

DNAガイドのナノ粒子結晶化：結晶化条件の最適化と構造解析

DNA-guided crystallization of nanoparticles: optimization of crystallization conditions and structure analysis

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Due to the programmability and self-assembly characteristic, DNA is a powerful tool for guiding the assembly of nanometer-sized particles into large-scale ordered structures. Recently, DNA-guided nanoparticle crystallization has been demonstrated and many types of crystal structures have been created^[1-3]. By changing particle size ratio, DNA length and complementarity, particles assemble into different types of crystals. By TEM imaging after silica encapsulations, Mirkin et al. confirmed that some of DNA-guided nanoparticle crystals assembled into wulff polyhedra in solution^[3]. However, it is crucially difficult to grow large-scale wulff polyhedral single crystal of DNA-functionalized nanoparticles (DNA-NPs) with size up to several microns. Herein, we demonstrate the optimization of solution composition on DNA-NP crystal growth, specifically the effect of sodium chloride concentration and polyethylene glycol (PEG) addition into DNA-NP colloidal solution.

Au nanoparticles of diameter 7 nm were functionalized with two different sequenced thiolated-DNA strands (DNAa and DNAb), respectively. These DNA-functionalized nanoparticles, DNA-NPa and DNA-NPb, were combined with linker DNA strands, which hybridize to complementary sequences of DNAa and DNAb (Figure 1). The mixed solutions were adjusted under different sodium chloride concentration (100, 300, 500, 700 and 900 mM) and by adding PEG with different molecular weights (1000, 4000 and 8000) and subsequently heated to 65 °C and then slowly cooled back to 25 °C. The crystal structure of assembled DNA-NP superlattices were analyzed by small angle x-ray scattering (SAXS) and imaged by scanning electron microscope (SEM).

We confirmed the crystal structure of assembled DNA-NP aggregates was bcc by analyzing peak positions of SAXS 1D curves. The differences were seen in SAXS and SEM data with different samples under different sodium chloride concentrations. Relatively large wulff polyhedral single crystals of DNA-NP superlattices were assembled in buffer solution under 500 mM NaCl. The significant differences were seen in SAXS and SEM data with different samples by adding PEG with different molecular weights. DNA-NP superlattices assembled in the buffer solution with PEG 1000 showed a clear SAXS 1D curve which indicates bcc structure (Figure 2) and a significant increase in average crystal size were confirmed by SEM (Figure 3).

We will also present the recent progress on the formation of lattice using DNA-NPs linked by DNA polyhedral constructs. DNA polyhedron has symmetrical connecting bonds and plays a determining role in the inter-particle bonding; that allows for control of particles coordination. DNA nanostructure-directed crystallization has a power of the proposed approach for assembly of complex nanoparticle lattices^[4].

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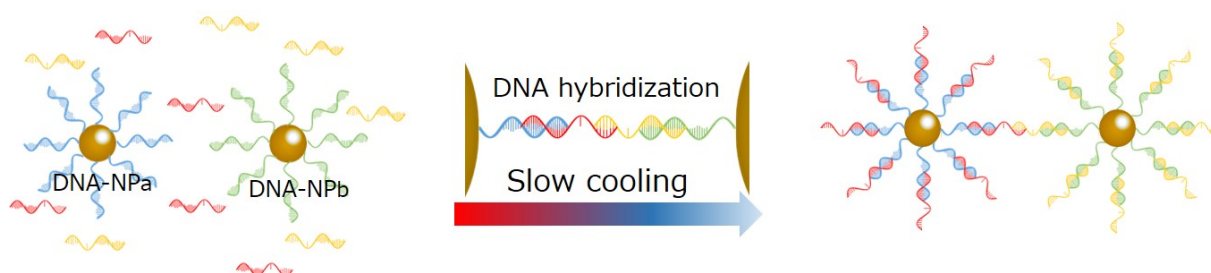


Figure 1. DNA-NP assembly

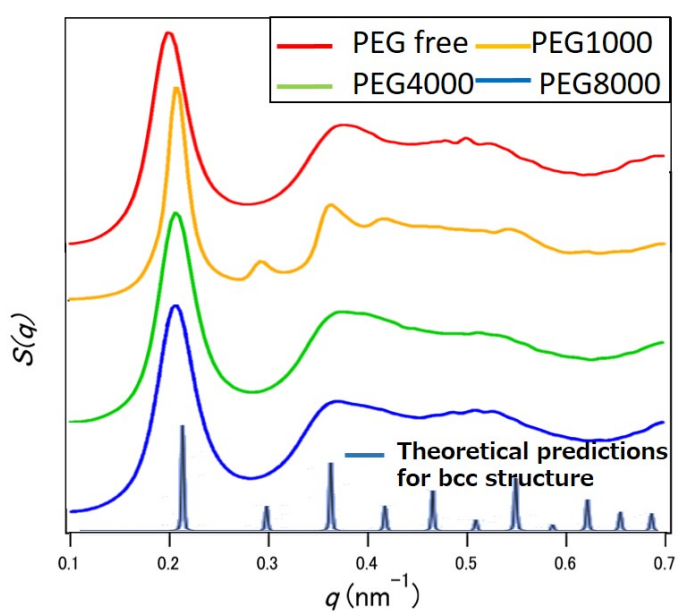


Figure 2. SAXS analysis of DNA-NP aggregates with PEG of different weights as additives

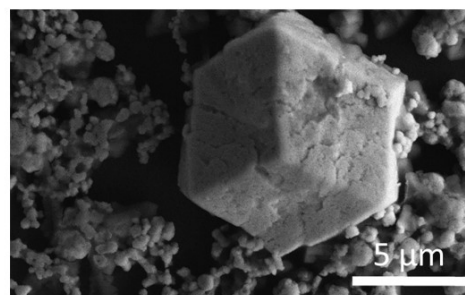


Figure 3. SEM image of DNA-NP superlattice