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PPS025-01

Room:101

Time:May 23 14:15-14:30

## 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

PPS025-02

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## 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 ~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

PPS025-03

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## 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

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## Morphology of presolar corundum grains from unequilibrated ordinary chondrites

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Corundum ( $\text{Al}_2\text{O}_3$ ) 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 corundum grains, we first made detailed observations of morphology and crystallography of corundum grains from residues of 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 cathodoluminescence 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 crystallographical 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  $^{17}\text{O}$  excesses and small  $^{18}\text{O}$  depletions; two grains show  $^{17}\text{O}$  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

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## Kinetics of spinel formation under circumstellar conditions

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Spinel ( $\text{MgAl}_2\text{O}_4$ ) 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 ( $\text{Al}_2\text{O}_3(\text{s}) + \text{Mg}(\text{g}) + \text{O}(\text{g}) = \text{MgAl}_2\text{O}_4(\text{s})$  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  $\sim 10^{-3}$ - $10^{-4}$  Pa, and then heated at desired temperatures (1640, 1590, and  $\sim 1450$  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  $\sim 0.02$  and supersaturation ratio (S) of  $\sim 5$  at 1450, 1590, 1640 degree Celsius. Therefore, we conclude that only  $\sim 2$  percent of colliding Mg gas can react to form spinel in the reaction of  $\text{Al}_2\text{O}_3(\text{s}) + \text{Mg}(\text{g}) + \text{O}(\text{g}) = \text{MgAl}_2\text{O}_4(\text{s})$  at a low super saturation ratio ( $S \sim 5$ ).

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.

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## 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

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## 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 O<sub>2</sub>. WO<sub>3</sub> 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,  $6.6 \times 10^6$ , which was determined from the interferogram. Surface free energy of WO<sub>3</sub> at 1100 K was calculated based on the classical nucleation theory and was  $1.38 \times 10^3 \text{ erg cm}^{-2}$ , which is within the reported values. Homogeneously condensed WO<sub>3</sub> nuclei initially maintain their temperature for ~5 ms and then cool down with a rate of  $\sim 5 \times 10^4 \text{ K/s}$ . 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.

[1] X. Y. Liu, K. Tsukamoto, M. Sorai, *Langmuir*, 16 (2000) 5499.

[2] J. L. Schmitt, G. W. Adams, R. A. Zalabsky, *J. Chem. Phys.* 77 (1982) 2089.

[3] G. W. Adams, J. L. Schmitt, R. A. Zalabsky, *J. Chem. Phys.* 81 (1984) 5074.

[4] K. K. Tanaka, Katsuyuki Kawamura, Hidekazu Tanaka, and Kiyoshi Nakazawa, *J. Chem. Phys.* 122 (2005) 184514.

Keywords: nucleation, crystal growth, nanoparticle, dust, interferometer, in-situ observation



PPS025-08

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## Molecular dynamics simulation of nucleation from vapor

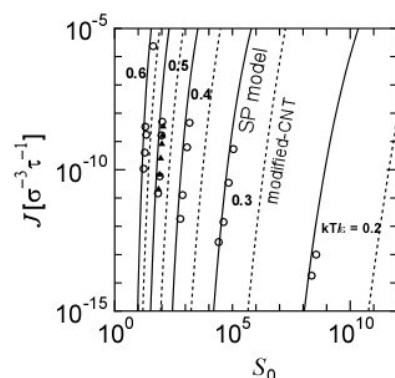
Kyoko Tanaka<sup>1\*</sup>, Hidekazu Tanaka<sup>1</sup>, Tetsuo Yamamoto<sup>1</sup>, Katsuyuki KAWAMURA<sup>2</sup>

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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.

[1]A.Dillmann,G.E.A.Meier,J.Chem.Phys.,94,3872(1991)

[2]K.K.Tanaka,K.Kawamura,H.Tanaka,K.Nakazawa,J.Chem.Phys. 122,184514 (2005)



Keywords: nucleation, condensation, MD simulation



PPS025-09

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## 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<sub>2</sub>O, CO<sub>2</sub>, H<sub>2</sub>CO, and CH<sub>3</sub>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.

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## 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-atoms 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-atoms 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 decrease on ASW by the spin conversion.

**Keywords:** hydrogen atom, deuterium atom, molecular hydrogen, amorphous solid water, surface diffusion

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## CO<sub>2</sub> formation through radical-molecule reactions on a solid surface inside dense molecular clouds.

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Solid CO<sub>2</sub> is ubiquitously distributed in icy grain mantles in molecular clouds. Since gas phase reactions cannot explain the observed abundance of CO<sub>2</sub> in those environments and CO is also abundant in the ice, CO<sub>2</sub> is considered to form on the surface of icy grains. It has been experimentally demonstrated that CO<sub>2</sub> is formed in interstellar ice analogues processed by UV, ions, or electrons. Recent astronomical observations found solid CO<sub>2</sub> in dense molecular clouds, where the UV field is weak, implying that there should be additional routes to the formation of CO<sub>2</sub> besides UV photolysis. We performed the experiment on surface reactions of CO with cold OH radicals to investigate a possible CO<sub>2</sub> formation route in dense molecular clouds.

OH radicals were produced by dissociating H<sub>2</sub>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<sub>2</sub> occurred at all temperatures investigated. Up to 10 % of CO was converted into CO<sub>2</sub> 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<sub>2</sub> in dense molecular clouds.

Keywords: interstellar molecular clouds, chemical evolution, carbon dioxide, radical-molecule reactions

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## 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 H<sub>2</sub> 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) H<sub>2</sub> line ratios towards T Tauri stars to diagnose the H<sub>2</sub> 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) H<sub>2</sub> 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

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## 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

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## 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], non 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<sub>2</sub> system (MgO = 28 wt.%, FeO = 22 wt.%, and SiO<sub>2</sub> = 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<sub>2</sub>-CO<sub>2</sub> gas mixture. The oxygen fugacity in this reduction experiment was -1.5 log unit above the IQF (Iron-Quartz-Fayalite) buffer ( $f_{O_2} \sim 10^{-23}$  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 clacks 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.

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## Alkali-rich Fragments in LL-chondritic Breccias

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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 $\sim$ 0.5 x CI, K $\sim$ 12 x CI, Rb $\sim$ 45 x CI, and Cs $\sim$ 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  $\mu$ m 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 ( $\sim$ 1  $\mu$ m in size) along with troilite ( $\sim$ 10  $\mu$ m in size) are commonly observed in the groundmass of K-rich glasses. Tiny Fe-Ni metal grains are identified together with troilite 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 SiO<sub>2</sub>. 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  $\sim$ 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.

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## 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 H<sub>2</sub>-H<sub>2</sub>O 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/H<sub>2</sub>O/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 H<sub>2</sub>O-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