

Laboratory synthesized silicate grains with mass dependent and independent oxygen isotopic fractionation

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To elucidate how grains were formed and grown in the early solar nebula, laboratory experiments have been performed. During the synthesis of silicate dust analogs using electrical discharges, we found that the refractory oxide particles produced, such as silica, iron oxide and iron silicate, have mass independent oxygen isotopic fractionation (MIOIF) [1]. Up to now, it was known that UV photodissociation processes can drive MIOIF, although no synthesis ever made solid particles with MIOIF using this process. Oxygen isotopic compositions formed by MIOIF processes in calcium-aluminium rich inclusions (CAIs) and chondrules produced in the early solar nebula have been discussed as the result of UV photodissociation processes [2]. After discovery of a new possible driving force, that is plasma, to produce grains with MIOIF, we obtained a chance to discuss an advanced historical process concerning material evolution in the early solar system in addition to the results of our typical smoke experiments. Here, we will show an experimental result concerning silicon oxide particles produced by radio-frequency plasma in addition to the previously produced refractory oxide particles using the smoke generator at GSFC [3].

To produce silica particles, the gas-evaporation method was used. Stainless-steel electrodes, 40mm square, were set with a gap of 10mm in parallel to produce a field of capacitively coupled plasma (13.56MHz) in the chamber. A v-shaped Ta boat, (70*5*0.1mm) charged with Si or SiO powder in the center, was set as an evaporation source 5mm under and parallel to the electrodes. The plasma field was generated with a power of 300W, the Ta boat was electrically heated in a gas mixture (20Torr) of He and oxidant (O₂, CO, H₂O or H₂O₂) and the powder was evaporated within 30s. Smoke particles were deposited on the electrodes. The evaporated vapor condenses after mixing with active species in the plasma and forms nm-sized silica particles. The samples were sent to the University of California, San Diego for isotopic analysis. As a result, silica was shown to be only mass dependently fractionation using radio-frequency plasma in contrast with the MIOIF formed by arc discharge. In the case of radio-frequency plasma, the temperature of the electrodes goes up typically around 500K during the experiment. In the case of the arc-discharge experiment, since it was observed that the MIOIF decreases in the higher temperature field above 400K [1], silica particles formed by radio-frequency plasma may show only mass-dependent fractionation. The trend of higher MIOIF observed for particles produced in the higher power of the electrical discharge. Therefore, the degree of MIOIF might be a compromise between the advantages of decreased temperature and the disadvantage of decreased density of charged particles from the central star and/or transferred gas by x-winds with heliocentric distance.

To see the effects of reactants on the fractionation the active species were changed by using different gas atmospheres. The plasma density and electron temperature of the plasma is known empirically to be 10⁸-10¹⁰cm⁻³ and 10⁴K, respectively. The plasma density roughly corresponds to that at 0.1AU in our solar system and the electron temperature is lower than the 10⁶K around the Earth. Although the plasma is not completely coincident with that in our solar system, we believe that it is suitable to discuss the origin of the MIOIF found in CAIs or chondrules. We will present temperature and gas species dependences for the observed fractionation processes combined with IR spectra and transmission electron microscope observations of the solids produced in these experiments.

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[1] Y. Kimura et al. 2007, *Meteoritics Planet. Sci.* 42:1429.

[2] e.g., H. Yurimoto & K. Kuramoto 2004, *Science* 305:1763.

[3] J.A. Nuth et al. 2002, *Meteoritics Planet. Sci.* 37:1579.