Biomolecular Machines as Functional Building Blocks in Programmed Nanostructures

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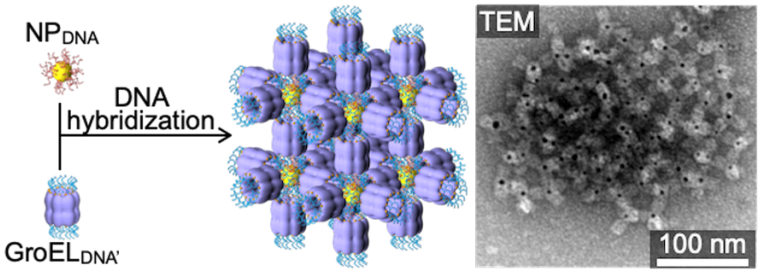
Biomolecular assembly is a requisite to the emergence and diversification of life on earth. However, considering the wide diversity of biomolecule structures and functions, relatively little has been applied to synthetic systems.[1]

Here we employ the biomolecular machine GroEL[2] as a supramolecular linker between nanoparticles to construct porous 3D clusters. GroEL is a large (800 kDa, 14.6 × 13.7 nm) barrel-shaped chaperonin protein that, in nature, assists in the refolding of denatured proteins with its hydrophobic cavities and ATP-dependant conformational motions.

We append DNA strands to the apical domains of GroEL[3] ($GroEL_{DNA}$) and complementary strands to 5-nm-diameter gold nanoparticles ($NP_{DNA'}$). The annealed mixture affords porous clusters with a single homeotropically-aligned GroEL unit linking NPs, as confirmed by TEM, cryoEM and cryoEM tomography. The small NPs accommodate a maximum of 6 $GroEL_{DNA}$ units, resulting in a 3D network with a NP-NP center-to-center distance of 33 nm. The large pores of the clusters, which are an order of magnitude larger than those of conventional supramolecular porous materials, permits the exploration of an expanded view of host-guest chemistry. The loading of a series of poly(ethylene glycol) (PEG)-appended GroELs as ultra large macromolecular guests is evaluated, showing a size-based guest selectivity on the order of several tens of nanometers.

Cluster growth could be monitored by dynamic light scattering (DLS) and manipulated in-situ by heating or adding either of the monomer species. Further, the GroEL units in the assembled state retain their ATPase activity, highlighting the dynamic nature of the clusters. Finally, the versatility of GroEL as a building block is demonstrated though the construction of porous clusters having 3 $GroEL_{DNA}$ units linking adjacent $NP_{DNA'}$.



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