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ScienceDirect

Procedia Engineering 168 (2016) 301 – 304

Procedia Engineering

www.elsevier.com/locate/procedia

30th Eurosensors Conference, EUROSENSORS 2016

The effect of thermal reduction and film thickness on fast response transparent graphene oxide humidity sensors

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Abstract

Resistive chemical sensors were developed having as sensitive film graphene oxide (GO) spin-coated on glass. Sensor electrical behaviour to room temperature relative humidity was evaluated. High sensitivity characterizes GO sensor. However, it lacks repeatability and long term stability. To this end, we developed spin-coated GO sensors with different active layer thickness, which were subjected to subsequent thermal annealing steps. It was found that the resulting reduced GO (rGO) sensors demonstrate excellent stability compromising, though, the sensitivity which mainly depends on the number of annealing steps and film thickness. Finally, we propose a rapid, transparent, low-power consumption rGO sensor, enabling its use in relevant applications.

Peer-review under responsibility of the organizing committee of the 30th Eurosensors Conference

Keywords: chemical, sensor, humidity, reduced graphene oxide, spin coating, transparent

1. Introduction

Humidity sensing is a significant issue in various applications including industrial, heating, ventilation, air conditioning (HVAC), medical and agriculture. Graphene, since its discovery, has been used as sensitive material for chemical and biological sensing due to the high surface-to-volume ratio and the exceptional quality of its crystal

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Peer-review under responsibility of the organizing committee of the 30th Eurosensors Conference
doi:10.1016/j.proeng.2016.11.201

structure, enabling on one hand the exposure of all atoms to the target gas and on the other hand reducing the electrical noise. GO, a functionalized form of graphene, has largely improved sensing properties as its defects further enhance the chemical interaction with the gas molecules [1]. Recently, there have been several efforts regarding the use of GO and rGO as humidity sensitive materials. However, GO sensors seem to lack of long-term stability and repeatability [1], and proposed rGO sensors may not be suitable for flexible substrates [2] or for industrial production implementation [3]. In this work, the effect of low temperature thermal annealing and GO film thickness were investigated in terms of electrical sensitivity to room temperature RH.

Nomenclature

GO	Graphene Oxide
rGO	reduced Graphene Oxide
XPS	X-ray Photoelectron Spectroscopy
RH	Relative Humidity

2. Experimental

GO solution in water with concentration of 5 mg/ml was procured and further diluted with DI water down to 2.5 mg/ml. The solution was ultra-sonicated for 1 h and stirred for another 1 hour at 250 rpm. Glass substrates were cleaned with acetone, isopropyl alcohol and DI water in ultrasonic bath and dried with nitrogen gas flow. GO was deposited on the substrate by spin-coating 2 ml of the GO solution. Different devices were prepared with two different spin-speeds at 3000 rpm and 6000 rpm for 60 s. Aluminum contacts were deposited with vacuum evaporation forming a GO channel of 0.9 mm length and 24 mm width. Figure 1(a) shows the transparent device with ITO contacts grown with the aid of DC-sputtering. The reduction of graphene oxide was achieved by thermal annealing up to 180 ^{o}C for 30 min in forming gas environment (5.2% H_2 in N_2). Reduction process was repeated for 6 times. In order to investigate the effect of the subsequent annealing steps to GO layer, XPS was performed after every process step. Electrical measurements were executed in a vacuum chamber in alternating vacuum-laboratory's humid air environment (30-45 % RH) at constant temperature (23 ^{o}C). The duration of the vacuum or humid air cycle was 100 s. Sensor response, R, was defined according to the formula:

$$R = \frac{I_{air} - I_{vacuum}}{I_{vacuum}} \quad (1)$$

where I_{air} and I_{vacuum} is the current at 7 V at air and vacuum respectively. Response time was defined as the duration elapsed from 10 % to 90 % of the signal's change from vacuum to humid air condition. It is important to note that none of the devices exhibited response when dry air was introduced in the vacuum chamber. Current-voltage measurements demonstrated Ohmic behavior for all devices. Finally, a commercial humidity sensor (HIH-4000-003) used as reference was placed in the vacuum chamber and its signal was monitored simultaneously with our devices via LabVIEW.

3. Results/discussion

3.1 Physical characterization

XPS was performed to evaluate the level of GO reduction after each step of thermal annealing. Figure 1(b) illustrates C1s XPS spectra of GO film grown at 3000 rpm of spin-coating. Three peaks at 284.5 eV, 286.5 eV and 288 eV are observed which are generally attributed to C-C, C-O and C=O bonds with a percentage of around 50%, 39% and 11% respectively. As shown in Figure 1(c), after the first annealing step, C-O and C=O bonds represent around 18% and 4% respectively of total C-bonds whereas C-C bonds more than 78%. After subsequent four annealing steps, Figure 1(d) demonstrates that C-C bonds peak reaches ca. 90% while C-O and C=O bonds peaks drop down to 6% and 4% respectively. It is therefore observed that the first annealing step efficiently reduces GO by