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Characteristic of graphene-based thick film gas sensor for ethanol and acetone vapor detection at room temperature

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ABSTRACT

Ethanol and acetone are volatile organic compound gases widely used in food processing. Health problems such as irritation of the eyes, nose, and throat can affect human health if exposed to these gases. Two graphene gas sensors were fabricated using a screen-printing technique onto a glass substrate to compare their performance to the acetone and ethanol vapors at room temperature. The graphene paste was prepared by mixing 95 wt.% of the binder with 5 wt.% of graphene nanoflakes. A silver paste was used as the interdigitated electrode of the gas sensor and became the first layer of the gas sensor. The silver paste was deposited on the glass substrate using a screen-printing technique and fired at 150 °C for 15 minutes. Next, the graphene paste was deposited onto the interdigitated electrode using a screen-printing technique and became the second layer of the gas sensor. The graphene was annealed at 200 °C for 30 minutes. Both graphene gas sensors responded well to ethanol and acetone vapor with an n-typed gas sensor at room temperature. As a comparison, the graphene gas sensor showed better characteristics in terms of response and recovery characteristics to ethanol vapor than acetone vapor at room temperature. The response and response time of the graphene-based thick film gas sensor to ethanol were approximately 21.89 and 24.08, respectively.

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1. INTRODUCTION

According to the 40 CFR 51,100 in CFR-code of federal regulations, volatile organic compounds (VOCs) are any carbon compound that takes part in atmospheric photochemical reactions, exception of carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate. Examples of VOC gases are acetone, benzene, cyclohexene, ethanol, formaldehyde, methanol, n-butanol, toluene, and 2-propanol. In traditional methods, VOC gasses have been identified using gas chromatography (GC) and liquid chromatography (LC). However, this technique requires time-consuming and different detectors [1]. In recent years, the chemical-based gas sensor has attracted incredible interest in sensing VOC gases with promising characteristics such as sensitivity and detection limit. Reported that various chemical-based gas

sensors such as TiO_2 [2], SnO_2 [3], [4], Cr_2O_3-In_2O_3 [5], In_2S_3 [6], and graphene [7] have been used to sense VOC gases in the literature.

Ethanol and acetone are also categorized under VOC gases, and these gases are the most used gases in many applications; thus, many studies have been conducted in gas sensing to detect these gases. Ethanol is used in biomedical, chemical, and food industry applications [8], while acetone is used as an essential solvent and raw material in various industrial fields, such as explosives, plastics, rubber, fiber, leather, and spray paint [9]. Ethanol and acetone gases are flammable and toxic, and it can affect to the human health and environment [9]. Therefore, a device such as a gas sensor that is able to quickly sense these gases is needed to monitor high concentrations and avoid gas leakage.

Besides that, ethanol and acetone also have been used as diseases biomarker based on exhaled from human breath. Exhaled human breath consists of more than 250 VOC gases, which can be used as a biomarker for diseases [5]. As an example, acetone vapor released from human breath can be used for diabetes, while ethanol vapor from human breath as identification for alcohol usage. Vasilescu *et al.* [10] ethanol and acetone were categorized as alcohol dehydrogenase and secondary alcohol dehydrogenase in biorecognition elements, respectively.

Recently, an intensive study has been done on carbon-based materials as sensing material of gas sensors, such as carbon nanotubes and graphene. Due to the enormous advantages of graphene in the thermal, mechanical, optical, and electrical properties make it suitable for nanoelectronics, nanophotonics, energy storage devices, solar cells, and biosensors [11]. Commonly, graphene was used as hybrid material in gas sensors to improve the performance of gas sensors [12], [13]. Herein, the graphene without hybridization was used as a sensing material for ethanol and acetone vapor detection to offer a low-cost and simple fabrication gas sensor.

Graphene as a sensing material for the gas sensor has been reported in many studies to sense various gases. The graphene derivatives include graphene quantum dots (GQDs), single-layer graphene, few-layer graphene, graphene oxide (GO), and reduced graphene oxide (rGO) [14]. Among them, rGO is the most graphene used in gas sensors because it is easy to fabricate, cheap to manufacture in large-scale production, and has unique functions and gas sensing properties [15]. However, rGO is less conductive compared to few-layer graphene. Graphene nanoflakes are categorized as few-layer graphene and reported that they have been used as gas sensors in literature [12], [16]. Therefore, graphene nanoflakes were used as a sensing material in this work to produce conductive gas sensors and also able to operate at room temperature.

This paper presents the fabrication of graphene-based thick film gas sensors using the screen-printing technique on the glass substrate. The gas sensor was exposed to ethanol and acetone at room temperature to compare its characteristics in terms of response, response time, and recovery characteristics. The results revealed that the graphene-based thick film gas sensor showed better characteristics in terms of response and response time to ethanol vapor than acetone vapor at room temperature.

2. METHOD

2.1. Preparation of binder and graphene paste

Graphene nanoflakes, ethyl cellulose, and α-terpineol were purchased from BTCORP, SIGMA-ALDRICH and, ACROS ORGANICS, respectively. All materials have been used without any purification. To make a thick film paste, two components were needed, which are a binder and a sensing powder. These components will be mixed using a magnetic stirrer until a homogeneous paste was obtained. In this work, the binder was prepared using ethyl cellulose and α-terpineol, while the sensing material is graphene nanoflakes powder. The binder which consists of ethyl cellulose (2 wt.%) and, α-terpineol (98 wt.%) was mixed using a magnetic stirrer for 24 hours. Next, a graphene paste was prepared by mixing the binder (95 wt.%) with graphene nanoflakes powder (5 wt.%) to make it as thick film paste. The graphene paste was stirred using a magnetic stir for about 24 hours to produce a homogeneous and viscous paste. A similar technique also was found in the literature to prepare the thick film paste [17].

2.2. Fabrication of the gas sensor using screen-printing

The thick film gas sensor consists of two layers: interdigitated electrode and a sensing layer. In this work, the interdigitated electrode will become first layer of gas sensor and sensing layer will become second layer of the gas sensor. The interdigitated electrode (silver paste) was deposited on the glass substrate using the screen-printing technique with a mesh thickness of stencil is 10 µm and fired at 150 °C for 30 minutes. Next, graphene paste was deposited onto the interdigitated electrode as a sensing layer (1×1 cm) using the screen-printing technique with a mesh thickness of stencil is 10 µm and annealed at 200 °C for 30 minutes. Lastly, a fine copper wire was used to make an electrical connection by attaching the wire to the leg of the IDE using silver paste. The fabricated graphene gas sensors is displayed in Figure 1. The fabricated graphene gas

sensors were labelled as SG(G)-1 as shown in Figure 1(a) and will be used for ethanol exposure, while SG(G)-2 is displayed in Figure 1(b) and will be used for acetone expsoure.

(a) (b)

Figure 1. Fabricated graphene gas sensor on glass substrate (a) SG(G)-1 and (b) SG(G)-2

2.3. Experimental setup for ethanol and acetone sensing

The gas sensor measurement of the graphene gas sensors was conducted in a gas chamber. The gas chamber was connected to the source meter (Keithley 6482), and the monitoring system was recorded using a LabVIEW program. 1V supply voltage was used to supply the voltage to the gas sensor. The graphene gas sensor was placed vertically and directly to the inlet gas hose to ensure the gas flow reached the sensing layer of the gas sensor, as shown in Figure 2. The ethanol solution was prepared by mixing 50 ml of acetone with 50 ml of deionized water and stirred for 5 minutes. The ethanol vapor was produced by heating the acetone solution on the hotplate at 100 ºC for 30 minutes. A similar procedure was also applied to the acetone solution. Initially, the current of the gas sensor was observed for 5 minutes to obtain a saturated current. Next, the acetone vapor will be flowed into the gas chamber for 5 minutes to monitor the response, and the response time will be recorded. After 5 minutes, the inlet hose gas will be disconnected from the gas chamber, and the current of the gas sensor will be observed again in 5 minutes to observe the recovery characteristic of the gas sensor. The experimental setup for ethanol and acetone vapor detection is shown in Figure 3.

Figure 2. Location of graphene gas sensor inside the gas chamber during ethanol/acetone exposure

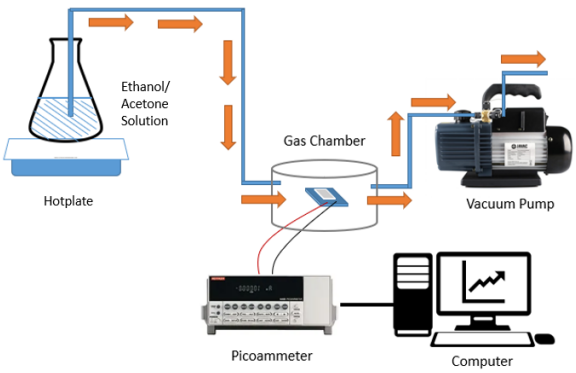


Figure 3. Experimental setup of graphene gas sensor to ethanol/acetone vapor

3. RESULTS AND DISCUSSION

3.1. Characterization of sensing layer using SEM and XRD

Scanning electron microscopy (SEM) and X-ray diffractometer (XRD) characterizations were carried out to the sensing layer after annealed treatment to verify that the sensing material on the surface of the gas sensor was graphene nanoflakes after the stirring process during making the thick film paste. Figure 4 shows the graphene nanoflake powder without purification. The raw structure of graphene nanoflakes is seen as a sheet, and this observation is also similar as reported in [18], [19]. It can be seen that the purchased graphene flakes were agglomerated. After the stirring process and annealing treatment, it can be seen that flakes were scattered on the sensing layer in Figure 5, as displayed Figure 5(a). The flakes become more significant after being combined with liquid, where this type of structure of graphene nanoflakes also can be seen in another study [20]. It can be observed that the size of graphene flakes varied. The thickness of the sensing layer was approximately in the range of 17-25 µm as presented in Figure 5(b), which this thickness was larger than the mesh thickness in the stencil screen-printing.

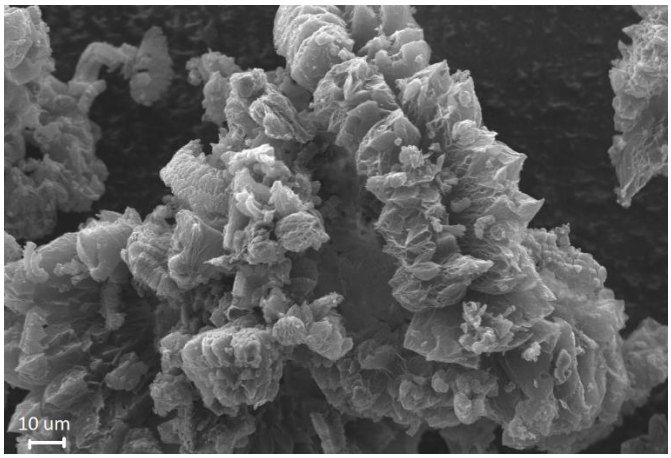
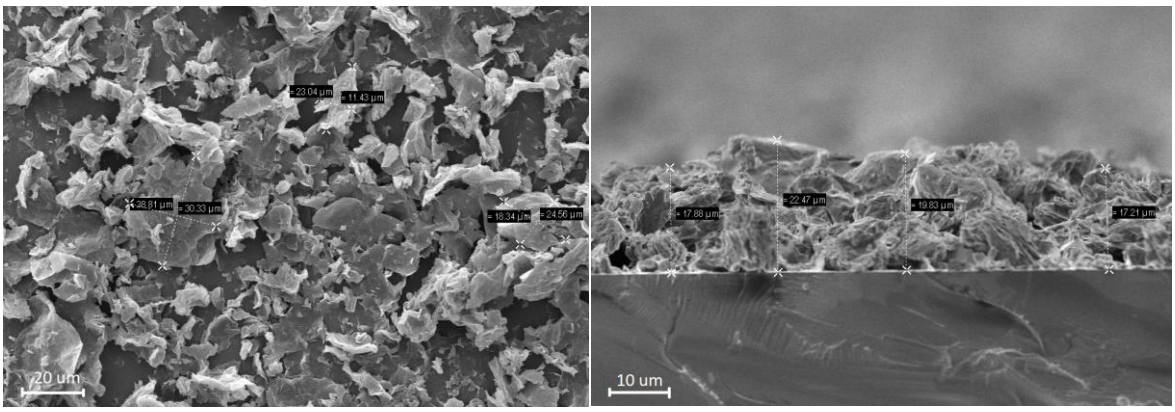


Figure 4. Graphene nanoflakes without purification



(a) (b)

Figure 5. Characterization of the sensing layer of the graphene gas sensor after annealing treatment using SEM (a) morpohology and (b) thickness

Figure 6 shows the XRD spectra of the sensing layer of the graphene gas sensor. One peak was detected in the XRD spectra at 2θ=23.78º, which this peak was contributed by C(002). A similar peak on graphene nanoflakes also has been reported in [21]. It also can be seen that the peak was broader, which indicates that few layers of graphenes were present in the sample [22]. Besides, broader graphene peak also can occur because of the low content and low diffraction intensity of graphene [23].

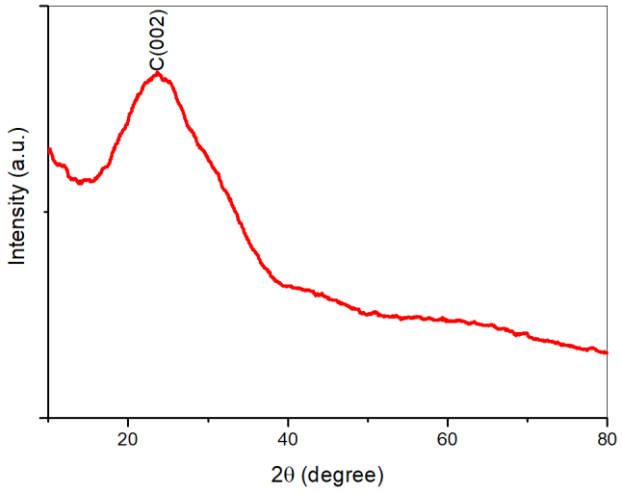


Figure 6. XRD spectra of the sensing layer of the graphene gas sensor

3.2. Current-voltage (I-V) characteristic of graphene gas sensors

I-V characteristics were carried out for the graphene gas sensors to check the conductivity of the gas sensor by applying a I-V supply voltage to both of the gas sensors. The I-V characteristic of the gas sensor produced must be linear to verify that the gas sensor followed Ohm's law and that it also was a resistance-based gas sensor. Only a conductive gas sensor can be exposed to the target gas in a resistance-based gas sensor. Figure 7 shows the I-V characteristic of the graphene gas sensor, and the results showed that all the graphene gas sensors were conductive based on a linearity graph, thus both gas sensors can be exposed to the ethanol and acetone vapor. The resistance values for both gas sensors also were almost similar with different percentage of 19%, which suggest that amount of graphene paste deposited using screen-printing technique on the both gas sensors were almost similar.

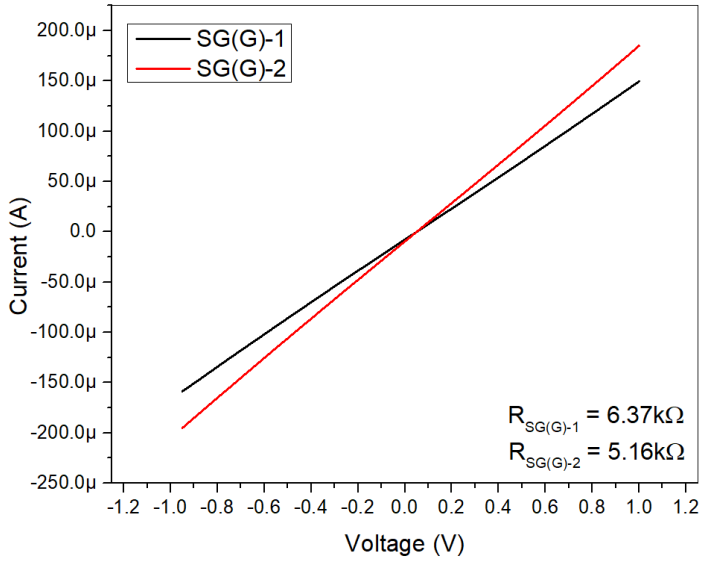
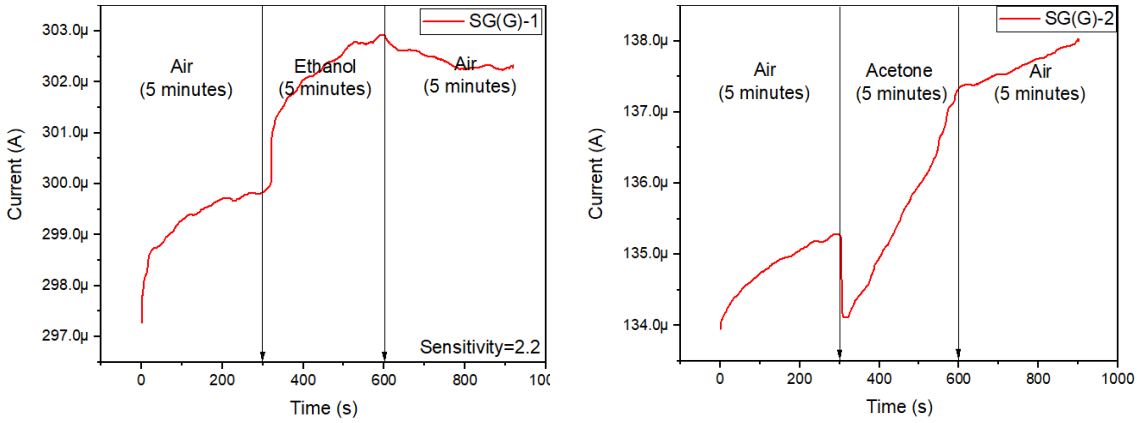


Figure 7. I-V characteristic of graphene gas sensors at -1V to 1V

3.3. Response graphene gas sensor to ethanol and acetone vapor

Figure 8 shows the response of fabricated graphene gas sensors, the SG(G)-1 graphene gas sensor to ethanol is shown in Figure 8(a), whereas the response of the SG(G)-1 graphene gas sensor to acetone vapor is shown in Figure 8(b) at room temperature. Both gas sensors showed increased current when exposed to the

target gas, which suggests that the graphene gas sensor was an n-typed gas sensor. This type of response is also similar, as reported in [24], [25]. It can be observed that the SG(G)-1 graphene gas sensor that exposed to the ethanol almost reached the saturated value in 5 minutes, while SG(G)-2 graphene gas sensor was decreased and after that increased sharply after exposed to the acetone vapor. Characteristics for both graphene gas sensors is displayed in Table 1. Both responses of the graphene gas sensors were almost similar and the responses also were unable to recover to its initial value during exposure to the air environment.



(a) (b)

Figure 8. The current measurement of graphene gas sensors to ethanol and acetone vapor (a) SG(G)-1 and (b) SG(G)-2

Table 1. Characteristics of graphene gas sensors on ethanol and acetone gas

Sample name	Response	Response time (s)	Recovery characteristic
SG(G)-1	21.89	21.89	Not recover
SG(G)-2	24.08	24.08	Not recover

4. CONCLUSION

The fabrication of thick film graphene gas sensors using the screen-printing techniques has been successfully developed onto a glass substrate and exposed to ethanol and acetone vapor. Both gas sensors responded well to ethanol and acetone vapor n-typed response at room temperature. The graphene gas sensor (SG(G)-1) showed a better response to ethanol than acetone vapor, with response and response time were approximately 21.89 and 24.08, respectively.

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