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Fabrication of Gas Sensor Based on Graphene for the Adsorption of Gases Produced from Waste Material in Kitchen and its Surrounding

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ABSTRACT

Graphene attracted to the researcher with huge attention due to their unique physicochemical properties including high specific surface area and high-speed electron mobility at room temperature. The graphene with several layers was synthesized through liquid phase exfoliation method using high shear force of the magnetic stirrer. This process was performed about 6 hour on the graphite powder and dimethyl-formamide solvent of 0.25 M, 0.5 M and 1 M concentration solution following the microwave treatment for 30 second and sonication for 2 hour at room temperature. The drop-casted exfoliated graphene into glass substrate had G peak and 2D peak. The graphene from 1M concentration had the better quality as compared to the graphene obtained from 0.25M and 0.5M concentration solution. The fabricated gas sensor device with two contact electrodes using exfoliated graphene as a channel material produced the different current (I)–voltage (V) characteristics. The current vs. voltage of bare graphene film without filling of waste harmful gases had the current shifted from 0 mA to 0.0652 mA when the maximum voltage was applied. The current increased nearly from 0.0652 mA to 0.2391 mA after harmful waste gases adjustment at maximum applied voltage. The current through few layers graphene channel after harmful gases filled was found 3.6 times higher than that of the current through the graphene channel without harmful waste gases. This result was due to the adsorption/absorption and interaction of more quantity of harmful waste gases by the exfoliated graphene. So, the device showing some current variation informed that the graphene gas sensor was sensitive to waste gases produced from home kitchen and its surrounding.

Keywords: Graphene, Liquid exfoliation, Dimethyl-formamide, Raman spectroscopy, Gas sensor.

INTRODUCTION

Graphene is ultra-thin two dimensional (2D) materials made entirely of sp^2 carbons arranged in honeycomb structure. It is basic constructional element for all other dimensions of graphitic materials and it can be wrapped up into zero-dimensional (0D) fullerene, rolled into one-dimensional (1D) carbon nanotube, or stacked into three-dimensional (3D) graphite [1, 2]. Graphene has unique physicochemical properties including high specific surface area (SSA) of 2600 m^2g^{-1} [3] and high-speed electron mobility of 200000 $cm^2V^{-1}s^{-1}$ at room temperature [1]. Geometrical modification [4] and chemical modification of this material allow different electronic and optical properties. The band gap is opened in hydrogenated [5] and fluorinated [6] graphene. In addition, graphene provides essentially infinite possibilities for the modification or functionalization of its carbon backbone through a variety of chemical interactions so as to form numerous graphene-based derivatives with desirable properties [7, 8, 9]. Graphene and its derivatives such as graphene oxide, reduced graphene oxide, functionalized graphene are promising candidates for novel functional materials. It has high potential application in sensor, biosensor, electrodes, gas separation, energy storage and conversion, super-capacitors, electronics and methanol adsorption [10, 11, 9]. The aggregations of graphene decrease its available surface area and further reduce its adsorption capacity. Functionalization of graphene with molecules has water solubility nature and affinity toward target analyses [12], so it will improve the selectivity of adsorbents or detection of devices as well as prevent the aggregation. Graphene and derivatives of graphene can be applied to environmental protection by adsorption of harmful gases such as hydrogen sulphide (H_2S), methane (CH_4), organic sulfides along with the detection of pollutants in waste material. Many gas sensors based on graphene detect toxic gases in waste air and waste water by attributing to the change in the conductance or resistance of the graphene. Such a change in conductance or resistance takes place by the charge transfer between adsorbed gas molecules and graphene sheet [13].

The environmental pollution due to the improper waste activities in home, industry, waste materials, polluted air and waste water that the people are doing day by day is a critical problem and it ultimately drawbacks development of nation. Along with the above reasons, pollution also occurs due to organic pollutants, toxic gas and heavy metals ions and they cause severe threaten to the human health and ecological balance. So the essential work is to detect and remove such toxic smelling and polluted air from surrounding waste materials (garbage) and wastewater. To overcome such problem, nano-material such as graphene and graphene derivatives made sensor has potential to be applied in environmental monitoring of toxic gases such as H_2S, CH_4 and others in air and waste water. The efficient adsorbents [14] based on high surface area and strong adsorption, electrochemical sensors [13] based on the excellent electrical conductivity have been constructed for the detection and removal of environmental pollutants. The graphene and graphene based materials have high adsorption capacity to kinds of organic pollutants in waste materials, air and waste water. In addition to this, graphene/graphene derivative based sensor with high limit of detection have been used to find the toxic gases and organic pollutants produced from waste water and air in the environment [12]. In contrast to these, detection of insight adsorption of harmful waste gases in the waste materials produced from our kitchen and its surrounding has not been carried out till now using the graphene sensor with simple electrodes. So, our objective is to develop sensitive, simple, and inexpensive sensor to detect adsorption of harmful waste gases in the pollutants.

MATERIALS AND METHODOLOGY

Compared with other techniques, liquid phase exfoliation process is important method in which there is high-shear mixing followed by microwave treatment and sonication. It is easy and possible technique to synthesize graphene for our experimental research work. For preparation of three different graphene samples, 0.06 gram of graphite powder with 20 ml Dimethyl-formamide (DMF) solvent, 0.12 gram of graphite powder with 20 ml of DMF solvent and 0.06 gram of graphite powder with 5 ml DMF solvent were mixed to make 0.25 Molar (M), 0.5 M and 1M concentration solution respectively. The graphite powder used in this graphene synthesis process is shown in figure 1(a) and it is 99% pure from Central Drug House (P) Ltd with product code: 025362. The DMF solvent used here belongs to Fisher Scientific Company and it has the purity of 99.5% with concentration of 73.09%. The resulting solution was vigorously stirred using high-shear force from the magnetic stirrer for the duration of almost around 6 hour. LABINCO L34 hotplate magnetic stirrer calibrating the device rpm (revolution per minute) unit of 2 (equivalent of 100 rpm) as shown in figure 1(b) was used. This resulted solution after high-shear mixing was treated with microwave for 30 second (microwave oven of CG Company). At last, the resulting solution was then sonicated using ultrasonic bath for almost 2 hour setting the temperature at 26 ºC. The digital ultrasonic cleaner used in this work is shown in figure 1(c) and is from JEKEN Company available in the lab of Patan Multiple Campus. The final resulted solution was drop-cast on the glass substrate then allowed to make dry for 1 hour. Now this sample was taken for Raman spectroscopic characterization. The Raman Spectrometer used for the graphene characterization belongs to Enspectr Company. The green laser of wavelength 532 nm produced from Nd-YAG (Neodymium-doped Yttrium Aluminum Garnet) material was used for the purpose of Raman spectroscopy characterization.

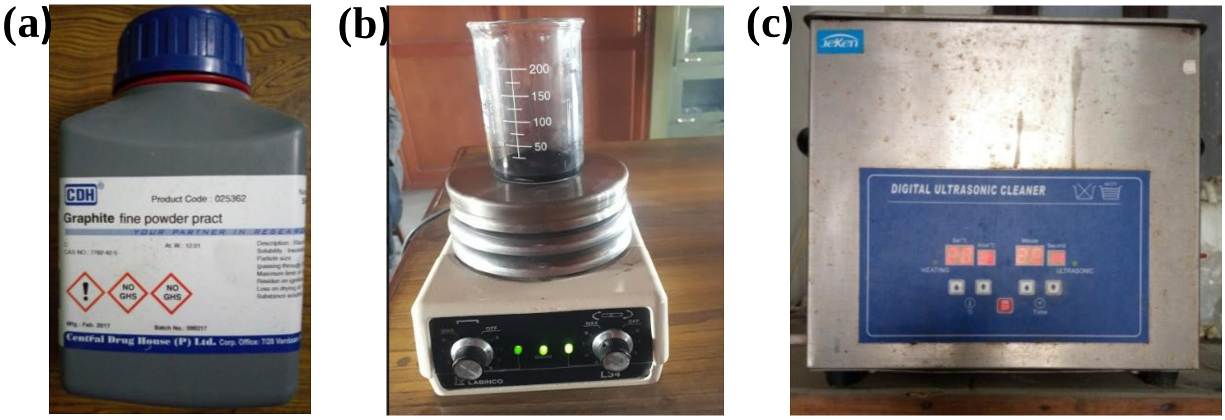

Fig. 1: (a) Graphite powder (b) magnetic stirrer for high-shear force (c) ultrasonic cleaner

Now, the Polyethylene terephthalate (PET) was lightly used to cover the graphene transferred on glass substrate and very small holes at some millimeter (mm) distance over the PET were made using small needles. The silver paste was put through the small holes to design the contact electrodes pad and then left for 1 hour to make it dry. Thus electrical contacts (source and drain) were made using silver paste keeping about 1 mm–2 mm spacing on the graphene film and finally the PET was taken out from graphene/glass substrate and used as gas sensor. The current voltage characteristics of graphene based gas sensor were measured inside a small glass box of dimension of 20 cm x 20 cm x 20 cm with and without harmful waste gases. This electrical measurement was performed with Keithley source meter model no. 2614B. Now, the glass box with gas sensor was filled with waste gases produced from waste materials packed from kitchen and its surroundings using the small pipe. The changed in the current (I)-voltage (V) due to the harmful waste gases around the graphene based gas sensor was observed and differences on such I-V values were used to analyze whether that the device detected the waste gases or not.

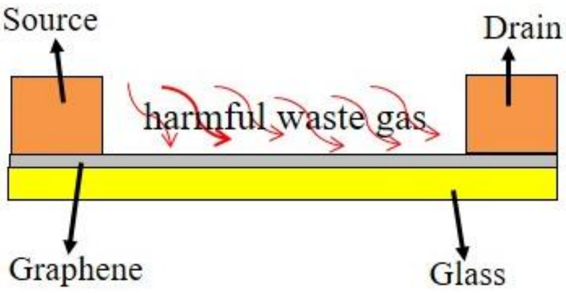

Fig. 2: Graphene based gas sensor.

RESULTS AND DISCUSSIONS

Raman spectroscopy is one of the most important and useful tool. It provides information about the number of layers in graphene stacks, atomic structure of graphene edges, disorder and defects, stacking order between different layers, effect of strain, and charge transfer. Graphene has three main features in its Raman spectrum, called the D, G, and 2D (also called G') modes. They appear at about 1350 cm^{-1}, 1580 cm^{-1} and 2700 cm^{-1} [7]. The D peak in the Raman spectroscopy represents presence of disorder or defects in graphene layer. If the D band is significant it means that there are a lot of defects in the material. The G band position is highly sensitive to the number of layers and thickness present in the sample. It is based upon the observed position of this band for a particular sample. The 2D band is the second order of the D band, sometimes referred to as an overtone of the D band. This band is also used to determine graphene layer thickness. In contrast to the G band position, the 2D band depends not only on band position but also on band shape [15].

The Raman spectrum shown in figure 3 belongs to the graphene in several layers from the 0.25 Molar (M) concentration solution of graphite with DMF solvent. The spectrum of graphene has D peak, G peak and 2D peak at 1345 cm^{-1}, at 1588 cm^{-1} and at 2695 cm^{-1} respectively where the G peak corresponds to the in-plane stretching of the C−C bonds of the graphitic sp^2 crystalline carbon atoms and the 2D peak is an indicator of graphene that originates from the second order double-resonance process related to a phonon near the K point in graphene [7]. The intensities of D peak, G peak and 2D peaks are 8892, 61257, 17282 arbitrary units (arb. units). The D peak shows the presence of defect produced in our synthesized graphene. The presence of G peak and 2D peak confirms that the presence of graphene in several layers i.e., more than 10 layers.

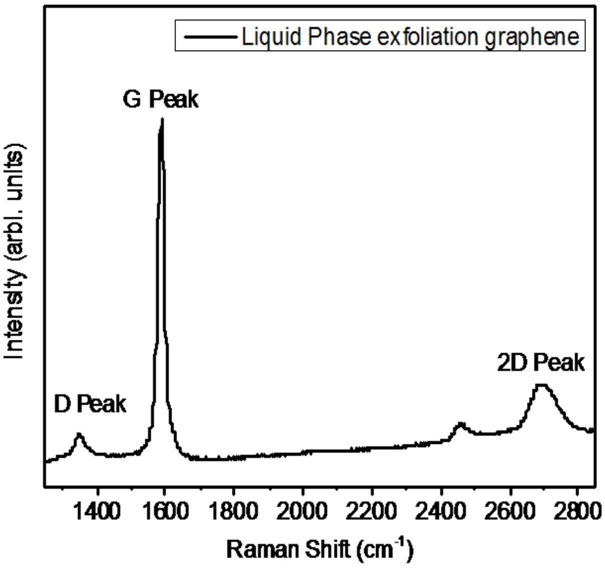

Fig. 3: Raman spectrum of liquid phase exfoliated graphene from 0.25 M concentration solution

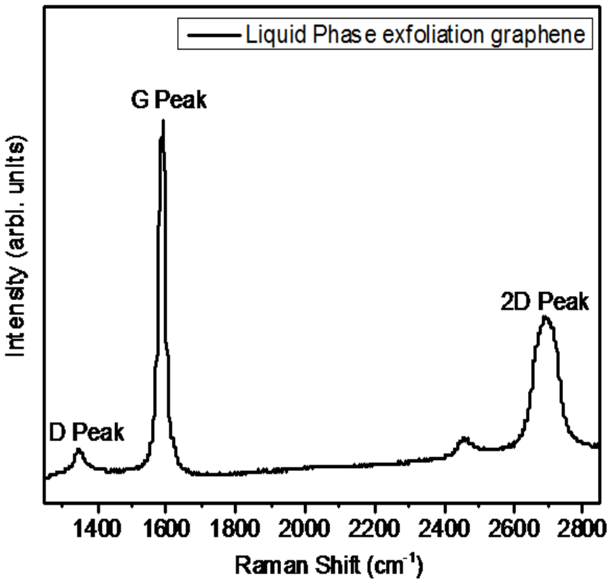

Fig. 4: Raman spectrum of liquid phase exfoliated graphene from 0.5 M concentration solution

The figure 4 shows the Raman spectrum of graphene (several layers) in better form from 0.5 M solution than the 0.25 M concentration solution. In this spectrum, the D peak is at 1345 cm^{-1}, the G peak is at 1587 cm^{-1} and 2D peak is at 2693 cm^{-1}. The intensities of D peak, G peak and 2D peaks are 8992, 60256, 30581 arbitrary units (arb. units) respectively. The presence of graphene in several layers can be predicted by the presence of G peak and 2D peak. The D peak intensity and 2D peak intensity of the graphene obtained from 0.5 M (figure 4) are somehow more than the D peak intensity and the 2D peak intensity of graphene obtained from the 0.25 M solution (figure 3). So, $2D_{0.5M}$ greater than $2D_{0.25M}$ informs that the number of graphene layers at 0.5 M are less than the number of layers of graphene from 0.25 M solution.

The Raman spectroscopy of graphene from 1 M concentration solution is shown in figure 5 and it has D peak at 1341 cm^{-1}, G peak at 1575 cm^{-1} and 2D peak at 2678 cm^{-1} with their respective intensities 13430, 58153 and 37281 arb. units. It can be clearly seen from the figure that D peak has high intensity than those of 0.25 M and 0.5 M concentration solution. This increasing defects is due to the decreasing number of graphene layers. The Raman spectrum of the sample in figure 5 shows a significant change in the shape of the 2D band signal. So, this informs that the obtainment of a better exfoliation of the graphite sheets as higher the initial concentration is. From the nature of G peak and the 2D peak intensity, the better graphene with less number of layers can be found in this 1 M concentration solution than the previous 0.25 M and 0.5 M concentration solution. With such result, this optimized thin film graphene (few layers) with source drain electrodes was used to make graphene gas sensor device for the detection of harmful waste gases adsorption/absorption/interaction.

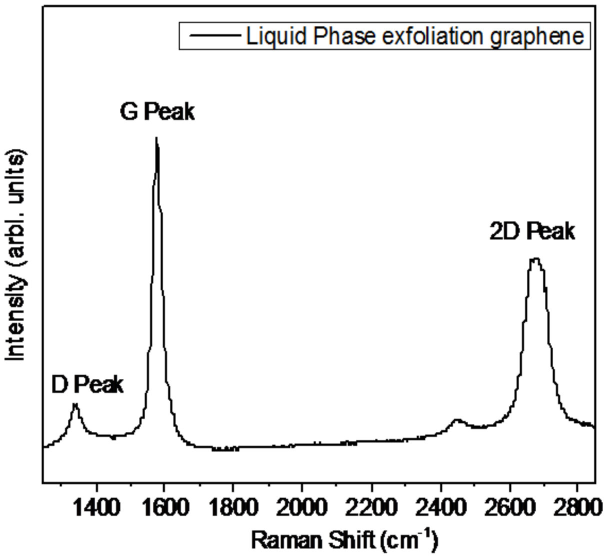

Fig. 5: Raman spectrum of liquid phase exfoliated graphene from 1 M concentration solution

Table 1: Different molar concentration of graphite powder in DMF and Raman spectrum of D peak, G peak, 2D peak position and intensity for graphene

S. N.	Molar Concentration Solution	D Peak		G Peak		2D Peak	
		Position (cm^{-1})	Intensity (arb. unit)	Position (cm^{-1})	Intensity (arb. unit)	Position (cm^{-1})	Intensity (arb. unit)
1.	0.25 M	1345	8892	1588	61257	2695	17282
2.	0.5 M	1345	8992	1587	60256	2693	30581
3.	1 M	1341	13430	1575	58153	2678	37281

Table 1 shows the comparative study of Raman spectrum of D peak, G peak, 2D peak position and intensity for the several layer graphene prepared from different molar concentration solution of graphite powder with DMF solvent. From the observed value of G peak and 2D peak position, we clearly see that the red shift occurs as the molar concentration increases from 0.25 M to 1 M. This red shift indicates that frequency of phonons interacting with the incident photon decreases. Such red-shift is observed due to the material's improved crystallinity.

Current (I) – Voltage (V) measurement:

In this case, electrical measurement was performed in two electrodes (source-drain) of graphene gas sensor (device) without harmful waste gases and with harmful waste gases filled from waste material produced from our kitchen and its surrounding. Due to the presence of harmful gases around on graphene/glass substrate (graphene gas sensor), the changed values of the current (I)-voltage (V) was observed. A linear relationship of the applied voltage and current in figure 6 shows an Ohmic contact between the electrodes and graphene film. Here, the red line in figure 6 shows the current vs. voltage of bare graphene film (i.e., without filling of waste harmful gases) in which current shifts from 0 mA to 0.0652 mA when the maximum voltage was applied. The black line in figure 6 shows that the current increases nearly from 0.0652 mA to 0.2391 mA after harmful gases adjustment at maximum applied voltage. Remarkably, the current through graphene film after harmful gases filled is 3.6 times higher than that of the current through the graphene film without gases filled in the glass box. So, the device showing some current changed indicates that the graphene material or structure is sensitive to waste gases produced from our kitchen and its surrounding. This result is due to the adsorption/absorption/interaction of more quantity of harmful waste gases by/with the exfoliated few layers graphene film. In this case, charge transferred is involved within the sensing mechanism of graphene based gas sensor [16].

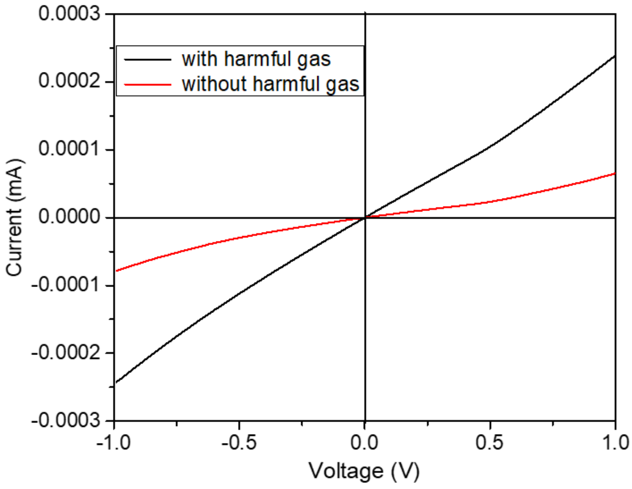

Fig. 6: Current-Voltage curve of graphene based gas sensor device without harmful waste gases (red line) and with harmful waste gases (black line)

Thus, the adsorption/absorption/interaction cause charges transfer between waste gases molecules and graphene such that there is rise in current. This phenomenon is also known as chemical doping by gas molecules.

CONCLUSION

0.25 M, 0.5 M and 1 M concentration solution were prepared from graphite powder and DMF solvent followed by the high shear force through the magnetic stirrer for about 6 hour, microwave treatment for 30 second and sonication for 2 hour at room temperature. Several layers graphene samples through liquid phase exfoliation method were characterized by Raman spectroscopy. We found different values of D peak, G peak and 2D peak of graphene and their corresponding values inform that the better graphene with less number of layers is obtained in 1 M concentration solution as compared to the graphene obtained from 0.25 M and 0.5 M. The fabricated two terminal gas sensor with exfoliated graphene as a channel material produces the different current (I)–voltage (V) characteristics. The current vs. voltage of bare graphene film without filling of waste harmful gases has the current shifted from 0 mA to 0.0652 mA when the maximum voltage was applied. However, the current increases nearly from 0.0652 mA to 0.2391 mA after harmful waste gases adjustment at maximum applied voltage. The current through graphene film after harmful gases filled is 3.6 times higher than that of current through the graphene film without gases filled in the glass box. This is due to the adsorption/absorption/interaction of more quantity of harmful waste gases by/with the exfoliated graphene having few layers. So, the device showing some current changed indicates that the graphene material or structure is sensitive to waste gases produced from kitchen and its surrounding.

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