

Electronic Supplementary Information for

Edge-Tailored Graphene Oxide Nanosheets-based Field Effect Transistors for Fast and Reversible Electronic Detection of Sulfur Dioxide

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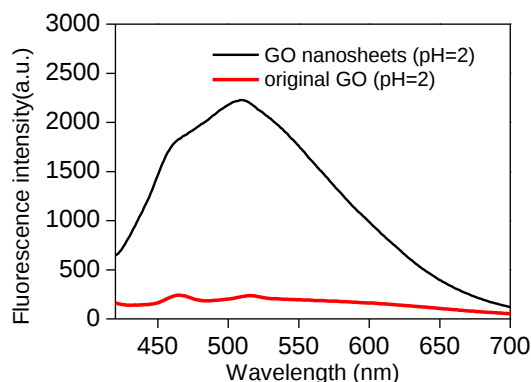
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1. Synthesis of edge-tailored GOnanosheets

10 Graphite powder was purchased from Alfa Aesar. Other chemicals were purchased from China National Medicine Corporation and used as received. As-prepared graphene oxide (GO) was prepared using a modified Hummers method at first.¹ Then 1 g of periodic acid were added into 1 mL of as-prepared GO dispersion (3.2 mg/mL) and kept at 50 °C for 24 h then cooled to room temperature. The resulting GO nanosheets were agglomerated from the solution with the addition of sodium chloride and centrifuged out from solution at 15,000 rpm for 30 min. The
15 obtained GO nanosheets were redispersed into water for the characterizations.

2. Spectroscopy characterization of edge-tailored GOnanosheets

Fig. S1 shows the fluorescence spectra of original GO and edge-tailored GO nanosheets. It can be seen that edge-tailored GO nanosheets is photoluminescent under acidic condition, whereas the original GO is not luminescent. The photographs of original GO and edge-tailored GO nanosheets dispersion illuminated by ultraviolet lamp (365 nm) also show that original GO is black but edge-tailored GO nanosheets are luminescent in green (Fig. S2).



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30 Fig. S1 Fluorescence spectra of original GO and edge-tailored GO nanosheets under excitation wavelength of 400 nm.

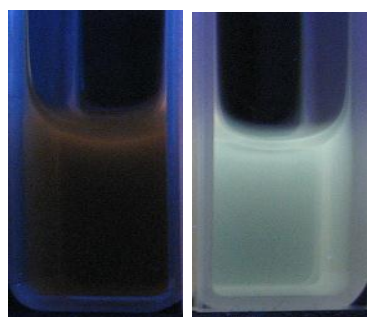


Fig. S2 Photographs of original GO and edge-tailored GO nanosheets dispersions illuminated by ultraviolet lamp ($\lambda = 365$ nm).

The FTIR and XPS spectra of original GO and edge-tailored GO nanosheets were measured as shown in Fig. S3 and Fig. S4. In the FTIR spectra, the band at 1735 cm^{-1} is corresponded to C=O. The band at 1627 cm^{-1} is corresponded to C=C bonding. It can be found that the relative strength of C=O band of edge-tailored GO nanosheets is stronger than original GO, indicating the number of C=O groups in edge-tailored GO nanosheets increases after 20 edge-tailoring. In the XPS spectra, the C1s curves are splitted into four peaks corresponding to C(O)O (carboxyl groups) at 289.0 eV, C=O at 287.5 eV, C-O at 286.6 eV, and C=C at 284.6 eV. It is calculated that the molar ratio of C=O/C=C for original GO and edge-tailored GO nanosheets are 0.73 and 1.12, respectively, indicating higher content of C=O bond in edge-tailored GO nanosheets than in original GO.

These data reveal that edge-tailored GO nanosheets possess abundant C=O groups. These C=O groups can reversibly interact with H^+ . This is the basis of using edge-tailored GO nanosheets for acid gas sensing.

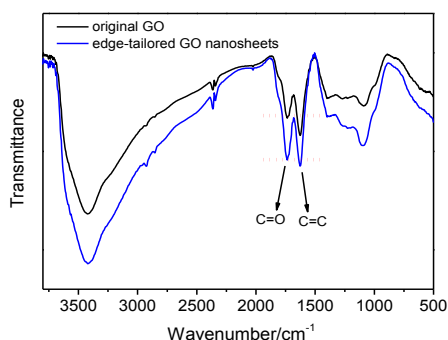


Fig.S3FTIR spectra of original GO and edge-tailored GO nanosheets

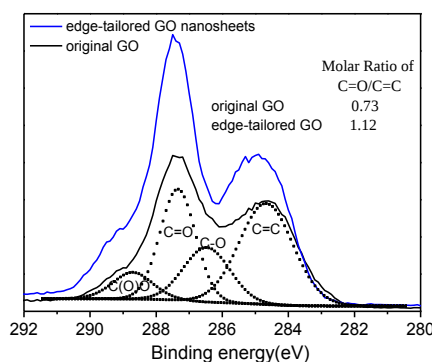


Fig. S4C1s XPS spectra of original GO and edge-tailored GO nanosheets

3. Sensor Device Fabrication and Characterization

The preparation of sensor array is carried out by lithographing technology as the procedure described earlier.² Interdigitated electrodes (Au/Cr, 200/20 nm, respectively) were vacuum sputtering coating on a Si/SiO₂ (500 nm SiO₂ layer) substrate. Then the interdigitated electrodes with 5 μ m gap were defined using lift-off techniques. Finally a 50 nL GO nanosheets suspension (0.05 mg/ml) was drop-casted onto the interdigitated electrodes using a microsyringe and dried at 40 °C under vacuum condition for 2 hours.

The electrical characterizations of original GO and edge-tailored GO nanosheets were monitored with Keithley 2602 Source Meter. Calculated $R = dV/dI$ versus V_{ds} dependence are shown in Fig. S5. In the test of edge-tailored GO nanosheets based FET, a homemade chamber was used to expose the device upon 5 ppm, 10 ppm HCl, respectively.

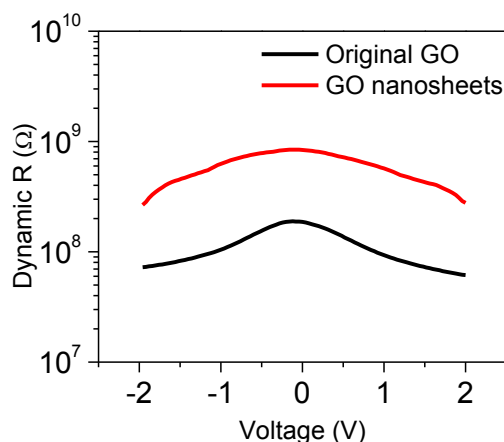


Fig.S5 Dynamic resistances of original GO and edge-tailored GO nanosheets based sensor devices.

4. Gas sensing procedure

The sensors were wire-bonded on a CERDIP chip carrier and each sensor was connected in series with a load resistor. A quartz gas flow chamber with two ports sealed the sensor chip for gas flow in or out. With a valve controlled system and mass flow controllers (MFCs, Sevenstar CS200, China), gases flow through the chamber can be switched quickly from analytes to carrier gas and regulated at a total gas flow of 200 std. cm³ min⁻¹. Dry air (purity: 99.99 %) was used as both carrier and diluting gas. Analyte gases of desired concentrations were generated by diluting dry SO₂, HCl and NO₂ with dry air in the gas sampling bag via MFCs. Relative humidity was adjusted by passing dry air through saturated sodium chloride solution. A custom Labview computer program was developed to continuously control and monitor the voltage of the circuit using Fieldpoint analog input and output modules (National Instruments, Austin, TX). In the SO₂ sensing experiments, the sensors were first exposed to moist air (RH=65%) to obtain the stable baseline resistance, then switched to SO₂ with desired concentration at same RH, and then backed to moist air purge to return the devices to their baseline resistances.

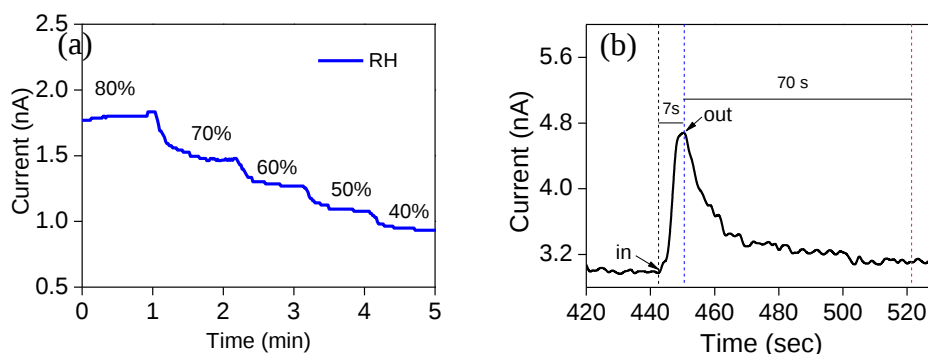


Fig.S6 (a)Real time response of edge-tailoredGO nanosheets based sensor to RH at room temperature. The sensor can work steady in certain temperature and damp condition.(b)Response time (less than 7 s) and recovery time (~ 70 s)when the sensor was exposed to 275 ppm of SO_2 .

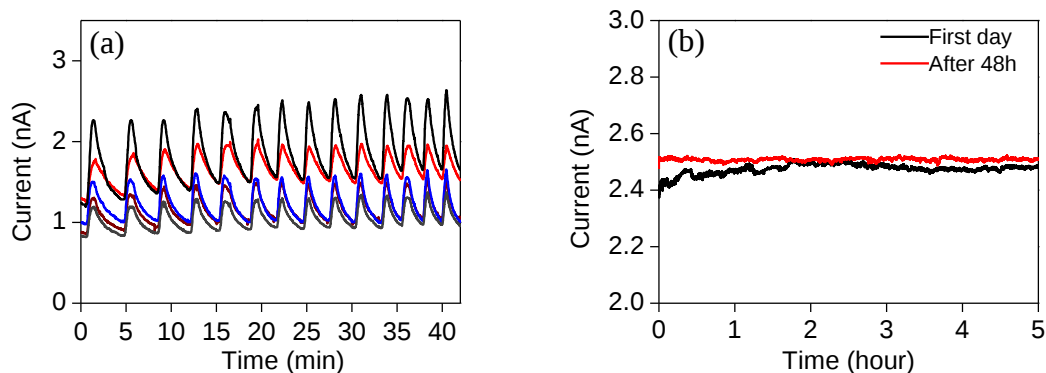


Fig.S7 (a) Response of five edge-tailored GO nanosheets based sensors to 14 cycles of 500 ppm SO_2 gas. The standard deviations of response curves were calculated to be 0.12, 0.05, 0.04, 0.06, and 0.05, respectively (from top to bottom). (b) The stability of sensor baseline over two days. Standard deviation of the baseline is calculated to be 1.2% over 2 days, which shows great stability.

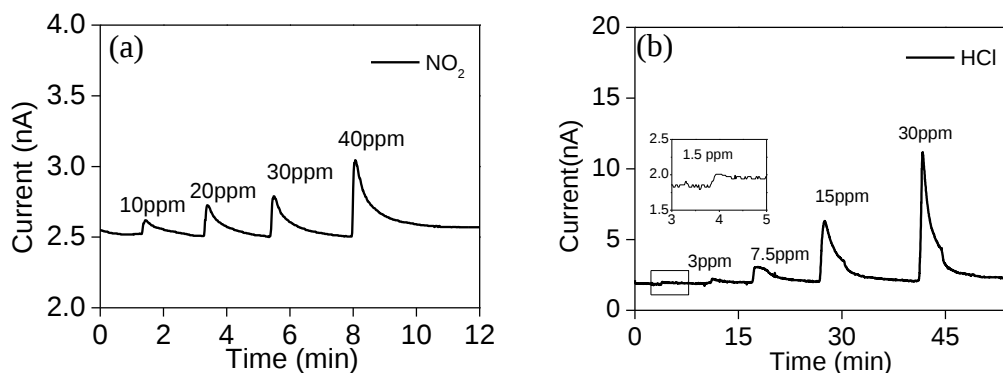


Fig. S8Real time responses of edge-tailored GO nanosheets based sensor to NO_2 (a) and HCl (b) under 65% relative humidity at 35 roomtemperature.

References

- 1 W. S. Hummers and R. E. Offeman, *J. Am. Chem. Soc.*, 1958, **80**, 1339-1339.
- 2S. Mubeen, T. Zhang, B. Yoo, M. A. Deshusses and N. V. Myung, *J. Phys. Chem. C*, 2007, **111**, 6321-6327.

