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Supporting Information

Graphene Oxide/DNA Duplex−Based Logic Gate and Sensor Mediated by RecA-ssDNA Nucleoprotein Filaments

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Chemicals and Materials. Graphene oxide (GO) was synthesized according to the modified Hummers' method. Graphite powder (99.99995%, 325 mesh) was purchased from Alfa Aesar. Other chemicals, $KMnO_4$, NaOH, KCl and NaCl, were from Beijing Chemical Company. Adenosine 5'-O-(3-Thiotriphosphate) (ATP γ S) was from Sigma. *E.coli* RecA protein (RecA) and RecA reaction buffer (70 mM Tris-HCl, pH 7.5, 10 mM $MgCl_2$, 5 mM DTT) were purchased from Mycombio Co. Ltd. (Beijing, China). λDNA and ATP were purchased from Dingguo Company (Beijing, China). All other chemicals were of analytical grade and obtained from Sinopharm Chemical Reagent Co. Ltd. The solutions were prepared using ultrapure water, which was obtained through a Millipore Milli-Q water purification system (Billerica, MA, USA) and had an electric resistivity >18.3 MΩ cm. The oligonucleotides used in this work were synthesized from Invitrogen Co. Ltd. (Shanghai, China).

Preparation of ssDNA-GO and dsDNA-GO complexes. In a typical procedure, 1 μM ssDNA and dsDNA were mixed with GO (0.1 mg mL^{-1}) aqueous solution by sonicating for 2 h and then incubated at room temperature for 24 h, respectively. To remove free unbound DNA, the solutions were centrifuged at 16,000 g, the supernatant was discarded and the precipitated ssDNA-GO and dsDNA-GO were then re-dispersed in 50 mM Tris-HCl buffer (pH 7.5) with 100 mM NaCl. This precipitation-redispersion process

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could be repeated several times to guarantee a complete removal of free DNA. Then the DNA-GO samples were used in the AFM and other measurement.

Synthesis of RecA-ssDNA nucleoprotein filament. In a typical procedure, 4 μM ssDNA was mixed with 4 μg/mL RecA at 37 °C in a commercial RecA reaction buffer solution consisting 75 mM Tris-HCl (pH 7.5) and 10 mM $MgCl_2$ for 1 min. Then, 4 mM ATP or ATPγS was added, and the reaction proceeded at 37 °C for 10 min. The mixture was incubated for 10 min at 37 °C to allow RecA protein to coat the oligodeoxynucleotide probes.

Fluorescent DNA Assays. In a typical DNA assay, 20 nM Cy3-ssDNA was hybridized with different concentration of cDNA (0 ~ 200 nM) in 0.1 M Tris-HCl solution (pH 7.5, 100 mM NaCl, pH 7.5) at 37 ^{o}C for 1.5 h. And then, 75 mg/mL GO was added with incubation for 30 min. Next, the mixture was incubated with ss-DNA-RecA nucleoprotein filament for 2 h at room temperature. The final concentrations of GO, RecA, ssDNA and ATP were 75 μg/mL, 1 μg/mL, 1 μM and 1 μg/mL, respectively. Finally, the fluorescence of the mixture was detected.

Characterizations.

Fluorescence measurements were conducted at F-7000 Hitachi spectrometer, and Fluorescence anisotropy was measured by FL3-P-TCSPC (JobinYvon, France). In a typical experiment, 100 μL sample was used. The fluorescence was monitored with excitation wavelength of 470 nm.

Circular dichroism (CD) measurements were taken with a JASCO J-715 spectrometer with temperature control (JASCO International Co. Ltd., Tokyo, Japan) at room temperature unless otherwise specified. Samples were freshly prepared. DI water was used in the dialysis step instead of buffer.

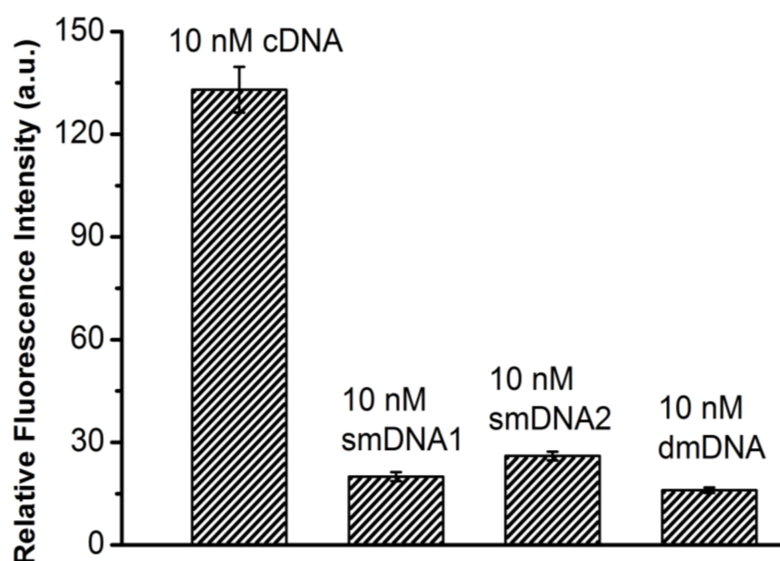

Figure S1. Relative fluorescence response over background of Cy3-ssDNA (20 nM) hybridized with 10 nM target cDNA or mismatched DNA in the presence of GO and then incubated with RecA-ssDNA and ATPγS. Single-base pair mismatched DNA (smDNA1: 5'- ATA TTG TTG ACA T**A**T ATT AAT AAT GTC CTC -3'; smDNA2: 5'- ATA TT**T** TTG ACA TCT ATT AAT AAT GTC CTC -3'), and double-base pair mismatched DNA (dmDNA: 5'-ATA TT**A** TTG ACA TCT ATT AAT AAT **T**TC CTC-3'). Excitation wavelength: 540 nm. GO: 75 µg/mL; RecA protein: 1 µg/mL; ATPγS: 1 mM; Buffer: 75 mM Tris-HCl solution (pH 7.5) containing 5 mM Mg^{2+}.