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Thermal reduced graphene oxide-based gas sensor for rapid detection of ammonia at room temperature

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ABSTRACT

Reduced graphene oxide (rGO) has attracted an enormous interest as a promising candidate for gas detection due to its large specific surface area, abundant oxygen-containing functional groups and scalable production. Although intense works have been conducted on rGO-based gas sensors, there is still much room for improvement on both their response speed and thorough exploration. In this work, thermal reduced graphene oxide (TRGO) is fabricated by spinning dilute graphene oxide (GO) suspension combined with subsequent in situ thermal reduction. TRGO sheets with size of more than 10 μ m are uniformly dispersed and smoothly coated on Au interdigitated electrodes. The ammonia sensing performance indicates that 250 °C-TRGO exhibits the shortest response time of 11 s to 100-ppm ammonia. Besides, 130 °C-TRGO shows strong response to low concentration ammonia with the calculated limit of detection (LOD) of 0.9 ppm. After being exposed to air for 3 months, it still maintains 74.23% of its initial responsivity demonstrating the excellent long-term stability. Moreover, the systematic investigation on the effect of annealing temperature to the sensing performance of TRGO elaborates that with the increase in thermal reduction temperature, the responsivity monotonically decreases while the response time will decrease initially and then gradually increase after the turning point of 250 °C. These results shed a light on developing TRGO-based ammonia sensors (TBASs) for future practical applications.

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Introduction

Ammonia (NH₃), a colorless and poisonous gas with pungent odor, can cause serious irritation and injury to human eyes and mucous membranes. Excessive inhalation of ammonia leads to lung swelling, even life-threatening in severe cases [1]. According to the regulation of occupational safety and health administration (OSHA), the exposure time to ammonia should be restricted to 8 h and 15 min with a concentration of 25 and 35 ppm, respectively [2]. Additionally, as one of the major causes of haze, ammonium salt aerosol in the atmosphere, which directly or indirectly resulted from the ammonia involved reaction, contributes 17% of PM 2.5 [3]. Non-invasively early stage diagnosis of some diseases can be achieved by detecting the concentration of ammonia in human exhaled gas [4]. Therefore, rapid, accurate and stable detection of ammonia is pressingly needed.

At present, commercial ammonia sensors are mainly made of metal oxides, which have the advantages of high sensitivity, easy integration and low raw material cost [5, 6]. However, they always require rather high operating temperature about 150–300 °C [7], which causes high-power consumption and largely limits applying scenarios such as the environment with explosives due to the potential safety problem of high temperature [8–10].

As a two-dimensional material, graphene has aroused much attention in gas sensing due to its extremely high specific surface area (2630 m^2g^{-1}) [11], superior room temperature conductivity and mechanical stability. Moreover, when the Fermi level is near the conical point, even a trace amount of charge transferring from the gas molecules to graphene leads to its significant change in conductance due to its unique Dirac conical band structure [12, 13], which gives it extremely high sensitivity in low concentration gas detection. Additionally, the very low electronic noise level of graphene further ensures the effective measurement of weak sensing signals [14]. Graphene is highly compatible with modern micro and nano processing technology, and it is easy to develop integrated gas sensor arrays with high efficiency and reliability. However, since the lack of dangling bonds on graphene, gas molecules are generally adsorbed through van der Waals forces. For this reason, the gas adsorption capacity and selectivity of pristine graphene is rather weak [15]. In

contrast, plenty of oxygen-containing functional groups and defects of rGO provide a large number of active sites for gas adsorption. Additionally, the same reasons also make rGO well dispersed in both aqueous and organic solutions [16] and facile to be tuned in sensing properties through surface functionalization [17]. Combing with aforementioned advantages and the low-cost massive production capability, rGO has been considered as one of the most promising materials for gas sensing that is capable to be operated at room temperature.

In 2008, Robinson et al. [18] reported the first study of rGO-based acetone vapor sensor, where the rGO membrane was made through the hydrazine vapor deoxidization. In the past decades, several GO reduction methods have been developed, including chemical [19, 20], thermal [21-23] and laser [24] reduction, and most of them have been applied to fabricate rGO-based ammonia sensors. In 2018, Li et al. conducted thermal reduction of GO with different layers of self-assembly, in which the response time of double-layer rGO to 50-ppm ammonia was 24 s [21]. Minitha et al. reported a systematic investigation on the ammonia sensing properties of hydrazine hydrate reduced GO and further unveiled the relation between reduction degree of GO and its sensing performance. It showed that at 35% relative humidity, the sample reduced for 15 h had the best repeatability to 400-ppm ammonia with response time of 30 s [19]. Later, Avik et al. [20] and Kavinkumar et al. [24] also evidenced the dependence of the ammonia sensing performance of rGO on the degree of reduction.

Among the current methods for GO reduction, almost all reducing agents, such as hydrazine, are environmentally unfriendly, and normally, the involved reactions are rather violent, which are unavoidably introduce a non-negligible number of holes and reaction residuals to the resulted graphene [25]. In addition, with the gradual decreasing of oxygen-containing functional groups, GO becomes less hydrophilic and tends to agglomerate [26], which is not conducive to maintain a large specific surface area. Another method for GO reduction is laser reduction; however, in order to reach expected energy uniformity of laser spot, the required hardware is rather costly; otherwise, the significant nonconsistency of fabricated material will be the most serious hurdle in it applications [27].

In contrast, thermal reduction of ultra-flat substrate supported GO film not only maintains its large specific surface area, but also realizes more uniform reduction which can both effectively avoid the introduction of defects through the appropriate control of heating temperature, and the method itself is very economical [28]. However, current ammonia sensors prepared by TRGO still suffer from long response time and poor stability in low concentration ammonia detection [29-31]. Therefore, a thorough exploration on the relation between reduction temperature and the sensing performance of TBASs is highly desirable, which could be essential for gain critical information to develop fast response and lowpower ammonia sensors for future practical applications.

Herein, smooth and uniformly dispersed TRGO sheets of large size are fabricated by spinning dilute GO suspension combined with subsequent in situ thermal reduction and used for rapid detection of ammonia at room temperature. This solution is more controllable and facile compared with the less reproducible drop-casting method. The uniformly dispersed GO sheets can be reduced into rGO with well-controlled reducibility by in situ heating. Therefore, it provides great advantages for developing TBASs. The effect of thermal reduction temperature on the content of defects, oxygen-containing functional groups and the type of semiconductor of TRGO is systematically investigated. Then, the relationship between these factors and the ammonia sensing performance is explored and interpreted further by relevant sensing mechanism. Consequently, highly stable and low-cost miniature TBASs are successfully fabricated that exhibit great sensing properties to ammonia at room temperature.

Experimental

Materials

Natural graphite powder (-325 meshes, 99.8%) was purchased from Alfa Aesar. Analytical grade concentrated sulfuric acid (H₂SO₄, 98%) and hydrochloric acid (HCl, 36%) were ordered from Tianjin Yuanli Chemical Co., Ltd. Sodium nitrate (NaNO₃, 99%) was acquired from Aladdin Co., Ltd. Analytical grade potassium permanganate (KMnO₄) was purchased from Tianjin Fengchuan Chemical Co., Ltd. Hydrogen peroxide (H_2O_2 , 30%v/v) was obtained from Tianjin Zhengcheng Chemical Co., Ltd. All reagents were used as received without further treatment. The testing gases were mixed by NH₃ and N₂ with concentration of 99.999% bought from Tianjin Ruixin Special Gases Institute and Tianjin Liufang gas company, respectively.

Fabrication of TBASs

The details of GO synthesis are presented in the Supporting Information. The SEM, XRD and AFM characterizations of GO are shown in Figure S1. The TBASs were fabricated on a SiO₂/Si substrate as schematically illustrated in Fig. 1. The procedure started from the gate electrode patterning through photolithography, followed by e-beam evaporation deposition to fabricate the gate electrodes which was made of 20-nm thick Cr and 20-nm thick Au sequentially. Then, 15-nm thick alumina was deposited as a dielectric layer by atomic layer deposition at 140 °C. Subsequently, interdigitated electrodes of 20-nm thick Cr and 20-nm thick Au with finger-width and inter-finger spacing of about 10 and $7 \,\mu m$, respectively, were fabricated. Finally, 20-µL GO dilute suspension (0.3 mg ml⁻¹) was spun onto the patterned channel area. These devices were treated in a home-built annealing furnace at 130, 165, 200, 250, 300 and 350 °C in Ar atmosphere at 8.2 mbar for 2 h, and the corresponding reduced GO was named as 130 °C-TRGO, 165 °C-TRGO, 200 °C-TRGO, 250 °C-TRGO, 300 °C-TRGO and 350 °C-TRGO, respectively. Before gas sensing test, the sensor was connected by Al wires from the electrodes to the chip carrier via silver paint.

Sensing performance characterization

The sensing tests were carried out on a home-built gas measuring system as shown in Figure S2, more detailed information can be found in ref.32. The specific concentration of gas setting was realized by tuning the flow ratio between the target gas and dry N_2 gas as a buffer. In the test, the experiment was conducted by alternatively flowing the target gas and dry N_2 into the chamber for testing and purging, respectively. Electrical signal acquisition was carried out using a PC controlled Keithley 2450 Source Meter though a homemade data acquisition program. In order to mimic the real applying environment, the





Spin Coating of GO

Figure 1 Schematic of fabrication process of TBASs.

sensing tests were conducted under different humidities, which was realized by changing the humidity of the injected air. The responsivity of the sensor is defined as follows:

$$\text{Responsivity}(\%) = \frac{R_{\text{g}} - R_0}{R_0} \times 100 \tag{1}$$

where R_0 and R_g are the measured resistances of the sensor before and after the exposure to NH₃, respectively. The response time is defined as the time taken for the sensor to achieve 60% of its maximum resistance change [32, 33].

The calculated LOD of the sensor is defined as follows:

$$LOD = \frac{3 \times RMS_{noise}}{Sensitivity}$$
(2)

where $\text{RMS}_{\text{noise}}$ is the noise level of the responsivity baseline before exposure to NH_3 [34]. The following discussion of LOD in this paper refers to the calculated value.

Characterizations

Scanning electron microscopy (SEM, SU3500, Hitachi, Japan) was used to image the morphology of GO and TRGO films. The X-ray diffraction (XRD) data were collected using a TD-3500 system (Dandong Tongda Co. Ltd., China) with Cu K α 1 X-rays of 0.157418 nm in the scattering range of 10°–80° with a rate of 2.4°/ min. The atomic force microscopy (AFM) image of

GO was obtained by a NX 10 system (Park Instrument, South Korea). Raman spectroscopy was performed on a Raman spectrometer (RTS-2, Titan Electro-Optics Co. Ltd., Hong Kong) using a 532-nm laser source. XPS spectra were collected with the ESCALAB 250Xi XPS system with an Al K α X-ray source.

Results and discussion

Characterization of TRGO

Raman spectroscopy is one of the most important measurements to characterize carbon materials. It has been widely applied to investigate the electronic structure, defects level and the functional group of graphitic materials [35]. Figure 2a exhibits that the Raman spectra of GO, 130 °C-TRGO and 350 °C-TRGO, more for TRGO annealed at 130-350 °C are shown in Figure S3. All of these spectra contain the following two peaks: (1) The G band at 1590 cm^{-1} is characteristic of sp^2 hybridization of carbon atoms and reflects the degree of graphitization of the TRGO. This band is due to the in-plane stretching vibration of carbon atoms. (2) The D band at 1350 cm^{-1} is derived from the in-plane breathing vibration of the six-membered aromatic carbon rings, which can reflect the degree of disorder and defects of graphene [27]. Figure 2b summarizes the I_D/I_G of TRGO



Figure 2 a Raman spectra of GO, 130 °C-TRGO and 350 °C-TRGO, b I_D/I_G of TRGO as a function of reduction temperature. C1s peak curve fittings of c 130 °C-TRGO and d 350 °C-TRGO.

reduced at different temperatures. It shows that with the reduction temperature increasing from 130 to 300 °C, the I_D/I_G decreases from 1.044 to 0.898, indicating the more reduction of GO. As the thermal reduction goes on, oxygen-containing functional groups on GO layers are gradually removed, and the content of sp^3 hybrid carbon atoms is gradually decreased, so the intensity of D band diminishes [36]. Consequently, the gradual restoration of sp^2 hybrid carbon atoms conjugate ring structure results in the enhancement of G band intensity. While, further thermal treatment leads to the increase in $I_{\rm D}/I_{\rm G}$. When the temperature exceeds 300 °C, the GO sheets are severely damaged by the deoxidation process and large amount of vacancy and edge defects result in its disordered structure [37].

The synthesized GO and TRGO are further analyzed by X-ray photoelectron spectroscopy (XPS) to evaluate their chemical components. Figure S4a shows that GO mainly has the characteristic peak of carbon and oxygen without other impurities. As shown in Fig. 2c, the C1s peak curve fitting of 130 °C-TRGO mainly consists of four major peaks. They center at 284.60, 286.62, 287.87 and 289.30 eV, which can be attributed to the C-C, C-O (hydroxyl and epoxy), C=O (carbonyl) and O–C=O (carboxyl) groups, respectively [38]. In Fig. 2d, the peak related to carboxyl nearly disappears in 350 °C-TRGO, and the contents of C-O and C=O are also low. The fittings of C1s peak in GO and TRGO reduced at the temperature range of 130-350 °C are shown in Figure S4b-f. With increasing thermal reduction temperature, the contribution of oxygen-containing functional groups (the sum of the peak areas of four functional groups divided by the full C1s peak area) gradually decreases from 26.53% (GO) to 9.88% (350 °C-TRGO), indicating its gradual increase in reduction degree.





Figure 3 a SEM image of the channel area of TBAS and white dashed lines is used to trace the edges of the TRGO sheets. b Current–gate voltage ($I_{DS} - V_G$) transfer characteristic of 130 °C-TRGO.

The whole structure of TBAS is shown in Figure S5a, where the surface of sensor is clean enough. Figure 3a shows the SEM image of TRGO sheets bridging the gaps of interdigitated electrodes. Most of TRGO sheets are larger than 10 µm and well dispersed on substrate forming a smooth morphology, which keeps the two-dimensional nature of graphene and the good contact between TRGO and electrodes. All of this ensures the largest extent exposure of TRGO sheets. With the increase in reduction temperature, the conductivity of TBAS improves as shown in Table S1 corresponding to the enhanced reduction degree. The I-V curve of 130 °C-TRGObased sensor is shown in Figure S5b, whose linearity and almost zero intercept to the Y-axil confirm the ideal ohmic contact between the TRGO and Au electrodes. Again, the $10^6 \Omega$ resistance of 130 °C-TRGO reflects the partial reduction of GO at low temperature with leftover of rather large amount of oxygen-containing functional groups. The transfer characteristic of 130 °C-TRGO measured under N2 purging is shown in Fig. 3b. A typical bipolar characteristic of graphene is observed with the measured neutral point at -0.095 V indicating a slightly n-typed semiconductor property.

NH₃ sensing properties of TBASs

Response time and responsivity are two key criteria for gas sensors. Figure 4a displays the dynamic response of 130 °C-TRGO-based sensor to NH_3 with various concentrations ranging from 5 to 200 ppm at room temperature. It shows that the electrical resistance of the sensor decreases rapidly with exposure to NH3 and gradually recovers after N2 purging. This can be attributed to the electrons transferring from ammonia to TRGO due to its nature of reducing agent, thereby increase the majority carriers (electrons) concentration in the n-type graphene and result in the decrease in its resistance. The responsivity and response time of 130 °C-TRGO-based sensor to 20-ppm NH₃ are about 0.81% and 35 s, respectively. And its response curve to 5-, 10- and 20-ppm ammonia is shown in Figure S6, in which 130 °C-TRGO-based sensor also exhibits apparent and stable response to other low concentration ammonia. Figure 4b shows the fit curves of the responsivity of this device as a function of ammonia concentration. It exhibits that the responsivity monotonically increases with increasing ammonia concentration, and there is a one-to-one correspondence between them. A LOD as low as 0.9 ppm of the sensor is calculated according to the data presented in Fig. 4b using Formula (2). Details of the calculation are shown in Figure S7. To check the reliability of the sensor, the response of 130 °C-TRGO-based sensor is tested in 100-ppm NH₃ as shown in Fig. 4c. The uniformly distributed TRGO sheets on substrate opt to ammonia molecules adsorption which results in excellent sensing performance of the devices.

In order to determine the optimal reduction temperature for the fabrication of TBASs and the mechanism of the rapid response, the sensing properties of TBASs reduced at different temperatures (130, 165,



Figure 4 a Dynamic response of 130 °C-TRGO to 5–200-ppm ammonia at room temperature. **b** Fit curves of the responsivity of 130 °C-TRGO as a function of ammonia concentration. **c** Response of 130 °C-TRGO of multi cycles to 100-ppm ammonia.



Figure 5 a Dynamic response curves and **b** sensing difference in response time and responsivity of TBASs reduced at different temperatures (130, 165, 200, 250, 300 and 350 °C) to 100-ppm ammonia. **c** The responsivity of TBASs reduced at different

temperatures (130, 165, 200 and 250 °C) as a function of ammonia concentration from 60 to 200 ppm. **d** Dynamic response and recovery curve of 250 °C-TRGO to 100-ppm ammonia and the enlarged inset indicates the response time of about 11 s.

200, 250, 300 and 350 °C) are systemically investigated. Figure 5a shows the dynamic response to 100-ppm NH_3 of TBASs reduced at different temperatures, and Fig. 5b shows the corresponding responsivity and response time. It is found that with the increase in thermal reduction temperature, the responsivity gradually decreases, and the response time decreases initially and then increases. Among



81.62% and 74.23% of its

these sensors, the responsivity of 130 °C-TRGO is the highest one-up to 1.97%, and the response time of 250 °C-TRGO is the shortest of 13.4 s on average. The relation between the responsivity of TBASs reduced at different temperatures (130, 165, 200 and 250 °C) and the ammonia concentration, ranging from 60 to 200 ppm, is shown in Fig. 5c. It is noticeable that 130 °C-TRGO shows the highest responsivity to all measured ammonia concentrations. Figure 5d exhibits the dynamic response and recovery curve of 250 °C-TRGO to 100-ppm ammonia, and the enlarged inset indicates that the response time is as short as 11 s.

It is worth noting that the response direction reverses from the resistance decline to rise when the sample reduction temperature is above 250 °C, which could be attributed to the flip of its semiconductor type. The GO reduced at low temperature (≤ 250 °C) is n-type semiconductor, while it becomes p-type when the reduction temperature is higher than 250 °C. The detailed transfer characterization of GO reduced at different temperatures is shown in Figure S8. In GO- and rGO-based devices, the variation of the ratio between electron acceptor (carboxyl and carbonyl groups) and donor (sp^2 hydroxyl, ether and epoxy) of oxygen-containing functional groups is the key that results in the electrical doping transition [39].

In order to test the durability of TBASs, the sensing performance to 100-ppm ammonia of 130 °C-TRGObased device kept under the ambient condition with humidity and temperature varying naturally for 3 months is tested. The results are demonstrated in Fig. 6a, the responsivity of the sensor remains at 92.45%, 81.62% and 74.23% of its initial value while the response time remains 123%, 138% and 117% for 50, 79 and 89 days, respectively. This demonstrates the good long-term stability of TBASs.

Considering the fact that the most common application environment of ammonia sensors is in refrigeration industry [40], so oxygen and water vapor in the air are the main interfering gases. The oxygen concentration is generally constant; in most cases, it can be treated as a flat background; therefore, the key interference comes from the humidity which has large variations with change of time and locations [41, 42]. To this point, it is critical to explore the influence of humidity on sensing performance and the dependence of output results on humidity. As shown in Fig. 6b, the responsivity is enhanced with the increasing RH (relative humidity) when RH is less than 30%, then it begins to decline. When the RH is higher than 60%, the responsivity is even lower than that one in the completely dry environment. Two main causes may account for these phenomena: (1) Under a relatively low RH, water is conducive to the deprotonation of carboxyl groups, which could promote the acid-base reaction between NH⁺₄ and carboxyl groups after ionization and enhance the specific adsorption of ammonia to carboxyl groups on the surface of graphene [43]. (2) When the RH is too high, the water molecules prevail ammonia molecules on rGO adsorption sites [44]. In practical applications of TBASs, one possible way to resolve this interference can be using such a type of systematic conducted measurements as a calibration with assistance of a real-time humidity information.



Figure 6 a Long-term stability of 130 °C-TRGO upon exposure to 100-ppm ammonia. b Responsivity to various ammonia concentrations of 130 °C-TRGO as functions of RH.

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In terms of the main interfering species in industrial cooling, the relevant selectivity test against oxygen, water vapor and another general gas (NO_2) is studied and presented in Figure S9, which indicates that the sensor has completely oppositely polarized response of NH_3 to all other tested gases.

A comparison between the ammonia sensing performance of the TBASs in this work and other results of graphene-based sensors is summarized in Table 1.

Gas sensing mechanism of TBASs

In this experiment, it is observed that with increasing thermal reduction temperature, the responsivity of TBASs to ammonia gradually decreases. This is due to the fact that gas molecules mainly bond to the carbon atoms of rGO, oxygen-containing functional groups and defects [53-55]. Among them, the rich oxygen-containing functional groups on rGO contain abundant hydrogen bonds which can largely promote ammonia adsorption on the polar functional groups [56, 57]. Therefore, the type and concentration of oxygen-containing functional groups mainly determine the gas sensing performance of TBASs. Low-temperature reduction of GO will result in relatively high concentration of residual oxygen-confunction groups, consequently taining more abundant active sites for adsorption of ammonia

molecules [58, 59]. However, when the reduction temperature is above 300 °C, most of functional groups on GO surface have been removed, which greatly weakens the responsivity of TBASs. As shown in Fig. 7a, with the increase in thermal reduction temperature, the responsivity decreases which is consistent with the content of residual functional groups.

Moreover, it is found that with increasing thermal reduction temperature, the response time of TBASs to 100-ppm ammonia decreases first and then increases. Maity et al. theoretically revealed the adsorption energy between different kinds of oxygen-containing functional groups and ammonia, the results showed that in all main functional groups of GO, the carboxyl group (-0.707 eV) had prominent higher adsorption energy than all others such as hydroxyl group (-0.406 eV), carbonyl group (-0.108 eV) and epoxy group (- 0.106 eV) [60]. Figure S10 shows the schematic of ammonia molecules adsorption on the TRGO film. The adsorption energy and charge transfer of ammonia molecules adsorbed on diverse sites of TRGO are different from each other. Since the higher the adsorption energy the fast the device response will be, as shown in Fig. 7b, the measured contribution of carboxyl group as a function of reduction temperature is nearly perfectly opposite to the result that the temperature-dependent response

Table 1 Performance of the presented NH₃ sensor in this work compared with the previous works

Material of sensor	Operating temperature	Concentration (ppm)	Responsivity (%)	Response time (s)	References
rGO (Hydrazine solution chemical reduced)	RT	300	3.1	28.5	[19]
rGO (Hydrazine solution chemical reduced)	RT	50	13.18	750	[45]
rGO (Hydrazine vapors chemical reduced)	RT	400	930	31	[20]
rGO (NaBH ₄ solution chemical reduced)	RT	1200	15.7	422	[46]
rGO (Glucose solution chemical reduced)	RT	800	12	505	[47]
rGO (Spray coated)	RT	80	~ 5	~ 300	[48]
rGO (LBL self-assembly and thermal reduced)	RT	50	5.7	24	[21]
rGO (Laser reduced)	30	50	~ 5	51	[24]
GO film	RT	50	4	30	[49]
Graphene (Aerosol jet printed)	RT	4.35	4.64	51.2	[50]
Graphene foam	RT	1000	22.5	347	[51]
rGO-Graphene hybrid film	RT	10	14.67	74	[52]
250 °C-TRGO (Thermal reduced)	RT	100	0.53	13.4	This work
130 °C-TRGO (Thermal reduced)	RT	20	0.81	35	This work



Figure 7 a Content of oxygen-containing functional groups according to the XPS C1s peak curve fittings and responsivity as a function of reduction temperature. b Content of COOH and response time as a function of reduction temperature.

time of TBAS afore-discussed. Noticeably, both the highest concentration of carboxyl and the fastest response appear at 250 °C-TRGO.

Conclusion

In this paper, we obtain smooth and well-dispersed TRGO film with flake size more than 10 µm by spinning dilute GO suspension combined with subsequent in situ thermal reduction processing. Using TRGO as sensing material, rapid responding, highly stable and low-cost miniature ammonia sensors are successfully developed. The testing results show that 250 °C-TRGO-based sensor has the fastest response to 100-ppm ammonia with the response time as short as 11 s, which can be attributed to the dominant content of carboxyl resulting in high adsorption energy to ammonia. The highest responsivity appears at 130 °C-TRGO-based sensor which is up to 1.97%-100-ppm ammonia with the response time of 23 s, and it has outstanding long-term stability benefiting from its rich oxygen-containing functional groups. The thermal reduction temperature is an effective parameter to be tuned for fabricating ammonia sensor according to the specific application scenario. The work reported here sheds a light on satisfying the needs of rapid responding and low-power ammonia sensors for the practical application at room temperature.

Supplementary information

Supplementary information including measurements for performance tests, characterization (SEM, XRD and XPS of the as-prepared samples) and supporting figures.

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Author contributions

XX worked in methodology, investigation, data curation and writing—original draft preparation. WJ helped in investigation and data curation. CT carrying out experiments of Raman and data curation. XQ carrying out experiments of photolithography and data curation. RL carrying out experiments of photolithography. YZ carrying out experiments of annealing. WZ carrying out experiments of XPS. XY contributed to writing—review and editing. XZ contributed to writing—review and editing. YM helped in validation, supervision and writing—review and editing. LM helped in conceptualization, methodology, validation, supervision, writing—review and editing and funding acquisition.

Data and code availability

The datasets generated or analyzed during this study are included in this published article and its supplementary information files.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical approval There are no experiments carrying out involving human and animal tissue in this manuscript.

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