

A new theory of impurity incorporation into a growing crystal

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Concentration of impurities (or additives) in crystal is an important factor that determines crystal quality and property. For example, the electric conductivity of semiconductor strongly depends on the impurity concentration. Perfection of protein crystals used for X-ray structural analysis is affected by incorporation of impurities. The impurity concentration and its distribution in minerals are important clues to speculate geophysical environments in which these minerals have formed. To understand what factor determines the impurity partition into growing crystals is important to control properties of industrial crystals and to elucidate formation process of rock-forming minerals.

The impurity concentration in crystals is characterized by a partition coefficient, which represents the ratio of impurity fractions in crystal to growth environment. The impurity fractions are defined by a ratio of concentrations of impurity molecules to host molecules. The partition coefficient being unity indicates that the impurity fraction in the growing crystal is the same as that in the growth environment. The partition coefficient depends on the growth condition in general. There are some theories that formulated the relationship between the partition coefficient and the crystal growth velocity such as the BPS theory [1] and Smith model [2]. Both theories have formulated the effect of the concentration field disturbed at around the growing crystal. However, the essence of the crystal growth is the incorporation of molecules to the crystal at its surface. The impurity molecules are also incorporated into the crystal after adhesion on the surface, migration along the surface, and finally incorporated at step edges. This suggests the importance of the surface process on impurity partitioning, but there is no theory that predicts what factors of the surface process affect the impurity distribution and how.

In this study, we propose a new theory of the impurity partition that takes the surface process of impurity molecules into consideration. The canonical theory of crystal growth that consider the surface process of molecules is the BCF theory [3]. This theory formulated the steady distribution of host molecules adsorbed on the crystal surface using a diffusion equation and derived their incorporation rate into step edges, namely, the advancing velocity of steps. What we should consider here is the same process for impurity molecules. The behavior of the impurity molecules on the crystal surface can be described by the same diffusion equation with the host molecules. The difference from the host molecules is the number density of molecules in the crystal; the density of host molecules is fixed, on the other hand, that of impurity molecules is variable. To determine the number density of impurity molecules in the crystal, we added another constraint on the impurity incorporation. We succeeded to formulate the partition coefficient as functions of parameterized quantities that characterize the surface process of impurity molecules. In the talk, we will introduce the outline of our theory and some interesting results on the impurity partition.

The theory that we proposed in this study is a straightforward extension of the BCF theory, so it is applicable for various crystals that exhibit layer-by-layer growth. For example, our theory would be useful to quantitatively analyze the experimental results of growth of semiconductors and protein crystals. Our theory might be able to clarify the importance of the surface process for impurity partition by applying to the experimental results that cannot be interpreted by the BPS theory or the Smith model.

References:

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