were formed on Si(111) 7×7 at 103-135 K with various deposition rate (0.08-0.8 nm/min) and various thickness(2.5-22 nm). From the observations of surface structure, we discuss the relation between the surface structure and the condition of ASW formation.

キーワード: アモルファス氷, 星間塵, 分子雲, 原子間力顕微鏡法
Keywords: amorphous ice, interstellar dust, molecular clouds, atomic force microscopy

模擬氷星間塵への真空紫外光照射による有機分子生成実験 Experimental approach to the formation of organic molecules following vacuum-ultraviolet irradiation of interstellar ice

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Cosmic gases and dust grains ejected from dying stars gradually assemble under the influence of gravity to form interstellar clouds. Among these gases, heavy elements such as magnesium (Mg) and silicon (Si) are incorporated in dust. Lighter and chemically active elements (e.g., hydrogen, carbon, oxygen, and nitrogen; H, C, O, N, respectively) play important roles in the chemistry of interstellar clouds. After the temperature and photon field decrease when the density of dust particles increases in interstellar clouds, atoms (e.g., H, O, C, N) and molecules (e.g., CO) deposit onto the dust surfaces. Cold-surface reactions proceed on the grain surface, and an ice mantle, which is predominantly composed of H₂O combined with other molecules such as CO, CO₂, NH₃, CH₄, H₂CO, and CH₃OH, is formed.

The ice mantles are also subjected to substantial energetic processing by the prevailing ultraviolet radiation during the lifetime of an interstellar cloud. Followed by repeated processing when cycling between diffused clouds and dense clouds, new refractory organic molecules are formed in the ice mantles. The ice mantles undergo further photon radiations upon the formation of protoplanetary disks, and finally evolve to non-volatile complex organic residues by irradiation and thermal processing. However, the detail of the chemical evolution of the organic molecules has still been ambiguous. Although the previous laboratory studies using infrared spectroscopy can provide the presence of polar compounds such as amines, carboxylic acid or amides functions, it often suffers from the low sensitivity and the difficulty to obtain precise identifications of molecular species due to the overlapping of broad solid-state bands. Since dust grains and ice mantles are the precursors of planetary material, studying the photoprocesses is essential to understanding the origin of our solar system, and more powerful analytical techniques are required to unveil rich chemistry of the ices in interstellar clouds and protoplanetary disks.

Here, we are going to present a talk about a new apparatus which is now under construction to shed light on the chemical evolution of organic molecules in interstellar clouds and protoplanetary disks. The apparatus consists of three basic parts, i.e., a vacuum system, a copper-substrate equipped with a closed cycle helium refrigerator, and a vacuum ultraviolet source. Multi-component interstellar ice analogues are created on the cold (10 K) substrate by vapor deposition, and subjected to irradiation by the ultraviolet. The irradiated ice is subsequently heated up to 800 K. The gas composition desorbed from the ice during heating is analyzed by a high-resolution quadrupole mass spectrometer in the vacuum chamber. The survived organic residue from heating are studied using gas chromatography coupled to mass spectrometry (GC-MS) and high performance liquid chromatography coupled to mass spectrometry (HPLC-MS).

キーワード: 分子雲, 原始惑星系星雲, 氷星間塵, 有機物

Keywords: interstellar cloud, protoplanetary disk, ice mantles, complex organic molecules

固体表面における C_2H_2 分子水素付加反応：彗星 C_2H_6 生成の解明に向けて Hydrogen addition reactions of C_2H_2 on cold grains; clue to the formation mechanism of cometary C_2H_6

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Volatiles incorporated into comets were formed in the pre-solar molecular cloud and probably chemically altered in the proto-planetary disk of the Sun. Although physico-chemical evolution from a molecular cloud to the disk is basically understood, detailed evolutionary processes are still in debate; e.g., the fraction of the materials originated in the molecular cloud incorporated into the disk without physico-chemical alterations (some fraction of materials might sublime via accretion shock) and physical conditions (temperature, densities of materials, etc.). To reveal those links, we focused on the molecules formed through grain surface reactions, which occurred under quite low temperature conditions like 10K. We discuss the origin of such molecules in comets (icy small body of the Solar system), which might preserve the information about chemical and physical conditions of proto-planetary disk. Cometary ethane (C_2H_6) and acetylene (C_2H_2) have been observed in multiple comets since 1996 and their abundances relative to H_2O (the major component of cometary ices) is $\sim 10^{-3}$ but with variations. This variation might be caused by the difference in the mixing ratios between the materials originated in the molecular cloud and the disk-processed materials. C_2H_6 has never been detected in the molecular cloud and the formation mechanism of C_2H_6 detected in comets is still in debate. One of the candidates of formation reactions of C_2H_6 is the hydrogen addition reaction of C_2H_2 on the cold grain surface ($C_2H_2 \rightarrow C_2H_3 \rightarrow C_2H_4 \rightarrow C_2H_5 \rightarrow C_2H_6$). In the previous experimental studies, those reactions were evaluated qualitatively and it was concluded that the reaction from C_2H_4 to C_2H_6 occurred more rapidly than the reactions from C_2H_2 to C_2H_4 and it would be a reason for the nondetection of C_2H_4 . To investigate these reactions more quantitatively in realistic conditions for molecular clouds, we performed the laboratory measurements of hydrogen addition reactions of C_2H_2 and C_2H_4 on the amorphous solid water (ASW), respectively.

The experiments were conducted by using laboratory setup for surface reaction in interstellar environment (LASSIE) at the institute of low temperature science, Hokkaido University³. A cryogenic aluminum substrate is located in the center of the main chamber and surrounded by a large copper shroud connected to a liquid-nitrogen reservoir. Atomic hydrogen used for the reactions were produced by the dissociation of H_2 molecules in microwave-induced plasma. The kinetic temperature of hydrogen atoms were ~ 120 K and the H-atom flux was $\sim 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$. The samples of pure solid C_2H_2 , C_2H_4 , and those on ASW were produced on the substrate at 10, 15 and 20K. Infrared absorption spectra of the ices were measured by FTIR before and during the exposure of H-atom.

Our measurements show basically the same trend as shown in the previous studies. We will discuss the temperature and thickness dependence of the time constant for the sample ices in the poster.

キーワード: 分子生成, 固体表面反応, 星間物質

Keywords: molecular formation, grain surface chemistry, Inter Stellar Medium

可視光応答性を持つ星間分子の実験室生成とその高分解能分光 High Resolution Spectroscopy of Laboratory-Produced Interstellar Molecule having Response to Visible Light

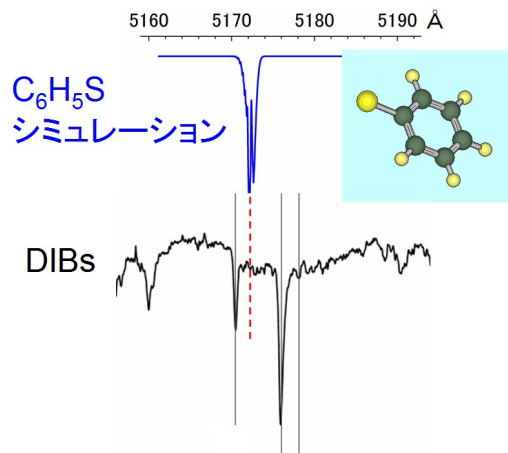
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地球と星の間の星間空間には光を遮断する希薄な分子雲が存在し、その分子雲の物質による可視光領域の吸収線 Diffuse Interstellar Bands (DIBs) が観測されている。これらは未同定の星間分子である。その起源となっている分子は、地球外生物の元となりうる地球外有機物であり、宇宙の分子進化のひとつの段階を示すものと考えられている。DIBs の候補であるためには、可視光領域に吸収線を持たなければならない。そこで、多環芳香族のラジカルが有力視されている。これらを同定するため、放電による星間分子の生成と測定を行っている。生成にはホロカソード放電を用いており、測定には、Cavity Ringdown 分光器と放電発光分光器を用いている。近年我々は、Cavity Ringdown 分光器により、フェニルチオラジカル (C_6H_5S) の電子遷移の高分解能測定を行った。また、放電発光分光器により、フェノキシラジカル (C_6H_5O) の電子遷移の測定も行った。これらの測定結果を用いて、星間空間で観測された DIBs スペクトルとの比較検討を行った。

キーワード: ぼやけた星間線, 星間分子, 分光, キャビティリングダウン, 分子雲, 放電

Keywords: Diffuse Interstellar Band, interstellar molecule, spectroscopy, cavity ringdown, molecular cloud, discharge



Murchison 隕石中の極性有機化合物の多様性 Variation of organic compounds in the polar extract of Murchison meteorite

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Various organic compounds have been found in carbonaceous chondrites, in which water-soluble bio-related organic compounds such as amino acids and carboxylic acids are fully studied mainly because of their great interests to origins of life in the universe and standards available for the analyses. The concentrations of these acids increase significantly after acid hydrolysis of the water extract. Even though a few studies attempted to identify the chemical structures of their precursors (Cooper and Cronin, 1995), the original structures remain largely unclear. The aqueous activity on the meteorite parent body also could proceed in a chemical oxidation (i.e. incorporation of oxygen by hydrous reaction), resulting in the alteration of original organic matter (Oba and Naraoka, 2009). The large abundance of carboxylic acids in the water extract may be attributable to such oxidation processes.

In contrast to the water extract, less polar methanol extract of chondrites has not been characterized well in spite of relatively high content of organic matter with the D- and ¹⁵N enrichment. Recently, ultrahigh-resolution mass spectral analysis on various solvent extracts of the Murchison meteorite (CM2) was performed by electrospray ionization (ESI) using Fourier transform-ion cyclotron resonance/mass spectrometry (FT-ICR/MS) to reveal significant chemical diversity to tens of thousands of different mass peaks having CHO, CHOS, CHNO and CHNOS elemental compositions (Schmitt-Kopplin et al., 2010). With the assumption for molecular formulae calculation and no chromatographic separation, however, the detailed chemical structures of the compounds cannot be determined. In particular, the organic compounds with their elemental compositions of CH and CHN were not discussed. In this study, we performed high-performance liquid chromatography/high resolution mass spectrometry (HPLC/HRMS) analysis of organic compounds in polar solvent extracts of the Murchison meteorite.

Homologous series of alkylpyridines have been identified in the polar solvent extracts of the Murchison meteorite by liquid chromatography/high-resolution mass spectrometry. The wide range of saturated- and unsaturated-alkylated (C1 to C21) pyridines is more diverse relative to that previously found, which could be produced by aldehydes and NH₃ through aldol condensation and Chichibabin-type synthesis on the meteorite parent body. This finding implies a high aldehyde activity under an alkaline condition with ammonia for the chemical evolution of organic matters in carbonaceous meteorites. In addition to the compound distribution of alkylpyridines, the occurrence of other compounds in the water extract will be also discussed with respect to their formation mechanisms in carbonaceous meteorites.

Keywords: polar organic compounds, carbonaceous meteorite, Murchison, high-resolution mass spectrometry

衝撃波による氷ダストの加熱と蒸発

Heating and evaporation of icy dust in transient heating events induced by nebular shocks

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分子雲から原始惑星系円盤へと至る際の化学進化において、固体氷は重要な役割を担う。固体氷表面ではCO分子へのH原子逐次付加反応によりホルムアルデヒドやメタノールなどの簡単な有機分子が形成する(Watanabe & Kouchi 2002)。さらにH-D置換反応により、これらの分子の重水素体が効率的に形成される(Nagaoka et al. 2005)。紫外線照射を受けた多成分氷を昇温させると、蒸発残渣としてアミノ酸などの複雑な有機分子が形成される(Munoz Caro et al. 2002)。固体氷が蒸発すると、中に閉じ込められていた多様な分子が放出されることでガスの化学組成が変化する。固体氷が経験する熱履歴は上記の化学進化に大きく影響するため、固体氷の熱進化を明らかにすることは重要である。

本研究では、円盤ガス中に生じる衝撃波による固体氷の過渡的加熱現象に着目した。ガス円盤では、分子雲からのガス降着やガス円盤内での惑星形成に伴い、様々な条件で衝撃波が生じる。固体氷が周囲のガスと共に衝撃波面を通過すると、周囲のガスが瞬間的に速度変化する一方、固体氷は慣性のために通過前の速度を維持するため、両者の間に大きな相対速度が生じる。ガス分子が高速衝突することで固体氷が加熱され、同時に蒸発が生じる。相対速度は時間と共に急激に減少するため、衝撃波による加熱は継続時間が極めて短い過渡的な現象である。このとき、固体氷が経験する熱履歴や蒸発率を、様々な衝撃波条件に対して系統的に調べた。

モデル：衝撃波後面のガスの温度や密度は一定だと仮定した。簡単のため、固体氷の形状は球であるとし、単一組成(H₂O or CO)からなるとした。また、固体氷の熱慣性は小さく、温度変化のタイムスケールは速度変化のタイムスケールに対して充分小さいと仮定した。以上の仮定の下では、個々の固体氷の蒸発率(初期半径に対してどの程度半径が減じるかの比)は、衝撃波後面のガスと固体氷の相対速度の初期値 v_0 と、衝撃波後面のガス密度 ρ_g のみに依存する。ある与えられた v_0 と ρ_g に対して、固体氷の運動方程式と蒸発に伴う半径変化の式を数値計算し、固体氷が経験する最高温度と蒸発率を求めた。

結果：定性的には、 v_0 と ρ_g が大きいくほど、固体氷は高温を経験し、蒸発率も大きくなる。その上で、以下のことが分かった。(i) v_0 と ρ_g を大きくしても、完全蒸発はなかなか起こらない。これは、蒸発率が激しくなると半径が小さくなり、ガスとの相対速度がすぐに減少することで加熱継続時間が抑えられるというnegative feedbackが作用するからである。(ii) 蒸発率が1%を上回るための臨界の $v_{0,c}$ が存在する。臨界速度は、純H₂O氷の場合は約3 km/s、純CO氷の場合は約1 km/sである。いくら ρ_g を大きくしても、 v_0 がこの臨界速度以下であればほとんど蒸発は生じない。(iii) 固体氷は、平衡状態であれば揮発してしまう温度(固気平衡温度)を上回る最高温度を経験し得る。これは、 ρ_g が大きいか場合は加熱継続時間が極めて短いため、完全に気化する前に加熱が終わるからである。

考察と結論：衝撃波の発生に伴う固体氷の熱履歴や蒸発率を、広い衝撃波条件にわたって系統的に明らかにした。これは、分子雲やガス円盤の天文観測で検出された化学組成の変化を、固体氷の蒸発現象と関連付ける上で有用であろう。固体氷が昇華温度を大きく上回る温度を経験することで、ガス円盤内においてアミノ酸などの複雑な有機分子が形成するかも知れない。高温では固体氷内でHとDの交換反応が期待できるため、これに伴う重水素濃集効果も興味深い。今後は、固体氷の過渡的加熱現象に伴う上記の過程の詳細について検討を進める予定である。

キーワード: 氷ダスト, 衝撃波加熱, 蒸発, 化学進化, 原始星円盤, 原始惑星系円盤

Keywords: icy dust, shock heating, evaporation, chemical evolution, protostellar disk, protoplanetary disk