Supporting Information

Wrinkled reduced graphene oxide nanosheets for high sensitive and easy recoverable NH₃ gas detector

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Figure S1 schematic of the sensor equipment.



Figure S2 XPS overall (a1, b1), C 1s (a2, b2), and O 1s (a3, b3) spectra of FG and WG.

X-ray photoelectron measurement

The detail XPS results of FG and WG materials are given in Figure S2. The surface concentration of carbon and oxygen was estimated from the ratio of the areas of the corresponding lines of the overall spectra. The intensity of C=O peak is lower while the C-O peak is higher in the FG (Figure S2a3) compared with that of WG (Figure S2b3). This difference is mainly due to the significantly introduced defect domains in natural graphite during ball-milling process, which is proven by the appearance of D-band at 1351 cm⁻¹ of the ball-milled graphite (Figure S3). The defect regions are more reactive, thus these carbon atoms performed much higher oxidative degree after chemical oxidation.

Structure characterization of ball-milled graphite



Figure S3 Raman curves of natural graphite and ball-milled graphite



Figure S4 Fourier-transform infrared spectra of FG and WG

To prove the same functional group distribution of FG and WG, Fouriertransform infrared spectroscopy was also carried out and the curves are given in Figure S4.

Calculation details



Figure S5 Curved and flat graphene fragments of C₅₄H₁₈ and C₆₂H₁₆ composition

Theoretical modeling of an ammonia molecule adsorption on flat and curved graphene surface was carried out within DFT using the three-parametrical hybrid functional of Becke [1] and Lee-Yang-Parr correlation functional [2] with a pair correction [3,4] accounting dispersion interactions (B3LYP-D3method) as it implemented in Jaguar 7.9 program package [5]. Atomic orbitals were described by $6-31G^{*+}$ basis set, including polarization and diffuse functions for all atoms except hydrogen. The FG and WG model fragments had a composition of $C_{54}H_{18}$ and $C_{62}H_{16}$ respectively where hydrogen atoms saturated the dangling bonds of the boundary carbon atoms. An ammonia molecule was initially placed at the starting distance of 1.5 Å above the centre of graphene fragment. Geometry of models was optimized by an analytical method to the gradient of $5 \cdot 10^{-4}$ atomic units for taking into account the shift of atomic position. The adsorption energy of an ammonium molecule was calculated as the difference between the total energy of graphene with the adsorbed NH₃ and the total energies of the isolated constituents.



Figure S6 sensing properties of WG towards NO₂ gas.

WG also demonstrates significant sensing performance to NO_2 exposure (Figure S6). The response reaches very high levels for different concentrations for 5 min expose; and another 5 min are required for its full recovery without any additional assistant treatments such as UV light or heating.

References:

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