NUMERICAL SIMULATION OF SOLIDIFICATION OF CHONDRULES: FORMATION OF OLIVINE BARS IN MG_2SIO_4-FE_2SIO_4 SYSTEM. H. Miura, *Department of Earth and Planetary Materials Science, Graduate School of Science, Tohoku University, Japan (miurah@m.tohoku.ac.jp)*, K. Tsukamoto, *Tohoku University, Japan*.

INTRODUCTION: Barred olivine (BO) chondrules show unique solidification texture characterized by parallel set(s) of olivine bars in a thin section, which have been considered actually platy in three-dimension [1, 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 bars, which connect to the rim [3]. BO chondrules are regarded as rapid crystallization products, where olivine crystals grow with morphologic instability, like dendritic growth [4]. Formation of BO texture was observed in situ in crystallization experiments using droplet levitation techniques [5, 6]. However, initial process of the morphologic instability that produces the olivine bars has not been investigated in details.

The aim of our study is to investigate the initial process of formation of BO texture based on numerical simulation of solidification of chondrule. As a first step, we consider Mg_2SiO_4-Fe_2SiO_4 system in which only olivine is considered as a mineral (the phase diagram was obtained by [7]). Other minerals observed in chondrules, such as pyroxene, silica, chromite, and so forth [3], are not considered for simplicity. We adopt a phase-field model as the numerical scheme [8]. In this paper, we investigate rim growth at the surface of molten chondrule and morphological instability that produces olivine bars.

MODEL: The thermodynamically consistent phase-field model for a binary alloy based on the entropy functional is adopted here [8]. In the phase-field model, a new variable, the phase filed ϕ, is introduced to keep track of the phase, taking on constant values indicative of each of the bulk phases and making a transition between these values over a thin transition layer that plays the role of the sharp interface. Another variable is a concentration C, which represents a molar fraction of Fe_2SiO_4. We give a temperature field with a constant gradient by hand, that is to say neglect the release of latent heat of crystallization (recalescence). Solving governing equations for ϕ and C allows us to simulate crystal growth process with partitioning of Fe at crystal-liquid interface and Fe diffusion in crystal and liquid phases. The derivation of the governing equations can be found in [8].

RESULT: Fig. 1 shows time evolution of crystal-liquid interface. We put a platy seed crystal at right side (gray region), which is in equilibrium with liquid (white region) at a temperature of 1900 K, then reduce the temperature at a constant rate of 5000 K s^{-1} to cause growth of crystal.[1] The crystal grows keeping the flat interface before 0.065 sec, then becomes unstable to form cell-like pattern. The initial stage in which the flat interface is maintained can be considered as growth of rim, and the cell-like pattern as olivine bars. In this case, the average width of bars is about 5 μm.

Fig. 2 shows concentration distribution at 0.0884 sec. We set C = 0.17 in the seed crystal, which corresponds to the equilibrium concentration at 1900 K. As the crystal grows, the concentration in crystal increases gradually and exceeds 0.6 at $x = -20$ μm. The flat interface shifted to the cell-like pattern as a result of morphological instability. There are some areas with high concentration of $C \approx 0.9$ around the root of cell-like pattern, as a result of rejection of Fe content from crystal. It is found that the concentration at core of each bar is almost constant at $C \approx 0.45$ along the direction of bar elongation (see Fig. 3). In contrast, the center of interstices of bars is enriched in Fe comparing with bars ($C \approx 0.8$).

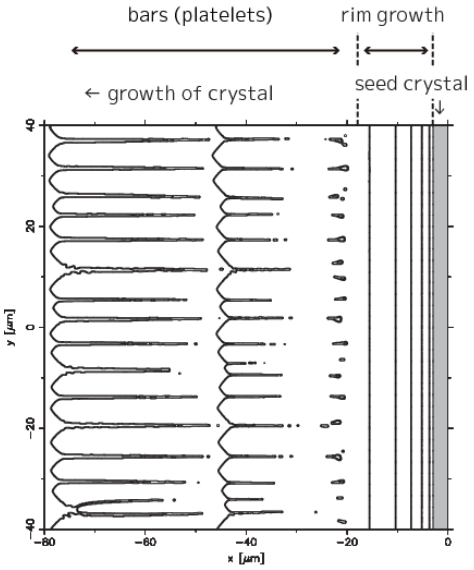

Figure 1: Morphological instability during solidification from a platy seed crystal. Solid curves show the shape of crystal-liquid interface every 0.013 sec. Cooling rate of the system is 5000 K s^{-1}. Periodic boundary condition is applied at top and bottom borders.

DISCUSSION: Olivine zoning shown in Fig. 2 has some similarities to the experimentally reproduced BO chondrules [4]. The experimental product showed that core composition of each bar is almost constant along the direction of bar elongation, which is similar to our numerical result. Formation of the zonal structure was considered as follows; the olivine crystal grew as a short and thin bar with relatively Mg-rich composition first, became long but still thin, and finally overgrowth of Fe-rich marginal part occurred [4]. However, our numerical simulation suggests that the elongation of thin bars and the overgrowth do not occur separately, but elongation of "thick" bar accounts for the zonal structure.

[1]The cooling rate is about 4 orders of magnitude larger than the canonical value of 1000 K hr^{-1} [4].

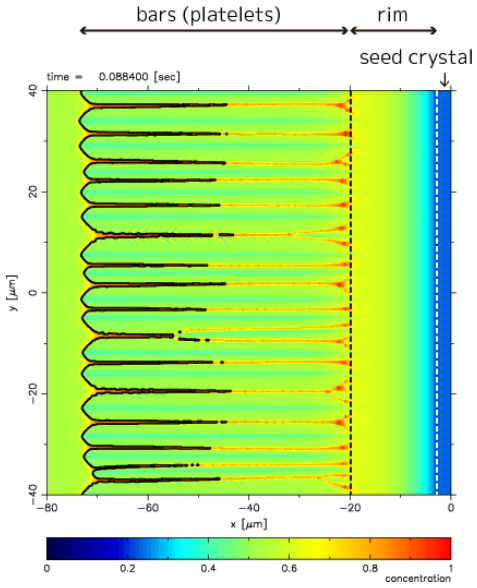

Figure 2: Concentration distribution at 0.0884 sec. Color shows molar fraction of Fe_2SiO_4. Solid curve shows crystal-liquid interface.

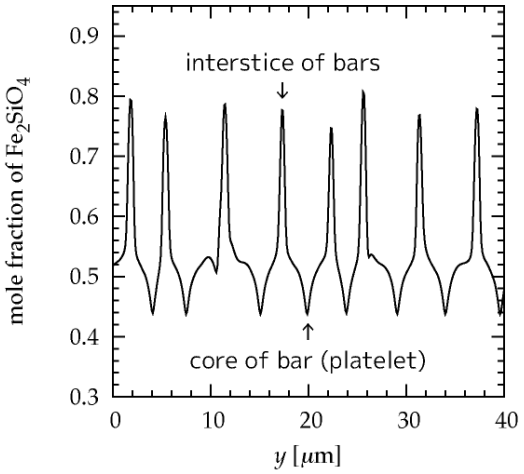

Figure 3: Concentration profile in direction perpendicular to parallel sets of bars (platelets) in Fig. 2; from $y = 0$ to 40 μm at $x = -40$ μm.

Bar width observed in natural BO chondrules varies from 5 to 100 μm [3]. Our numerical result matches to the minimum value (see Fig. 2). Since it is known that the bar width increases as the cooling rate decreases [9], the cooling rate of 5000 K s^{-1} would be an upper limit to reproduce the proper bar width. A phase-field simulation of solidification of Ni-Cu alloy showed that the bar spacing λ is inversely proportional to the square of cooling rate [10]. If the bar width also has the same dependence on cooling rate, the cooling rate of 45,000 K hr^{-1} is required to reproduce olivine bars of 100 μm in width. The required cooling rate is much larger than the canonical value inferred from the crystallization experiments of BO textures [4].

Major phases in BO chondrules except of olivine are pyroxene and feldspathic glass that mainly exist at interstices of olivine bars [3]. These phases could be formed at the interstices as a result of partitioning of incompatible elements, such as Ca and Al, from olivine crystal. In order to consider formation of these phases at the interstices, we need to extend our numerical model from Mg_2SiO_4-Fe_2SiO_4 system to MgO-FeO-SiO-CaO-Al_2O_3 system with the formation of minerals except of olivine. The extension will allow us to simulate formation of BO texture in more realistic situation. We will leave this challenging theme in future.

Formation of olivine rim is also unsolved problem. We numerically simulated growth of rim from a platy seed crystal in this paper. However, We did not answer a question how the rim covered entire (or partial) surface of BO chondrule. The rim formation requires olivine growth along the surface after nucleation at surface of completely molten chondrule [4]. They considered that olivine grows along the surface that is cooler than the interior by the latent heat of evaporation, mainly of FeO. On the other hand, it was showed that the formation of rim structure can occur by radiative cooling at the surface of molten chondrule [11]. It was suggested by [4] that the change of liquidus temperature along the surface by evaporation of FeO plays minor role for the rim formation, however, this mechanism is worth being investigated in details based on numerical simulation as shown in this paper.

CONCLUSION: The numerical simulation of solidification of chondrule gives us a deep insight about the formation process of solidification texture of chondrules. This is a very powerful tool to investigate the formation condition of chondrules by a different viewpoint from chondrule reproduction experiments. The Mg_2SiO_4-Fe_2SiO_4 system adopted in this study might be not enough to simulate solidification process of real chondrules, however, the phase-field model has a great potential for understanding the formation process of chondrules.

REFERENCES: [1] Tsuchiyama et al. (2000) *LPSC* **31,** 1566. [2] Noguchi (2002) *Antarct. Meteorite Res.* **15,** 59-77. [3] Weisberg (1987) *J. Geophys. Res.* **92,** E663. [4] Tsuchiyama et al. (2004) *GCA* **68,** 653-672. [5] Tsukamoto et al. (2001) *LPSC* **32,** 1846. [6] Srivastava et al. (2010) *J. Appl. Phys.* **107,** 114907. [7] Bowen & Schairer (1935) *Ame. J. Sci.* **29,** 151. [8] Bi & Sekerka (1998) *Physica A* **261,** 95. [9] Lofgren & Lanier (1990) *GCA* **54,** 3537. [10] Takaki et al. (2005) *J. Crys. Growth* **283,** 263. [11] Miura et al. (2010) *J. Appl. Phys.* **108,** 114912.