Supporting information

Ultrafast room-temperature NH₃ sensing with positively-gated reduced graphene oxide field-effect transistors

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Experimental Methods

<u>Preparation of R-GO</u>: The R-GO suspension was prepared using a procedure previously reported.¹ First, graphite oxide was produced by the oxidative treatment of purified natural graphite (SP-1, Bay Carbon, MI) using a modified Hummers method.² A stable suspension of graphene oxide sheets was then obtained by adding graphite oxide in water (3 mg graphite oxide per ml H₂O) followed by 1 hour (h) ultrasonication. The presence of oxygen functional groups makes graphene oxide sheets highly hydrophilic and the aqueous dispersion stable.³ After that, dimethylformamide (DMF) was added into the graphene oxide suspension at a volume ratio of DMF : H₂O = 9, resulting in a graphene oxide concentration of 0.3 mg/ml. Hydrazine monohydrate was subsequently used to chemically reduce the graphene oxide suspension for 12 h at 80 °C with stirring, which led to a homogeneous suspension of R-GO platelets.

<u>*R-GO Sensor Fabrication:*</u> The *p*-doped silicon wafer covered with a 200-nm-thick thermally formed oxide layer was used as the substrate on the top of which the interdigitated electrodes were fabricated using an e-beam lithography process. The Au fingers (50 nm thick) of the electrode are $\sim 1 \,\mu\text{m}$ wide and $1 \,\mu\text{m}$ apart. A 2-nm-thick Cr layer was used as an adhesion layer between the Au and the Si wafer. A few drops of the R-GO suspension were cast onto Au interdigitated electrodes, resulting in a network of R-GO sheets (by bottom-contact with the substrate) left on the Au fingers after solvent evaporation.

<u>*FET Characterization:*</u> Transport properties of R-GO FETs were characterized using a Keithley 2602 source meter that has a voltage range of ± 40 V.

<u>Gas Sensing Characterization</u>: The gas sensing performance of R-GO devices was characterized under nearly practical conditions (i.e., room temperature, atmospheric pressure, and dry, clean air as reference gas environment) against low-concentration NH₃ diluted in dry air, both of which were controlled by mass flow controllers. Gas cylinders with certified analyte concentration (1% NH₃) were purchased from Praxair. The R-GO device was mounted in an air-tight test chamber with electrical feedthroughs.⁴⁻⁶ Variations in the electrical resistance of the R-GO device were monitored when the device was periodically exposed to clean air and NH₃– laden air. In a test cycle, the R-GO device was exposed successively to (1) clean air flow to acquire a base value of the sensor resistance, (2) NH₃ in air to register a sensing signal, and (3) clean air again to recover the device. The Keithley source meter was used to maintain a low constant dc current I_{ds} (100 nA) between the source and drain electrodes of the device and to simultaneously record the change in the dc source-drain bias V_{ds}. V_g was intentionally adjusted for revealing the gatedependent NH₃ sensing of R-GO devices. Note that V_g was changed between test cycles but was kept constant during a single cycle.

<u>SEM:</u> The morphology of R-GO devices was characterized using a field-emission SEM (Hitachi S 4800), which has a resolution of 1.4 nm at 1 kV acceleration voltage.

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Fig. S1. Schematic of the R-GO device on an FET platform.



Fig. S2. R-GO FETs were irresponsive to air or Ar flow. (a) I_{ds} -V_{ds} curves for an R-GO FET in open air, in air flow (2 lpm), in pure Ar flow (2 lpm), and in 1% NH₃ flow (diluted in air) (2 lpm); curves were obtained after the gas flow was turned on for 5 min. (b) The dynamic behavior of the R-GO sensor remained unchanged when air flow was turned on or off. (c) The R-GO FET showed no response to Ar flow at $V_g = 0$ or +40 V.



Fig S3. The source-gate current (I_{gs}) vs. V_g curve recorded simultaneously with the I_{ds} - V_g curve shown in Fig. 1b. I_{gs} (leakage current) was with ±1 nA and considerably lower than I_{ds} .



Fig. S4. Repeatable and fast responses of R-GO to 1% NH₃ under V_g =+40 V (*n*-mode). Six test cycles were performed; the sensitivity appeared similar for all the cycles. For each test cycle, the R-GO sensor was successively exposed to air flow for 5 min, 1% NH₃ flow for 3 min, and air flow for 12 min; these time intervals and gas flow conditions are the same as those in cycles # 1 and 2 (V_g =+40 V) in Fig. 2. (Note that the 'spike' in the signal at the beginning of each test is due to the fluctuation caused by the resetting of the source meter.)