

High Sensitive NO₂ Gas Sensor Based on Self-assembled Monolayer Graphene Decorated with Ag Nanoparticles

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Abstract:

We present a high performance NO₂ gas sensor based on neat monolayer graphene film decorated with Ag nanoparticles. The RGO/Ag nanocomposites were bound to the electrode by the 3-Aminopropyltriethoxysilane (APTES) assisted self-assembly technique. In the hybrid material, the sensing film with few overlapping and gaps provides large sensing area per unit volume. Besides, with the catalytic activity of the uniformly loaded Ag nanoparticles, it exhibits high sensitivity. This sensor can work at room temperature, which makes it available for practical application. Furthermore, it can fully recover to the initial state without the treatment of heating or light irradiation, which shows good reversibility.

Key words: high performance, monolayer, self-assembly method, room temperature, fully recovery

Introduction

Nowadays, with the overwhelming development of technology, an increasing number of nitrogen dioxide (NO₂) have been produced, which can cause acid rain and respiratory problems for human beings[1]. Compared with other sensing materials, graphene is appealing because of their room temperature carrier mobility, low electrical noise and large surface area, which makes it sensitive at room temperature.[2]

Even so, graphene-based gas sensors still face some critical challenges on the sensing performance, which are the response time and recovery time. From the theoretical aspect, it can be elucidated that monolayer graphene maximizes the sensing effect because its whole volume exposed to surface adsorbates. Among various monolayer graphene synthesis method such as CVD[3] and epitaxial growth[4], RGO derived from graphene(GO), has the merits of scalable production and easy processability, which make it a very attractive material for graphene-related electronics applications.[5] Nevertheless, it is difficult to control the morphology and the thickness of the film due to the inevitable agglomeration.[6]

In this work, in order to solve above problems, effectively enhanced gas sensitive monolayer graphene /Ag nanosheets were fabricated using the APTES assisted self-assembly technique, which contributes to the performance of the sensor. Besides, the loading of AgNPs catalysts can enhance the response and this sensor can detect NO₂ at room temperature with high sensitivity and short response time.

Experiments

The RGO/Ag composites were prepared by one-step chemical reduction method. 100ul, 600ul, 1000ul AgNO₃ (0.1mol/L) solution were slowly added into the 20cm³ stirring GO suspension (1mg/mL) named S₁, S₂, S₃, respectively. Next, 50ul NH₃·H₂O and 10ul N₂H₄·H₂O were added to the above suspension under ultrasonic processing for 30 min. The solution was heated to 90°C for 1.5h with stirring to perform the reduction and Ag⁺.

The scheme of the fabrication process of monolayer RGO/Ag gas sensor is shown in Fig.1. The substrate was treated by UV-ozone treatment for 10 min to make the surface of the substrate hydrophilic. Then the interdigitated electrode was immersed into the 2% APTES solution. Last, the spin-coating method was used to assemble the sensing materials on the interdigitated electrodes.

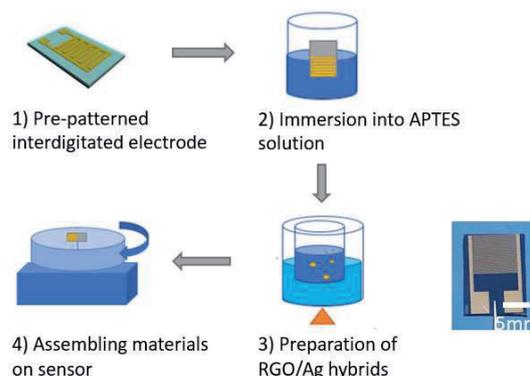


Fig.1. Schematic fabrication process of the gas sensor with the monolayer RGO/Ag sensing film.

Results and discussions

The mechanism of the self-assembly method is shown in Fig.2. APTES serves as the linkage, which immobilized on the substrate through the interaction of silane on APTES with the –OH groups. Because the RGO of our sensing materials is not fully reduced, there still many –OH or–COOH groups on the surface of the nanosheets, which can make covalent bond with the –NH₂ groups.

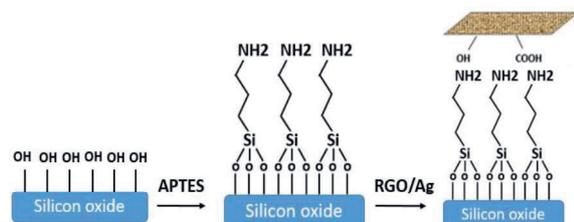


Fig.2. Schematic illustration of the principle about the self-assembly method.

The Fig.3.(a) shows a SEM image of the RGO sheets which bound to the interdigitated electrode. Fig.3.(b) gives a SEM image of the RGO/Ag nanocomposites, showing the ultrathin RGO sheets just like a chiffon with the decoration of AgNPs. Silver nanoparticles are uniformly dispersed on the RGO sheets with almost similar sizes, ranging from 30nm to 40nm.

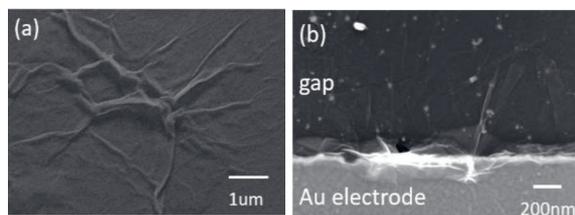


Fig.3.(a) Typical SEM image of an RGO membrane using the self-assembly method.(b) Typical SEM image of an RGO/Ag membrane using the self-assembly method.

Fig.4.(a) illustrates the real-time sensing response of the RGO-based sensing advices(S₁–S₃) in sequence. At the NO₂ concentration from 0.5ppm to 10ppm, the S₂ sensor shows the best performance and the maximum sensitivity achieves 26.9% ppm⁻¹. The saturated response time is shorter than 680s. Besides, nearly no drift of baseline is observed during the multiple cycle process, which implies the good recovery of the sensor without the UV/IR light or thermal assistance. The good performance of our sensor is due to the morphology of the sensing film made by the self-assembly method. According to the absorption model developed by the Langmuir, we fit our results in Fig.4.(b) and it shows good agreement with the model.

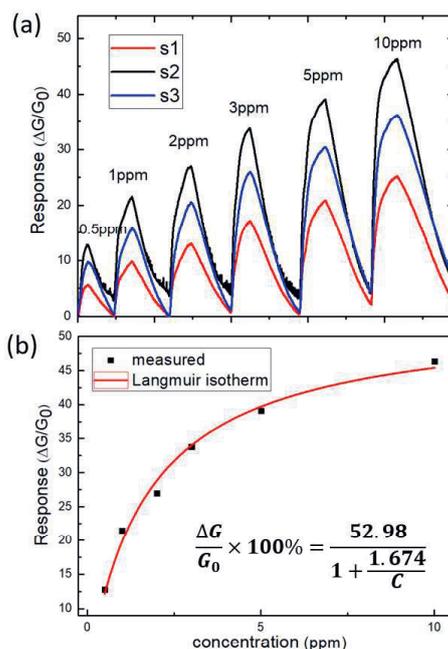


Fig.4.(a) Response of sensor_{1,2,3} upon exposure to NO₂ concentrations ranging from 0.5ppm to 10ppm.(b) Response vs NO₂ concentration applied to the sensor showing an agreement between the measured values and the fitted Langmuir isotherm.

Conclusion

In this paper, monolayer RGO/Ag sensing film was fabricated by the APTES assisted self-assembly method. The sensor exhibits a fast and sensitive response at room temperature to NO₂, of which maximum sensitivity achieves 26.9% ppm⁻¹. Our sensor can overcome the shortcoming of traditional graphene-based gas sensor. Besides, it is easy to fabricate, which shows the prospect of practical application.

References

- [1] Huan Liu, Min Li, et al. Physically Flexible, Rapid-Response Gas Sensor Based on Colloidal Quantum Dot Solids. *Adv. Mater.* 2014, 26, 2718–2724; doi:10.1002/adma.201304366
- [2] Xiao Huang, Zongyou Yin, et al. Graphene-Based materials: Synthesis, Characterization, Properties, and Applications. *Small.* 2011, 7, 1876; doi: 10.1002/smll.201002009
- [3] Amin Salehi-Khojin, David Estrada, et al. Polycrystalline Graphene Ribbons as Chemiresistors. *Adv. Mater.* 2012, 24, 53; doi: 10.1002/adma.201102663
- [4] Md.W.K. Nomani, Razib Shishir, et al. Highly sensitive detection of NO₂ using epitaxial graphene on 6H-SiC. *Sens. Actuators B.* 2010,150,301; doi: 10.1016/j.snb.2010.06.069
- [5] Nantao Hu, Yanyan wang, et al. Gas sensor based on p-phenylenediamine reduced graphene oxide. *Sens. Actuators B.* 2012. 163. 107; doi:10.1016/j.snb.2012.01.016
- [6] M. Acik, G. lee, C. Mattevi, et al. Unusual infrared-absorption mechanism in thermally reduced graphene oxide. *Nature Materials.* 2010 ,9, 840-845; doi: 10.1038/nmat2858