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Study of DNA coated nanoparticles as possible programmable self-assembly building blocks

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Abstract

Nanoparticles coated with single stranded DNA have been shown to efficiently hybridize to targets of complementary DNA. This property might be used to implement programmable (or algorithmic) self-assembly to build nanoparticle structures. However, we argue that a DNA coated nanoparticle by itself cannot be used as a programmable self-assembly building block since it does not have directed bonds. A general scheme for assembling and purifying nanoparticle eight-mers with eight geometrically well-directed bonds is presented together with some preliminary experimental work.

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Keywords: Nanoparticles; Nanostructure; DNA coated nanoparticles; Programmable self-assembly; Gold particles

1. Introduction

DNA functionalized nanoparticles have been a prospect material for the construction of self-assembled structures since first demonstrated [1,2]. So far, the main interest in these particles has been to use them in detection methods for small amounts of specific DNA [3]. The structures formed in these, and other [4] experiments, are mainly periodic, or random agglomerations of particles. To fully exploit the potential of self-assembly of DNA coated nanoparticles one needs to address the problem of assembling non-periodic structures.

To implement programmable self-assembly (PSA) one needs building blocks of a certain complexity [5–9]. In short, the requirements are: (1) unspecific bonds, that can bind to several types of different building blocks, (2) an incoming building block must interact with at least two other, already assembled blocks and (3) at least four bonds on each block to avoid blocking of the assembly. For general discussions on criteria for PSA see for example [9–11]. Using a diamond like structure, it is possible to build three-dimensional structures using building blocks with only four bonds, but a more practical approach is to use triangular prisms or cubes having at least 5, respectively, 6 bonds (6, respectively 8 bonds, if the bonds are situated at the corners).

Building blocks that have a size of micrometers can be manufactured and functionalized using conventional micro-technology, one example of a such a top-down approach for the functionalization of two distinct areas of 1 μm particles can be found in [12]. However, with reducing dimensions, it is increasingly difficult to functionalize different parts of the building blocks with different functions making a top-down procedure no longer feasible. A new approach is needed. We here propose such a new method for the fabrication of nanoscale PSA-building blocks using only bottom-up methods.

2. Making PSA-building blocks from nanoparticles

Two sets of nanospheres of a suitable material are functionalized each with two different types of single stranded DNA (ssDNA). For example, for gold [13,14] and SiO_2 [15] nanoparticles, there are standard protocols for this. These spheres are then mixed in solution together with linker

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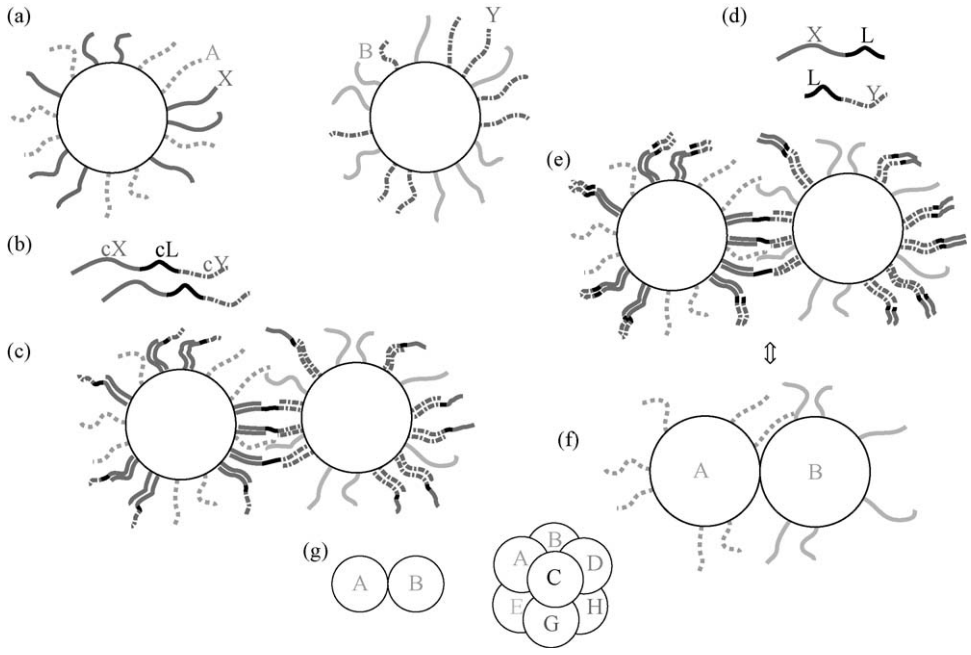

Fig. 1. Scheme for nanoparticle PSA-building block assembly. (a) Two different sets of nanospheres are each coated with two (or more) single stranded DNA sequences (A–X and B–Y). (b) Linker molecules. (c) Dimers can be extracted by mass separation. (d) Passivation of remaining sticky ends by complementary molecules (X–L and L–Y). (e) The only single stranded DNA left is of type A on one sphere and of type B on the other so the dimer in (e) is functionally equivalent to the one in (f). Using several iterations of this process will produce eight-mers (g) with eight separate and specific binding sites to be used for PSA.

molecules (Fig. 1a–e). By controlling the temperature of the solution we can promote the formation of dimers. After a while the linker DNA is rinsed away, stopping any further aggregation of the nanoparticles. The dimers are then separated from the rest of the aggregates by mass separation. The dimers produced in this way constitute simple PSA-building blocks with ssDNA of types A and B sticking out at different ends of the dimers (Fig. 1f and g). By starting out with particles coated with four specific DNA sequences and using several iterations of this technique one could make four-mers and eight-mers to be used as more advanced PSA-building blocks (Fig. 1g).

Following the above method one can create different basic building blocks with specific sticky ssDNA on different faces of the blocks. As the basic building blocks are mixed together with blocks having complementary DNA self-assembly will occur (Fig. 2).

Programmable self-assembly require simultaneous binding of two bonds to uniquely produce the desired structures. For example, in Fig. 2, the blocks 5 and 1 can connect to the 2 block. The right position is determined by matching two bonds. The A DNA's of both blocks 1 and 5 could, in principle, make a single bond to the cA DNA of block 2 but the alignment of the connected blocks would probably be wrong. This could be avoided by setting the temperature and/or the amount of stirring to values where blocks bonded by one bond are unstable structures whereas blocks bonded by two bonds are stable.

3. Materials and methods

Gold colloids with mean diameters ranging from 10 to 50 nm were purchased from G.Kisker GbR. Typical size

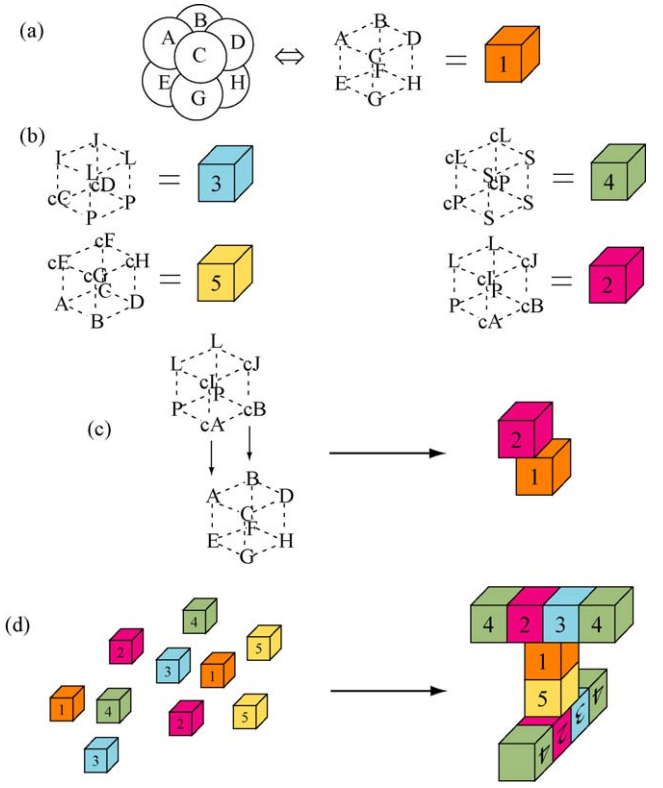

Fig. 2. Scheme of progammable self-assembly. (a) The eight-mers have eight specific ssDNA binding sites. (b) Four more eight-mers are depicted. (c) Example of binding: the cA and cB of block 2 binds to the A and B strands on block 1. (d) Structure formation. The "S" DNA of block 4 has no complement on the other blocks so it acts as a stop-block.