EXPERIMENTAL CONDENSATION OF METALLIC IRON AT CONTROLLED SUPERSATURATION.
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Introduction: Dust particles are formed by condensation from cooling gas in protoplanetary disks and around evolved stars. Kinetics of condensation and evaporation determines time scale of evolution of dust particles in the universe, and causes chemical and isotopic fractionation between condensates and gas as well.

When surface reactions are rate-limiting processes, the net condensation (growth) flux $J_c$ is expressed by the Hertz-Knudsen equation based on a classical kinetic theory of gases,

$$J_c = \frac{\alpha_c p - \alpha_e p^{*v}}{\sqrt{2m_k T}},$$

where $p$ is the pressure of condensing gas species, $p^{*v}$ is the equilibrium vapor pressure, $m$ is the weight of the condensing gas molecule, $k$ is the Boltzmann constant, and $T$ is the temperature. $\alpha_c$ and $\alpha_e$ are condensation and evaporation coefficients, respectively, that are measures of kinetic hindrance of condensation and evaporation, respectively. They should be equal to each other at equilibrium, but not necessarily under non-equilibrium conditions. Moreover, they can be a function of pressure, temperature, and gas compositions because kinetics of surface processes should depend on such physical and chemical conditions. For instance, [1] showed that $\alpha_c$ for evaporation of metallic iron depends on temperature and pressure of iron vapor. However, in many model calculations for dust evolution in the universe, $\alpha_c$ and $\alpha_e$ were assumed to be unity or constant [e.g., 2, 3].

In order to understand kinetics of evaporation of minerals and melts, many evaporation experiments have been carried out. However, there have been few condensation experiments under controlled physicochemical conditions aiming to determine condensation kinetics. In this study, we carried out condensation experiments of metallic iron under a controlled pressure of iron vapor to understand dependence of condensation kinetics on iron-vapor pressure. Metallic iron is one of the major minerals in the universe, and some Fe-Ni grains in CH chondrites have been reported to be condensates from the nebular gas [e.g., 3]. It is thus important to know kinetics of metallic iron condensation to understand evolution of solid particles in protoplanetary disks and in circumstellar environments.

Experiments: We improved experimental techniques in condensation experiments of metallic iron performed by [4], where a metallic iron plate in the center of a tungsten-mesh heater was heated and evaporated gas was condensed onto a molybdenum substrate placed in a cooler region of the vacuum chamber. However, the position of the substrate was changed for experiments with different temperatures, making it difficult to control the pressure of iron vapor independently from temperatures. Moreover, molybdenum diffused into condensates to form Mo-Fe alloy.

In this study, a metallic iron pellet was evaporated at 1300°C through an alumina tube, and the incoming iron flux onto a substrate was controlled by changing the inner diameter of the alumina tube (3-6 mm), i.e., the larger incoming flux, the larger diameter of the tube.

An alumina disk (10 mm in diameter and 1.5 mm in thickness) was used as a substrate to avoid diffusion of elements in the substrate into condensates. The substrate was placed at the distance of 13 mm from the exit of the alumina tube, at which the temperature of the substrate was 962°C (1235 K). An experimental duration ranged from 6 to 48 hours. The pressure in the vacuum chamber was kept less than 10⁻⁶ bar during experiments by continuous evacuation with a turbo-molecular pump and a rotary pump.

The weight loss of the iron pellet and the weight gain of the alumina substrate were measured. The surface and cross-section of the condensates were observed by using reveal SEM and chemical compositions and crystallinity of condensates were analysed with EDS and EBSD (electron back-scattered diffraction).

A supersaturation ratio $S (= p/p^{*v};$ a ratio of the pressure of iron vapor to the equilibrium vapor pressure of metallic iron) at the surface of the substrate was calculated based on the flux distribution of iron vapor emerged from the tube and the measured evaporation rate of source. Because the incoming flux to the surface of the substrate has a relatively uniform distribution, $S$ appears not to have a large variation at the surface of the substrate.

Results: EDS and EBSD analyses showed that all the condensates are $\alpha$-Fe, which was probably transformed from $\gamma$-Fe during quench after heating, except for a small amount of refractory oxides and refractory metals that were from an alumina tube and a heater.

SEI images of surfaces and cross-sections of condensates ($S \sim 30$) are shown in Fig. 1. Characteristic surface structures on the surface are continuous growth steps with an interval of $\sim 10$ nm. It is also seen that
condensates form a compact layer for experimental duration longer than 12 hours and that the thickness of the compact layer increases with time.

\[ \alpha \approx \alpha_c = 1 \]  \hspace{1cm} (c)

**Discussion:** Nucleation and growth of metallic iron on the alumina substrate occurred for runs with heating durations shorter than 6 hours, while metallic iron grew on Fe metal itself for longer heating duration. Based on weight changes of substrates heated for longer than 12 hours, we obtained the condensation (growth) rates of metallic iron at 962°C with \( S \) of \( 20 \) or \( 30 \).

The obtained condensation flux was compared with the ideal condensation flux calculated using Eq. (1) with \( \alpha_c = \alpha_c = 1 \) (Fig. 2). Although the obtained condensation fluxes are slightly higher than the ideal flux, they appear to be close to the ideal flux within experimental uncertainties. Because \( S = p/p^* \) is large in the present experiments, the effect of evaporation from condensates can be small, which indicates that \( \alpha_c \) should be close to unity in the present experimental conditions (\( S \sim 20 \) to \( 30 \) at 962°C). This means that almost all the colliding molecules incorporate into the condensates without any kinetic hindrances.

**Fig. 2.** Condensation flux of metallic iron as a function of \( S \). The ideal condensation flux (Eq. (1) with \( \alpha_c = \alpha_c = 1 \)) is also shown for comparison.

Because the condensation temperature of 962°C (1235 K) is close to the equilibrium condensation temperature of metallic iron in the system of solar abundance [e.g., 5], the present results indicate that condensation of metallic iron proceeded with an ideal rate as long as \( S > 20 \) as assumed in [3]. However, more experiments at lower \( S \) and at different temperature conditions are needed for detail discussion on iron dust formation in the protoplanetary disks and around evolved stars.


**Fig. 1.** FE-SEM images of surfaces (left) and cross-sections (right) of condensates formed at 962°C with \( S \sim 30 \). In the cross-section images, bottom porous regions represent alumina substrates and light gray regions above the substrates are condensed metallic iron. (a) 6 hours, (b) 12 hours, Acicular or euhedral grains (-0.1-1 µm) seem to nucleate on nano-scale aluminum oxides. (c) 24 hours. Acicular or euhedral grains (-0.1-1 µm) seem to nucleate on contaminants. (d) 48 hours.

Weight gains of the substrates increased linearly with time, suggesting that surface processes control condensation kinetics and that the Hertz-Knudsen equation can be used to discuss condensation kinetics.