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**Principle of the unique adhesion mode in protein crystallization**

Jindrich Hasek, *Institute of Macromolecular Chemistry, Academy of Sciences, 16206 Praha 6, (Czech Republic).* E-mail: hasek@imc.cas.cz

Analysis of crystal packing and of the role of ligands built into protein crystals [1,2] lead to very natural concepts helping in rational growth of high quality crystals:

1) The principle of a dominating protein-protein adhesion mode says that high diffraction quality protein crystals require a unique driving force of crystallization guaranted by a single dominating adhesion mode.
2) The protein surface shielding molecules block competitive adhesion modes leading thus to a unique deposition of protein molecules into the growing crystal.

Protein molecules in solution have always number of mutual adhesion modes because of their generally large surface areas. In the case, that crystallographicaly incompatible adhesion modes combine during crystallization, the growing crystals are full of stacking faults. In extreme cases it leads to so called phantom crystals – optically well looking crystals without diffraction. The protein surface shielding (PSS) molecules can temporarily block possibly incompatible adhesion modes lowering thus the probability of irregular stacking of molecules in the crystal lattice.

The good PSS molecules should bind only to very specific places on the protein surface, and only with low affinity so that they can leave the protein surface during deposition of other protein molecules into the growing crystal and the PSS molecules dissolve again in the crystal cavities filled by solvent. Due to low affinity, the PSSA molecules are experimentally observed on the protein surface rarely (often disordered and with lower occupancies). In few cases they are observed also trapped at the protein-protein interfaces contributing to stability of crystal.

Different PSS molecules have different binding characteristics, they bind to different places on protein surface and thus they can force protein to crystallize in different space groups. It is useful, because the structure determination of identical protein in different molecular environments provides more complete description of its hydration shell and also shows possible tiny changes of the surface structure induced by protein associations.

Even low concentration of PSS molecules can bring the desirable effect, so that their use generally does not change dramatically the existing crystallization screens.

The hypothesis presented above is supported by numerous experimental observations of polyethyleneglycol molecules selectively trapped on protein surfaces in structures deposited in the PDB. The classification of experimentally observed binding of the PEG-type polymers [1] is instructive for understanding the PSS effect of the hydrophilic polymers.

These concepts give also more natural and less conflict explanations of many mysterious methods including the “lysine methylation”, “surface entropy reduction”, “silver bullets”. They offer several new alternative ways how to improve current crystallization methods.

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**A Crystal Growth Approach for DNA Nano-Structure Formation**

Hitoshi Miura,[a] Yuya Ueno,[a] Shogo Hamada,[b,c] Satoshi Murata,[c] Katsuo Tsukamoto,[a] [a]*Departament of Earth and Planetary Materials Science, Tohoku University (Japan).* [b]*Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology (Japan).* [c]*Graduate School of Engineering, Tohoku University (Japan).* E-mail: miurah@m.tohoku.ac.jp

The DNA molecule is now attracting attention as a new self-assemble material. Many nano-structures have been produced using DNA, for example, DNA tile [1]. 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 DNA tile combines with another one via the complementary sticky end to form an ordered nano-structure. The formation of the nano-structure occurs in the solution just by cooling the temperature, like as crystal growth in solution. The self-assembly of the DNA tiles has a calculation capability, however, the misfit between the complementary sticky ends is not a negligible problem [2]. To reduce the assemble misfit, we carried out experimental study of the DNA tile self-assembly and analyzed the result based on crystal growth theory.

We chose T-motif as DNA tile, which is able to grow on the electrically-charged Mica surface like two dimensional crystal [3]. We synthesized the T-motif ordered nano-structure on the Mica surface under a constant temperature $T$, and observed it using atomic force microscope. We found that the nano-structure was formed when $T$ is lower than about 41.5℃, not depending on the initial solute concentration significantly in a range of 2 – 10 nM. When $T$ is relatively high, most of the nano-structures have polygonal shape. As $T$ decreases, the dendrite-shaped nano-structure was observed. We also found that the number density of nano-structures observed on the Mica surface monotonically increased as $T$ decreased.

We analyzed our experimental results based on the theory of crystal growth assuming that the T-motif self-assembly can be considered as melt growth. We calculated the step free energy $\beta$ and the melting point $T_m$ under the assumption that the number density of nano-structures is proportional to the two-dimensional nucleation rate. When the melting point was assumed to be $T_m = 50$℃, we obtained the step free energy of $\beta = 4.2\text{x}10^{-13}$ J/m. Using the calculated step free energy, we derived the critical undercooling $\Delta T_{dend}$ above which the dendrite-shaped nano-structure was formed. For small undercooling, many T-motif units bind only at a site with two sticky ends. In contrast, for large undercooling, the T-motif unit can bind to anywhere. We calculated the difference of Gibbs free energy in two cases, one match bond of sticky end or two matches, and derived $\Delta T_{dend}$ as a function of $\beta$ and $T_m$. The theoretical consideration matched the morphological change of the nano-structure qualitatively.

Our study indicated that the DNA self-assembly could be interpreted as the crystal growth. We could propose DNA self-assemble methods with fewer misfit based on the crystal growth approach.

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