Supporting Information

Holey Reduced Graphene Oxide Nanosheets for High Performance Room Temperature Gas Sensing

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Figure S1. TEM images of GO sheets.





Figure S2. TEM images and hole size distribution of RGO 120 °C (a), RGO 150 °C (b) and RGO 180 °C (c), respectively.





Figure S3. N_2 adsorption-desorption isotherms and the BJH Adsorption corresponding pore size distribution curve of RGO under different preparation conditions (a) 120 °C 6h; (b) 150 °C 6h; (c) 180 °C 6h.



Figure S4. XPS C1s (left, 1) spectra and O1s (right, 2) of (a) GO and RGO produced from hydrothermal treatment under different temperature (b) 120 °C, (c) 150 °C, (d)

180 °C. The XPS C 1s spectrum of GO can be fitted into three peaks centered at 284.6 eV, 286.7 eV and 287.5 eV, which could be indexed to the sp2 hybridized carbon, the carbon in C-O and C=O bonds,¹ respectively. Curve fitting analysis of the XPS C1s spectra of the RGO samples affords three peaks at 284.6 eV (sp2 C-C), 285.6-286 eV (C-O) and 289.7-290.3 eV (O=C-O), respectively. The fitted O 1s peaks centered at 532-532.6 eV and 530.7 are attributed to the C–OH group and C=O or O=C-O bonds.²



Figure S5. Raman spectra of GO, RGO produced from hydrothermal treatment under different temperature (a) 120 °C, (b) 150 °C, (c) 180 °C and RGO annealed at 150 °C for 6h in air.



Figure S6. Current versus voltage of RGO obtained from hydrothermal treatment with different temperature (120 °C, 150 °C and 180 °C) at ambient condition.



Figure S7. Four successive cycles of exposure to 12.5 ppm NO₂ of the porous RGO fabricated sensor at different preparation temperature: (a) 120 °C, (b) 150 °C and (c) 180 °C; (d) non-porous RGO from anneal at 150 °C for 6h in air.



Figure S8. (a) Dynamic response of holey RGO 150 °C to 5 ppm NH₃; (b) Sensitivities of RGO prepared at different temperature exposed to 5 ppm NH₃ for 5min.



Figure S9. XPS N1s spectrum and O 1s spectrum of the holey RGO adsorbed with NH₃. The XPS N 1s spectrum of GO can be fitted into two peaks centered at 399.6 eV and 402 eV. The former could be indexed to the NH_2^- groups bonded to the negative sites (–OH) on the holey RGO.^{3,4} The latter peak is stemmed from the NH₃ adsorption.⁴ This experiment definitely verifies that a measurable fraction of NH₃ molecules (2.68 atom% of N content based on the XPS data) is strongly binding on the holey RGO even under severe vacuum conditions as the ones imposed by an XPS experiments. Further, compared with the holey RGO (Figure S4c2), a decrease of 0.5 eV is observed. The interaction of NH₃ molecule to –OH on the RGO generates H₂N–H–O– bond, this gives rise to an increase in the electron density on O because the electronegativity of O atom is higher than that of N atom, which manifests as a decrease in the O 1s binding energy.⁵ In return, the shift confirms that a covalent bond between NH₃ and HO– formed.



Figure S10. Dynamic response of holey RGO 150 °C to 100 ppm and 500 ppm H₂S.



Figure S11. Dynamic response of holey RGO 150 °C to 750 ppm CO.



Figure S12. Dynamic response of holey RGO 150 °C to 1000 ppm and 5000 ppm H₂.

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