Effect of crystallization conditions for the enlargement of the size of DNA-functionalized nanoparticles crystals

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By applying DNA as a ligand, nanoparticles can be ordered and grown into Wulff polyhedral single crystals due to the programmability based on the selective binding of DNA. However, it is difficult to grow a single crystal exceeding several μm. The reasons for inhibiting crystal growth are various. In this study, we investigated the effect of using deuterium oxide (D_2O) in the crystallization solution on crystal growth, D_2O has similar chemical properties to water, making the structure of DNA stable, but differs from water in physical properties. Furthermore, we demonstrated that larger DNA- functionalized nanoparticles (DNA-NP) crystals could be grown by adjusting the DNA-NP concentration and salt concentration.

Different DNA-NP crystallization experiments were conducted by solvent (water or phosphate buffer adjusted with D_2O), DNA-NP concentration, and salt concentration (0.3M, 0.5M, 1M, 1.5M, 2M) for binding the nanoparticles, respectively. The shape and size of the grown crystal were evaluated by an optical microscope and a scanning electron microscope (SEM), and the crystal structure was evaluated by small-angle X-ray scattering (SAXS) (Fig. 1).

The SAXS structure analysis, SEM observation of the crystal surface, and optical microscopy of DNA-NP crystals crystallized in water or phosphate buffer adjusted with D_2O are shown (Fig. 1). DNA-NP crystals crystallized with D_2O showed better crystal quality than water (Fig. 1a, b), and crystals with a size exceeding 50 μm were successfully synthesized (Fig. 1c). This study revealed that the type of buffer solution, DNA-NP concentration, and salt concentration had a significant effect on crystal size and crystal quality. Furthermore, the viscosity, density and surface tension of the solvent are thought to affect the frequency of nucleation and step growth, and more detailed mechanisms need to be elucidated.

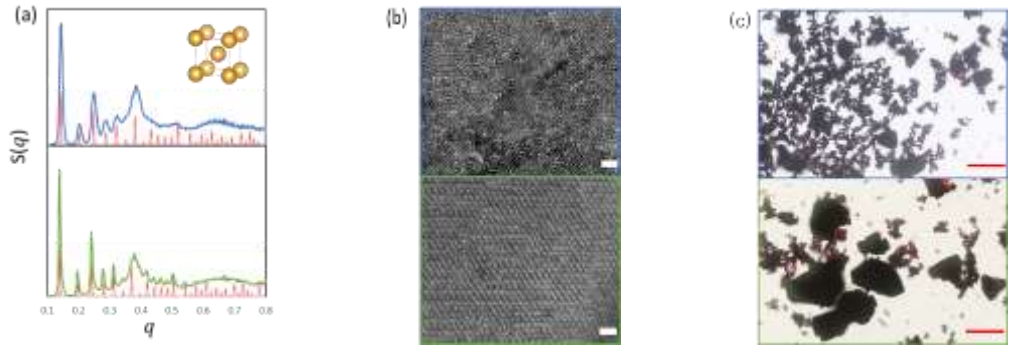

Fig. 1 Comparison of crystals grown in H_2O (upper) solution and D_2O solution (lower): (a) SAXS profiles of DNA-NP crystals (0.3M NaCl) $q = (4\pi/\lambda)sin\theta$ (scattering angle, 2θ); (b) SEM images of DNA-NP crystals after silica encapsulation. Scale bar is 100 nm. (c) Optical microscope images of DNA-NP crystals. Scale bar is 50 μm