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Research Article

The influence of thickness on ammonia gas sensitivity of reduced graphene oxide films

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ABSTRACT

Graphene is a single carbon layer in a two-dimensional (2D) lattice. Its delocalized π bonds give rise to unique electronic properties, but these π bonds are easily influenced by the environment. Meanwhile, many publications present that the sensitivity of graphene is not only necessarily intrinsic to this material but also by external defect. In this study, we produced reduced Graphene Oxide (rGO) sensors based on random rGO plates. We analyzed the ammonia (NH_3) sensitivity of such sensors as a function of thickness of rGO films (in terms of change in transparence) at room temperature. When the thickness of rGO films decreased, a maximum response was observed for the thinnest rGO film (the transparence was 84 %), with a sensitivity up to 38 %. Our results suggest that the dependence of NH_3 sensitivity on rGO films thickness is dictated by the fully exposed surface area for thinnest films and by 2D charge carrier hopping through edge defects.

Key words: Graphene, Ammonia gas sensing, Reduced Graphene Oxide, Defects

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INTRODUCTION

Many researchers have shown that the sensitivity of rGO film can be decreased by oxygen-containing groups (epoxy groups, hydroxyl groups, *etc.*) [1,2], and by surface and edge defects of rGO [3,4]. The effects of the oxygen-containing groups on the gas-sensing signal can be controlled by the reduction process from GO to rGO films (dependent on the reducing agent). Moreover, as reported by Lili Liu *et al.* [3], structural defects can also affect gas sensitivity signals. When the defects are in the rGO lattice, they will naturally have impacts on the electronic structures, such as bond lengths in the strain fields of the defects, the local re-hybridization of sigma and π-orbitals, and the scattering of electron waves [3].

In this study, we investigated ammonia (NH_3) gas sensitivity with different thickness of rGO films by two steps. Firstly, rGO films were synthesized by the chemical method with different thickness through different volumes of rGO solution [5], and secondly, these rGO films were investigated for NH_3 gas sensitivity at room temperature [6]. It is important to note that the effect of the oxygen-containing groups on the sensitivity of rGO films was fixed by the stable reducing condition. In the study herein, we focus on the structural defects (surface and edge defects) that directly affect gas sensitive signals when the rGO films are overlapped. These defects can be controlled by the different thicknesses of rGO films because the electronic properties of two-dimensional (2D) lattices strongly depend on the thickness of materials [3,5,7].

METHODS

Synthesis of the reduced graphene oxide (rGO) and fabrication of gas sensor

The fabrication process of gas sensor based on the reduced graphene oxide (rGO) material was performed by the following protocol. Firstly, the graphite (Sigma-Aldrich, India) was exfoliated by microwave irradiation and then, the exfoliation graphite was oxidized to GO by chemical method- with the mixture of 0.8g $KMnO_4$ /16ml $H_2 PO_4$ /0.1g $NaNO_3$ (modified Hummers method): $KMnO_4$ (Duc Giang Detergent – Chemicals JSC, Vietnam), $H_3 PO_4$ (Xilong Scientific Co., Ltd, China), and $NaNO_3$ [1,8]. Secondly, GO material was deposited directly on spaced inter-digitated silver electrodes patterned on the clean (1 cm^2) substrate by using spin coating method (**Figure 1a**). During this period, we used different volumes of GO solution (from 0.04 ml to 0.25 ml) with the aim of changing the thickness of the achieved rGO films. Then, these GO films were exposed with hydrazine agent at 80^0 C and heated quickly at 350^0 C to reduce GO films to rGO films. Finally, we investigated the NH_3 gas sensitivity as a function of the thickness of rGO films at room temperature. Additionally, we used different spaced inter-digitated silver electrodes (space between lines was 1 mm and 1.5 mm) (**Figure 1b**).

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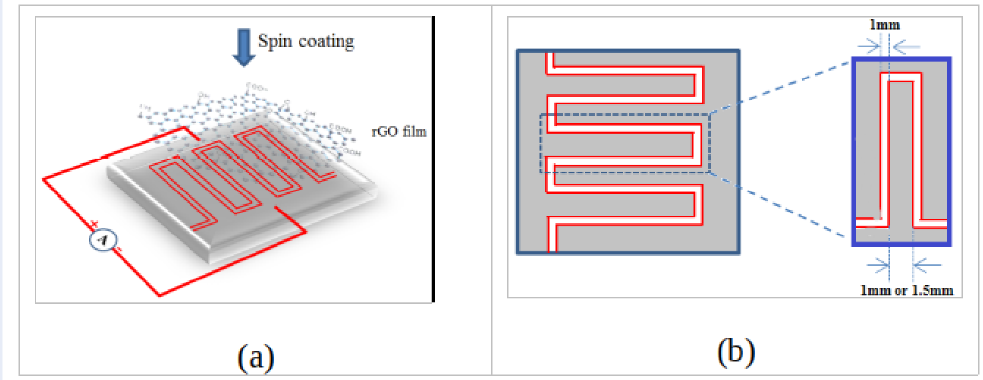

Figure 1: **The gas sensor.** (a) The spaced inter-digitated substrate with rGO film ; (b) space between lines was 1 mm and 1.5 mm.

Based on the rGO films used, we had two sensing samples which were named "rGO- space -volume". For example, rGO-1.0-0.04ml sample was fabricated on 1.0 mm spaced inter-digitated silver electrodes with 0.04 ml of GO solution. Tell what the 2^{nd} sensing sample was.

The measurement system

The gas sensor was connected to two probes in the test chamber and the signal was displayed on the screen computer by the transducer through the LABVIEW software. The measurement consisted of two processes that were called absorption and desorption. In the absorption process, the NH_3 gas flowed into the test chamber for the period time and the change in resistance of sensor was recorded during that time[2]. In the desorption process, the argon (Ar) gas was pumped into the test chamber to re-establish the initial resistance of rGO[2].

RESULTS

Investigating the change of thickness of rGO films

Caterina Soldano *et al.*[6] showed that graphite crystal becomes highly transparent when thinned down to a graphene monolayer (using Chemical Vapor Deposition method). Indeed, in the visible light region, the transparency of graphene monolayer was 97.7 % and it decreased linearly when the thickness of graphene was increased to five layers. However, as the thickness of graphene film continually increased, the transparency of graphene film should decrease nonlinearly[6,9].

Herein, we investigate the different thickness of rGO films using the transparency spectra by ultraviolet-visible (UV-vis) and Stylus method, as described in **Figure 2**.

Interaction of ammonia gas with the rGO films

After preparation of the gas sensor, we measured NH_3 gas sensitivity *($\Delta R/R0$)* of rGO films. For the spaced inter-digitated silver electrodes of 1.5 mm (i.e. rGO-1.5 sample), as shown in **Figure 3a**, the thinnest rGO film (rGO-1.5-0.04ml) demonstrated the highest sensitivity (34 %).

When the volume of the GO solution was increased from 0.04 ml to 0.25 ml, the sensitivity decreased from 34 % to 4.5 % (**Figure 3b**).

The result of rGO-1.0 in **Figure 4a** was similar to the result of rGO-1.5 in **Figure 3a**. When the volume of GO solution was increased, the thickness of rGO films became thicker and the sensing signal of rGO films decreased (**Figure 4b**). However, from **Figure 3a** and **b**, it can be seen that the NH_3 gas sensitivity of rGO-1.0 (38 %) was higher than that of rGO-1.5 (34 %).

Comparing our experimental results with the results of other research groups on the gas sensitivity of two-dimensional (2D) materials, there was some similarity. Therefore, the gas sensitive signals of 2D materials are optimal when their thickness are decreased to monolayer[5,7].

DISCUSSION

By ultraviolet-visible (UV-vis) setting, when the volume of GO solution was increased in the range of 0.04 ml to 0.25 ml (**Figure 2a**), the transparency of rGO films was decreased in the range of 84 % (rGO-0.04 ml sample) to 74 % (rGO-0.25 ml sample) at λ = 550 nm, as shown in **Figure 2b**. The result of the transparency of the rGO films was similar with the variation of thickness from 151 nm to 784 nm (**Figure 2b**),

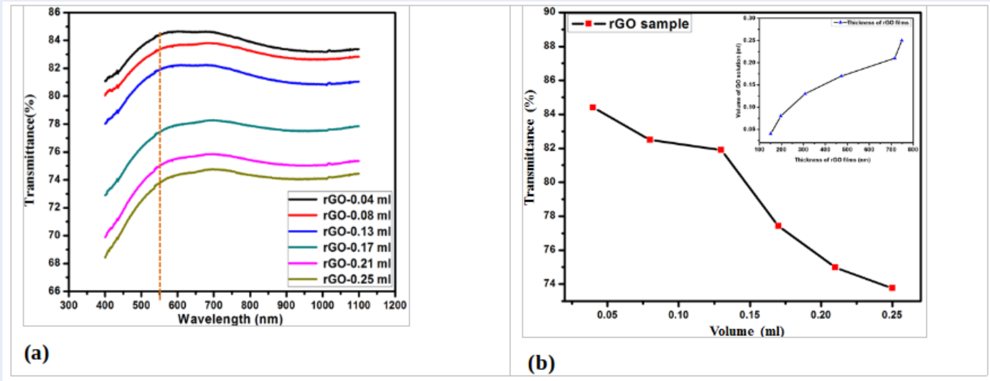

Figure 2: **The transparency spectra**. (a) the different thickness of rGO films, (b) dependence of transmittance on the GO volume (at λ = 550). In the inset: the different thickness of rGO film on GO volume.

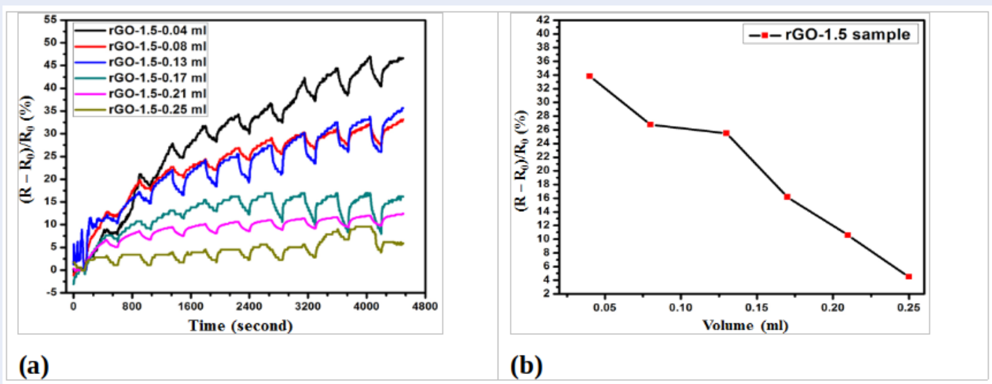

Figure 3: **The characteristic of NH_3 gas sensitivity.** (a) the ΔR/R0 value of rGO-1.5, (b) the *ΔR/R*0 value with different rGO-1.5 volume.

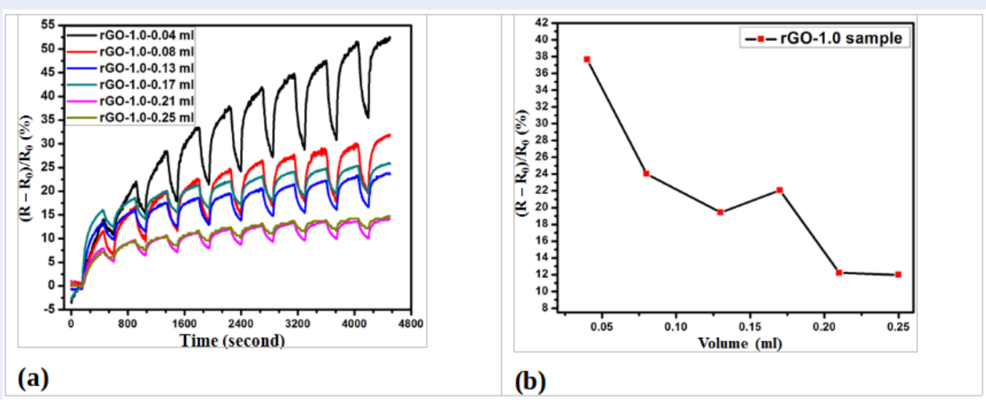

Figure 4: **The characteristic of NH_3 gas sensitivity.** (a) the ΔR/R0 value of rGO-1.0, (b) the ΔR/R0 value with different rGO-1.0 volume.

by the Stylus method. These results demonstrate that the transparency of rGO films is strongly affected by the thickness of the rGO film.

The transport of electrons of the gas sensor- based on rGO material- directly affected the sensitivity signal ($\Delta R/R0$) of the device. In **Figure 3**, when the rGO film was thinner, its gas sensitivity increased. This result could be explained by the fact that the rGO sheets in the rGO film were arranged in the most uniform manner and there was less overlap in the thinnest rGO film [3]. This produced the convenience for interaction between the NH_3 gas molecules and rGO sheets, not only on the planar sheet but also on the edge defects [3,4]. Hence, the surface resistance of the rGO film changed significantly.

This problem could be overcome by reducing the thickness of the rGO film and the distance between the electrode lines. In **Figure 4**, the sensitivity signal of this device was improved (from 34% to 38%). This can be explained by the fact that as the space between inter-digitated silver electrodes were decreased, the electron trajectories were shorter. This was easy for transmitting sensing signals to the measurement equipment. From our results, we suggest that when the space between electrode lines is continually decreased to micrometers, one rGO sheet can be used for making gas sensor and the response signal of the devices can be made more optimal.

CONCLUSION

When the rGO film was thinner, its gas sensitivity increased remarkably as follows: the rGO film decreased 5-fold, and the response signal of the device increased 3.2-fold. At that time, the distance between electrode lines decreased 1.5-fold, and the response signal increased ~1.2 times. However, our study has also shown the limitations of the thickness film; we fabricated the gas sensor substrate with a large electrode distance (millimeter). Moreover, we deposited the rGO film by chemical method which led to the rGO sheets being dispersed non-uniformly and overlapping together. In future experimental studies, we will decrease the electrode distance to yield the lowest rGO sheets, and the thickness of rGO films would be made thinner.

COMPETING INTERESTS

No conflict of interest declared.

AUTHORS' CONTRIBUTIONS

Tran Quang Nguyen implemented the experiment about the fabrication of gas sensor and the investigation of ammonia (NH_3)gas sensitivity based on reduced graphene oxide (rGO). Huynh Tran My Hoa synthesized rGO material from graphite flakes. We proposed the experiment plan and wrote the manuscript together. Tran Quang Trung helped us evaluated the stability of ammonia (NH_3) gas sensitivity based on rGO films.

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