Supplementary Material for Chemical Communications

Supplementary data

Capture of Double-Stranded DNA in Stacked-Graphene: Giving New Insight into the Graphene/DNA Interaction

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Reagents and Instruments

DNA samples were purchased from Takara Biotechnology Co. (Dalian, China) purified by high-performance liquid chromatography (HPLC). The DNA sequences are as follows: 5'-Cy3-AATGTTCGATGCTGACGGTCCATATGGACCGTCAA-3'; 5'-TTGACGGTCCATATGGACCGTCA GCATCGAACATT-3'. All reagents were of analytical reagent grade and purchased from the Kemiou Agent Co., Tianjin (China). Ultrapure water obtained from a Millipore water purification system (resistivity > 18.0 MΩ cm^{-1}, Laikie Instrument Co., Ltd, Shanghai, China) was used throughout the experiments. Phosphate buffer solution (PBS, 20 mM) with pH 7.4 was prepared by mixing the stock solution of Na_2HPO_4 and NaH_2PO_4. FL measurements were performed using a Hitachi F-4500 spectrofluorimeter with a scan rate at 2400 nm/min. The excitation wavelength was set at 540 nm. The slits for excitation and emission were set at 5 nm/10 nm. Zeta-potential and Z-average diameter were measured by Zetasizer nano series Nano-ZS90 (Malvern) at 25 °C with a detector angle 90°. Zeta potentials were recorded in a cell containing 8 μg/mL or 16 μg/mL in 20 mM PBS buffer (pH 7.4, 50-400 mM NaCl) at 25 °C. The morphology and height profile of stacked-CCGs were measured by atomic force microscopy (AFM, Agilent PicoPlus II) in liquid phase.[1] The samples were prepared by dropping by dropping colloidal stacked-graphene on a positively charged mica surface which was firstly modified by immersing into 0.1% cationic polyethylenimine solution. As shown in the following Fig. S3, typical morphologies of stacked-CCGs were able to be observed with non-uniform height and size. As noted, dsDNA can even be observed in stacked-CCGs rather than in CCGs despite of the broad height distribution of stacked-CCGs.

Preparation of Chemically Converted Graphene Sheets (CCGs) and Colloidal Stacked-CCGs

Graphene oxide was firstly prepared by our reported method.[2] An environment-friendly hydrothermal route was employed to convert graphene oxide to CCGs (0.15 mg/mL).[3] In the following, the resultant CCGs colloids with different concentrations was added to PBS buffer solution (20 mM, pH 7.4) in the presence of NaCl (0-500 mM). Zeta-potential and Z-average diameter of the assembled stacked-CCGs were then measured, respectively.

Preparation of Cy3-labeled dsDNA

For the preparation of dsDNA, 2.8 μM Cy3-labeled ssDNA and its perfectly complementary target (2.8 μM) were mixed in 20 mM PBS buffer (pH 7.4). Then the reaction was performed in a thermal cycle under the following conditions: 10 cycles of 95 °C for 30 s and 25 °C for 120 s. The obtained Cy3-labeled dsDNA were stored at 4 °C.

Capture of Double-Stranded DNA in Stacked-CCGs

In a typical experiment, Cy3-labeled dsDNA was firstly incubated in PBS buffer (20 mM, 50-500 mM NaCl, pH 7.4, 25 °C), then colloidal CCGs (8 μg/mL or 16μg/mL) were added to the mixture for the time-dependent fluorescence measurement at $\lambda_{ex}/\lambda_{em}$ = 540/568 nm. The control experiment was carried out under the same condition without the presence of NaCl.

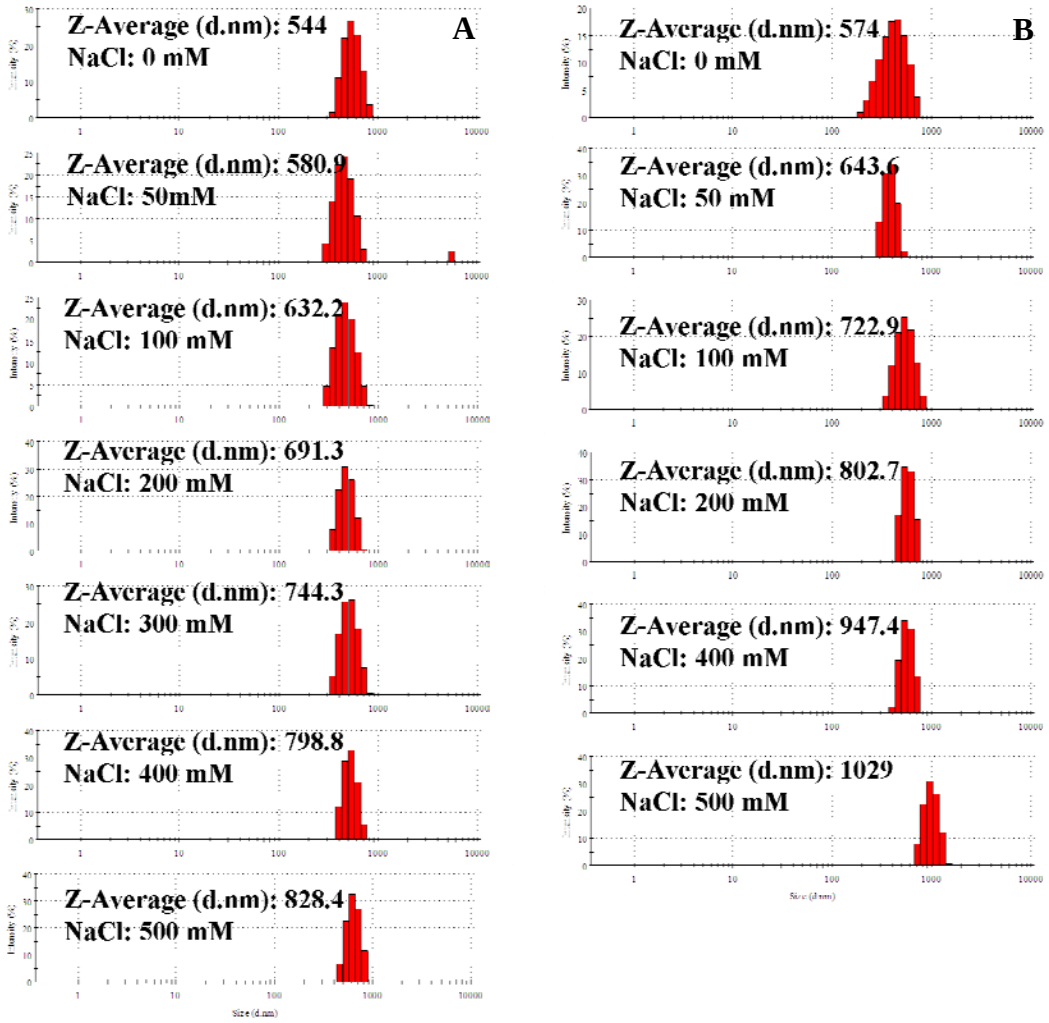

Fig. S1 Size and size distribution of stacked-CCGs with the concentrations of (A) 8 μg/mL and (B) 16 μg/mL in the NaCl concentration range of 0-500 mM.

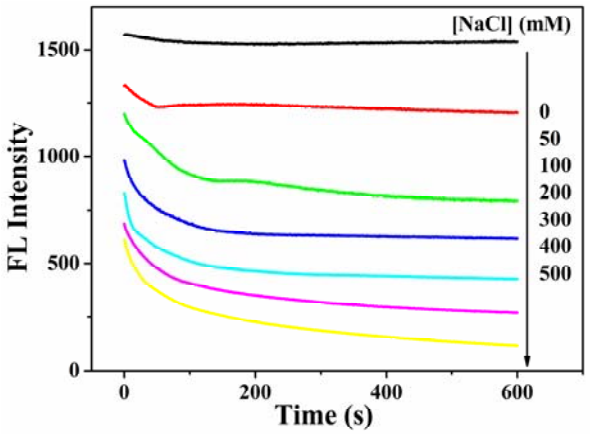

Fig. S2 Kinetics study for the FL changes of Cy3-labeled dsDNA induced by stacked-CCGs colloids at varying NaCl levels. Reactions were carried out in 20 mM PBS buffer (pH 7.4, 25 °C) containing 8 μg/mL CCGs, 58 nM dsDNA and 0-500 mM NaCl, excitation wavelength: 540 nm, emission wavelength: 568 nm.

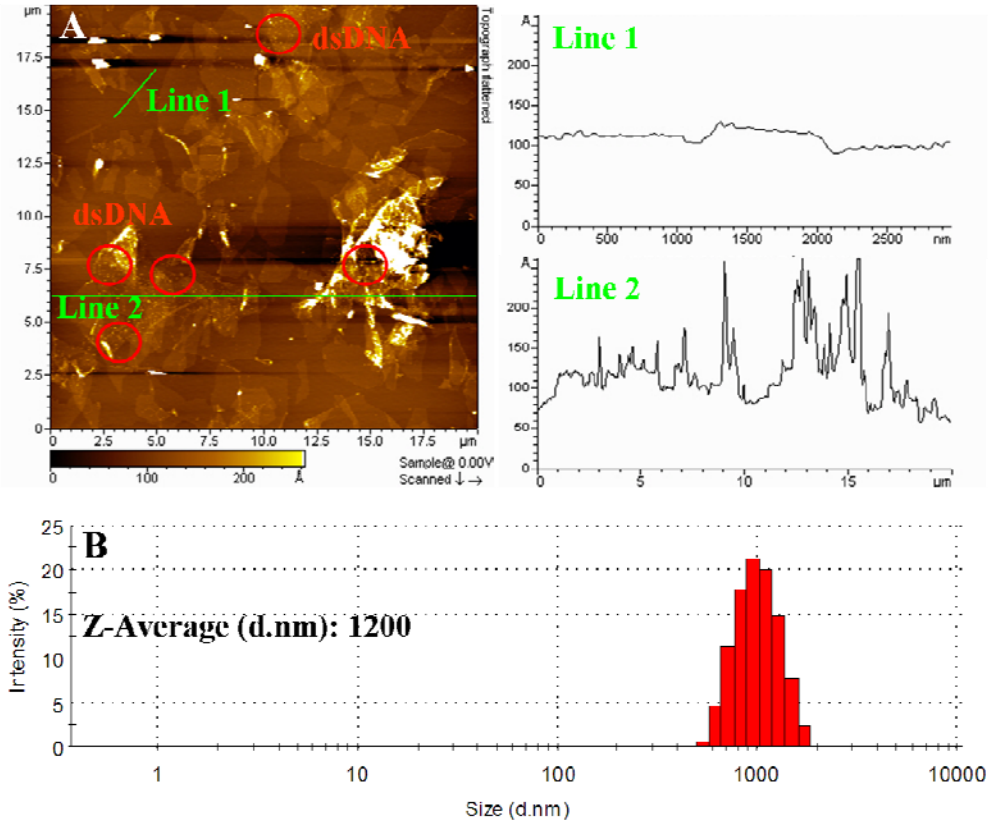

Fig. S3 (A) Typical AFM image and height profile (Line 2) of stacked-CCGs carried out in liquid phase. Reactions were carried out in 20 mM PBS buffer (pH 7.4, 25 °C) containing 16 μg/mL CCGs, 58 nM dsDNA and 500 mM NaCl. Line 1: height profile of CCGs. (B) Z-average diameter of stacked-CCGs. Note that the date was calculated according to a sphere model.[4] So the obtained diameter value could not accurately reflect the absolute size of stacked-CCGs. However, such value might be close to the relative size of stacked-CCGs (AFM image indicated that the size of stacked-CCGs was around several micrometers).

References

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