Supplementary Material (ESI) for Chemical Communications

Self-sorting in DNA Self-assembly

Yu He, Ye Tian, Yi Chen, Alexander E. Ribbe and Chengde Mao*

Purdue University, Department of Chemistry, West Lafayette, Indiana 47907, USA

Supplementary Information

Experimental section:

Oligonucleotides. The oligonucleotides were purchased from Integrated DNA Technologies, Inc., and purified by denaturing polyacrylamide gel electrophoresis. Strand blue: 5'-Agg CAC CAT CgT Agg TTT TCT TgC CAg gCA CCA TCg Tag gTT TTC TTg CCA ggC ACC ATC gTA ggT TTT CTT gCC Agg CAC CAT CgT Agg TTT TCT TgC C-3'; Strand black: 5'-Agg CAC CAT CgT Agg TTT CTT gCC Agg CAC CAT CgT Agg TTT CTT gCC Agg CAC CAT CgT Agg TTT CTT gCC Agg CAC CAT CgT Agg TTT CTT gCC-3'; Strand red: 5'-ACT ATg CAA CCT gCC Tgg CAA gCC TAC gAT ggA CAC ggT AAC g; Strand green: 5'-CgC gCg TTA CCg TgT ggT TgC ATA gTC ATg-3'.

DNA self-assembly. Mixed strands with different molar ratios were combined in a Tris-Acetic-EDTA-Mg^{2+} (TAE/Mg^{2+}) buffer followed by slowly cooling from 90°C to room temperature over 48 hours. The buffer contained: 40 mM Tris base, pH 8.0, 20 mM acetic acid, 2 mM EDTA and 5 mM $Mg(Ac)_2$. The sum of the concentrations of the blue and black strands was kept constant at 0.6 μM. The detailed DNA concentrations in each sample are listed below:

I:**II**	Blue strand	Black strand	Red strand	Green strand
0:100	0	0.60 μM	1.80 μM	1.80 μM
5:95	0.03 μM	0.57 μM	1.83 μM	1.83 μM
10:90	0.06 μM	0.54 μM	1.86 μM	1.86 μM
20:80	0.12 μM	0.48 μM	1.92 μM	1.92 μM
30:70	0.18 μM	0.42 μM	1.98 μM	1.98 μM
40:60	0.24 μM	0.36 μM	2.04 μM	2.04 μM
80:20	0.48 μM	0.12 μM	2.28 μM	2.28 μM
100:0	0.60 μM	0 μM	2.40 μM	2.40 μM

AFM imaging. DNA samples (drops of 4 μL solution) were deposited onto freshly cleaved mica (Ted Pella,Inc.) and left to adsorb to the surface for 3 min. 20 μL TAE/Mg^{2+} buffer was then added to the mica surface. Imaging was performed in a fluid cell in tapping mode on a Multimode NanoScope IIIa (Digital Instruments) with NP-S tips (Veeco,Inc .). The tip velocity was kept at 10 μm/s, otherwise a scan frequency of 1 Hz or lower was used. The tip-surface interaction was minimized by optimizing the scan set-point.

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Figure S1. DNA nanostructures generated from pure cross motif at low concentrations. The zoom in images on the right show the double layered structures.

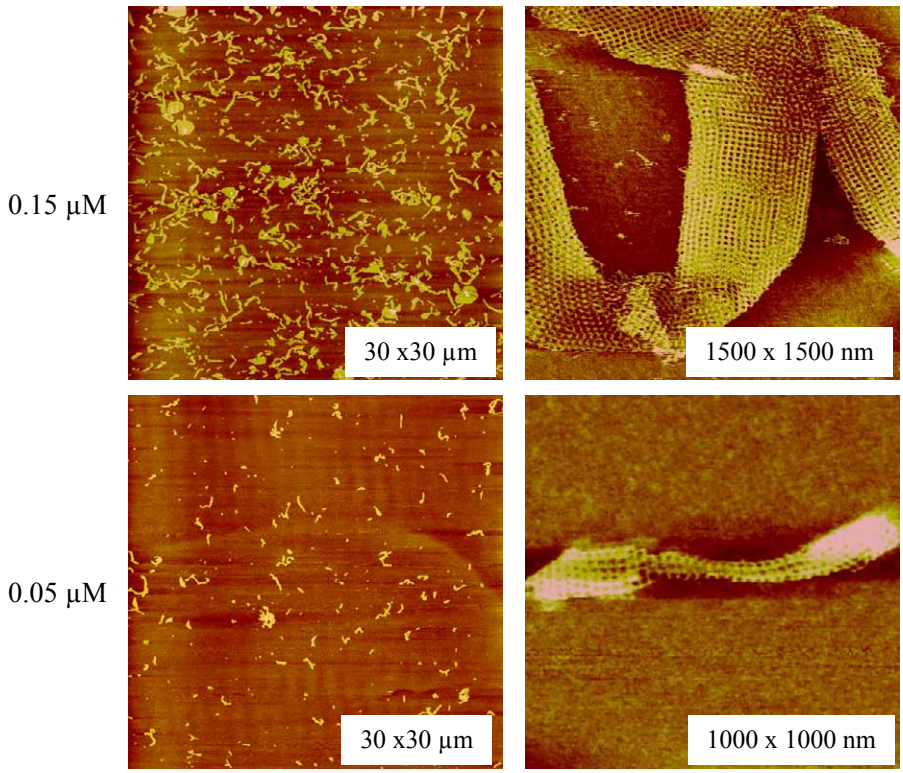

Figure S2. Foreign tiles are incorporated at the edge as indicated by arrows. This is a sample with a **I**:**II** ratio of 20:80.