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

S. Papamatthaiou[1], D.-P. Argyropoulos[1], F. Farmakis[1], A. Masurkar[2], K. Alexandrou[2], I. Kymissis[2], N. Georgoulas[1]

1. Department of Electrical and Computer Engineering, Democritus University of Thrace, 67100 Xanthi, Greece
2. Department of Electrical Engineering, University of Columbia, 10027 New York, USA

Summary

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. Despite GO sensors' high sensitivity, they lack of long-term stability and repeatability. To this end, we developed spin-coated GO sensors with different active layer thickness, which were subjected to consequent thermal annealing steps. It was found that reduced GO (rGO) sensors demonstrate excellent stability but with lower sensitivity that depends on annealing steps and film thickness. In particular, for increasing annealing steps, rGO sensors' sensitivity is decreased which is in accordance with the proposed humidity adsorption models as analyzed by X-ray Photoelectron Spectroscopy (XPS). In addition, rGO sensors present a response time of less than 3 s, which is better compared to industrial HIH4000 humidity sensor. Finally, we propose a rapid, transparent, low-power consumption rGO sensor to human's exhale, enabling its use in relevant applications.

Motivation and results

Humidity sensing is a significant issue in various applications including industrial, heating, ventilation, air conditioning (HVAC), medical and agriculture. 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 relative humidity. The sensors were fabricated as follows: GO films were deposited via spin-coating (GO solution of 2.5 mg/ml spun at 3000 and 6000 rpm, alternatively) on glass and Aluminum (or alternatively ITO) contacts were grown with the aid of PVD methods through a shadow mask. The sensors were then subsequently annealed at 180 ^{o}C for 15 min in forming gas (5.2% H_2 in N_2) and electrical measurements were performed in a vacuum chamber in order to examine its sensitivity to laboratory's humid air (35-45 %RH) at constant temperature (23 ^{o}C). Finally, GO and rGO films were analyzed by XPS. Fig.1 shows GO sensor high sensitivity on humid air but it is also observed that the resistance fails to remain constant over several air/vacuum cycles. As expected, when GO sensors are thermal annealed at above-mentioned conditions, the current (at V=7 V) largely improves and after the third annealing step (or second step for the thinner samples) it remains rather constant (Fig.2). Fig.3 illustrates that sensors' sensitivity is decreased for increasing annealing steps until the third step (or second step for the thinner samples) and then it remains constant and also that, in all cases, thicker rGO sensors exhibit higher sensitivity and (shown in Fig.4) faster response to humidity (<3 s for six times annealed rGO). Finally, by comparing our rGO sensor and a commercial one (Fig.5), it is observed that our sensor's signal appears stable over time, with excellent repeatability and fast response time, with power consumption less than 3.5 $\mu W/cm^2$ while the commercial sensor consumes around 45.5 mW/cm^2. **Word count:** 499

References

[1] G. Naik, et al. (2016), *Graphene*, vol. 05, 1-13.

[2] D. Phan, et al. (2015), *Sensors and Actuators B: Chemical*, vol. 220, 1050-1055.

[3] L. Guo, et al. (2012), *Carbon*, vol. 50, 1667-1673.

Corresponding author

Sotirios Papamatthaiou, Democritus University of Thrace, Building of Electrical and Computer Eng. (Room 13), Vas. Sofias 12, 67100 Xanthi, Greece, +30 25410 79936, soter155@windowslive.com

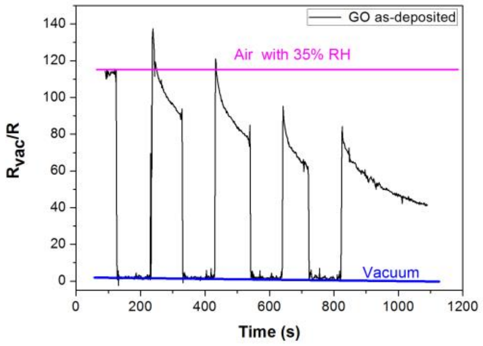

Figure 1: Response of spin-coated GO sensor to humidity (spin speed: 6000 rpm).

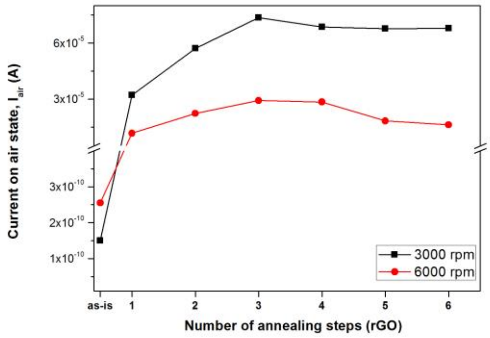

Figure 2: The effect of thermal annealing and spin speed of GO film on sensor's current (for V=7 V) on air state.

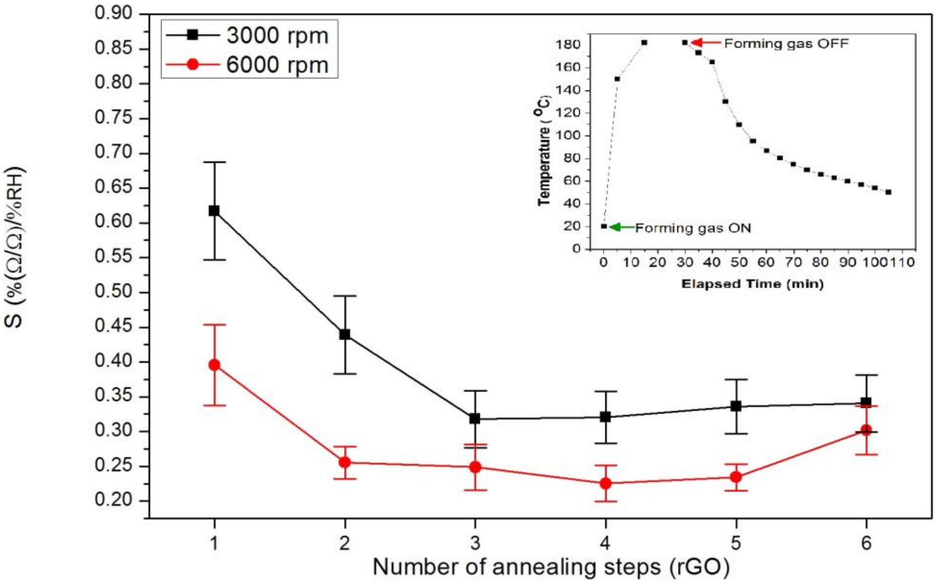

Figure 3: Sensitivity ((Rair-Rvac)/Rvac/%RH) comparison of 3000 and 6000 rpm for six annealing steps and schematic of annealing process.

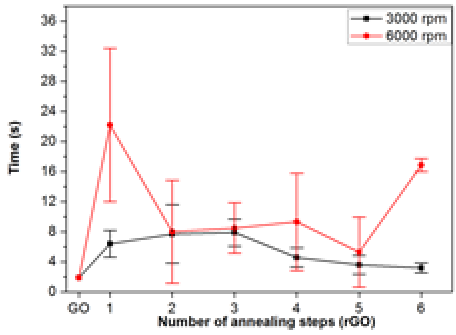

Figure 4: Comparison of response time to humidity for GO and rGO spin-coated at 3000 and 6000 rpm as a function of the annealing steps.

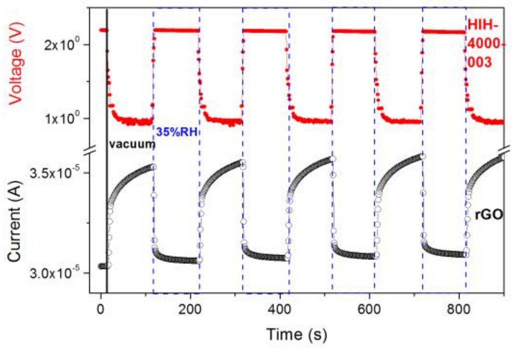

Figure 5: Response comparison of rGO sensor (3000 rpm, annealed steps:6) to commercial humidity sensor HIH4000 for 30% RH.