Supporting Information

Characterization of partially reduced graphene oxide as room temperature sensor for H_2

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Experimental section

Synthesis of Graphene Oxide and reduced graphene oxide (RGO)

Graphene oxide (GO) was synthesized accordingly to a modified Hummers' method.[1,2] Typically, 1.5 g graphite obtained from Beijing Chemical agents Co. was pretreated in a concentrated sulfuric acid (35 ml) solution added with sodium nitrate (0.75 g) at room temperature. 16 hours later, 4.5 g $KMnO_4$ was added to the mixture at around 0 °C which was assisted by an ice bath and continuously stirred for 2 hours. After the mixture was allowed to be stirred at room temperature for another 3 hours, the slurry was pureed into 200 ml ice water. Then the solution was further reacted at 98°C for 15 mins. After that, the excess hydrogen peroxide (3%) was added to the mixture followed by overnight stirring. The mixture was then filtered through Nylon Millipore filter and washed by 10% HCl solution and excess water to remove the acid. Finally, the solid was vacuum dried at 80 °C overnight to obtained GO.

Reduced graphene oxide was produced by heating the as-prepared GO at different temperature (300°C, 500°C, 700°C, 900°C) to obtained RGO-300, RGO-500, RGO-700, RGO-900 respectively, in a tube furnace. The ratio of H2/Ar is 1:3 controlled by a mass flow controller.

Charaterization

The microscopic features of the samples were characterized by transmission electron microscopy (TEM, JEOL JEM-1011) and high-resolution TEM (HRTEM, JEOL JEM-2010). For the XPS analysis, a Kratos AXIS 165 multitechnique electron spectrometer was used. XANES experiments were performed on the Soft X-ray Magnetic Circular Dichroism (SXMCD) station of National Synchrotron Radiation Lab (NSRL) at Hefei. The samples were loaded in an ultrahigh-vacuum chamber at a vacuum of <5 × 10-7 Pa. All spectra were acquired in the total electron yield (TEY) mode with an experimental resolution of 0.2 eV at room temperature. All 13C and 1H magic angle spinning nuclear magnetic resonance (MAS NMR) spectra were recorded on a Bruker Avanc II WB 400 MHz spectrometer with a 4 mm Bruker MAS NMR probe. Each spectrum was acquired using a $\pi/2$ pulse, pulse delay time of 5 s at the 13C resonance frequency of 100.6 MHz and 1H resonance frequency of 400.1 MHz. The spinning speed is 6 kHz or 10 kHz. All the 13C and 1H spectra were referenced to tetramethylsilane (TMS).

The gas-sensitivity measurements were conducted in a home made sensor testing system, and the sensor film were prepared by spin coating a drop of as-prepared N,N-dimethylformafide suspension of 5μl RGO (1 mg/ml) on a commercial sensor electrode (UST, Germany). Hydrogen, carbon monoxide and methane were used as the testing gas and were introduced into the testing chamber by mass flow controller with a concentration of 500 ppm, 2500 ppm and 5000 ppm, respectively. For the investigation of the influence of the temperature to the sensitivity, the measurements were carried out at 25 °C, 47 °C and 132 °C, respectively. The electronic conductivity is measured by sourcemeter (KEITHLEY, Model 2400). The conductivity of the sensor film was measured by two probe method. In the sensor device, a 21 v voltage was applied and the current flow was measure. The resistance of the RGO sensor films was obtained according to the formula below:

R=U/I

The thickness of the sensor films were characterized by scanning electron microscopy (SEM, JSM 6701F). Four silicon chip sliced with the same size of the sensor electrode mentioned above were used as the substrates for the measurement. Subsequently, 5μl N,N-dimethylformafide suspension of RGO-300, RGO-500, RGO-700 and RGO-900 were spin coated on the silicon chips, respectively.

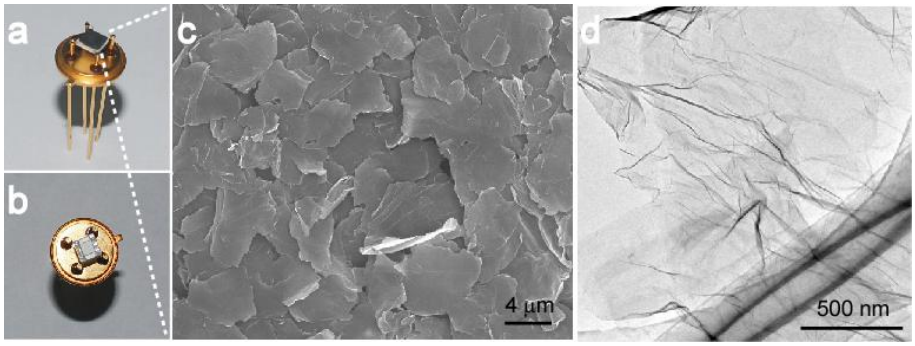

Figure S1 (a) sensor device coated with RGO-300. (b) Bare sensor device. (c) SEM image of the RGO-300 coated on the device. (d) TEM image of RGO-300.

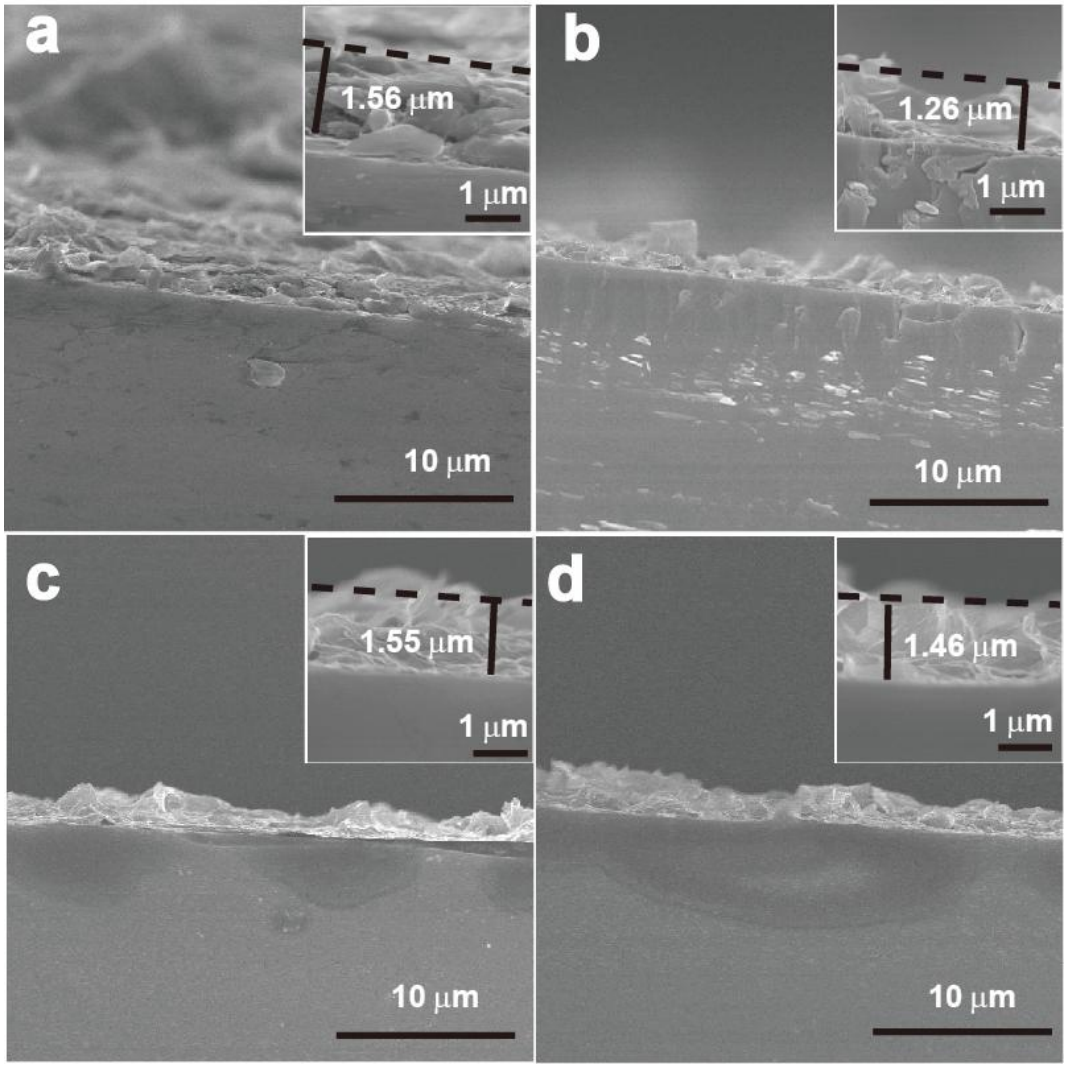

Figure S2 SEM images of the film thickness of (a) RGO-300, (b) RGO-500, (c) RGO-700 and (d) RGO-900, respectively.

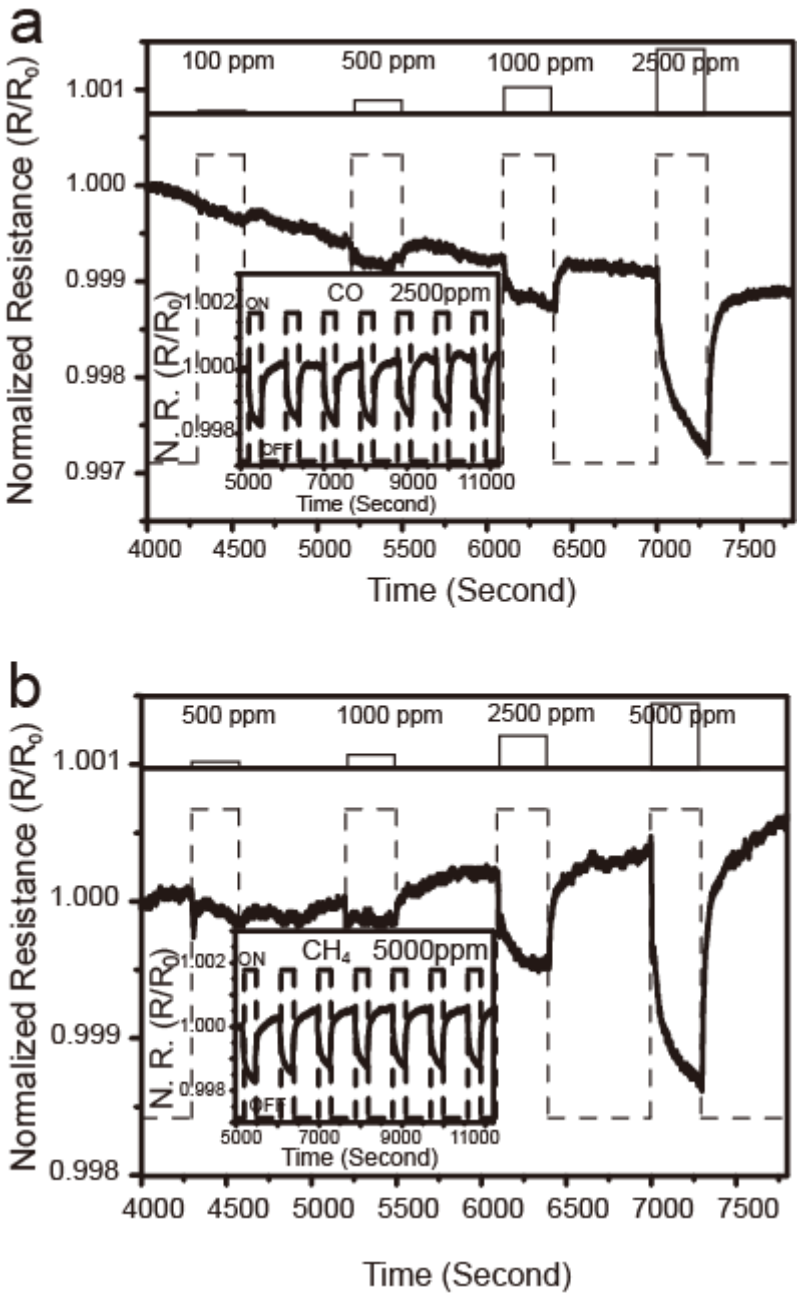

Figure S3 Response of RGO-300 at different (a) CO and (b) CH_4 concentration, inset shows the reproducibility, respectively.

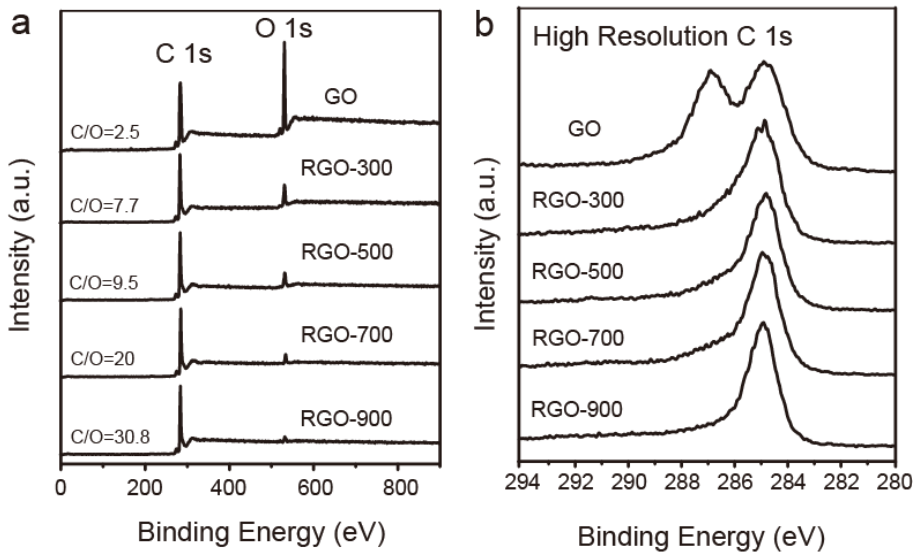

Figure S4. (a) XPS and (b) High resolution C 1s spectra of GO, RGO-300, RGO-500, RGO-700, RGO-900.

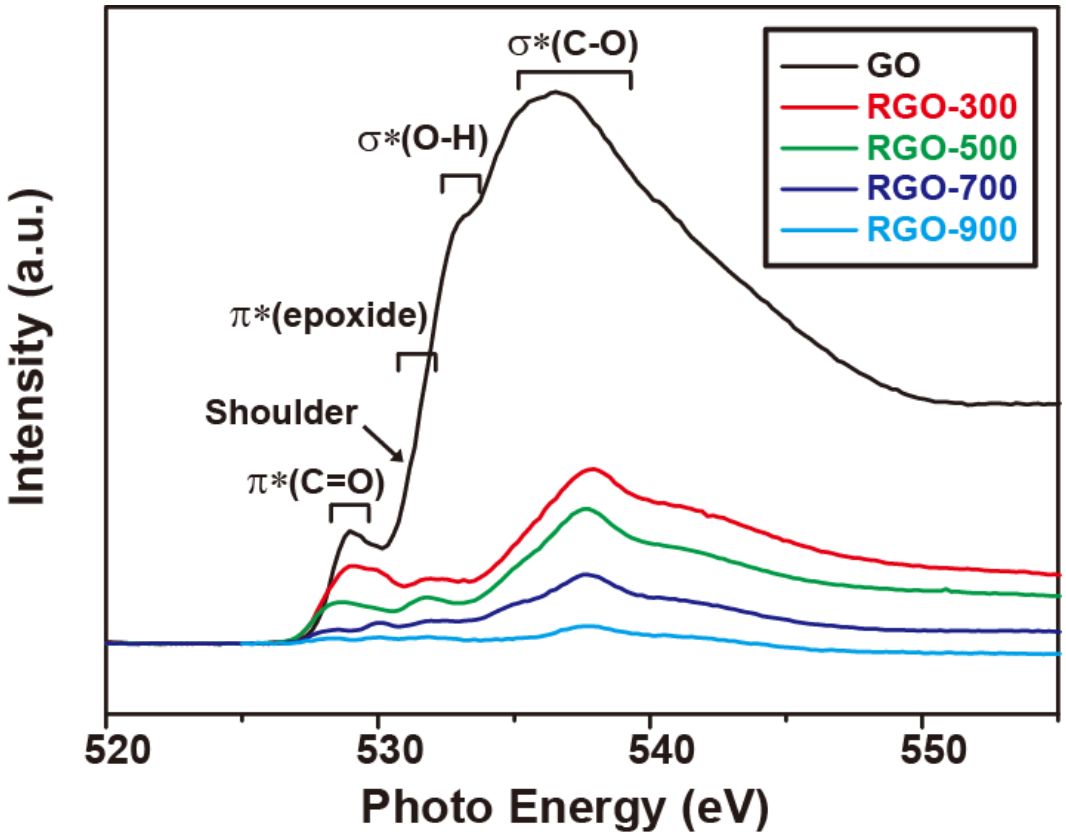

Figure S5. O K-edge XANES spectra of GO, RGO-300, RGO-500, RGO-700 and RGO-900, respectively.

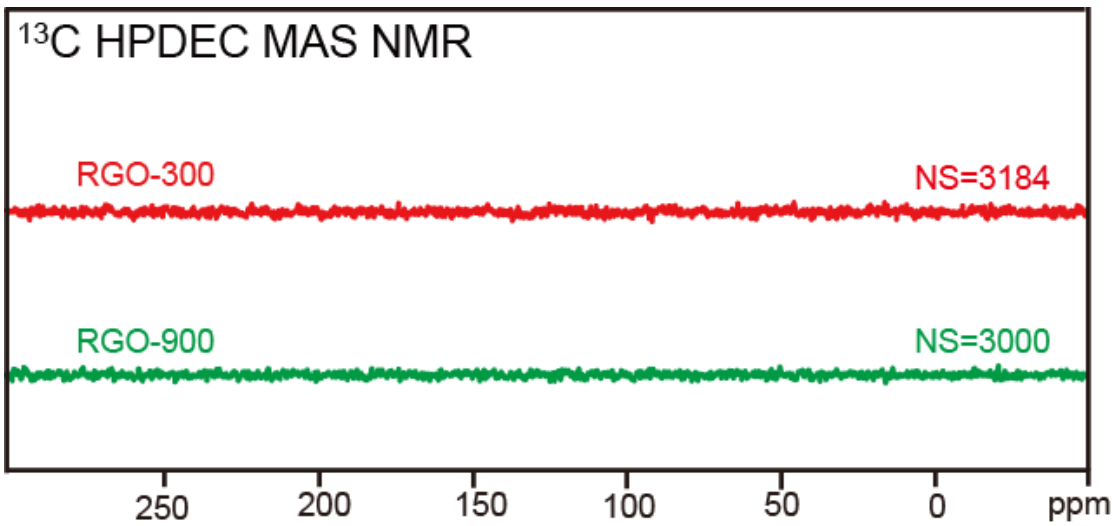

Figure S6 ^{13}C MAS NMR of RGO-300 and RGO-900

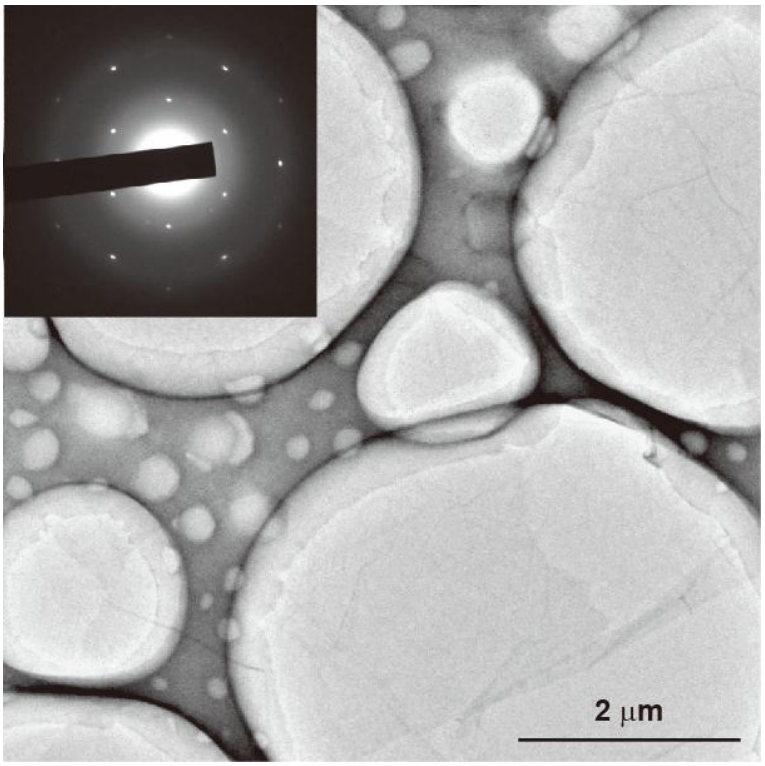

Figure S7 TEM image of GO single sheet, which looks transparent. Inset shows the SAED pattern

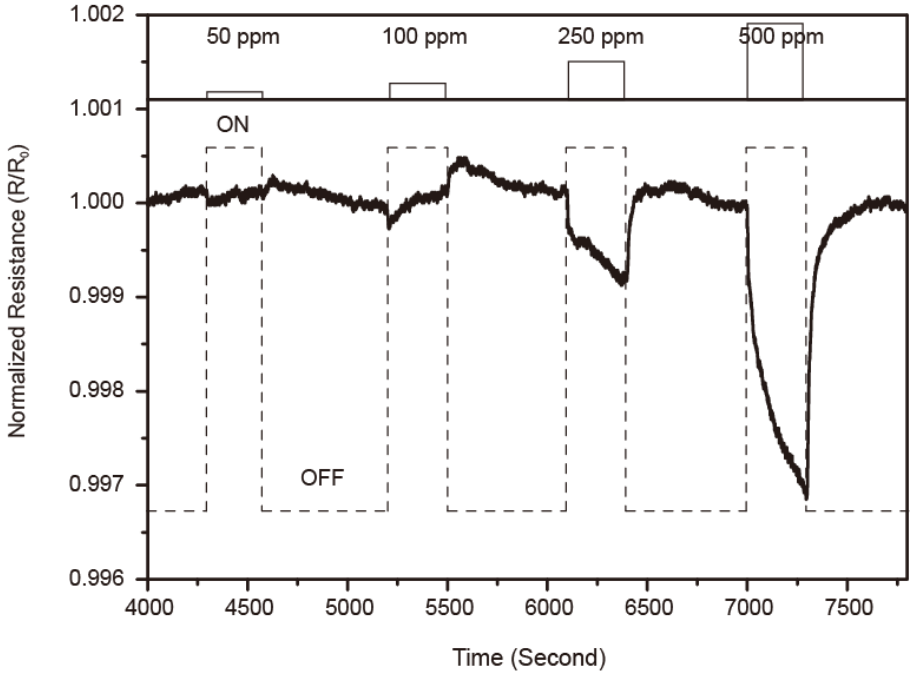

Figure S8 Response of RGO-300 at different H_2 concentration.

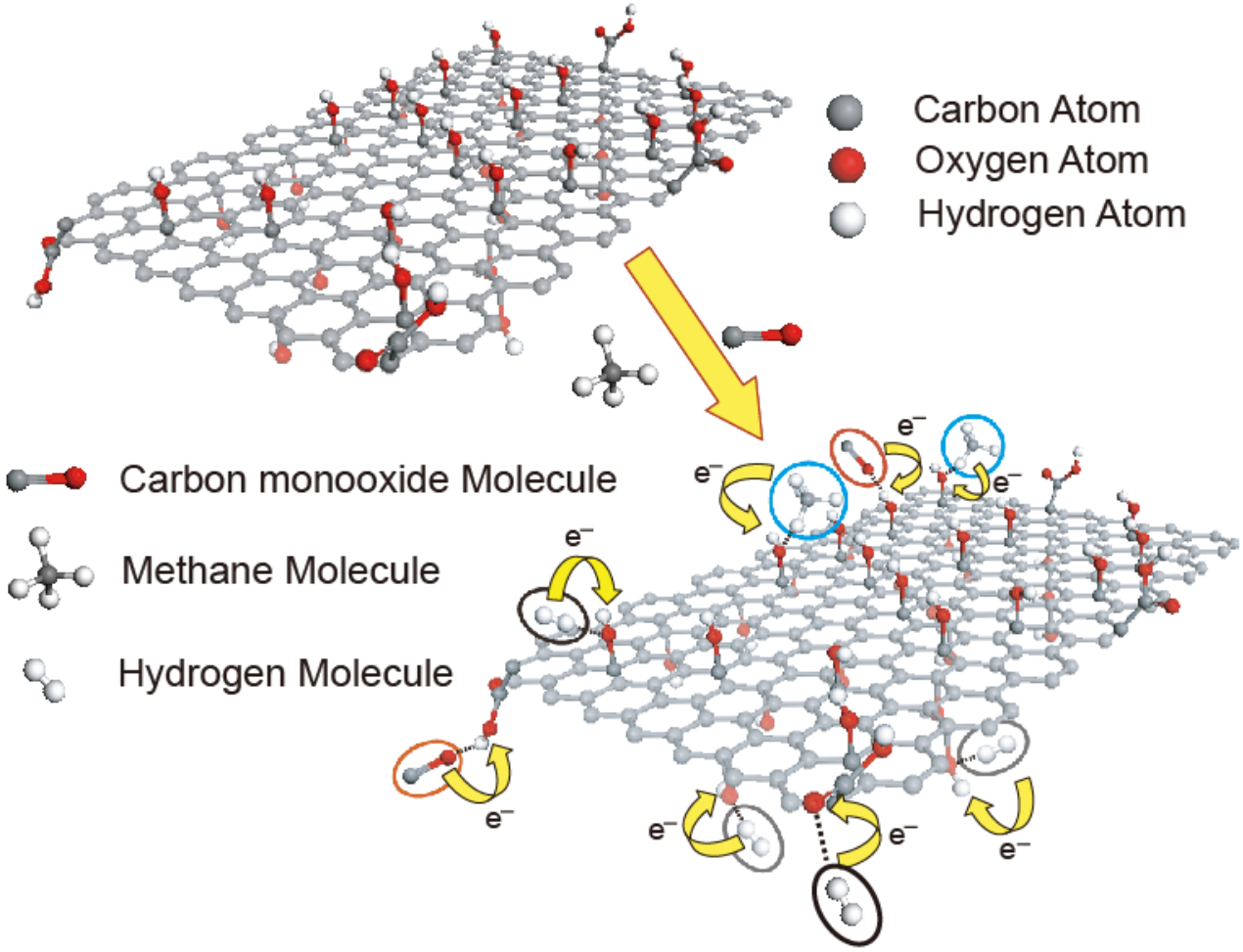

Figure S9 Sensing process of the RGO sample toward hydrogen, carbon monoxide and methane

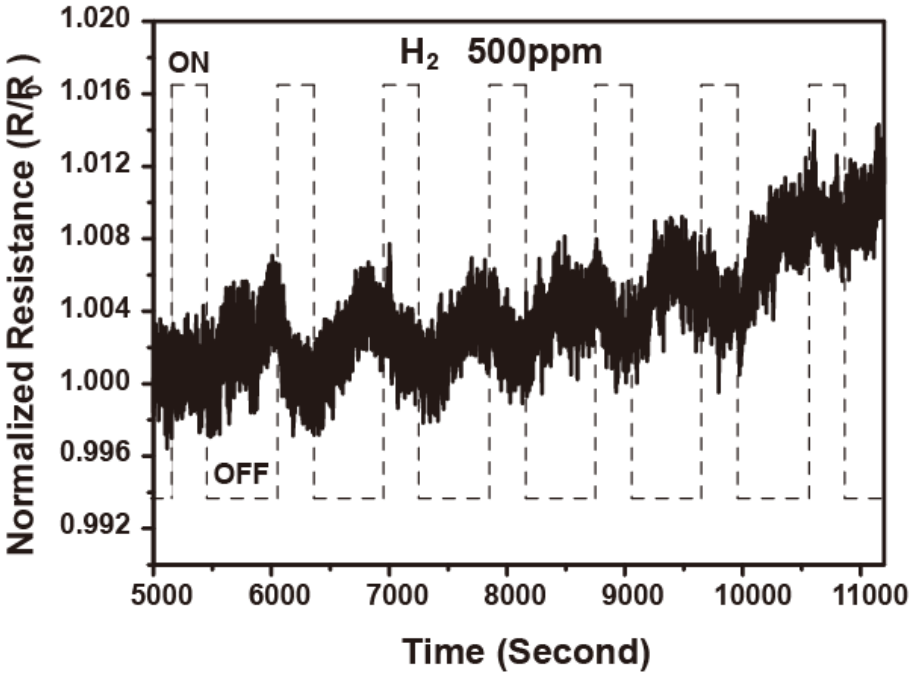

Figure S10 Response of RGO-200 at a H_2 concentration of 500 ppm

Table S1 Content of oxygen in RGO samples (atom %)

Sample	Oxygen content
GO	28.14
RGO-300	11.39
RGO-500	9.45
RGO-700	5.01
RGO-900	3.58

Reference

[1] Li, Y. G.; Wu, Y. Y. *J. Am. Chem. Soc.* **2009**, *131*, 5851-5857.
[2] Hummers, W. S.; Offeman, R. E. *J. Am. Chem. Soc.* **1958**, *80*, 1339.