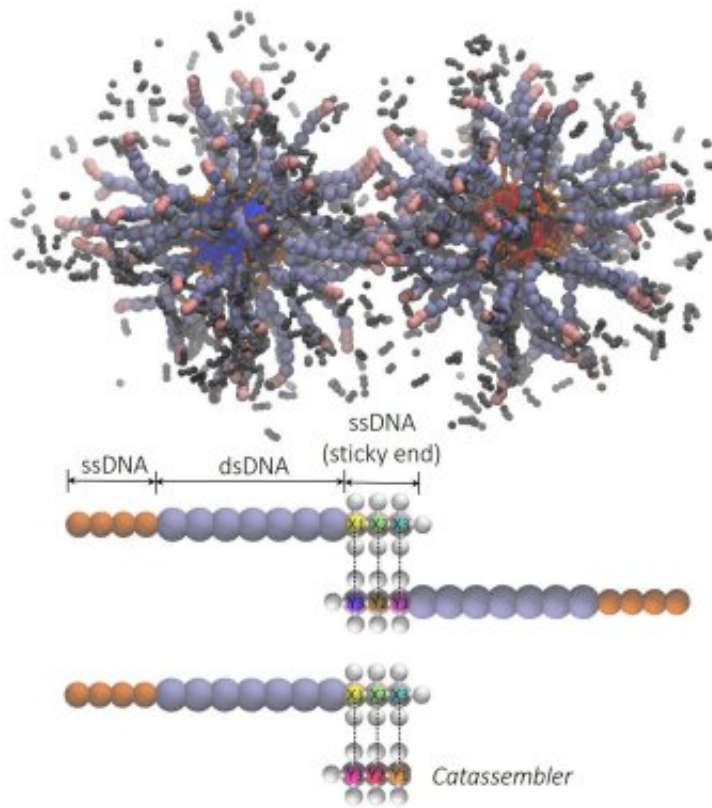


DNA facilitates escape from metastability in self-assembling systems

May 24 2023, by Liu Jia



Top: Coarse-grained model of two building blocks of the hybridized PAEs using the catassembly strategy. For clarity, only the backbone segments of sticky ends are shown. Bottom: Detailed sketch of the interaction between the non-self-complementary sticky ends on two types of PAEs (X1-Y3, X2-Y2, and X3-Y1) and the interaction between the non-self-complementary sticky ends on PAEs and Catassembler (X1-Z3, X2-Z2, and X3-Z1). Credit: *Proceedings of the National Academy of Sciences* (2023). DOI: 10.1073/pnas.2219034120

Prof. Liang Haojun from the University of Science and Technology of China (USTC) of the Chinese Academy of Sciences (CAS) proposed a new catalytic assembly approach to escape from metastable states in a far-from-equilibrium system of DNA-functionalized colloids. The study was published in the *Proceedings of the National Academy of Sciences*.

Self-assembly refers to the process in which assembled primitive elements (molecules, nanoparticles, etc.) spontaneously form ordered structures through non-covalent interactions. The excellent capacity of the system to create new materials has drawn attention. In an ideal assembly process, the system will reach a thermodynamically stable state with the lowest free energy and form a high-quality assembly structure. However, for the assembly system far away from the [equilibrium state](#), the system is prone to be stuck in metastability where the local free energy is extremely small, blocking the formation of a high-quality assembly structure.

How to circumvent metastability in a far-from-equilibrium system is regarded as a challenging conundrum in the field of [self-assembly](#). For DNA-functionalized nanoparticle assembly, a typical far-from-equilibrium system, the entropy-controlled thermal annealing strategy constitutes a traditional and generally adopted way to escape from metastability. Nevertheless, the aggregation and dispersion of nanoparticles usually occur over a narrow temperature span during annealing. In correcting misconnected non-covalent bonds, [thermal energy](#) is not selective. Thermal annealing is not conducive to the assembly of biologically active particles or under physiological conditions.

Inspired by the concept of "catassembly" proposed by academician Tian Zhongqun from Xiamen University, Prof. Liang and his team presented a new way to achieve catalytic-assembly of DNA-functionalized colloidal nanoparticles in a far-from-equilibrium system. Based on their

prediction on theoretical simulation and previous research results on constant enthalpy control strategy for nanoparticle assembly, they employed a removable molecule named a "catassembler," which serves as a catalyst, to adjust imperfect linkages and assist the system to escape from metastability while preserving assembled framework.

In this strategy, the short DNA strand acting as the accelerator has a direct competitive effect with the bonding end on the surface of the nanoparticles inside the assembly structure, and the non-covalent bond of the wrong connection could be corrected by the transient DNA strand replacement reaction, assisting the system to escape from the metastability. During the process, the accelerator would not destroy the overall skeleton of the assembly structure, and it could be removed from the final assembly structure. Moreover, by changing the structural design of the accelerator, it could even reduce the dosage of the accelerator and improve its efficiency.

On the basis of the same principle, superlattice structures with different crystal symmetries can be obtained by changing the kernel type of nanoparticles in a two-component system and directly adding the corresponding DNA accelerator after the design of the DNA sequence. This strategy makes the implementation of the assembly of [nanoparticles](#) easy to carry out as the chemical reaction occurs at a constant temperature.

Furthermore, this DNA accelerator regulation strategy is simple and effective enough that the "solid–solid" phase transformation between different colloidal crystals becomes easier to achieve, after breaking through the constraints of the temperature regulation and the initial phase state free energy. It displays its application potential in structurally reconfigurable "solid–solid" phase transformation bio-inorganic composites.

As a general method for regulating non-covalent interactions within assembly structures, the [accelerator](#) strategy proposed in this study is expected to be extended to the controlling and devising of assembly processes for other soft material systems (polypeptides, [block copolymers](#), etc.) that are far from equilibrium.

More information: Dongbao Yao et al, Catalytic-assembly of programmable atom equivalents, *Proceedings of the National Academy of Sciences* (2023). [DOI: 10.1073/pnas.2219034120](https://doi.org/10.1073/pnas.2219034120)

Provided by Chinese Academy of Sciences

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