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Theoretical and experimental study on the combination of bullets of snow

Natsuki Niekawa^{1*}, Masao Kitamura¹, Norimasa Shimobayashi¹, Akira Miyake¹, Masaki Takaya¹, Yu Kodama¹

¹Kyoto-Univ. Sci.

Combination of bullets is a kind of aggregation snow crystals constructed by single crystals of ice Ih; the stable phase of ice. The formation process of these crystals were considered as that ice Ic, which is the metastable phase of ice, was formed at first, and later the stable phase ice crystals (Ih) nucleated and grew on the surfaces of the preexisting metastable crystal (Takahashi, 1982; Takahashi and Kobayashi).

The phenomenon that the thermodynamically metastable phase is born at the beginning of crystallization and later the stable phase crystal nucleates and grows is called the Ostwald's step rule. The crystallization processes governed by the Ostwald's step rule has been observed in many experiments with various materials (Ostwald, 1897; Barrer, 1988). However, in spite of the universality of the Ostwald's step rule, the mechanism has not been sufficiently elucidated so far. Most studies on the Ostwald's step rule are nothing but reports of the observed phenomenon with respect to specific materials, or the partial understanding of crystallization process governed by this rule.

In this study, for the purpose of the comprehensive understanding of crystallization process governed by the Ostwald's step rule, both theoretical and practical study were conducted.

I. Theoretical study

The fastest nucleation process against the values of the chemical potentials of the metastable and stable phase was derived from the comparison of the free energy barrier of all possible nucleation process (homogeneous 3D nucleation of the stable or the metastable phase, heterogeneous 2D nucleation of either phase on the other phase, and homogeneous 2D nucleation of the stable or metastable phase). Then, on the basis of the results, a diagram which shows the nucleation process against the condition of the system (nucleation mode diagram) was obtained. The path on this diagram which the actual system will trace enables to know the subsequent nucleation process, from beginning to end of the crystallization.

The necessary condition of the surface energy and driving force of the nucleation for crystallization governed by Ostwald's step rule will take place was revealed. It can be thought as that the metastable phase crystal was born under high supersaturated condition at first, and later the decrease of the degree of supersaturation due to the nucleation and the crystal growth made the stable phase crystals to nucleate on the surfaces of the existing metastable crystals.

II. Experimental study

On the basis of the present theoretical study, the experiments for artificial combination of bullets was conducted.

In the cold room at Nishiborieizaburo kinen Explorer Museum in Higashiomi, Shiga Prefecture, water droplet less than 1 micrometer in diameter were introduced into the vessel cooled with liquid nitrogen to be frozen and grew. After that, they were recovered for microscopic observation.

As a Result, combination of bullets in nature was recreated for the first time. This success is owe to the present theoretical study, thus the validity of the present theory was roughly confirmed.

Keywords: combination of bullets, Ostwald's step rule