DEMONSTRATION OF CHONDRULE MELT SOLIDIFICATION BY NUMERICAL SIMULATION. H. Miura, *Department of Earth and Planetary Materials Science, Graduate School of Science, Tohoku University, Japan (miurah@m.tains.tohoku.ac.jp)*, E. Yokoyama, *Gakushuin University, Japan*, K. Nagashima, *Osaka University, Japan*, K. Tsukamoto, *Tohoku University, Japan*.

INTRODUCTION: Chondrules are millimeter-sized, once-molten, spherical-shaped grains mainly composed of silicate materials. They have various types of internal textures; porphyritic, barred-olivine, radial pyroxene, and cryptocrystalline. Many authors have been carried out dynamic crystallization experiments to constrain the formation mechanism of each texture ([1] and references therein). Barred-olivine (BO) chondrules are characterized by parallel set(s) of olivine bars in a thin section. It has been considered that olivine bar crystals are actually platy in three-dimension [2]. A BO chondrule usually has an olivine crystal that covers the chondrule surface. This olivine rim has the same crystallographic orientation as inner olivine platelets, which connect to the rim.

The textures close to that of BO chondrule have been reproduced successfully in some experiments, however, the formation condition was so different at each experiment. Tsukamoto et al. carried out crystallization experiment of forsterite melt which cools very rapidly ($\sim 100 - 1000\,\mathrm{K/s}$) by using aeroacoustic levitation system and in-situ observation of the crystallization process [3, 4]. They found that the melt crystallized in a very short period of time (less than 1 sec) at temperature much lower than melting point by a few hundred kelvins or more. The internal texture showed rim and parallel bar structure in thin section. On the other hand, Tsuchiyama et al. succeeded in reproducing textures close to classic BO together with olivine rim by evaporation in vacuum [5]. BO textures close to natural chondrules were formed by heating at superliquidus temperature of about $100 - 200\,\mathrm{K}$ for 10 min and cooled at $1000\,\mathrm{K/h}$.

To clarify the formation mechanism and condition of rim and parallel plate structure, we have to get back to the general mechanism of crystallization in supercooled chondrule melt. We carry out numerical simulation of crystallization from highly-supercooled melt sphere based on the theory of crystal growth. For simplicity, we consider a single-component melt composed of forsterite, as the same as experiments by [3, 4]. In addition, we limit the system in two-dimension for reducing computational cost. In this paper, we report one successful simulation result in which crystallization pattern very similar to texture of BO chondrule was reproduced.

NUMERICAL SIMULATION METHOD: One of the key subjects to understand the solidification process is how crystal growth speed V is determined. For crystallization of a pure material from its melt, it is known that V relates to undercooling at interface ΔT_k as (Wilson-Frenkel low [6])

$$V = \mu \Delta T_k, \tag{1}$$

where μ is proportional constant (kinetic coefficient). The interfacial undercooling is determined by balance between release of latent heat of crystallization and the heat transfer.

The phase-field model is one of the powerful models to solve crystallization process with taking into account time-dependent field parameters like temperature and concentration [7]. In this model, crystal and liquid are distinguished by using a filed parameter ϕ, which continuously changes from zero to unity; $\phi = 1$ for liquid, $\phi = 0$ for solid, and $0 < \phi < 1$ for transition layer, say, crystal-liquid interface. The governing equation for ϕ is given by

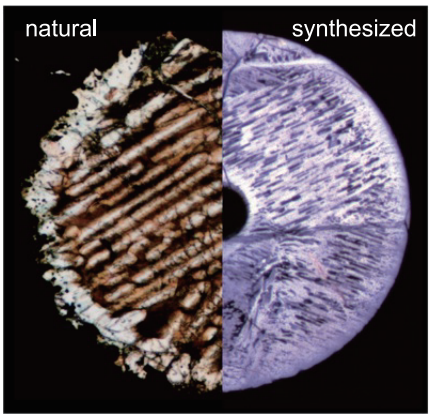

Figure 1: Internal texture of BO. **Left:** natural BO chondrule. **Right:** synthesized by using aeroacoustic levitation system [3, 4].

$$\frac{\delta}{\mu}\frac{\partial \phi}{\partial t} = \frac{\sqrt{2}}{12}\frac{T_\mathrm{M}}{T}p'(\phi)(T - T_\mathrm{M}) - \frac{T_\mathrm{M}\Gamma}{4\delta}g'(\phi) + T_\mathrm{M}\Gamma\delta\triangle\phi, \tag{2}$$

where T is the temperature, T_M is the melting point, $\Gamma \equiv \gamma_\mathrm{s}/L_0$ is the capillary length, γ_s is the surface tension, L_0 is the latent heat of crystallization, and δ is the thickness of solid-liquid interface. The governing equation for T is given by

$$c_0\frac{\partial T}{\partial t} + L_0 p'(\phi)\frac{\partial \phi}{\partial t} = \boldsymbol{\nabla}\cdot(k\boldsymbol{\nabla}T), \tag{3}$$

where c_0 is the specific heat and k is the thermal conductivity. The second term of left hand side gives the temperature increase due to release of latent heat of crystallization (recalescence). The expressions of functions $p(\phi)$ and $g(\phi)$ are shown in reference.

SIMULATION SETTING: We consider a completely-molten forsterite sphere which cools at the surface. There is no crystal nucleus inside the sphere, so crystallization will be triggered by nucleation or seeding when the sphere achieves some degree of supercooling. There should be a temperature gradient inside the sphere that center is hotter than surface. The difference in temperature between center and surface is

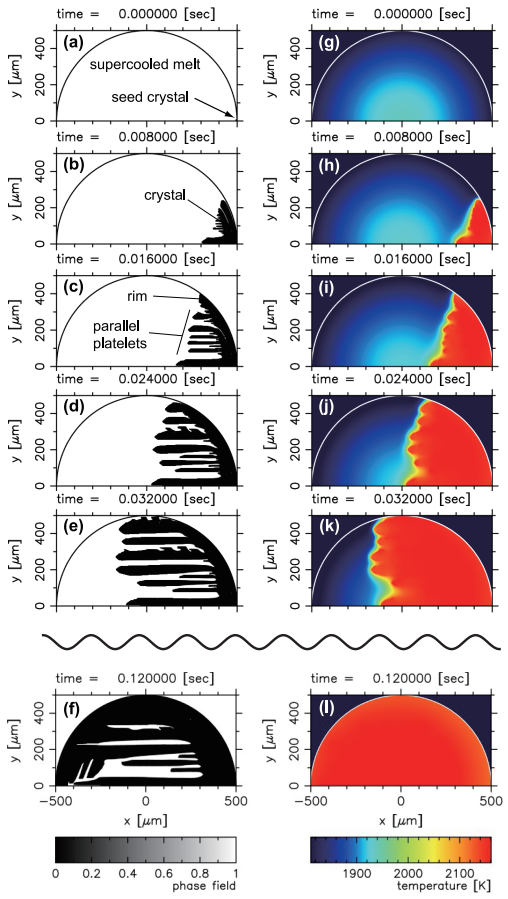

Figure 2: Snapshots of rim and parallel platelets formation inside sphere of $500\,\mu$m in radius at every 8 msec. **Left:** phase ϕ. **Right:** temperature T. Initial undercooling is 350 K at surface. The cooling rate at surface is $q_{\rm s} = 10^9\,{\rm erg\,cm^{-2}\,s^{-1}}$.

given by

$$T_0 - T_{\rm s} = \frac{q_{\rm s} r_{\rm d}}{2\kappa} = 125 \left(\frac{q_{\rm s}}{10^9\,{\rm erg/cm^2/s}}\right)\left(\frac{r_{\rm d}}{500\,\mu{\rm m}}\right)\,{\rm K}, \quad (4)$$

where T_0 and $T_{\rm s}$ are temperatures at center and surface, respectively, $q_{\rm s}$ is the cooling rate at surface, and $r_{\rm d}$ is radius of sphere. In this paper, we adopt $q_{\rm s} = 10^9\,{\rm erg\,cm^{-2}\,s^{-1}}$, which corresponds to black body radiation at 2000 K.

RESULT: Figure 2 shows snapshots of simulation at every 8 msec except of panels (f) and (l). Panels (a)-(f) show phase ϕ and (g)-(l) show temperature T. We put one seed crystal at surface as suggested by arrow in panel (a). The undercooling at surface is $\Delta T_{\rm s} = 350\,{\rm K}$. Crystal grows from the seed crystal along the surface (panel (b)), and temperature increases up to melting point by recalescence. As a result, a very steep *negative* temperature gradient appears at crystal-melt interface (panel (g)). The term *negative* means that crystal is hotter than liquid. It is known that a flat interface growing under such *negative* temperature gradient is unstable with respect to small undulation [8]. We found that many bars appear at inner interface of the rim (panels (b)-(c)).

It should be noted that these bars grow keeping their parallel structure (panels (d)-(e)). The reason is in anisotropy of kinetic coefficient μ adopted in our simulation, with which crystal growth in vertical direction is strongly suppressed at low supercooling. The anisotropic kinetic coefficient was introduced to phase-field model to simulate facet formation during crystal growth [9]. Unfortunately, no data of μ for pure forsterite melt has been reported. In this study, we used an interim model for μ to imitate facet formation of forsterite crystal. The measurement of μ is indispensable for more realistic simulation.

The parallel bars grow continuously passing through the center of sphere, and finally arrive at opposite side. The panels (f) and (l) show snapshot at 120 msec. The snapshot is very similar to texture of BO chondrule. The formation of rim and parallel bar structure completed within $\sim 0.1\,{\rm sec}$, which is consistent with levitation experiment [3, 4]. At that time, the temperature in whole sphere increases up to melting point by recalescence. The unsolidified liquid region among the parallel bars will solidify as the latent heat is removed to outside. The cooling timescale is much longer ($\sim 1\,{\rm sec}$) than that of rim and parallel bar formation. The difference in crystallization timescale might be recorded in the completely solidified sphere as internal texture.

DISCUSSION: It was suggested that BO chondrules are regarded as rapid crystallization products, where olivine crystals grow with morphological instability, like dendritic growth [5]. However, they did not mention how the instability occur during solidification. Our simulation result clearly showed that the morphological instability occurs due to the very steep *negative* temperature gradient at inner interface of rim (see panel (g) in Fig. 2). In order to generate such steep temperature gradient at interface, crystal growth is required very rapid. The width of thermal diffusion layer at interface is estimated as $\delta_T \sim \kappa/V$, where κ is the thermal diffusivity. δ_T should be much smaller than chondrule radius $r_{\rm c}$, otherwise the temperature gradient will be diffused away. This condition can be rewritten by $r_{\rm c}/V \ll r_{\rm c}^2/\kappa$, which means that timescale of crystal growth ($\sim r_{\rm c}/V$) should be much shorter than the thermal diffusion timescale ($\sim r_{\rm c}^2/\kappa$). The thermal diffusion timescale is evaluated as $r_{\rm c}^2/\kappa \simeq 0.7\,{\rm sec}$, so the formation of rim and parallel bar structure should be completed within a very short period of time (1 sec or shorter) as shown in our simulation.

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