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DNA nano-structure formation and the interpretation based on crystal growth theory

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The DNA molecule is now attracting attention as a new self-assemble material. The reason why DNA is used for self-assembly is that DNA molecule has calculation capability. Many nanostructures have been produced using DNA, for example, DNA tile (Winfrey and Seeman, 1998). DNA tile is a complex molecule, which is composed of some single strand DNA (ssDNA). Each tile has some sticky ends (a part of some exposed bases). The complementary sticky end spontaneously makes hydrogen bond and grow to large ordered structure (DNA tile crystal) as the solution cools down. This process is termed as self-assembly, in other words, crystallization. Although DNA tile has potential as computer, there are some problems. The most important matter is presence of error (misfit crystallization). In order to reduce this error, many types of DNA tiles have been designed. However, it is difficult to completely prevent the assemble error. For synthesis of DNA tile crystal without the assemble error, we carried out experimental study of DNA tile formation and interpreted the result based on crystal growth theory.

First, we chose T-motif as DNA tile, which is able to grow on the electrically-charged Mica surface like two dimensional crystal, and measured its growth rate using DNA origami as a seed crystal with atomic force microscopy. The growth rate of T-motif crystal on Mica substrate was about 4.30 [monomers/minute].

In the second experiment, we observed T-motif crystals synthesized on the Mica surface for various conditions (temperature and concentration). We found that the nucleation temperature of the T-motif crystal was about 41.5 [deg. C.], which did not depend on the concentration significantly in a range of 2~10 [nM]. From this result, we assumed that the T-motif crystal growth can be considered to be melt growth. In order to understand the growth mechanism, we calculated the step free energy β [J/m] and melting point T_m [deg. C.] under the assumption that the number density of crystals on the Mica surface is proportional to the two-dimensional nucleation rate. When the melting point is assumed to be 50 [deg. C.], the calculated step free energy was about 4.21×10^{-13} [J/m]. The calculated step free energy of T-motif crystal is similar to that of Lysozyme crystal (8.9×10^{-13} [J/m]).

We also observed the morphological change of T-motif crystals depending on the growth condition. In the highest supersaturation condition in this study (T-motif concentration is 10 [nM] and temperature is 38 [deg. C.]), the morphology of the T-motif crystal was similar to dendrite crystal. In other cases, the morphology was found to be polygonal shape. The reason why the T-motif crystal becomes dendritic in the highest supersaturation condition is considered to relate to the thermal stability of the sticky end binding. In low supersaturation, many T-motif units bind only at a site with two sticky ends. In contrast, in the highest supersaturation, the T-motif unit can bind to anywhere. We calculated the difference of Gibbs free energy in two conditions; one match bond of sticky end or two matches. We found that when the driving force exceeded the critical point, the T-motif crystal has possibility to be formed as dendrite. Using the critical driving force, we calculated the temperature at which the T-motif dendrite crystal was formed. In this calculation, the temperature of dendrite formation is 41.8 [deg. C.] when the melting point is assumed to be 50 [deg. C.]. On the other hand, experimental result shows that T-motif dendrite form in 38 [deg. C.]. The inconsistency between the theory and the experiments should be resolved in the future, however, our study is an important first step to describe the growth mechanism of the T-motif crystal based on the theory of crystal growth from melt phase.

Keywords: DNA tile, Crystal growth