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Self-activated Graphene Gas Sensors: A Mini Review

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Abstract

Graphene has been widely considered a promising candidate for high-quality chemical sensors, owing to its outstanding characteristics, such as sensitive gas adsorption at room temperature, high conductivity, high flexibility, and high transparency. However, the main drawback of a graphene-based gas sensor is the necessity for external heaters due to its slow response, incomplete recovery, and low selectivity at room temperature. Conventional heating devices have limitations such as large volume, thermal safety issues, and high power consumption. Moreover, metal-based heating systems cannot be applied to transparent and flexible devices. Thus, to solve this problem, a method of supplying the thermal energy necessary for gas sensing via the self-heating of graphene by utilizing its high carrier mobility has been studied. Herein, we provide a brief review of recent studies on self-activated graphene-based gas sensors. This review also describes various strategies for the self-activation of graphene sensors and the enhancement of their sensing properties.

Keywords: Gas sensors, graphene, self-heating, microscale patterning, surface modification

1. INTRODUCTION

Graphene, which is a representative two-dimensional (2-D) material, has been widely studied for varied applications such as electronics, optoelectronics, energy storage devices, and sensors owing to its intrinsic properties, namely high carrier mobility, tunable band gap, high chemical stability, and transparency with flexibility [1-7]. In particular, graphene has been considered the next-generation gas-sensing material, as its large specific surface area of 2-D planar carbon atoms is highly sensitive to the adsorption of gas molecules even at room temperature i.e., 25 °C [8-15]. However, graphene-based gas sensors suffer from very slow responses, and they do not fully recover to the initial state after a reaction with gas molecules at room temperature [16-18]. The gas sensing mechanism is based on the electrical resistance changes of the sensing materials caused by the adsorption and desorption of gaseous species [19-21]. The adsorption and desorption are temperature activated process, thus dynamic

sensing properties such as response and recovery times depend significantly on the operating temperature [22]. Moreover, other important factors of surface coverage, chemical decomposition, charge-carrier concentration, and work functions also depend on the temperature [23]. Thus, thermal energy should be provided at elevated temperatures to enhance the overall sensing properties. According to Fowler et al., reduced graphene oxide (rGO)-based gas sensors have to operate at 150 °C for repetitive NO₂ gas sensing [24]. An external heater such as a tube furnace is the most common way to increase the operating temperature, but it has the limitations of a large volume, high power consumption, and thermal safety issues. Although new sensing device structures such as rear-side heaters and microelectromechanical system structures have been used, the power consumption is not sufficiently low for these device structures to be adapted for portable devices [25]. In addition, these methods are based on metal-based heaters, which require brittle and non-transparent materials, hindering practical applications for flexible and transparent devices. Thus, a self-activating graphene-based gas sensor platform, operating at room temperature without an external heater, has been studied to solve this problem. The selfheating graphene-based gas sensor generates heat by the Joule heating effect using the extremely high electrical conductivity of graphene with micro-/nano-scale patterning techniques [26]. Joule heating is the process by which the energy of an electric current is converted into heat as it flows through a resistance. The heat is generated at the narrow graphene region by increasing the bias

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voltage, and the gas sensor can be activated without external heating [27-29].

This paper reviews recent studies on various self-activated graphene-based gas sensors. The synthesis of graphene, the fabrication of the sensor devices, and strategies to maximize the self-heating effects in each study are described. Furthermore, the self-heating abilities of the sensors are analyzed with respect to various factors, such as the type of graphene, tailored shapes and widths, and the number of graphene layers. The effects of selfheating on sensitivity, response time, and recovery time, which are important gas-sensing properties, are compared. Moreover, the power consumption and mass production, which are important for practical applications, are investigated. In addition, strategies to obtain high sensitivity to various specific gas molecules are addressed for maintaining the self-heating effect of graphenebased sensors.

2. SELF-ACTIVATED GRAPHENE-BASED GAS SENSORS

2.1 Gas molecule sensor integrated with sensing and heating graphene layers

Self-activating graphene gas sensors without an external heater were first developed by integrating a sensor and heater consisting of graphene. Choi et al. reported an NO₂ gas sensor based on sensing and heating graphene layers [30]. Fig. 1 shows real images of a selfactivated graphene gas sensor based on single-layer graphene (SLG) and bi-layer graphene (BLG). Graphene layers for gas sensors were grown on various substrates via chemical vapor deposition (CVD). CVD is the most favorable method for depositing graphene on a metal substrate, demonstrating excellent flexibility and transparency. The SLG layer was grown on a preannealed Cu foil via the CVD method at 1000 °C under a CH4 and H₂ gas atmosphere, followed by a rapid cooling process under an Ar atmosphere. The SLG film was transferred to a polyestersulfone (PES) substrate using polymethylmethacrylate (PMMA). After removing PMMA, an infrared laser with a resolution of 30 µm was employed to separate the sensing and heating regions. The BLG film was prepared on the PES substrate via the layer-by-layer stacking of SLG films by employing the same transfer procedure. The SLG layer was utilized as a channel located at the center, whereas the BLG layer was utilized as a heater located at the sides of the PES substrate. By adopting this structure, the SLG layer showed a good response toward NO₂ gas, and the BLG layer



Fig. 1. (a) and (b) real image (c) optical microscope image of transparent and flexible SLG sensor. (d) NO₂ sensing curve without heating. (e) recovery time constant as a function of temperature. (f) temperature distribution of sensor-heater device (g) NO₂ sensing curve at 165 °C. Reprinted with permission from [30]. Copyright © 2014 John Wiley & Sons, Inc.

significantly reduced the recovery time of the graphene-based gas sensor. Moreover, the electrical resistance was controlled, and the temperature of the device reached 250 °C under an applied voltage of 30 V, demonstrating an electrical power consumption of 1.7 W. At room temperature without the heating layer activation, full recovery after NO₂ gas sensing required approximately 3000 s as shown in Fig. 1 (d). However, the recovery time constant was reduced to 55 s and the response reached -39% at 250 °C due to the thermal energy flow through the lateral pathway from the heater to the sensor when the heating layer was activated. Moreover, the temperature was controlled by the distance between the heaters and the channel width. The temperature was increased when the distance was short; however, this resulted in an increase in the resistance of the SLG layer. Thus, in consideration of the temperature and the resistance, the optimization of the distance and the channel width is required. The result is depicted in Fig. 1 (g). It is shown that a channel width of 6 mm and a distance of 20mm between the heaters are the optimal values.

2.2 All-graphene gas sensor

Through the integration of a graphene sensor and a graphene



Fig. 2. (a) Schematic diagram and real image of self-activated graphene gas sensor. (b) Thermographic image thermal characteristics under bias voltage. (c) Response curves of patterned graphene sensor to 5 ppm of NO₂ Reprinted with permission from [31]. Copyright © 2015 American Chemical Society.

heater, a sensor platform driven only by the self-heating of graphene without external heaters was developed. However, a separate power source was required for sensor measurement and heater operation, and the power consumption was too high to embed the sensors in portable or wearable devices. The following studies developed self-activated graphene-based gas sensors using only one power source by designing the sensing area in micro- to nano-scale through various patterning methods. Kim et al. reported a graphene-based NO₂ gas sensor self-activated by Joule heating in a micropatterned graphene channel area [31]. Threelayer CVD graphene was patterned on a Cu foil with a width of 5 µm and a narrow channel of length 5 mm via photolithography and oxygen plasma etching treatment. After patterning the graphene microchannels, PMMA was spin-coated on the graphene patterns. The Cu foil was etched, and the PMMA-coated graphene patterns were scooped with a polyimide substrate. Subsequently, the PMMA was removed to complete the transfer of the patterned graphene (Fig. 2 (a)). Using an infrared camera, the temperature of the active region in the patterned sensor was measured to be 73.4 °C at 60 V, whereas the temperature of the non-patterned sensor remained below 30 °C even at 60 V as shown in Fig. 2 (b). The power consumption of the sensor was 14.2 mW when the applied voltage was increased to 60 V. The patterned graphene sensor showed a dramatic enhancement in its response to 5 ppm NO₂ with increasing bias voltage, whereas the non-patterned graphene sensor showed no enhancement until 10 V (Fig. 2 (c)). Upon increasing the bias voltage from 1 to 60 V, the response of the micropatterned graphene sensor increased from 4.47% to 12.49%, the response time decreased from 328 to 89 s, and the recovery time decreased from 1941 to 579 s. The

responses of the sensor to 50 ppm NH₃, C_2H_5OH , CH_3COCH_3 , and 50% relative humidity were 5.4%, 1.71%, 0.17%, and 0.9%, respectively. The sensor also showed a high endurance to a humid condition (less than 5% degradation at 50% relative humidity) and stable responses under high mechanical bending strains (3% degradation under a bending radius of 1 mm). These results suggest that the self-activation of graphene reduces the effect of humidity, and the flexible characteristic of the graphene-based gas sensor enables reliable sensing under a high bending strain.

2.3 Metal-decorated graphene gas sensor

Surface functionalization is an effective way to control the selectivity of a gas sensor. Pristine graphene has good response to NO2 gas; however, decorating metal nanoparticles affects its gas response. Yokoyama et al. reported a Pd-functionalized suspended-graphene-nanosheet-based H₂ gas sensor by Joule selfheating as shown in Fig. 3 (a) [32]. The sensor device was fabricated by the deposition of Ti, Pt, and Au metals via electron beam deposition. The Ti layer enhanced the adhesion between the Pt and SiO₂ layers. The Pt layer has good endurance against the needle probes for electrical measurement. The role of the Au layer was to inhibit the catalytic reaction between Pt and H₂ gas. The suspended graphene layer was grown on top of the Au layer via a thorough transfer process using a polydimethylsiloxane (PDMS) stamp. Finally, Pd nanoparticles were deposited on the suspended graphene layer via electron beam deposition. The Pd-nanoparticlefunctionalized suspended graphene film acted as a sensing layer



Fig. 3. (a) Optical and SEM image of Graphene and Au contact. (b) Raman spectrum of Graphene and Au layer. (c) Cross-sectional TEM image of Pd/Graphene on Au layer. (d) Gas response comparison of self-heating at 0.9 V and heating by hot chuck at 135 °C at 0.01 V. (e) Effect of hot chuck temperature and self-heating power on response time. (f) Relationship between self-heating power and temperature. Reprinted with permission from [32]. Copyright © 2018 American Chemical Society.

with enhanced self-heating capacity. The small contact area between the Pd/graphene and Au layer resulted in a large thermal contact resistance and the suppression of heat dissipation to the electrodes. If the gap between the Pd/graphene and Au increases, the transfer of phonons from graphene to the Au electrode is degraded; this leads to a high thermal contact resistance. By maximizing the self-heating capacity, the temperature was raised to 180 °C with a power consumption of 0.93 mW/µm per unit graphene. The gas-sensing characteristics were significantly improved; the response time was reduced to 15 s under 100 ppm of H₂ gas. When the temperature exceeded 100 °C, the gas response was enhanced using an external heater than through selfheating. In the self-heated graphene, charged-carrier scatterings with H-induced potentials were suppressed under high electric fields. In contrast, under a humid atmosphere where the relative humidity is above 50%, the sensor showed a response to 10 ppm of H₂.

H₂ detection using Au-nanoclusters (NCs)-decorated graphene sensors self-activated in the graphene microchannel was demonstrated by Kim et al [33]. A three-layered graphene film was deposited on a Cu foil using CVD. The film was micropatterned via photolithography and transferred onto a polyimide substrate using the conventional PMMA transfer method. Thereafter, PMMA was removed to finish the transfer process (Fig. 4). Au NCs were decorated on the prepared graphene micropatterns via an electron beam evaporator and the self-activation of the graphene channel. The Joule heating effect on the microchannel caused the agglomeration of the deposited Au film and formed the Au NCs, which improved the gas-sensing properties. When the sensors were exposed to 50 ppm H_2 , the responses of the Au-NCs-decorated graphene sensor were 0.98% at 1 V and 5.46% at 60 V, and the response of the pristine graphene sensor was 0.09% at 60 V. From an infrared camera test, the temperature of the sensing area in the Au-NCs-decorated sensor was 91.0 °C at 60 V, whereas the temperature of the sensing area in the pristine sensor reached 120 °C at 60 V. A comparison between the pristine graphene sensor and the Au-NCs-decorated graphene sensor under exposure to various gas molecules demonstrated that the self-activated Au NC decoration selectively increased the H₂ sensing performance. By increasing the applied voltage from 1 to 60 V, the H₂ sensing response time of the Au-decorated sensor decreased from 374 to 16 s. Discrete Fourier transform (DFT) calculations were performed to investigate the H₂ sensing of the Au-NCs-decorated graphene gas sensor for various geometries. The calculations demonstrated that hydrogen was not likely to bind to the Au or graphene surface



Fig. 4. Real image of pristine, Au-decorated sensor and Au-decorated sensor on a nickel coin. (b) Raman spectrum of sensor devices. Optical images of Au-decorated Graphene sensor (c) before (d) after self-activation. (e) Response curves to H₂, H₂S, C₂H₅OH and NH₃. (f) Response curves under 1 V and 60 V. Reprinted with permission from [33]. Copyright © 2019 Royal Society of Chemistry.

individually, however, the hydrogen atom was strongly bound to Au NCs at the interface vertex and vertex sites, which enhanced the H_2 sensing properties.

2.4 Graphene-based hybrid composite gas sensor

А graphene hybrid composite improves gas-sensing characteristics by establishing a heterojunction between the materials. Yang et al. investigated a gas-sensing platform based on a self-heated stretchable laser-induced graphene (LIG) pattern [34]. The self-heating capability of the LIG patterns enables the integration of the sensor device and the external heater. The LIG patterns were applied using a 10.6 µm CO₂ laser on a polyimide substrate attached on a glass slide. Subsequently, LIG/polyimide was transferred to a soft substrate and two serpentine lines with a width of 2 mm were formed. The linewidth of the LIG sensing region was varied from 120 to 240 µm. Ag ink was coated on the serpentine lines, which reduced the electrical resistance. Finally, various gas-sensing materials such as MoS₂, rGO/MoS₂, and ZnO/ CuO were drop-cast on the center of the serpentine lines. The LIG pattern is a suitable alternative to conventional interdigitated



Fig. 5. (a) schematic illustration of experimental procedure. (b) Response curves of rGO/MoS_2 nanoflower on LIG platform at 60°C with different width. (c) Gas response to 1 ppm of NO₂ at various self-heated temperature of small petal. Reprinted with permission from [34]. Copyright © 2019 Royal Society of Chemistry.

electrodes owing to its low-cost and environment-friendly fabrication process. The LIG pattern consists of the sensing region where a sensing material is coated and the serpentine interconnect region where a thin metal layer is coated. The electrical resistance of the serpentine interconnect region is much lower than that of the sensing region, which induces Joule heating to be localized in the sensing region and causes low power consumption as well. Under an applied voltage of 12 V, the temperature of the LIG sensing region reached 80 °C. The relative resistance of the LIG sensing region with respect to that of the serpentine region was higher at a smaller linewidth and longer length. An rGO/MoS2 nanoflower with a small petal synthesized via the solvothermal method was utilized as the sensing material, and it showed better gas-sensing characteristics owing to its large specific surface area and more uniform distribution. Furthermore, the device showed a higher response when the temperature of the edge of the LIG sensing region was lower and the linewidth was larger. However, the recovery rate increased at a higher temperature due to thermal activation. This indicates that the simultaneous enhancement of both response and recovery rates is not feasible by controlling the operating temperature. Thus, the optimal operating temperature was 60 °C, which showed a response time of 360 s and a recovery time of 720 s.

3. CONCLUSIONS

In this article, we reviewed the recent studies on self-activated graphene-based gas sensors. The changes in the self-heating performance of the graphene sensors depending on the synthesis and patterning methods of the graphene sheets and the significant enhancements in their sensing characteristics through surface modifications were described. The self-activation of the graphene sensor using Joule heating enables its operation at room temperature without an external heater, and metal decoration or hybrid composite formation allows the sensors to react selectively to various gases. The results indicated that the self-activated graphene sensor is a suitable sensing platform for the fabrication of high-performance gas-sensing devices operating in diverse environments, such as humid conditions and under high mechanical bending strains. We believe that the self-activated graphene-based gas-sensing device will significantly expand the practical use of gas sensors to next-generation technologies, such as flexible, transparent, and wearable electronics.

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