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COMMUNICATIONS

Published on Web 02/05/2004

Tensegrity: Construction of Rigid DNA Triangles with Flexible Four-Arm DNA Junctions

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This paper reports a physical realization of tensegrity at the nanometer scale. A stable tensegrity structure comes from a combination of tense and integrity,[1] instead of rigidly joining rigid building blocks. Ever since tensegrity structures were built by R. B. Fuller, they have fascinated many people.[2] This concept has been explored in structural applications. In this paper, we extend the concept of tensegrity to construct nanometer scale geometric structures, equilateral triangles, out of DNA.

Being able to control structures at the nanometer scale would facilitate the design and preparation of nanostructured materials and devices.[3,4] Biomacromolecules, DNA in particular, have long been recognized for their potential applications in nanoconstruction.[5,6] Intuitively, geometric structures or periodic arrays will result from rigidly joining rigid building blocks. This idea motivated searching for rigid branched DNA structures.[7a,b] Tensegrity, an alternative strategy, involves rigid struts and flexible tendons. Struts push outward, and tendons pull inward. The balance between these two forces leads to stable, rigid structures. A tensegrity strategy has been explored for construction of DNA triangles,[7c,d] imagining stiff DNA duplexes as struts and flexible single-stranded DNA loops as tendons. However, a ligation-closure analysis and unsuccessful attempts of 2D self-assembly suggested that the resulting triangles were not rigid, presumably because the tendons are not tense enough in that particular design.[7c,d] More recently, a similar method has been used to construct DNA tetrahedra,[7e] whose rigidity still remains to be examined. Here, we reexamine the tensegrity strategy with a different design to construct triangles.

Three four-arm junctions are fused together in a DNA nanotriangle (Figure 1a−c). Each vertex of the triangle consists of a four-arm junction, and each side of the triangle is a DNA duplex. Each individual four-arm junction has four helical arms, which stack into two pseudo-continuing DNA duplexes.[8a] Each pseudo DNA duplex is rigid, but the interhelical angle, the angle between the two pseudo DNA duplexes, is very flexible.[8b] The angle has an equilibrium value of ∼60°. In the triangular design, we take advantage of the flexibility of the interhelical angle. The crossovers in the four-arm junctions are used as tendons to join the rigid struts (pseudo-continuing DNA duplexes). The lengths of the three DNA duplexes will fully define the triangle geometry, including its three inner angles, which are also the interhelical angles of four-arm junctions. The inner angles of an equilateral triangle are 60°, the same as the equilibrium value of the interhelical angle of a four-arm junction. Thus, four-arm junctions in the context of an equilateral triangle still remain in their native conformations, which would increase the triangle stability.

The designed DNA triangles were formed by slowly cooling an equimolar mixture of the component DNA strands. The triangles were well-behaved. We characterized this triangle structure first with native gel electrophoresis (Figure 2). The DNA triangles appeared as a single sharp band, indicating that the constructed DNA triangle was a stable structure. Thermal denaturation analysis

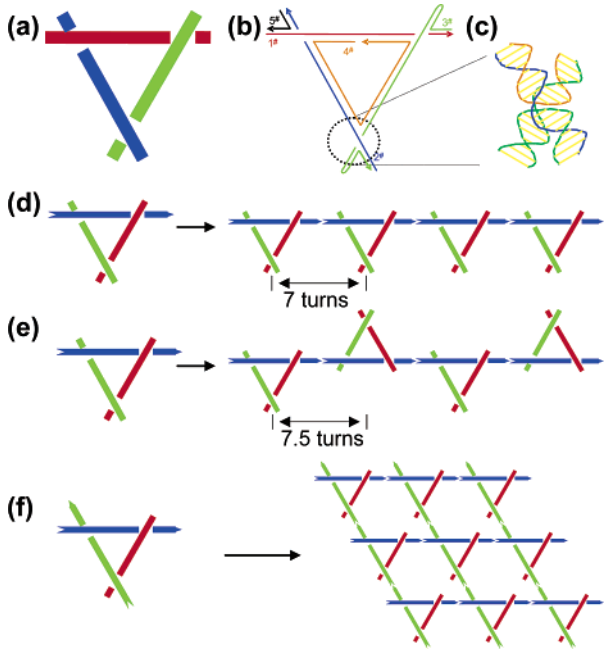

Figure 1. The design of a DNA triangle. (a) A DNA triangle contains three DNA duplexes, shown as rods with different colors. (b) Strand structure of a DNA triangle. Each thin line represents a single DNA strand. An arrow indicates the 3′ end of a DNA strand. (c) The detailed structure of a triangle vortex. (d and e) 1D self-assembly of DNA triangles. (f) 2D self-assembly of DNA triangles. The DNA sequence complementary is represented with the geometric complementary.

further demonstrated that the triangle was a stable structure. The optical absorption at 260 nm remained unchanged up to 60 °C (see Supporting Information).

Atomic force microscopy (AFM) imaging provided more direct information about the DNA triangle structure. Individual triangles appeared as scattered particles with uniform size and triangular shape (see Supporting Information), which suggested that the particles had the same structure. The detailed triangular structure was not able to be resolved due to the limited AFM resolution.

Self-assembly of the triangles into arrays further demonstrated our design strategy. A pair of sticky ends was added to one duplex, so the triangles could associate with one another into one-dimensional (1D) arrays (Figure 1d,e). Figure 3a and b shows the AFM analysis of such arrays. The observed spacings between the triangles are consistent with the designed values. Besides linear arrays, some cyclic assemblies exist. This phenomenon is expected, because the rigid triangles are linked by more flexible, doubly nicked duplexes. To reduce cyclic assemblies, the duplex linkers should be replaced by stiffer structures, for example, double crossover molecules. The relative twists between two consecutive triangles are designed to be 0° (Figures 1d and 3a) and 180° (Figures 1e and 3b), but not optimized, thereby accounting for the phase

10.1021/ja031754r CCC: $27.50

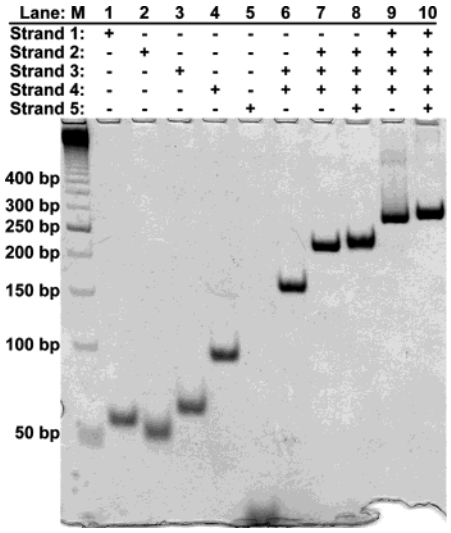

Figure 2. Native gel electrophoretic analysis of the formation of individual DNA triangles. Each lane contains an annealed equimolar mixture of DNA triangle component strands, which are indicated at the top. Lane M contains linear DNA duplex size markers.

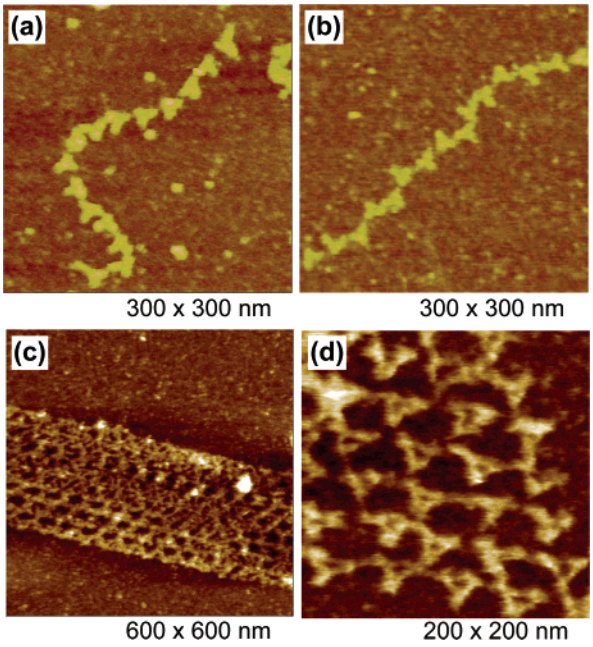

Figure 3. AFM images of DNA triangle arrays: 1D arrays with a periodicity of 7 (a) and 7.5 turns (b), and 2D arrays (c and d).

variation observed. Sample−substrate interaction during AFM sample preparations also complicated the 1D array morphologies.

Rigidity of the constructed DNA triangle was further confirmed by 2D self-assembly into periodic arrays. When two pairs of complementary sticky ends were added to two duplexes, respectively, rigid DNA triangles could assemble into 2D arrays (Figure 1f). Otherwise, irregular aggregates would form. AFM analysis clearly proved our hypothesis: this DNA triangle is rigid. Periodic arrays formed (Figure 3c). These arrays usually were several micrometers long, but not very wide (see Supporting Information). Occasionally, we reached very high resolution, and each individual triangle was well resolved (Figure 3d).

In summary, we have constructed nanotriangles out of DNA with a tensegrity strategy. These structures expanded the available DNA structures for nanoconstructions. Future goals include the construction of nonequilateral triangles, use of the nanotriangles to build sophisticated structures, and the design of triangles for 3D self-assembly. The structure presented here differs from previously reported DNA tiles by being nonplanar (Figures 1a and S6 in Supporting Information). The three component duplexes extend in three directions, much like the axes of a 3D coordinate system. Thus, it is possible to use DNA duplexes to guide DNA array growth in three discrete directions, thereby enabling 3D self-assembly, a key goal of nanotechnology.[5] The tensegrity strategy developed here may become an important strategy for designing biomimetic nanomaterials.[9]

Acknowledgment. We thank Prof. N. C. Seeman and P. E. Constantinou for intriguing discussions, Dr. A. Ribbe for help with AFM imaging, and Prof. D. R. McMillin for use of a UV−vis spectrophotometer. This work is supported by DARPA (MDA-972-03-10020), NSF (EIA-0323452), and Purdue University.

Supporting Information Available: Experimental methods, DNA sequences, thermal profiles, and AFM images (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.

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JA031754R