Mechanism determining dust content in galaxies

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The origin of dust in galaxies is a fundamental issue in planetary science. In this talk, I will introduce the mechanism determining dust content in galaxies based on the "galactic chemical evolution model" which describes material circulation among four components of stars, interstellar medium, metals (elements heavier than helium), and dust in a galaxy.

Keywords: galaxy, dust formation, dust growth, dust destruction
Properties of Dust Formed around Wolf-Rayet Binary System based on Infrared Observations

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We have carried out the mid-infrared multi-epoch observations of periodically dust-forming Wolf-Rayet binary system WR140 with Cooled Mid-Infrared Camera and Spectrometer (COMICS) onboard Subaru telescope. WR140 is one of the nearest Wolf-Rayet binaries and the secondary is known to come across the periastron of the primary Wolf-Rayet star with a period of $\sim$8 years accompanied by distinct variations in light curves at various wavelengths, often termed as the ”spectroscopic events”. In particular, the variations in infrared light curves are supposed to be closely related to the dust formation event during the periastron. Therefore, multi-epoch mid-infrared high-angular resolution observations of WR140 using 8m-class telescopes provide us quite unique and valid opportunity to investigate the process of dust formation and its evolution associated with the active mass ejection by evolved massive stars. In this presentation, we discuss the properties (e.g., compositions, mass, temperature) of the expanding dust clouds of WR140 formed during the periastron events at 2001 and 2009 based on our multi-epoch mid-infrared imaging and spectroscopy with Subaru/COMICS.

Keywords: circumstellar dust, silicate, carbonaceous dust, mid-infrared observation, subaru Telescope
Estimation of s-process condition based on the isotopic composition of heavy elements

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Approximately half of the amount of elements heavier than the iron-group in solar-system material originates from the slow neutron-capture process (s-process). The main component of the s-process is known to be produced in Asymptotic Giant Branch (AGB) stars, i.e. evolved intermediate or low mass stars. The chemical abundances, especially, isotopic ratios around branching points of s-process provide an unique information on the neutron density and temperature of the s-process site (Terada et al. 2006).

Here, we report the dependency of heavy element isotopic composition on temperature and neutron density, and compare them with previously reported isotopic composition of various planetary materials to constrain on the possible s-process condition in AGB star.

Keywords: Nuclear synthesis, Isotopic composition, meteorite, AGB star, s-process, Neutron capture process
Morphology of presolar corundum grains from unequilibrated ordinary chondrites

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Corundum (Al₂O₃) is one of the first refractory minerals that condense directly from gas of the solar composition. Presolar corundum grains, which have highly unusual oxygen isotopic compositions compared to solar-system materials, are condensates in outflows from oxygen-rich AGB stars and/or supernovae. It is therefore important to understand the formation conditions of corundum grains in order to understand the first stage of dust formation around evolved stars. The morphological and crystallographic features of presolar corundum grains should reflect the formation conditions and subsequent thermal history of the grains. In order to understand morphology and crystallographic orientation of presolar corun-dum grains, we first made detailed observations of morphology and crystallography of corundum grains from residues unequilibrated ordinary chondrites using field-emission scanning electron microscopy (FE-SEM) and electron back-scattered diffraction (EBSD) and the oxygen isotopic compositions of the grains were then measured to identify the circumstellar condensates.

The acid-residues of ordinary chondrites, Semarkona (LL3.0), Bishunpur (LL3.1), and Roosevelt County (RC) 075 (LL3.2) were used for this study. Corundum candidate grains were found with cathodo-luminescence imaging, and 198 corundum grains were confirmed by EDS equipped to a FE-SEM. For individual grains, secondary electron images were taken from four different directions and crystallographic information was obtained by EBSD. Oxygen isotopic compositions of 111 grains were measured with UH Cameca ims-1280 ion microprobe. The details of analytical technique are described in Makide et al. (2009). After the isotopic measurements, the presolar corundum grains were reexamined by FE-SEM and EBSD.

The 198 corundum grains discovered were classified into three types according to their morphology. Type A grains have smooth surfaces (73 grains), type B grains are irregularly shaped and have rough surfaces with 10-nm-sized fine structures without crystal faces (62 grains). Intermediate grains that cannot easily be classified into either type A or B are type C (73 grains). The 111 corundum grains, of which oxygen isotopic compositions were measured. Nine presolar corundum grains were found: seven grains have oxygen isotopic compositions with positive ¹⁷O excesses and small ¹⁸O depletions; two grains show ¹⁷O depletions. The presolar grains consist of 6 type B, 2 type A, and 1 type C grains.

The observed presolar/solar corundum ratio of 8% is higher than that of 1% reported by Makide et al. (2009). This difference could be attributed to the difference in size of grains analyzed. The size of grains measured in our study is about 1 micron on average, but only larger grains (1-5 micron) were measured by Makide et al. (2009). Because larger grains have a higher possibility of survival during thermal events in the early solar system, these observations may imply that circumstellar corundum grains are dominantly <1 micron in size. Hoefner (2008) showed that corundum dust grown to ~1 micron could be easily ejected by mass-loss winds due to radiation pressure, indicating that further growth could be suppressed by rapid cooling, which may support our observation.

Choi et al. (1998) argued that presolar grains have irregular surfaces or are aggregates of smaller grains. This is consistent with the dominance of type B presolar corundum grains in this study. We obtained 10 EBSD patterns for different spots on the type B presolar grain. The EBSD patterns of all spots were the same, which are also the same as that taken prior to isotopic analysis, suggesting that the grain is not an aggregate of small grains but is a single alpha-corundum crystal. The present results may suggest common formation of fluffy and fine-structured corundum grains around various evolved stars.

Keywords: presolar grain, circumstellar dust, evolved star, oxygen isotope
Kinetics of spinel formation under circumstellar conditions

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Spinel (MgAl\textsubscript{2}O\textsubscript{4}) is one of the most abundant presolar oxides found in primitive chondrites with highly unusual oxygen isotopic compositions compared to solar-system materials. Presolar corundum and spinel grains are considered to be condensates in outflows from oxygen-rich AGB stars and/or supernovae. It has been reported that Mg/Al ratios of some presolar spinel grains are lower than the stoichiometric ratio (Choi B.-G.- et al., 1998). Spinel can form through the reaction between pre-existing corundum and Mg gas, and the non-stoichiometry of presolar spinel and the presence of presolar corundum may imply that corundum and Mg gas did not react completely. In order to discuss the origin of presolar spinel and evolution of refractory materials in circumstellar environments, it is important to understand the kinetics of spinel formation under low-pressure circumstellar conditions. In this study, we conducted spinel formation experiments through a reaction between corundum and Mg+O gas (Al\textsubscript{2}O\textsubscript{3}(s) + Mg(g) + O(g) = MgAl\textsubscript{2}O\textsubscript{4}(s) in vacuum.

An MgO pellet as a gas source of Mg and O was put in the bottom of an alumina crucible. The alumina tube was set in a vacuum chamber and evacuated to $10^{-3}-10^{-4}$ Pa, and then heated at desired temperatures (1640, 1590, and 1450 degree Celsius) for desired durations (6-695 h). The wall of the alumina tube was almost isothermal at 1640 and 1590 degree Celsius, and a small temperature gradient existed at 1450 degree Celsius, where it was 1470 and 1420 degree Celsius at the bottom and top of the tube, respectively. The inner wall surfaces and the cross sections of reacted alumina tubes were observed with FE-SEM, and their chemical compositions were determined by EDS and EPMA. Thicknesses of the reacted layer were measured at different heights from the bottom.

A spinel layer was formed on the inner wall of the alumina tube under all the experimental conditions. The thickness of the layer was largest at the bottom, and became smaller with increasing the distance from the bottom. The thickness of the spinel layer increases linearly with time. The Mg/Al compositional profile of the spinel layer showed that the layer was depleted in Mg, and the typical ratio of Mg and Al was Mg:Al = 0.72:2.18, 0.61:2.26 and 0.59:2.27 at 1450, 1590, and 1640 degree Celsius for O=4, respectively.

The linear increase of the thickness of the spinel layer with time suggests that the spinel formation rate is controlled by the surface chemical reaction, i.e., the reaction rate can be expressed by the Hertz-Knudsen equation. In order to obtain the condensation coefficient for spinel formation, we developed a model for steady-state diffusion of Mg gas inside the alumina tube under the molecular flow conditions. We fitted the growth rates of the spinel layer with the model, and obtained the spinel reaction coefficient of $0.02$ and supersaturation ratio ($S$) of $5$ at 1450, 1590, 1640 degree Celsius. Therefore, we conclude that only $2$ percent of colliding Mg gas can react to form spinel in the reaction of $\text{Al}_2\text{O}_3(s) + \text{Mg}(g) + \text{O}(g) = \text{MgAl}_2\text{O}_4(s)$ at a low super saturation ratio ($S<5$).

The obtained condensation coefficient was applied to spinel formation in an expanding mass-loss wind around a cool-low-mass evolved star with gas of the solar composition. The mass-loss velocity where spinel grains form may be much lower than the terminal velocity of the wind ($\sim 10$ km/s) and vary with time. The reaction efficiency between corundum and Mg+O gas changes with the wind velocity: corundum grains (1 micron) react almost completely with gas to form spinel in the case of wind velocity of 0.01 km/s, while little reaction occurs in the case of faster wind (1 km/s). Spinel formation under kinetic conditions in mass-loss winds with various velocities therefore may result in the variation of circumstellar oxides, which is consistent with the presence of both presolar spinel and corundum in the solar system materials.
SiO dust formation from CO gas as a reactant

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Circumstellar dust of silicates and other oxides finding in carbon rich star (C/O $> 1$) suggests the possibility that the stellar atmosphere underwent an oxygen rich (C/O $< 1$) environment of previous mass loss rate phase. This is based on the thermal stability of CO molecule. When this ratio is larger than unity, all oxygen will be trapped in the very stable CO molecule.

Since CO is one of the most abundant molecule in the dust-forming region, we considered that the reaction pathway of using CO molecule as a reactant for oxidation of silicon. Silicon oxides are the main components of silicate dust and precursor substances.

Our laboratory experiments of grain formation in a CO gas atmosphere show that amorphous SiO grains can be directly produced from silicon oxidation. SiO smoke produced by the evaporation of a fragment of Si (10 mg) from the tungsten boat at a gas pressure of 10 kPa of CO.

The color of the collected grains was yellow brown and their electron diffraction pattern showed a halo. No existence of tungsten in the produced SiO grain was detected by energy dispersive x-ray spectroscopy. The mid-IR feature of the SiO grain embedded in KBr pellet is similar to that of beta-cristobalite, i.e., the grain may be composed of microcrystallites of Si and beta-cristobalite. Moreover, the residue of evaporation was examined by transmission electron microscopy and turned out the beta-SiC which has cubic structure of high temperature phase. Graphite layer was formed to be parallel to the surface of \{111\} SiC planes.

Demonstration of formation of circumstellar oxide dust on oxidative or reductive condition requires the introduction of gas species other than oxygen controlled partial pressure.

Keywords: carbon monoxide, silicon monoxide, circumstellar dust, grain formation, transmission electron microscopy, infrared-spectroscopic analysis
Homogeneous nucleation and coalescence growth of dust analogs in supersaturated vapor

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To investigate the homogeneous nucleation and growth process of nanoparticles in vapor phase, interferometric observation was attempted for the first time to the gas evaporation method, which has been a commonly accepted physical production method of nanoparticles. Using the gas evaporation method, fine particles with the size of several nm to ~1 micron are directly produced from the gas phase and recognized nanoparticles have a crystalline habit similar with the bulk crystal even in such tiny particles. When an evaporant is initiated in an inert gas, the evaporated vapor subsequently cools and condenses in the gas atmosphere, i.e., solid grains are obtained via homogeneous nucleation from the vapor phase. Therefore it can be assumed that nucleation occurs far from the equilibrium state, but it is not obvious how far condensation takes place. However, there is no report concerning nucleation and limited study in view of crystal growth for smoke experiment, although significant numbers of smoke experiments have been performed so far. Although there has been reports concerning homogeneous and heterogeneous nucleation from solution phases [1], there has been few reports concerning homogeneous nucleation from a vapor phase in recent years. Homogeneous nucleation rates of droplets were measured as a function of temperature and supersaturation using ethanol and nonane [2, 3]. The nucleation rates were different in several orders of magnitude from the classical nucleation theory.

In preliminary experiment, Tungsten oxide was evaporated by electrical heating of a tungsten wire in a mixture gas of Ar and O2. WO3 nanoparticles were formed via homogeneous nucleation and growth during a gas cools following a thermal convection produced by the evaporation source. The degree of supersaturation for nucleation was extremely high, 6.6×10^6, which was determined from the interferogram. Surface free energy of WO3 at 1100 K was calculated based on the classical nucleation theory and was 1.38×10^3 erg cm^-2, which is within the reported values. Homogeneously condensed WO3 nuclei initially maintain their temperature for ~5 ms and then cool down with a rate of ~5×10^4 K/s. A part of the difference between actual formation rate of produced nanoparticles, which were determined based on a transmission electron microscope, and calculated values based on the semi-phenomenological nucleation theory [4] were well explained if we adopt the idea of coalescence growth.


Keywords: nucleation, crystal growth, nanoparticle, dust, interferometer, in-situ observation
Molecular dynamics simulation of nucleation from vapor

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We performed molecular dynamics (MD) simulations of nucleation from vapor at temperatures below the triple point for systems of $10^4$-$10^5$ Lennard-Jones (L-J) type molecules in order to test nucleation theories at relatively low temperatures. Simulations are done for wide ranges of the initial supersaturation ratio ($S_0 = 10^{-10}^8$) and the temperature ($T = 24$-72 K for argon). Clusters are nucleated as supercooled liquid droplets because of their small size. Crystallization of the supercooled liquid nuclei is observed after the slow down of their growth. The classical nucleation theory (CNT) extremely underestimates the nucleation rates (or the number density of critical clusters) at the low-T region. It is found that the semi-phenomenological (SP) model [1], which corrects the CNT-prediction of the formation energy of clusters using the second virial coefficient of vapor, reproduces the nucleation rate and the cluster size distributions with a good accuracy in the low-T region as well as in the higher-T cases in our previous study [2]. The sticking probability of vapor molecules onto the clusters are also obtained in our MD simulations. By using the obtained values of the sticking probability in the SP model, we can further refine the accuracy of the SP model.


Keywords: nucleation, condensation, MD simulation
Grain-surface reactions: chemical evolution and deuterium fractionation in molecular clouds

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Surface processes on interstellar icy grains play an important role in chemical evolution in molecular clouds. Until now, there have been many experimental works which report the synthesis of many molecules by energetic processes, such as UV, electron and ion bombardments, in interstellar ice analogues. Even formation of amino acid was observed after such energetic processes in ices. However, in dense core of molecular cloud where the radiation field is very weak, since the energetic processes are relatively suppressed, nonenergetic processes like neutral-neutral surface reactions become important. In this context, our group has performed a series of experiments regarding surface reactions of cold H(D)-atoms and OH radical with molecules to produce H\textsubscript{2}O, CO\textsubscript{2}, H\textsubscript{2}CO, and CH\textsubscript{3}OH. We have also tackled an important issue, deuterium fractionation of molecules in molecular clouds and demonstrated for the first time that tunneling surface reaction on interstellar ice is the key for the deuterium enrichment of some organic molecules. In the presentation, we will review our series of works described above and talk briefly about the very recent experiment for H-atom diffusion on the ice surface.
Diffusion of hydrogen and deuterium atoms and molecular hydrogen production on amorphous solid water

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In interstellar dense clouds where a radiation field is very weak, nonenergetic reactions of hydrogen atoms on dust grains, including quantum tunneling reactions, play an important role in chemical evolution. For example, it has been widely accepted that molecular hydrogen which is the most abundant molecules in the Universe is formed by recombination of H-adatoms on the dust surface. Successive reactions of H- and D-atoms with carbon monoxide are also key molecular steps in the synthesis of many organic molecules observed in the ice mantles, such as formaldehyde and methanol.

These reactions proceed through the sequence of elementary processes of H-atoms, i.e., adsorption, diffusion, and encounter with another adsorbed species. Therefore, to understand formation of not only molecular hydrogen but also complex molecules, above-mentioned physical and chemical properties of H- and D-atoms on cold surfaces should be clarified.

In this talk, we report a spectroscopic approach for the behavior of H-atoms on the surface of amorphous solid water (ASW) using photo-stimulated desorption and resonance enhanced multiphoton ionization methods.

The diffusion rate of H-atoms was directly measured after H atom deposition on ASW at 8 K. In addition, we measured the ortho/para nuclear spin ratio (OPR) of nascent molecular hydrogen formed via recombination, and observed the spin conversion of molecular hydrogen adsorbed on ASW.

Efficient molecular hydrogen formation was observed on ASW during the H-atom deposition on ASW at 8-15 K, while some fractions of H-atoms were successfully detected even after H-atom deposition at 8 K. These results show the presence of at least two types of potential sites on ASW. The analysis of attenuation curve of H-adatoms at 8 K provides the two different activation energies of H-atom surface diffusion with about 20 meV and >50 meV. Quantitatively similar results were obtained in the case of deuterium atom, suggesting that the thermal hopping mechanism better explains the diffusion rather than tunneling diffusion, because a large isotope effect should be observed if it is tunneling.

The nuclear spin temperature, which is defined by OPR of hydrogen molecules, of nascent hydrogen molecules formed from H-atoms on ASW at 8 K is very close to that of adsorbed molecular hydrogen directly from the gas phase. However, when we left the hydrogen molecules on ASW, it was found to decreases on ASW by the spin conversion.

Keywords: hydrogen atom, deuterium atom, molecular hydrogen, amorphous solid water, surface diffusion
CO$_2$ formation through radical-molecule reactions on a solid surface inside dense molecular clouds.

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Solid CO$_2$ is ubiquitously distributed in icy grain mantles in molecular clouds. Since gas phase reactions cannot explain the observed abundance of CO$_2$ in those environments and CO is also abundant in the ice, CO$_2$ is considered to form on the surface of icy grains. It has been experimentally demonstrated that CO$_2$ is formed in interstellar ice analogues processed by UV, ions, or electrons. Recent astronomical observations found solid CO$_2$ in dense molecular clouds, where the UV field is weak, implying that there should be additional routes to the formation of CO$_2$ besides UV photolysis. We performed the experiment on surface reactions of CO with cold OH radicals to investigate a possible CO$_2$ formation route in dense molecular clouds.

OH radicals were produced by dissociating H$_2$O molecules in microwave-induced plasma and cooled to 100 K before reaction. CO and OH radicals were continuously codeposited onto an Al substrate at 10-40 K. Reaction products were monitored in-situ by FTIR. We found that the formation of CO$_2$ occurred at all temperatures investigated. Up to 10% of CO was converted into CO$_2$ under the present experimental conditions. We propose that surface reactions of CO with non-energetic OH radicals are potential pathways to the formation of CO$_2$ in dense molecular clouds.

Keywords: interstellar molecular clouds, chemical evolution, carbon dioxide, radical-molecule reactions
Probing dust evolution in protoplanetary disks by near-infrared line ratios of molecular hydrogen emission

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It is believed that planets are being formed from dust and gas in protoplanetary disks. Recent high spectral resolution and high sensitivity observations have made it possible to detect transition lines of molecular hydrogen in the disks. Meanwhile, historically, near-infrared H2 line ratios have been used as a tool to derive the physical properties of various astronomical objects.

In this work we have measured the 2-1 S(1)/1-0 S(1) H2 line ratios towards T Tauri stars to diagnose the H2 excitation mechanisms and the evolutionary status of dust grains in protoplanetary disks. By using Subaru/IRCS+AO188, we observed the 2-1 S(1) and 1-0 S(1) lines simultaneously with sufficiently high sensitivity and high spectral resolution. As a result, we have succeeded in constraining an upper limit of 0.14 for the 2-1 S(1)/1-0 S(1) H2 line ratio. Our result suggests that the molecular hydrogen is excited by thermal collisions, that is, the gas temperature is sufficiently high in the disk surface. The high gas temperature means that there will be enough amount of small dust grains which heat the gas via photoelectric effect induced by FUV photons. Comparison between the observational result and our model calculations suggests that dust-to-gas ratio in the disk surface relative to that in molecular clouds should be larger than 0.1. Results of our calculation of dust evolution show that this condition is satisfied if we take into account migration of dust particles from the outer disk towards the central star, which are coupled with viscously accreting gas. Also, we show that in this case dust particles accumulate at a particular point at the disk midplane, which makes a favorable condition for planetesimal formation.

Keywords: dust evolution, protoplanetary disks
Material mixing in a protoplanetary disk formed by the collapse of a molecular cloud core

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Almost all the rocky samples taken from meteorites and comets have the isotopic compositions of refractory elements nearly identical to those of the terrestrial rocks in spite of the difference in the original locations of their formation. In the parent molecular cloud core, on the other hand, the isotopic composition may be significantly heterogeneous because the traces of short-lived radio nuclides found in primitive meteorites support the external injection of super nova ejecta or gas wind from AGB star just prior to the core collapse. These facts imply that the isotopic homogenization had occurred in the early protoplanetary disk from which the solar system was formed. Furthermore, the observations of cometary dust and extra-solar protoplanetary disks indicate that crystalline silicate dusts are contained in them with significant proportions, implying radial mixing of dust from the inner disk region to the outer one. Such mixing might be induced by the same disk mechanism which causes the homogenization of isotopic abundance.

This study therefore explores the possible mixing processes in an accreting protoplanetary disk including the stage of the collapse of parent molecular cloud by using a disk model with the 1D axial symmetry and the alpha parameterization of turbulent viscosity. Mixing of disk gas is formulated by the advection and turbulent diffusion of gas components discriminated by their infall age and maximum temperature in the disk. Because the difference in the infall age corresponds to that of original position in the core, the mixing of different gas components may represent the isotopic homogenization.

According to the parameter study, the isotopic homogenization is found to occur within the timescale of $10^6$ yr when the viscosity parameter alpha is larger than $10^{-2}$. On the other hand, mixing of high temperature gas toward the outer disk region becomes ineffective with increasing the angular momentum of the parent molecular cloud core. Given the angular momentum consistent with the observations and single star formation, the calculated mean crystallinity of silicate dust is in the range of 1-30\% among disks at the stage of 99\% completion of isotopic homogenization. The correlation between disk mass and crystallinity is basically consistent with the observation of protoplanetary disks. These results contain examples which are consistent with the properties of the solar system, that is, the total disk mass required for making the all planets, the observed isotopic homogeneity among solid materials, and the crystallinity in the cometary dust. The calculated timing of homogenization also implies that the oldest refractory inclusions in primitive meteorites are formed about 1 My after the beginning of the collapse of the parent molecular cloud core.

Keywords: protoplanetary disk, molecular cloud core, material mixing, primitive meteorite, isotopic anomaly, crystallinity
Heating experiments on the reductive condition of amorphous silicates with the mean composition of GEMS

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GEMS (glass with embedded metal and sulfides) are spherical objects of < 500 nm in diameter and characteristically included in anhydrous IDPs (Interplanetary Dust Particles), which are considered to be of the cometary origin. They have nanometer-sized (10-50 nm) Fe-Ni metals and sulfides embedded in the matrices of amorphous silicate. In spite of several propositions for the origin of GEMS [e.g., 1, 2], none of them were widely accepted. Infrared astronomical observations strongly suggest that interstellar silicates are almost amorphous [3]. If the amorphous interstellar silicates were brought into the early solar nebula and heated, the reduction of the silicates might occur. It is proposed that metallic Fe in GEMS are reduction products of FeO-bearing amorphous silicates with carbonaceous materials based on reduction experiments of thin amorphous olivine foils in a reducing atmosphere [4]. However, detailed discussion of metal formation process in the glass was not made. In this experiment, in order to study possible GEMS origin by reduction of interstellar silicates, we synthesized amorphous silicates with the mean composition of GEMS and performed heating experiments under reducing atmosphere.

The amorphous silicates as the starting material of the reduction experiments were prepared by quenching the melt of the mean composition of GEMS in a simple MgO-FeO-SiO$_2$ system (MgO = 28 wt.%, FeO = 22 wt.%, and SiO$_2$ = 50 wt.%). The quenched glass was cut and shaped for cubes of about 2 mm on a side. The starting materials were heated at 923 K and 973 K for 3 hours, and at 1023 K for 1-48 hours at one-atmosphere in a gas mixing furnace using a H$_2$-CO$_2$ gas mixture. The oxygen fugacity in this reduction experiment was -1.5 log unit above the IQF (Iron-Quartz-Fayalite) buffer (fO$_2$ ~ 10$^{-23}$ atm).

The X-ray diffraction (XRD) analysis shows that clino-pyroxene was crystallized in the heated samples. Many cracks (typically ~10µm in length and ~1µm in width) were observed both on the surfaces and cross sections, and metallic Fe grains of a few microns in size were recognized nearby cracks under a field emission-scanning electron microscope (FE-SEM) observations. Metal grains present in the cracks have euhedral shapes. Magnetite or maghemite grains of 50-100 nm in size were also observed inside of the sample under FE-SEM and a transmission electron microscope (TEM). For the TEM observation an ultra-thin sections was made by focused ion beam (FIB).

These results suggests that cracks were formed by volume change of the pyroxene crystallization and metallic Fe grains were formed on the surface or along the cracks by reaction with the reducing gas. The crystallization of the metallic grains and the magnetite or maghemite nano-particles shows that reduction did not occur inside of the glass but only near the glass-gas interface.

If metallic iron grains were formed by the reduction of FeO-bearing amorphous silicates for GEMS, metallic nano-grains can be formed only around the interface. In contrast, TEM observation for natural GEMS shows that metal grains are uniformly embedded in amorphous silicates. So, the present study suggests that metal grains in GEMS is not reduction products.


Keywords: amorphous silicate, GEMS, reduction experiment
Alkaline elements, Na, K, Rb, and Cs, are classified as moderately volatile elements, and large fractionations are expected as a result of evaporation/condensation processes. K-rich igneous fragments were identified in brecciated LL-chondrites, Kraehenberg (LL5) [1], Bhola (LL3-6) [2], and Yamato (Y)-74442 (LL4) [3,4], and show characteristic fractionation patterns (e.g., Na~0.5 x CI, K~12 x CI, Rb~45 x CI, and Cs~70 x CI [5]). In order to understand fractionation processes of moderately volatile elements as well as origin of alkali-rich fragments in chondritic breccias, we have undertaken mineralogical and petrological studies on K-rich fragments in Kraehenberg, Bhola, and Y-74442. Rb-Sr isotopic studies on the K-rich fragments in Bhola and Y-74442 are in progress.

Kraehenberg, Bhola, and Y-74442 consist of mineral fragments, K-rich fragments, impact-melt clasts, and chondrules. Kraehenberg and Bhola contain large K-rich fragments (1-2 cm in size). Small K-rich fragments (1-2 mm in size) are heterogeneously distributed in Bhola and Y-74442. These K-rich fragments show quenched textures and are composed largely of olivine (50-100 um in size) and groundmass of brown glasses which are highly enriched in alkaline elements. The boundaries between K-rich fragments and their hosts are sharp, and no reaction relation is observed along the boundaries. Dendritic Ca-pyroxene and chromite (~1 um in size) along with troilit (~10 um in size) are commonly observed in the groundmass of K-rich glasses. Tiny Fe-Ni metal grains are identified together with troilit in K-rich glasses.

Chemical compositions of olivine in the K-rich fragments fall within the compositional range of equilibrated LL-chondrites (Fa26-32 [6]). Data points of groundmass glasses of the K-rich fragments are overlapped when plotted on a ternary diagram of Na+K+Al-oxides, Ca+Mg+Fe-oxides, and SiO2. These K-rich fragments are almost identical to their host matrix in major element compositions except Na and K.

Although grain sizes of olivine are somewhat different, textures and constituent phases of the K-rich fragments in Kraehenberg, Bhola, and Y-74442 are indistinguishable.

Abundances of Rb in the whole-rock (WR) samples of Bhola and Y-74442 [7] (50-100 mg in weight) are three to ten times of chondrites, suggesting that K-rich fragments are heterogeneously distributed.

Similarities in textures, chemical compositions, and fractionation patterns of K-rich fragments in LL chondritic breccias suggest that they might be formed from identical precursor materials with related processes. Alkaline elements are also classified as large ion lithophiles and are partitioned into residual phases during crystallization. The K-rich fragments in Kraehenberg and Bhola possess flat REE patterns, which are different from the GRA 06128/06129 meteorites (alkali-rich early planetary objects showing LREE/HREE fractionations [8]). Geochemistry (i.e., solid/liquid fractionation process) could not be responsible for the enrichments of heavier alkalis in the Kraehenberg, Bhola, and Y-74442 fragments. Taking into account the lack of K isotopic fractionation [9] and the old formation ages of ~4.56 Ga [1,10], an alkali-rich component of the K-rich fragments could have formed during the early stages of solar system evolution.


Keywords: chondrite, breccia, alkali elements
Condensation experiments of magnesium silicates under protoplanetary disk conditions

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Magnesium silicates are one of the most abundant materials condensing in the system of solar abundance. Condensation from vapor is the fundamental process for formation of Mg-silicates, and there have been many experimental studies to simulate Mg-silicate formation. However, quantitative discussion on condensation kinetics, especially on the condensation coefficient that reflect the efficiency of condensation of gas species colliding with the surface of condensate, has not yet been made due to experimental difficulties.

In this study, we conducted condensation experiments of magnesium silicates at low pressures in the H2-H2O atmosphere to simulate condensation of magnesium silicates under controlled conditions of pressure, temperature, and gas chemistry close to protoplanetary disk conditions and to discuss condensation kinetics of magnesium silicates quantitatively.

Several important findings were made in this study:

(1) Crystalline forsterite condensed on the substrate of forsterite under controlled conditions (1340 K; total pressure of 1 Pa; Si/H2O/H ratios of ~0.07/70/1 relative to the solar ratios; supersaturation ratio of ~7), which are much more similar to protoplanetary disk conditions and better controlled than those in previous studies.

(2) The condensation coefficient of forsterite under the above condition was estimated to be <0.2 (or possibly <0.01).

(3) Forsterite (possibly olivine) condensed on the surface of metallic iron under the same condition, but not on molybdenum and corundum. Metallic iron also condensed on the substrate of forsterite with a similar supersaturation ratio.

(4) The evaporation rate of forsterite was suppressed under H2O-rich conditions.

These findings, for instance, suggest that forsterite and metallic iron can nucleate and grow mutually in protoplanetary disks but with different efficiencies, and they will make huge contribution to discussion on dust evolution and chemical fractionation in protoplanetary disks and on physical properties (especially thermal structure) of disks as fundamental kinetic data for condensation of magnesium silicates.

Keywords: protoplanetary disk, dust, condensation, silicate, kinetics, heterogeneous nucleation
Strength contrast between plagioclase and olivine and rheological structure of the terrestrial planets

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It is thought that plate tectonics is a product of the localized brittle failure in the lithosphere and viscous flow in the asthenosphere, and strength profile is a key to understand tectonics of terrestrial planet (Burgmann and Dresen, 2008). Physical properties, such as temperature and pressure and stress, and the chemical compositional layering between crust and mantle result in a strong rheological layering in the planet interior. It has been estimated by previous experiments that the brittle-ductile transition occurs in the planet interior and deformation mechanisms can be changed with increasing depth. In the present study, we evaluate rheological variation in the crust-mantle transition based on new series of deformation experiments, and discuss why plate tectonics doesn’t exist in the other terrestrial planets except the Earth.

In case of the earth, two different models on the strength profile in the continental crust have been proposed. The first is the ”jelly sandwich” model that had been embraced for the past two decades. This model is that a weak middle and lower crust are sandwiched between strong upper crust and strong mantle lithosphere just like a jelly sandwich (e.g., Chen and Molnar, 1983). The second one is the ”creme brullee” model, in which the upper mantle is significantly weak, and consequently region for viscous deformation continues into the mantle depth (Jackson, 2002). These two models of strength profile are given by extrapolating frictional strength and viscous flow law of each material to temperature and pressure corresponding to interior of the Earth.

In this study, we performed experiment to directly determine the relative strength between plagioclase and olivine without any extrapolating of flow law; the crustal materials consist predominantly of plagioclase that largely control deformation of the crust, whereas deformation of the upper mantle is largely controlled by olivine. These samples are together sandwiched between alumina pistons in simple shear geometry and we used the hot-pressed samples and performed deformation experiments using solid-medium deformation apparatus. The experimental conditions were ranging 1GPa and 400-800 degrees, corresponding conditions to Moho of the Earth under water-rich conditions. The experimental results show that plagioclase and olivine are expected to show almost no difference in strength at temperatures of the continental Moho of the Earth, ca. 500-600 degrees. Moreover, we found the change of relative strength contrast between plagioclase and olivine at low temperature; plagioclase becomes stronger than olivine at 400 degrees. Plagioclase is generally believed to be weaker than olivine (Brace and Kohlstedt, 1980). However, our experimental results indicate that olivine can be weaker than plagioclase (Azuma et al., 2010). In materials with a relatively strong chemical bonding such as silicates, Peierls mechanism becomes dominant at low temperatures (Tsen and Carter, 1987). Based on deformation mechanism map, deformation of olivine could be controlled by this type of flow mechanism under our low temperature experiments. Thereby, the strength contrast between plagioclase and olivine are reversed. Consequently, our result of this experiments supported ”creme brullee” model (e.g., Jackson, 2002), as continental strength profile and showed us that flow law can not be applied for low temperature conditions.

In the future, we are going to conduct experiments under dry condition to evaluate strength profile of terrestrial planets like dry Venus. Venus has been thought as a similar planet to the Earth because of closet to the Earth in mass, density, size (Taylor and McLennan, 2008). However, Venus has extraordinary crustal features and plate tectonics does not seem to work. This can be a result of different rheological property on the Venus. We are going to report our new result of deformation experiments under dry conditions, and their tectonic difference.

Keywords: strength profile, terrestrial planet, rheology, olivine, plagioclase
Petrographic observations of hibonite-bearing inclusions from Murchison using SEM-EDS.

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Hibonite is one of the minerals which condense from the solar nebula at the highest temperature ranges. Therefore, hibonite-bearing refractory inclusions may have important information in the earliest history of the solar system formation. Hibonite-bearing inclusions found in Murchison (CM2) meteorite are morphologically classified into several groups such as SHIBs (Spinel-HIBonite inclusions), PLACs (PLAty Crystals), BAGs (Blue AGgregates) (Ireland, 1988), and each group has its own significant isotopic characteristics (Ireland, 1988; Liu et al., 2009). PLACs and BAGs show large isotopic anomalies in Ca and Ti, and to less extent in Mg (delta-25Mg), but their inferred initial 26Al/27Al ratios are low or even negative. On the contrary, SHIBs show almost canonical 26Al/27Al ratios (~4.5x10E-5) but almost no anomalies in Ca and Ti. These characteristics may reflect the presence of distinct isotopic reservoirs and their mixing processes in the early solar system.

In order to better understand these isotopically distinct reservoirs and their mixing processes, we recovered about ~30 of hibonite-bearing inclusions from the Murchison (CM2) meteorite. About 10 grams of the Murchison chips were disaggregated using the freeze-thaw method. Then we applied size separations, magnetic separations, and density separations (using methylene iodide: ~3.3 g/cm3). Candidates of hibonite-bearing inclusions (containing blue or light blue minerals) were hand-picked under an optical microscope from non-magnetic, dense fractions of the separated grains. After preliminary examinations of these grains with SEM-EDS, they were fixed on a glass slide with epoxy and were examined using an optical microscope. Finally the glass slide was polished so that surfaces of most of the grains were exposed together. They were petrographically analyzed with SEM-EDS. In the present study, we have recovered 21 SHIBs, 3 PLACs, 2 BAGs, 3 grains either SHIBs or BAGs, and 2 unidentified ones. In addition, some hibonite-free inclusions, especially spinel-rich ones, and a few large spinel grains were also recovered. In the present report, we will show petrographic characteristics of these hibonite-bearing grains in detail and compare them with those of previously reported grains. We will make isotopic analyses (e.g., Al-Mg isotope analysis) on these grains in near future.


Keywords: hibonite, refractory inclusion, isotopic anomaly, Al-Mg chronology, Murchison meteorite
Evolution of infrared spectra in crystallization by heating of amorphous magnesium silicates

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Comparison between infrared spectra of astronomical observations and laboratory measurements revealed that circumstellar and interstellar dust has been investigated, crystalline silicates (e.g., olivine ((Mg;Fe)₂SiO₄), and pyroxene ((Mg;Fe)₂SiO₃) exist in circumstellar environments around oxygen rich young and evolved stars (e.g., Waelkens et al. 1996; Waters et al. 1996). It is possible that the crystalline silicates are formed by crystallization by heating from amorphous silicate in the circumstellar environments. For example, a precursor material for the crystalline silicates in circumstellar regions around young stars is considered to be interstellar amorphous silicate dust, which is believed almost completely amorphous (Kemper et al. 2004). And, in circumstellar regions around evolved stars, it is considered that amorphous silicates condense from out flow gas and are partially crystallized by heating. In order to reveal the conditions of circumstellar environments, it is important to understand crystallization process of the silicates. In recent years, distributions of minerals and crystallinity in protoplanetary disks around the T Tauri stars are estimated by comparison between the 10 μm infrared emission arising from inner warm regions in the protoplanetary disks and the 20 μm emission arising from more distant regions (e.g. Olofsson et al., 2010). In order to discuss the properties of the circumstellar dust, it is necessary to investigate evolution of infrared spectra at each wavelength region particularly.

As starting materials of heating experiments, amorphous silicates with the enstatite composition (Mg/Si =1) and the forsterite composition (Mg/Si=2) were synthesized using the radio frequency thermal plasma processing at Nisshin Engineering Co. Ltd. The amorphous samples were heated at various temperatures for various durations, and clinoenstatite (MgSiO₃) and forsterite (Mg₂SiO₄) were crystallized from the starting amorphous materials in the Mg/Si ratio of 1 and 2, respectively. By analyses of infrared absorption spectroscopy and x-ray powder diffraction of the heated samples, the degrees of crystallization were estimated. Then, we investigated the relation between the degree of crystallization and change of the infrared spectral features at each wavelength regions. At results, the infrared spectral features of the samples with forsterite component at around 20μm change at a more rapid rate than those at around 10μm in the crystallization process. On the other hands, the infrared spectra of the samples with enstatite composition have no such trend. By comparing between the results and astronomical observation of T Tauri stars, we discuss crystallization from amorphous silicate in circumstellar environments.

Keywords: infrared, dust, crystallization, amorphous silicate
Density Estimation from Impact Track Morphology in Silica Aerogel: Application to Dusts of Comet 81P/Wild 2

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Cometary dust particles of Wild2 have been successfully collected and returned in Stardust Mission [Brownlee et al. (2006) Science 314, 1711-1726]. Hypervelocity capture (6.1 km/s) of those particles mandated various degrees of heating, fragmentation and evaporation of the projectiles during their capture process in silica aerogel [Zolensky et al. (2006) Science 314, 1735-1739]. Nevertheless, an impact track formed by each particle can be an indicator of its original properties [Horz et al. (2006) Science 314, 1716-1719]. Particle size dependence of track properties has been studied in several papers [Burchell et al. (2009) Planet. Space. Sci. 57, 58-70; Horz et al. (2009) Meteo. Planet. Sci. 44, 1243-1264] and impact tracks in Stardust aerogel formed by several sized soda lime glass beads of different sizes were used for calibration of Wild2 dust size distribution [Burchell et al. (2008) Meteo. Planet. Sci. 43, 23-40]. In the work of Iida et al. [(2010) Meteo. Planet. Sci. 45, 1302-1319], three-dimensional structures of Stardust impact tracks were analyzed and Wild2 dust density was estimated based on their track formation model. However, density dependence of track properties has not been investigated precisely yet. Therefore, we carried out impact experiments into silica aerogel (20 mg/cc) using projectiles of several densities in order to clarify the relation between projectile properties (size and density) and track morphology. The experiments were carried out with a two-stage light-gas gun at ISAS, JAXA. The projectiles we used were bubble glass (0.5 g/cc) polystyrene (1.06 g/cc), sintered silica (~1.3 g/cc), soda lime glass (2.5 g/cc), alumina (3.9 g/cc), and copper (8.9 g/cc). All the projectiles except for sintered silica were spherical in shape. Size of these impactors ranged from ~0.03 to ~0.1 mm in diameter and they were fired into 20 mg/cc silica aerogel at ~6 km/s to simulate the capture of Wild2 dust. All the individual impact tracks were observed with an optical microscope. The results show that track length (L\textsubscript{t}) depends on projectile size and density while maximum track width (D\textsubscript{m}) mainly depends only on projectile size. Therefore, aspect ratio (L\textsubscript{t}/D\textsubscript{m}) does not change with projectile size, but only with projectile density. This means that when we estimate projectile properties from a track shape, L\textsubscript{t}/D\textsubscript{m} is a good indicator of projectile density. This can be applicable for Stardust impact tracks; densities of Wild2 dust particles are estimated by examining the relation between projectile density and aspect ratio of a track in Stardust aerogels.
We simulated the bow shock excited around the planetesimal moving with supersonic velocity relative to the nebula gas, and examined whether dust grains are heated enough to melt and become chondrules.

Chondrules are millimeter sized spherical silicate particles that constitute up to 80% of chondrite in volume. Although they must have experienced heating and then melting to account for their spherical shapes and their textures, the heating source remains to be solved. Some kind of heating events must have happened in their formation age, about 4.56 billion years ago, because the temperature of the nebula gas was a few hundred kelvins at that time, which is too low to melt dust grains.

The shock wave heating model is the one of the ideas for the heating mechanism, which explains the observational constraints for chondrule formation properly. However, no reliable sources of shocks are still confirmed.

In this study, we focused on the planetesimal bow shock as the source of the shock. The idea of the bow shock excited by the supersonic planetesimal with respect to the nebula gas is offered by Hood (1998) and Weidenschilling et al. (1998) and the only numerical study so far for the planetesimal bow shock is conducted by Ciesla et al. (2004). They simulated the bow shocks in two-dimensional Cartesian coordinate system, regarding the planetesimal as a cylinder, and calculated the thermal history of a dust grain in one dimensional shock model by using the shock properties given by their simulation.

In order to analyze quantitatively the possibility of the planetesimal bow shock for chondrule formation, we conducted hydrodynamic simulations in axisymmetric spherical coordinate system, regarding a planetesimal as a sphere, and calculated the thermal history of a dust grain along its trajectory with various impact parameters. The flow around the supersonic planetesimal was simulated by using the ZEUS-2D code (Stones & Norman 1992) with various velocities, densities and planetesimal sizes.

As a result, we restricted the possible chondrule formation region in the gas density - gas relative velocity parameter space. In addition, we found the possible impact parameter range in which dust grains could melt. By using these results, we estimated the total amount of chondrules that could be made by planetesimal bow shocks. About one earth mass of dust grains could be heated to melting point by bow shocks under the scenario that supersonic planetesimals with high eccentricity was excited by Jovian resonances (Marzari & Weidenschilling 2002). We concluded that the planetesimal bow shocks are still possible chondrule formation site.

Keywords: planetesimal, shock wave, chondrule
UV-CPL irradiation experiment of lactic acid: photostability, racemization, and asymmetric decomposition

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Introduction: Since L-amino acid enantiomeric excesses (e.e. = 0.4 - 9.1%) were found in carbonaceous chondrites (Cronin and Pizzarello, 1997), the study on the origin of homochirality in space has been developing. In particular, detection of circular polarized light (CPL) in the star forming regions (Bailey, 1998; Fukue, 2010) has provided an opportunity for a number of the UV-CPL irradiation of amino acids or its precursors to detect e.e. experimentally (e.g., Takano et al. 2007; Takahashi et al. 2009). However, the production mechanism of molecular homochirality and a possibility of homochirality production of the other extraterrestrial organic molecules than amino acids have been infrequently investigated. Recently, e.e. of L-lactic acid (3 - 12%) from carbonaceous chondrites have been newly reported (Pizzarello, 2010). We have conducted the UV-CPL irradiation experiment of lactic acid in aqueous solution, as a starting study to understand the homochirality of lactic acid in space.

Experimental: 0.02 mM DL lactic acid (D:L = 1:1) in aqueous solution, 0.01 mM L-lactic acid in aqueous solution and 0.01 mM D-lactic acid in aqueous solution were prepared. Four ml of in quartz cell was irradiated with UV-CPL. Left and right UV-CPL at 215nm from a free electron laser produced at BL5U, UVSOR. Power of irradiation was 10-200mWh. After irradiation, 100 micro L of the sample solution was analyzed by High Performance Liquid Chromatography (HPLC) with UV detector (254 nm). For the optical resolution of D- and L- lactic acids, ligand exchange chiral HPLC column (D-penicillamine ODS silica, SUMICHIRAL, OA-5000) was used. 1 mM copper sulfate aqueous solution was used for the mobile phase. Identification and quantification of compounds were made by comparison of peak retention times on HPLC chromatograms and peak areas, respectively, with those of standard compounds.

Results and discussion: For every sample, the concentrations of lactic acid exponentially decreased with increasing irradiation power, indicating the progress of photodecomposition of lactic acid. After 100 mW irradiation, the concentration of lactic acid decreased to less than 1% of the initial concentration. There was no difference in the concentration change between left and right CPL. Irradiation of D-lactic acid yielded L-lactic acid, and the ratio of D to L came close to 1:1 with increasing irradiation power. The opposite result was obtained by irradiation of L-lactic acid. There was no difference in the ratio change between left and right CPL. These results are probably reflected by deprotonation and racemization of a lactic acid molecule. A small e.e. was detected after irradiation of DL-lactic acid in this study. However, at this stage, it is difficult to determine whether the value is a true e.e. or analytical error. To be summarized, photodecomposition, racemization, and asymmetric decomposition of lactic acid occur simultaneously during UV-CPL irradiation, which gives a final e.e., if any. If the obtained e.e. in this study is a true value, CPL would have likely played a role of inducing the initial small asymmetry of lactic acid, which can be consistent with the past studies about amino acids (e.g., Flores et al. 1977).

References:

Keywords: homochirality, circular polarized light, lactic acid, photostability, racemization, asymmetric decomposition