

Supporting Information

A reversible fluorescent DNA logic gate based on graphene oxide and its application for iodide sensing

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1. Materials

Graphene oxide (GO) was synthesized from natural graphite powder by modified Hummers method. Prior to the experiments, the GO powder was dissolved in Milli-Q water and then sonicated for 5 h to give a homogeneous brilliant yellow solution (Fig. S1). The AFM image showed GO sheets with a thickness of *ca.* 0.6-0.8 nm, which is well consistent with that of the single-sheet graphene reported in the literatures.¹ The nucleic acids (*Probe*: 5'-FITC-TTC TTT CTT GGG TTG TTT GTT3') was synthesized and purchased from Songon Inc. (Shanghai, China). Other chemicals were reagent-grade and purchased from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China). 10× Tris-acetate buffer (500 mM Tris, 500 mM MgAc, pH7.4) was prepared using metal-free reagents in distilled water purified by a Milli-Q water purification system (Millipore Corp., Bedford, MA) with an electrical resistance of 18.2 MΩ.

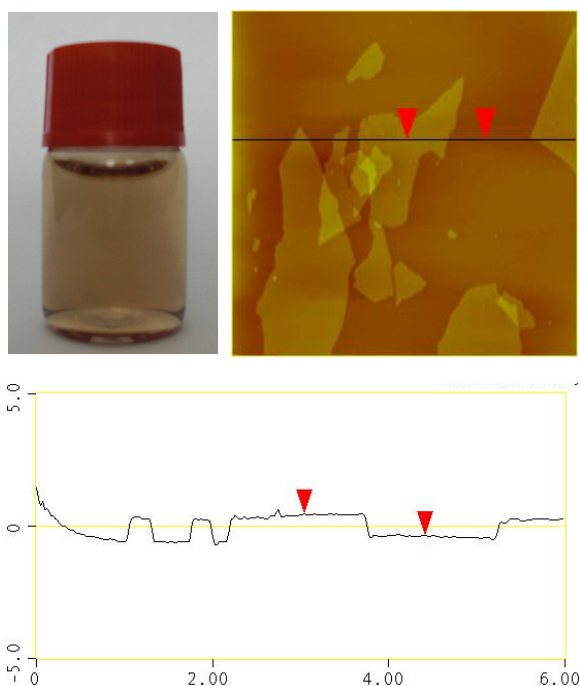


Figure S1. Photo of GO solution (0.2 mg/mL) after ultrasonic reaction for 5 h, and AFM images of GO (6×6 μm, vertical scale 0-10 nm) nano layer on a freshly cleaved mica surface.

2. Instrumentation

Fluorescence was measured in a fluorescence microplate reader (Bio-Tek Instrument, Winooski, USA) using a black 384 well microplate (Fluotrac 200, Greiner, Germany).

3. Procedures for Fluorescence Detection

An aliquot of 100 nM *Probe* in the Tris-acetate buffer was mixed with GO to form *Probe*-GO complex and incubated for 5 min at room temperature prior to the addition of metal ions. After that, an aliquot of the tested metal ion or ddH₂O (as blank sample) was added to the *Probe*-GO mixture. The final concentration of GO was 0.02 mg/mL. The mixture was vortexed to mix all the reagents, and then incubated for 12 min at room temperature and after that, an aliquot of 0.1 mL mixture was placed in the microplate to measure the fluorescence intensity. The experiments of optimization of sensing conditions were carried out under identical conditions. For the detection of iodide, first, an aliquot of 10 μM Hg²⁺ was added to the *Probe*-GO solution and incubated at RT for 12 min. After that, the fluorescence emission spectra of the mixture were recorded. Then an aliquot of iodide or other anions (as control samples) for the selectivity experiment was added to the above solution, and the mixture was allowed to incubate for another 12 min RT, while the emission spectra were recorded in the same way.

We tested the kinetic behaviors of *Probe*-GO solution with Hg²⁺ by monitoring the fluorescence intensity as a function of time. Figure S2A illustrates that the time for the formation of *Probe*-Hg²⁺ complex and its release from GO took from 0 to 12.5 min. Similarly, we also studied the kinetic behaviors of the *Probe*-GO solution with *Probe*-Hg²⁺ complex when adding iodide by monitoring the fluorescence intensity as a function of time. Figure S2B clearly shows that the *Probe*-Hg²⁺ complex was disrupted by the interactions of Hg²⁺ and iodide, causing fluorescence quenching within 12 min.

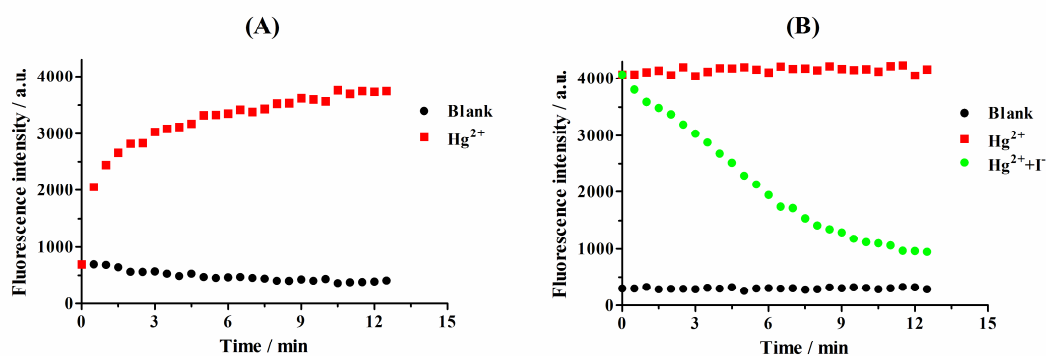


Figure S2. (A) The kinetics of fluorescence restoration of *Probe*-GO solution upon added Hg²⁺ (10 μM). (B) The kinetics of fluorescence quenching of *Probe*-GO solution with Hg²⁺ upon the addition of iodide (10 μM).

4. Data Analysis

The GraphPad Prism 5.0 software (GraphPad Software, San Diego, CA) was employed to perform the data processing. Each sample was repeated in duplicate, and data were averaged.

References

- (a) D. Li, M. B. Mueller, S. Gilje, R. B. Kaner, G. G. Wallace, *Nat. Nanotechnol.*, 2008, **3**, 101;
(b) K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, A. A. Firsov, *Science*, 2004, **306**, 666.