

CONDENSATION AND GAS-SOLID EXPERIMENTS OF MINERALS IN PROTOPLANETARY DISK CONDITIONS: IMPLICATIONS FOR FORMATION OF CHONDRITIC COMPONENTS. S. Tachibana¹, A. Takigawa¹, H. Nagahara¹ and K. Ozawa¹ ¹Dept. Earth Planet. Sci., Univ. Tokyo, 7-3-1 Hongo, Tokyo 113-0033, Japan (tachi@eps.s.u-tokyo.ac.jp).

Introduction: Condensation from vapor and gas-solid reaction may have been responsible for dust formation in the high-temperature region or during high-temperature events in the early solar system. Some chondritic components such as fluffy Type-A CAIs, AOAs, chondrules, and metals in CH chondrites show, in fact, the signatures of condensation and/or gas-solid reaction during their formation [e.g., 1-3]. Moreover, these reactions may also be responsible for the isotopic homogenization of metallic elements and for the volatility-controlled elemental fractionations recorded in chondritic meteorites.

Equilibrium condensation models predict thermodynamically equilibrium mineral assemblages as a function of temperature in a system of fixed pressure and gas compositions [e.g., 4-6]. The minerals predicted in the models for the system of solar abundance are commonly found in chondrites, and thus equilibrium calculations provide a useful theoretical framework for formation of solids in the early solar system. However, dust formation processes do not necessarily proceed in equilibrium conditions. For instance, if the cooling timescale of the system is much faster than the timescale of a chemical reaction, the reaction may cease at a certain temperature and the reaction product should have chemical and/or mineralogical features at the temperature where the reaction was quenched because chemical reaction rates generally have strong dependence on temperature, which could be the case for preservation of refractory components such as Type-A CAIs and AOAs. The elemental fractionation due to physical separation of early condensates, which may be responsible for chemical fractionations in chondrites, should also be tested considering both timescales of chemical and physical processes. Physical properties of condensed materials, such as size of individual components and textural relationship in a mineral assemblage, are also important because they may change the efficiency of physical separation of dust and the interaction between dust and a radiation field, i.e., the thermal condition of the dust-forming environment. These properties are determined by reaction processes, but equilibrium calculations cannot deal with processes of reactions.

It is thus crucial to understand condensation and gas-solid reaction processes of minerals and their kinetic aspects to understand the evolution of solar system materials.

Kinetics of condensation and gas-solid reaction in protoplanetary conditions: The reaction rate is controlled by the slowest step of the process (the rate-limiting step). In the case of direct condensation of solid from low-pressure disk gas, the rate-limiting step is generally the surface atomistic process and the rate is expressed by the Hertz-Knudsen equation;

$$J = \frac{\alpha_c p_i - \alpha_e p_i^{eq}}{\sqrt{2\pi m_i k T}}, \quad (1)$$

where p_i is the partial pressure of a condensing gas species i , p_i^{eq} is the equilibrium vapor pressure of the species i , m_i is the molecular (or atomic) weight of i , k is the Boltzmann constant, T is the absolute temperature, and α_c and α_e are the condensation and evaporation coefficients ranging from 0 to 1 to represent kinetic hindrances for condensation and evaporation, respectively [7]. When condensation and evaporation proceed without kinetic hindrances, α_c and α_e are equal to unity, and Eq. (1) simply represents the gaseous flux onto the surface of a condensate. Note also that α_c and α_e should be equal to each other at equilibrium, but they are not necessarily identical in non-equilibrium conditions.

In the case of the gas-solid reaction, where a preexisting solid phase reacts with gas to form a new phase, the reaction product may surround the preexisting phase and diffusion of reactive components through the reaction layer could be the rate-limiting step. However, the rate-limiting step could be the surface process if diffusion in the reaction layer is faster than the surface-reaction kinetics (Eq. (1)). Such a situation is likely to occur in low-pressure conditions.

Condensation and gas-solid reaction experiments: There have been many experimental studies on evaporation of major minerals in chondrites such as forsterite, enstatite, metallic iron, and troilite [e.g., 8-13], while it has not been easy to carry out condensation and gas-solid experiments under low-pressure conditions for quantitative discussion on kinetic processes due to experimental difficulties. However, recent progresses of experimental studies have made it possible to determine the growth kinetics of minerals in chondrites. Here we report our recent condensation and gas-solid reaction experiments and the growth kinetics of minerals from vapor obtained in the ex-

periments. Although there have been many experimental studies on homogeneous nucleation of solid particles in a hot vapor [14 and references therein], it is difficult to obtain nucleation kinetics under disk conditions in this type of experiments due to experimental difficulties. We thus focus on the growth kinetics of minerals obtained in low-pressure experiments.

Condensation of metallic iron. Condensation experiments of metallic iron were carried out under known pressure conditions of metallic iron vapor, which was generated by evaporation of metallic iron heated in a vacuum furnace [13]. It was found that α_c is almost unity at 1235 and 1337 K for a supersaturation ratio S of >10 . This suggests that metallic iron grows without large kinetic hindrance. In the experiments, metallic iron nucleates easily on a corundum substrate, indicating that metallic iron can form without significant delay for homogeneous nucleation. More kinetic data, especially for $S < 10$, will be presented at the workshop.

Condensation of forsterite. We recently reported the condensation experiments on forsterite at ~ 1340 K and at 1 Pa of the H_2 - H_2O mixed vapor [15], where vapor evaporated from a gas-source forsterite condensed on a forsterite substrate placed at a cooler part of a vacuum furnace. The experimental conditions in [15] were much more similar to protoplanetary disk conditions and better controlled than those in previous studies. We succeeded to obtain 10-100-nm-sized crystalline forsterite as condensates on the forsterite substrate. We further made the experiments with a much larger flux from the gas-source forsterite and obtained 1- μ m-sized crystalline forsterite as condensates. The α_c of forsterite estimated in these experiments was <0.2 (or possibly <0.01). We will report more quantitative α_c in the workshop.

Formation of spinel. Spinel can form via a reaction between preexisting corundum and Mg gas in the disk conditions. The spinel formation experiments [16] showed that the thickness of the spinel layer increased linearly with time, suggesting that the surface reaction is the rate-limiting step. This is most likely to be due to fast diffusion of metallic elements in the spinel layer that has a non-stoichiometric chemical composition. By applying Eq. (1) to the experimentally obtained reaction rate, we estimated α_c of 0.01-0.02 for spinel formation.

Formation of troilite. Troilite formation experiments in the low-pressure (1 Pa) He- H_2S mixed gas showed that the formation rate of troilite is controlled by the surface reaction and that α_c is in the order of 0.02 at ~ 770 K [17]. The obtained α_c is consistent with that predicted by a simple collision theory by [18].

Formation of enstatite. In the experiments on the reaction between forsterite and SiO-O gas, vaporized from silica glass, at 1300 K in vacuum (10^{-3} Pa), only amorphous SiO was obtained on the surface of forsterite and no enstatite formation occurred [19]. The experiments under protoplanetary disk-like pressures (~ 1 Pa) should be done to conclude that enstatite cannot form through the reaction between forsterite and SiO-rich gas in protoplanetary disks.

The kinetic condensation model including these kinetic data sets has been developed [e.g., 20], where the condensation path of minerals in a cooling gas, the grain size distribution of each phase, and the change of bulk compositions of condensates and residual gas are calculated. It has been suggested that the formation kinetics and the supersaturation ratios required for heterogeneous nucleation of phases largely affects the condensation behavior of the system, and that different sets of mineral assemblages could be formed in systems cooled at different rates. We will also present new results from the kinetic condensation model at the workshop.

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