Supplementary Material (ESI) for Chemical Communications

## Subtractive Assembly of DNA Nanoarchitectures Driven by Fuel Strand Displacement

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### Material and methods:

#### **Structure Design**

The sequences of the 4-armed cross-shaped tiles and the 8-helix bundle tiles were copied from our previously published work (S1). Random sequences of 7-base or 8-base were chosen as toe-hold regions. 15-base long fuel strands were designed to be fully complementary to the sticky-end 3' and the toe-hold region. The strand sequences used are given below.

#### Fixed-size DNA Array Assembly

DNA strands were purchased from Integrated DNA Technology (www.idtdna.com) and purified via denaturing PAGE. The concentration of each strand was estimated by measuring  $OD_{260}$ . Each individual tiles were assembled by mixing a stoichiometric quantity of the strands involved in the tile in 1xTAE/Mg buffer (20 mM Tris, pH 7.6, 2mM EDTA, 12.5 mM MgCl<sub>2</sub>). The final concentration of each strand was 1.0  $\mu$ M. The DNA mixtures were cooled slowly from 90°C to room temperature in 2L water placed in a styrofoam box over 16 hours to facilitate hybridization. Non-denaturing PAGE gel was used to confirm the assembly of each individual tiles. Then a stoichiometric volume of the tiles were mixed with the final concentration 0.11  $\mu$ M for each 9-tile array and 0.04  $\mu$ M for each 25-tile array. The mixture was program cooled from 40 to 10°C in a PCR machine. The cooling was cycled 5 times in a 5°C step at a speed of 0.2 degree per minute. The initial moderate heating and cycled slow cooling was chosen to balance the needs between avoiding disassembly of the tiles, and eliminating the possible mismatches among the different sticky ends of the tiles.

#### Subtractive Assembly

Subtractive assembly was executed by adding 10 times quantity of the fuel strands to the assembled arrays in 1xTAE/Mg buffer. The reaction mixtures were incubated at room temperature for 20 hours before AFM imaging.

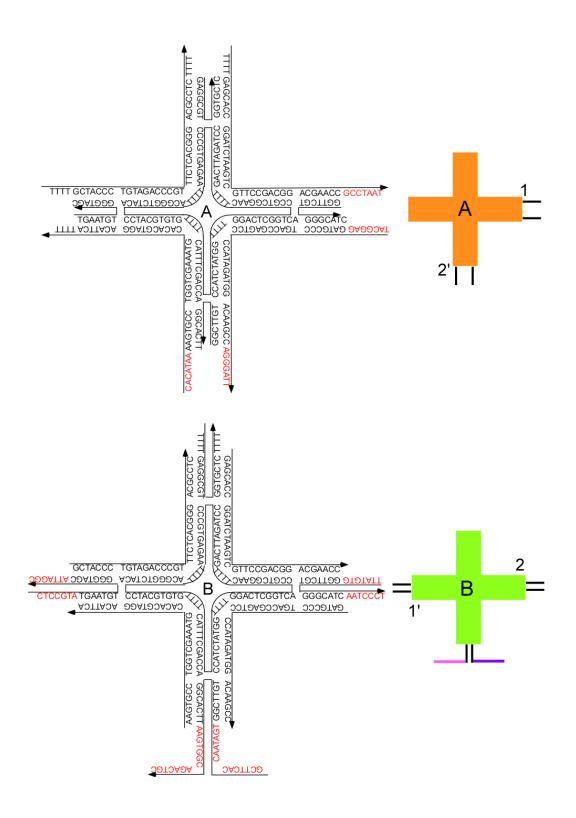
#### **AFM Imaging**

To prepare the sample for AFM imaging, 2  $\mu$ L of 1 mM NiCl<sub>2</sub> solution was first spotted on a piece of freshly cleave mica (Ted Pella, Inc.), and left to adsorb on the surface for 2 min. The Ni<sup>2+</sup> adsorbed on mica surface can help the DNA array stay on the surface during the scanning (S2). Then 2  $\mu$ L of the sample (10 times diluted in 1xTAE/Mg buffer) was added to the spot and left for binding to the mica surface for 2 min. Finally, 30  $\mu$ L 1xTAE/Mg buffer was added onto the mica. Imaging was performed under 1xTAE/Mg buffer on diMultiMode V AFM (Veeco Inc.) in TM AFM mode, using the tip on the thinner and shorter cantilever of the NP-S tips (Veeco Inc.).

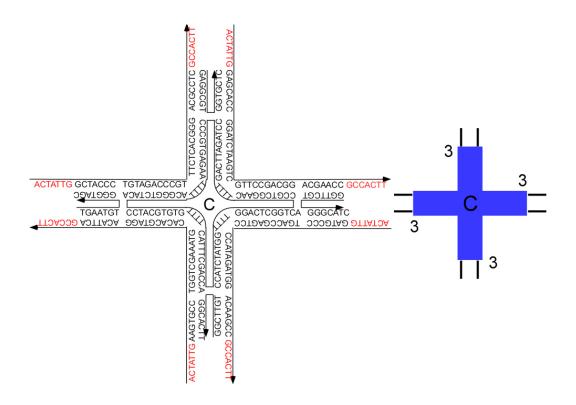
#### **Reference:**

- S1 Liu, Y.; Ke, Y.; Yan, H. Journal of the American Chemical Society 2005, 127, 17140-17141
- S2 Hansma, H.G.; Laney, D.E. Biophysical Journal, 1996, 70, 1933-1939





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Sequences of two fuel strands used to remove the C tile in the 3x3 tile array:

5'-CACTATTGGTGAAGC;

5'-GCAGTCTGCCACTTA.

# DNA strand structure and sequences used in Fig. 2

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#### Supplementary Material (ESI) for Chemical Communications This journal is (c) The Royal Society of Chemistry 2008

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Sequences of the two fuel strands used to remove A tile in the 5x5 tile array: 5'-AGTCAGCCTTATGTG; 5'-AGGGATTTCAACTTC.