TILE BASE DNA LATTICE GROWTH FOR NANODEVICE APPLICATIONS

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ABSTRACT:: *The development of programmable growth patterns with predictable self-assembly at the molecular scale is a key target of nanotechnology. We present here a DNA nanostructure, Scaffold Cross Tile consisting of 4×4 arm junction with a square aspect ratio. The 8 ssDNA oligonucleotides are designed in a way that the adjacent strands with sticky ends association can produce continuous arms both in X and Y direction representing tile-based origami. The self-assembled scaffold cross tiles result in two-dimensional nanogrids with periodic square cavities. The Scaffold cross tile lattice structure is not only assembled in free solution but also grown on mica substrate. A mica-assisted crystal growth phase diagram is analyzed by atomic force microscopic images with rational growth analysis by annealing SXT in different concentrations. These 4x4 scaffold nanogrids may prove to be useful as a nanoscale contact mask for transferring metallic and ferromagnetic square patterns onto a silicon substrate through vapor deposition.*

Keywords: DNA, Self assembly, Nanotechnology, Mica, Substrate Growth

1. INTRODUCTION

We are living in the age of nanotechnology, where great research and development is in progress in almost all fields of life including materials, photonics, medicine, biotechnology and electronics. This requires a wide variety of materials to fulfill the researchers and industrial growing demands. DNA is the key component in this contest because of its programmable and predictable self-assembly and great potential in the fabrication of nanoscale structures and devices [1-3]. Over the last three decades, DNA has been under research and development for assembling periodic 1D, 2D and 3-D patterns and their manipulation in nanodevices and molecular electronics [4-9]. Highly ordered DNA motifs and structures have a great potential to provide ultra-fine frameworks for the next generation nanofabrication. All of these applications may be predicated on the complementarity of two DNA strands, embedded in the hydrogen-bonded base-pairing linking adenine (A) and thymine (T), guanine (G), and cytosine (C) [10,11]. DNA provides a smart route for the creation of nano architectures with programmable and predictable patterns. DNA strands twist along one helix for a number of bases before switching to the other helix by passing through a crossover junction. The association of two crossovers holds the helices in their parallel and also grips the DNA helices tightly together, allowing the assembly of larger structures [12-15]. Because of its incredible chemical and physical properties, it stands at the forefront in the current research and development scenario.

Two different approaches are adopted for larger nanostructure fabrication; tile-based assembly [16] and a single long DNA strand folding into multi-shapes or dimensions called DNA origami [17,18]. Here we present a unique DNA 4×4 scaffold tile structure comprised of 8 ssDNA oligonucleotides; four strands include a sticky-end part for binding with neighboring tiles and the four strands act as inner crosslinkers to shape a square pattern. Each DNA strand is positioned by a half-full turn (HFT) base-pair length that is 0.34nm. The recurring distance of ~20nm is calculated from the DNA model assuming 0.34 nm/base pair for the pitch and 2 nm for the diameter of a DNA duplex, respectively. The scaffold cross tile (SXT) is designed in a way that it can grow incessantly two-dimensional (2D) large DNA grid structure in both X and Y direction as shown in figure 1. We have placed T4 loops at each of the four corners within the tile center allowing the strands to twist in four different directions (figure 1). The complex SXT design aids the formation of larger structures impersonating DNA origami, unlike the cross tile motif presented by Hao Yan et. al. [19]. The scaffold design of SXT overcomes the binding rate-limiting factor that prevents the previous cross tile to assemble the rigid and larger structures in a continuous fashion. Also, we are introducing DNA crystal growth characterization by measuring its optical properties as ssDNA oligos and later annealed SXT in free solution.

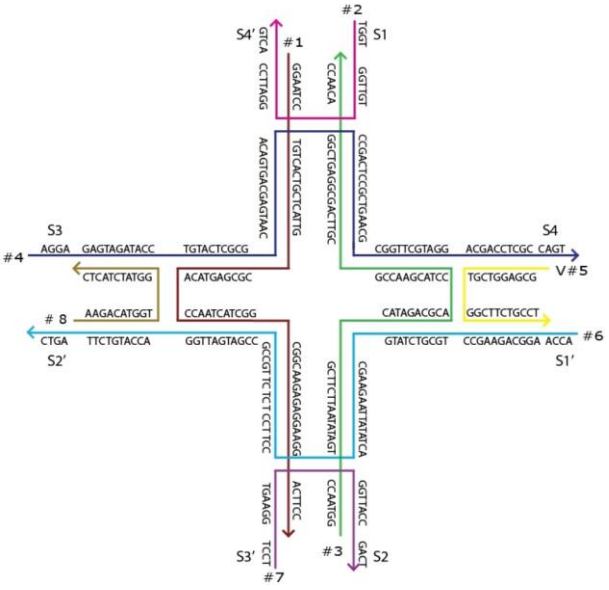

Fig. 1. The Scaffold Cross Tile (SXT) Structure. The sequence map of the SXT with the details of base pairs (bp) number along strand direction with sticky ends (S) and complementary sticky ends (S`).

Fabrication Technique.

2.1 SXT free solution annealing

The DNA base sequence for 4×4 SXT was designed with the SEQUIN [20] program to reduce undesired complementary association during structure fabrication. The base sequences of the eight oligonucleotides are explained in supporting information in figure S1 and table S1. Synthetic oligonucleotides, purified by High-Performance Liquid Chromatography (HPLC), were obtained from the Integrated DNA Technologies, IDT (Coralville, IA, USA). SXT complexes were assembled with 200nM concentration by mixing a stoichiometric quantity of each strand in physiological 1×TAE/Mg^{2+} buffer [Tris-Acetate-EDTA (40 mM Tris (pH 8), 1 mM EDTA) with 12.5 mM magnesium acetate]. For SXT nanogrid annealing, the equimolar mixtures of 8 strands were placed at 95 °C and then slowly cooled down to 25 °C (room temperature) in 1.5 L of boiled water in a Styrofoam box for at least 24 hours to facilitate hybridization. After annealing, the samples were stored at 4°C for structure stabilization and Atomic force microscopic (Nanoscope III, Veeco, USA) analysis in liquid tapping mode under 1×TAE/Mg^{2+} buffer. The SXT design with sticky ends cohesion, the SXT motifs self-assembled into extended, periodic square arrays. The nanogrid arrays showed highly ordered fabrication, as analyzed by the atomic force microscopy (AFM) images (figure 2 (a-d)). The height of SXT normal to the plane is 1.2nm and both the vertical and horizontal planes are ~20nm in length.

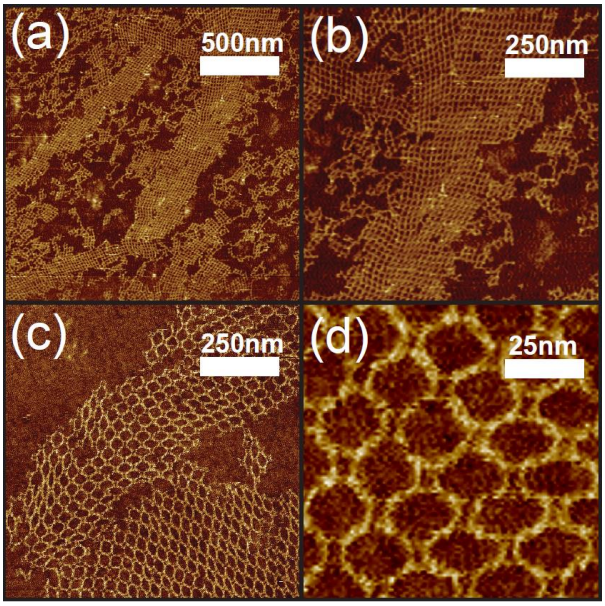

Fig. 2. Atomic Force Microscopic (AFM) images of SXT in free solution (a) Self-assembled SXT grid lattice with a scan size of 2µm×2µm.

2.2 SXT mica assisted growth (MAG)

For the assembly of homogeneous, large, and logically controlled structures on a functional substrate with a logical analysis of the growth pattern, we have grown the SXT nanogrid on mica substrate [21]. For the demonstration of the growth pattern, five different oligos concentrations were employed to assemble SXT nanogrid on mica substrate. Mica assisted Growth (MAG) has a unique fabrication scheme,; starting from seeding, tile formation, and growth of well-established lattice structure with optimum concentration. For SXT-MAG, after preparing the DNA oligos in desired concentration a mica sheet of 0.5cm x 0.7cm was also inserted into the test tube. These mica-containing oligos were annealed in a Styrofoam box with hot water at 95°C and cooling down to room temperature (25°C) and stored at 4°C for AFM. At 95°C the DNA base pair hydrogen bonds are broken and the DNA strands exist in individual phases. During the cooling process, a number of single strands initiate seeding on the mica substrate randomly, and subsequently the complementary strands hierarchically assemble to the seeds for lattice formation. The electrostatic interaction between DNA oligos and mica sheets creates a favorable environment for lattice growth. The MAG process is schematically explained step by step in figure3 (a).

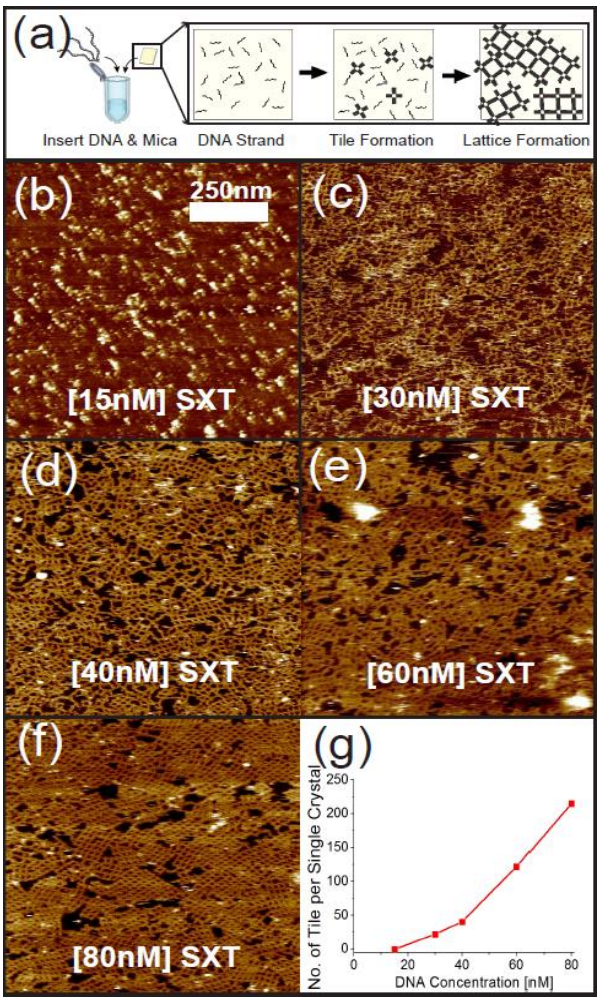

Fig. 3. Mica assisted growth (MAG) method for SXT growth on mica substrate with rational growth pattern analysis (a) SXT mica assisted growth pattern explanation (b) Single strands seeding on mica surface with a 15 nM DNA oligos concentration (c) The single strands start tile formation at 30 nM concentration. (d) The SXT grid starts appearing as the oligos concentration increase up to 40nM. (e) Large size lattice appeared at 60 nM concentration but still, a precise pattern is not seen (f) the optimum concentration for SXT lattice growth is 80nM, where a nice grid structure is observed. (g) The analysis of SXT growth pattern on mica surface with DNA concentration increase and the number of tile participating in the structural growth.

MAG technique is designed to obtain a large-scale growth and to cover the mica substrate with controlled lattice growth. However, when we applied this method with SXT, we were able to get DNA structure in polycrystalline form, not a large single crystal. This type of crystal growth is a result of the MAG crystal growth mechanism. With an increase in DNA oligos concentration, the tile seeding on the mica surface is also increased causing hierarchically assembled small structures rather than large size single lattice structures. We made a systematic analysis of the number of tiles per single crystal of SXT from AFM images (figure 3(b-f)) and obtained a trend crystal growth curve for SXT as shown in figure 3(g). At 15nm DNA oligos concentration, the mica surface seeding initiates, and with an increase in concentration up to 30nM the SXT nanogrid starts to appear on the mica surface. The DNA SXT nanogrid covers the mica surface with an exponential increase in concentration and at 80nM DNA is the optimum concentration with ~215 tile/crystal and we can see almost fully covered polycrystalline SXT lattices.

3. SXT growth characterization

3.1 Experiment setup.

In this paper, we are also introducing the optical characterization technique for DNA structure growth verification. To validate the SXT growth optically, a lab-developed, polymer optical fiber (POF) with a micro cuvette-based spectroscopic system was used for the highly sensitive analysis. The schematic diagram of the experimental setup is illustrated in supporting information in figure S2. A standard quartz micro cuvette with a path length of 1 cm is used for the ssDNA oligos and SXT nanogrid structure, both with a DNA 200 nM concentration. A polymer optical fiber (POF, Mitsubishi Rayon, Japan) is used to transmit the light into the micro cuvette and deliver the transmitted light through the DNA samples to the spectrometer (Avaspec - 2048, Avantes). The POF has a core diameter of 980 µm and a cladding thickness of 20 µm. The spectrometer with a wavelength range of 200 nm - 1100 nm was connected to the computer for the evaluation of intensity, absorbance, and transmittance. All experiments were performed in a dark room at room temperature.

4. Summary

In this research article, we have explained DNA lattice growth in free solution and on mica surface to create a DNA nanogrid. Scaffold cross tile lattice growth has great potential in device fabrication for nanosensor and molecular machine applications. This scaffolding structure is expected to have a profound impact on future molecular device fabrication and nanoelectronics applications.

REFERENCES

[1] Gothelf K V and LaBean T H 2005 DNA-programmed assembly of nanostructures *Org Biomol Chem* **3** 4023-37

[2] Park S H, Yan H, Reif J H, LaBean T H and Finkelstein G 2004 Electronic nanostructures templated on self-assembled DNA scaffolds *Nanotechnology* **15** S525-S7

[3] Pistol C and Dwyer C 2007 Scalable, low-cost, hierarchical assembly of programmable DNA nanostructures *Nanotechnology* **18** -

[4] Dittmer W U, Reuter A and Simmel F C 2004 A DNA-based machine that can cyclically bind and release thrombin *Angew Chem Int Edit* **43** 3550-3

[5] Goodman R P, Heilemann M, Doose S, Erben C M, Kapanidis A N and Turberfield A J 2008 Reconfigurable, braced, three-dimensional DNA nanostructures *Nat Nanotechnol* **3** 93-6

[6] Park S H, Barish R, Li H Y, Reif J H, Finkelstein G, Yan H and LaBean T H 2005 Three-helix bundle DNA tiles self-assemble into 2D lattice or 1D templates for silver nanowires *Nano Lett* **5** 693-6

[7] Reif J H and LaBean T H 2007 Autonomous programmable Biomolecular devices using self-assembled DNA nanostructures *Commun Acm* **50** 46-53

[8] Turberfield A J, Mitchell J C, Yurke B, Mills A P, Blakey M I and Simmel F C 2003 DNA fuel for free-running nanomachines *Phys Rev Lett* **90** –

[9] Yin P, Yan H, Daniell X G, Turberfield A J and Reif J H 2004 A unidirectional DNA walker that moves autonomously along a track *Angew Chem Int Edit* **43** 4906-11

10] Sa-Ardyen P, Vologodskii A V and Seeman N C 2003 The flexibility of DNA double crossover molecules *Biophys J* **84** 3829-37 [11] Seeman N C 2003 DNA in a material world *Nature* **421** 427-31

[12] Chelyapov N, Brun Y, Gopalkrishnan M, Reishus D, Shaw B and Adleman L 2004 DNA triangles and self-assembled hexagonal tilings *J Am Chem Soc* **126** 13924-5

[13] Ding BQ, Sha R J and Seeman N C 2004 Pseudohexagonal 2D DNA crystals from double crossover cohesion *J Am Chem Soc* **126** 10230-1

[14] Niemeyer C M 2000 Self-assembled nanostructures based on NA: towards the development of nanobiotechnology *Curr Opin Chem Biol* **4** 609-18

[15] Shen Z Y, Yan H, Wang T and Seeman N C 2004 Paranemic crosover DNA: A generalized Holliday structure with applications in nanotechnology *J Am Chem Soc* **126** 1666-74

[16] Lin C X, Liu Y, Rinker S and Yan H 2006 DNA tile based self-assembly: Building complex nanoarchitectures *Chemphyschem* **7** 1641-7

[17] Rothemund P W K 2006 Folding DNA to create nanoscale shapes and patterns *Nature* **440** 297-302

[18] Rothemund P W K 2006 Scaffolded DNA origami for nanoscale shapes and patterns *Abstr Pap Am Chem S* **231** –

[19] Yan H, Park S H, Finkelstein G, Reif J H and LaBean T H 2003 DNA-templated self-assembly of protein arrays and highly conductive nanowires *Science* **301** 1882-4

[20] Seeman N C 1990 De novo design of sequences for nucleic acid structural engineering *J Biomol Struct Dyn* **8** 573-81

[21] Hamada S and Murata S 2009 Substrate-Assisted Assembly of Interconnected Single-Duplex DNA Nanostructures *Angew Chem Int Edit* **48** 6820-3