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Arsat, R., Breedon, M., Shafiei, M., Kalantar Zadeh, K., Wlodarski, W., Gilje, S., Kaner, R., & Arregui, F. (2008). Graphene-like nano-sheets/36deg LiTaO3 surface acoustic wave hydrogen gas sensor. Proceedings of the 7th IEEE Conference on Sensors, 188–191.
<https://doi.org/10.1109/ICSENS.2008.4716414>

Published Version: <https://doi.org/10.1109/ICSENS.2008.4716414>

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Graphene-like Nano-Sheets/36° LiTaO₃ Surface Acoustic Wave Hydrogen Gas Sensor

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Abstract— Presented is the material and gas sensing properties of graphene-like nano-sheets deposited on 36° YX lithium tantalate (LiTaO₃) surface acoustic wave (SAW) transducers. The graphene-like nano-sheets were characterized via scanning electron microscopy (SEM), atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS). The graphene-like nano-sheet/SAW sensors were exposed to different concentrations of hydrogen (H₂) gas in a synthetic air at room temperature. The developed sensors exhibit good sensitivity towards low concentrations of H₂ in ambient conditions, as well as excellent dynamic performance towards H₂ at room temperature.

I. INTRODUCTION

Interest in H₂ gas sensing has enjoyed a skyrocketing popularity, as hydrogen has been touted as a panacea for clean energy production. It is highly flammable and explodes in air at concentrations as low as 4% [1]. Furthermore, H₂ has a low ignition energy, posing a significant risk of deflagration. H₂ sensing also finds important applications in aerospace, chemical refining and biomedical systems [2, 3].

A great deal of attention has been drawn to graphene due to its unique electrical and mechanical properties. As a monoatomically thick two-dimensional layer of sp²-bonded carbon, graphene can be considered as the basic building block of all graphitic materials, arranging itself into large planar sheets with repetitious benzene ring motif [4, 5].

To date, several synthesis methods have been developed by researchers in order to obtain a perfect single layer of graphene. Novoselov et al. [4-6] have micromechanically cleaved highly oriented pyrolytic graphite (HOPG) to obtain a single layer of graphene. The technique was repeated by Joshi et al. [7], who exfoliated the graphene onto doped silicon substrates. Furthermore, they [7] explored the potential of a single graphene layer for chemical sensing applications. Alternatively, graphene can be produced by the conversion of nanodiamond [8] and reduction of SiC [9], both of which are of high cost.

Rapid progress for the electronic applications of graphene is anticipated. As graphene is an inexpensive lightweight material, it has been employed in room temperature operating transistors [10] and tunable storage media [11]. In addition, as it is a two-dimensional material, graphene has a large surface area with respect to volume. This large surface area should increase the interaction area available for hydrogen molecules, thus enhancing sensitivity. Schedin et al. [6] and Joshi et al. [7] have pioneered the investigation of graphene based chemical sensors. Joshi et al. [7] have fabricated and tested graphene based field effect transistors towards different gases. These have been supported by the experiment performed by Schedin et al. [6] to assess the effect of gaseous chemicals on graphene devices. Their Hall measurements have revealed that NO₂ has acted as acceptor and CO as donor. Thus illustrating that upon exposure, the graphene surface has become p-type. Electrical baseline measurements and Raman spectroscopy [6, 7] confirm that analyte gases readily desorb from graphene based sensors.

In this paper, we have deposited graphene-like nano-sheets on the active area of 36° YX LiTaO₃ SAW transducers and measured the frequency shifts towards different concentrations of H₂ in dry synthetic air. To the best of authors' knowledge, sensing performance of SAW gas sensors based on graphene-like layer towards H₂ is reported for the first time. The dynamic response of various concentrations of H₂ gas at room operating temperature (25°C), are presented in this paper.

II. EXPERIMENTAL

A. Fabrication

The SAW transducers were fabricated on a 36°YX LiTaO₃ substrate using photolithography. It consists of 38 electrode pairs in input and output interdigitated transducers (IDTs) with a periodicity of 40 μm. These devices also have 160 reflectors in each side, and a 700 μm aperture width. The center-to-center distance between the IDTs is 1920 μm.

Authors thank Ministry of Higher Education, Malaysia and University of Technology Malaysia (UTM) for the financial support.

Metallization double layer of Ti (20 nm) and Au (80 nm) were patterned to form the device's IDTs and reflectors.

The graphene-like nano-sheets were chemically synthesized via the reduction of spray-coated graphite oxide (GO) with hydrazine [12]. GO was synthesized by Hummers method after which the GO was then dispersed in water and the spray-coated onto a preheated device. The as deposited GO device was placed in a flow cell and heated at 80°C under flowing helium gas. Hydrazine vapor was supplied via a bubbler to the system. The produced graphene layer was then characterized via SEM, AFM and XPS.

B. Gas Sensing Measurement

The sensor was placed in a computerized multi-channel gas calibration system. The system was used for generating 5 pulses of H₂ gas (0.06%, 0.125%, 0.25%, 0.50%, and 1%) in a synthetic air. This sequence was utilized to expose the sensor at room operating temperature. The response to the H₂ gas was monitored using a frequency counter (Fluke PM6680B) as change in frequency of the SAW sensor. The total flow rate of the gas was kept constant at 200 sccm, and synthetic air was used as the reference (baseline) gas and the carrier gas.

III. RESULTS & DISCUSSIONS

SEM was conducted on graphene-like nano-sheets deposited onto SAW transducers. It can be seen in Fig. 1, that the graphene-like sheets (visible as pale grey regions) cover the area of the SAW device. Complementing this is in Fig. 2, is an AFM image which depicts numbers of graphene layers. It is estimated that the graphene-like sheets are monoatomically thick.

Figure 2: SEM image of graphene-like nano-sheets deposited on SAW transducers

Additionally, XPS (Fig. 3) has been provided. The C1s XPS spectrum has signified the existence of C-C at 284.5 eV. Other carbon bonding schemes arising from different functional groups were observed in the spectrum. This is ascribed to the adventitious carbon present on the sample and the incomplete reduction of GO with hydrazine. It is believed that the incomplete reduction of GO with hydrazine results in the absence of the C=C bond.

As such, we employed the nomenclature of graphene-like, rather than graphene. The deposited sensing layer displays graphene-like characteristics such as thickness, but XPS has concluded that the GO precursor has not been completely reduced to ideal, stoichiometric graphene.

Figure 1: AFM image highlights the thickness of the graphene-like layers

Figure 3: XPS analysis of graphene-like sheets

Figure 4: Response of graphene-like nano-sheets/SAW sensor towards H_2 pulse sequence at room temperature

Fig. 4 shows the dynamic response of the graphene-like nano-sheets/SAW sensor to a sequence of different H_2 gas concentrations in synthetic air at room temperature. The measured response at room temperature was 5.8 kHz towards 1% H_2 . This low temperature operation also is favorable in many applications, especially those involving low power operations and inflammable environment.

We also have measured the frequency response towards NO_2 gas, and the frequency decreased upon exposure to NO_2 gas and increased after the gas being purged out [13]. NO_2 is an oxidizing gas whereas, H_2 is one the important reducing gas. The opposite frequency responses show p-type response. This agrees with the report by Schedin et al. [6].

The frequency shifts of different H_2 gas concentrations at room temperature are plotted in Fig. 5. It is observed that the frequency shift of H_2 at this temperature increases almost linearly with the increase in H_2 concentration.

The plotted graph in Fig. 6 shows the response and recovery time of measured SAW sensor towards H_2 gas at room temperature. Exposed to 0.06% H_2 , response and recovery time of 76 s and 128 s were observed, respectively. It is known that at elevated temperature, the probability of desorption increases [14]. Therefore, the slow recovery (~ 9 minutes) for 1% H_2 at room temperature is expected.

Figure 5: Frequency shifts for graphene-like nano-sheets/SAW sensors at room temperature

Figure 6: Response and recovery time vs H_2 gas concentrations

IV. CONCLUSION

We have successfully fabricated graphene-like nano-sheets/ $LiTaO_3$ SAW sensors and tested its sensing performance towards H_2 gas. The combination of graphene-like nano-sheets and SAW device technology has promising environmental applications, owing to its room temperature operation. Such devices may find applications for low temperature H_2 sensors, as our experiments have demonstrated that the graphene/SAW device shows good sensitivity towards H_2 gas.

ACKNOWLEDGMENT

Authors acknowledge Associate Prof. Dr. Johan du Plessis for help acquiring XPS spectrum.

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