Numerical method for growth and dissolution processes of stoichiometric mineral crystals based on a phase-field model

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Various silicate crystals are found in rocks and chondritic meteorites. Most of them have formed from magmas that have been cooled at generally unknown conditions. Their formation conditions can be speculated from their properties, such as shapes, chemical compositions and sizes. Since the chemical compositions of crystals are different from the host magma in general, the elemental partitioning is unavoidable between the growing crystals and magma. When the crystal growth is extremely slow, each composition can be regarded as homogeneous (equilibrium crystallization). However, the elemental diffusion in solid is generally so slow that the composition of the growing crystal is inhomogeneous. In this case, the compositional change of magma is recorded as a zoning profile in the crystal. If the elemental diffusion in magma is slower than the crystal growth, the incompatible elements are concentrated around the growing crystal to form a boundary layer. The formation of the boundary layer will make the planar crystal surface unstable to form dendrites [1], and affect the chemical composition of fluid inclusions [2]. Therefore, it is important to understand how the boundary layer affects the crystal shapes and chemical compositions.

In this study, we propose a numerical method for growth and melting of stoichiometric silicate compounds based on a phase-field (PF) model. We consider a binary system composed of two end members, A and B, with a crystal phase composed of only component A and a liquid phase assumed as a regular solution. We reformulated a binary PF model proposed by Kim et al. [3] in order to deal with a stoichiometric compound. We adopted a forsterite-SiO_2 system and gave the free energy as functions of composition and temperature so as to agree with the phase diagram obtained experimentally [4]. We combined the free energy with the reformulated PF equations and carried out the numerical calculations in one-dimensional plane-parallel geometry.

First, we examined cases of diffusion-limited growth. With supercooled initial conditions, the incompatible component B was removed from the growing crystal to form the boundary layer. The liquid composition at the crystal-liquid interface was almost equal to the equilibrium value. Finally, the crystal growth ceased when the liquid composition reached equilibrium homogeneously. The growth rate was inversely proportional to the square of the elapsed time, which is consistent with the diffusion-limited condition. With under cooled initial conditions, the crystal melted to reach equilibrium like growth conditions. Second, we examined cases of kinetic-controlled growth. The growth rate is proportional to the driving force of crystal growth. In all calculations stated above the total free energy was monotonically decreased with time, which is consistent with the second law of thermodynamics.

The PF method can treat any systems if the free energy is formulated. Since some thermodynamics databases are available in silicate system (e.g., MELTS), our numerical method that can treat a stoichiometric compound enables us to investigate various situations considered in Earth and planetary science. In the future, we will extend our model to ternary or more system.

References: [1] W. W. Mullins and R. F. Sekerka, 1964, J. Appl. Phys. 35, 444. [2] D. Baker, 2008, Cont. Miner. Petro. 156, 377. [3] S. G. Kim et al., 1999, Phys. Rev. E 60, 7187. [4] N. L. Bowen and O. Andersen, 1914, 37, 487.

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