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Electrical and humidity-sensing properties of a composite material of RGO and CeO_2 that was fabricated by one-pot technique

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ABSTRACT: Novel impedance-type humidity sensors that are based on reduced graphene oxide and cerium oxide (CeO_2) (RGO/CeO_2) composite film were fabricated using one-pot procedure. The electrical and humidity-sensing properties of such films to which various amounts of RGO were added to CeO_2 were measured in detail as functions of relative humidity, to elucidate the contribution of RGO to the humidity-sensing capacity of the RGO/CeO_2 composite film. The humidity sensor that was made of RGO/CeO_2 composite film exhibited high sensitivity, good linearity and good long-term stability.

Keywords: **Humidity sensor; RGO/CeO_2; one-pot; impedance analysis.**

I. INTRODUCTION

Fabricating a good humidity sensor is rather complicated topic it must meet various requirements, including a linear response, high sensitivity, short response and recovery times, small hysteresis, chemical and physical stability, a wide range of operating humidities and low cost. Many materials have been used to fabricate humidity sensors such as polymers [1,2], ceramics [3,4] and composites [5,6]. Ceramic humidity sensors have attracted great interest due to their intrinsic characteristics, such as good reproducibility of the electrical properties, mechanical strength, chemical and physical stability [4]. CeO2 has been considered as a promising sensing material for detecting gas due to its good resistance to chemical corrosion, non-toxicity, safety and reliability. Recently, Wang et al. [7] fabricated an impedance-type humidity sensor that was made of CeO2/ionic liquid hybrid material. Xie et al. [8] fabricated an impedance-type humidity sensor that was made of CeO2 nanoparticles.

Graphene oxide (GO) sheets have been fabricated by the oxidation of graphite using strongly acidic oxidants, with hydroxyl and epoxide functional groups on their basal planes and carboxyl groups at their edges [9,10]. These groups are responsible for the high hydrophilicity and electrical insulation of GO, which makes it unsuitable for use in resistive sensors. GO can be reduced using reductants such as hydrazine and sodium borohydride, greatly increasing its the electrical conductivity by the removal of some of the oxygen functional groups to form reduced GO (RGO) [11,12]. To the best of our knowledge, no work has been done on the fabrication of an impedance-type humidity sensor that is based on composite film that is made by a one-pot process. This work elucidates the electrical and humidity-sensing characteristics, as functions of relative humidity (RH), of RGO/CeO_2 composite films that are fabricated by one-pot technique. The humidity-sensing performance of RGO/CeO_2 composites in which various amounts of RGO were loaded into the CeO_2 matrix, was studied.

II. EXPERIMENTAL

A. FABRICATION OF HUMIDITY SENSORS

The graphene oxide (GO) that was used herein was prepared by oxidation of graphene powder using Hummer's

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method [9]. RGO/CeO_2 composite was prepared using a method similar to our previous study [13]. In a typical synthesis, 0.05 g $CeCl_3$ and the required amounts of GO were added into 20 mL water/ethanol solution and then 150 µL HCl (36-38%) was added slowly under stirring for 1 h to form a homogenous solution. CeO_3 was used as a reducing agent in the reduction of GO to RGO in the one-pot process. This method typically generates CeO_2 particles that grow in situ homogeneously on the surface of the RGO. Then, the gel product were coated on an alumina substrate with a pair of comb-like electrodes by dip-coating, followed by thermal treatment at 60 °C for 0.5 h, thereby a humidity sensor of impedance-type was obtained. The amounts of GO dopant were varied 0.08, 0.1 and 0.12 g. Figure 1(a) displays a picture of the structure of humidity sensors fabricated on an alumina substrate.

B. Instruments and analysis

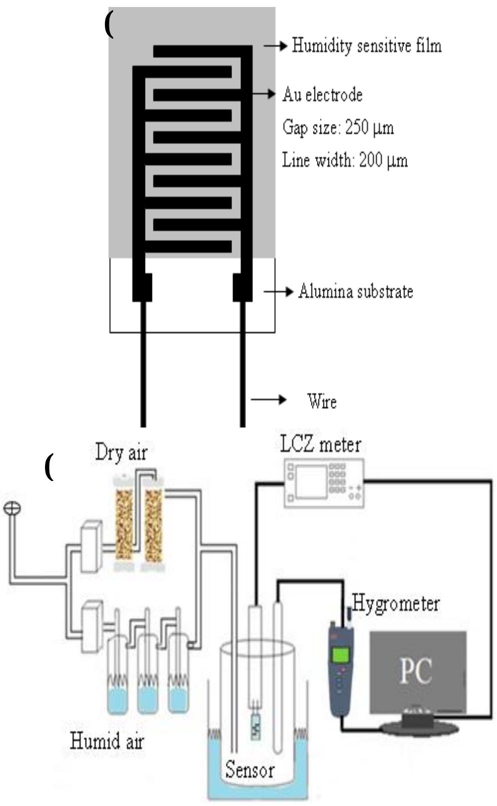

Fig. 1(a) Structure of humidity sensor, (b) humidity measurement system.

The impedance of the sensor was measured as a function of RH using an LCR meter (Philips PM6306) in a test chamber under the conditions of a measurement frequency of 1 kHz, an applied voltage of 1 V, an ambient temperature of 25 °C. As shown in Fig. 1(b), a divided humidity generator was used as the principal facility for producing the testing gases. The required humidity was produced by adjusting the proportion of dry and humid air generated by the divided flow humidity generator under a total flow rate is 10 L/min. The RH values were measured using a calibrated hygrometer (Rotronic) with an accuracy of ±0.1% RH.

III. RESULTS AND DISCUSSION

A. Structural characteristics of RGO/CeO_2 composite films

Figures 2(a) and 2(b) present the SEM of the pure CeO_2 and RGO/CeO_2 composite film, respectively. The size of the pure CeO_2 ranged from 0.5 to 3.0 µm. Compared with pure CeO2, it was obvious that the RGO sheets were attached to CeO_2 particles.

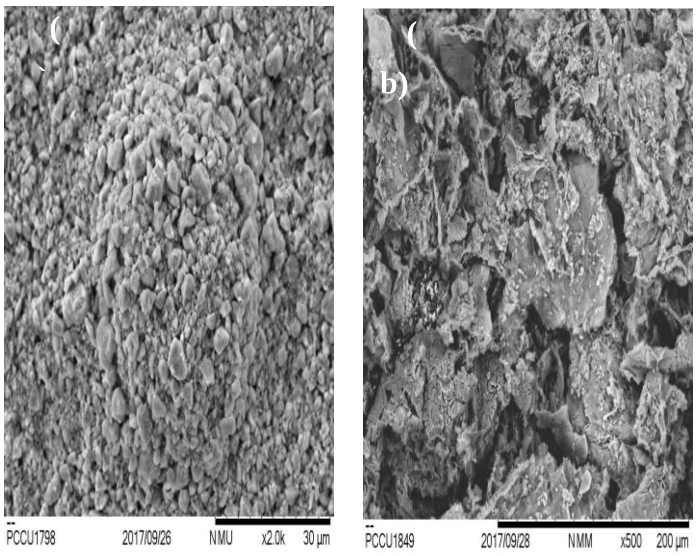

Fig. 2 SEM of CeO_2 and RGO/CeO_2 composite film

Figure 3 presents the the XRD spectra of the GO and RGO/CeO_2 composite that were prepared by sol-gel processes. GO yielded a sharp diffraction peak (002)

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appears at 10.75°, revealing that most of the graphite powder was oxidized to GO with an increase in the d-spacing to 6.70 Å [14]. The XRD patterns of CeO_2 include main peaks at 28.6°, 33.2°, 47.5°, 56.4°, 59.1° and 69.4°, corresponding to (111), (200), (220), (311), (222) and (400) of CeO_2, respectively. It is in good agreement with cubic fluorite structure CeO_2. The RGO/CeO_2 composite exhibited no appreciable difference between the orientations and phases of CeO_2 probably owing to the fact that RGO was embedded in the CeO_2 matrix and so could not be easily detected by X-ray diffraction. Additionally, the diffraction peaks of the RGO/CeO_2 composite were very broad, indicating a disordered and poor crystalline structure.

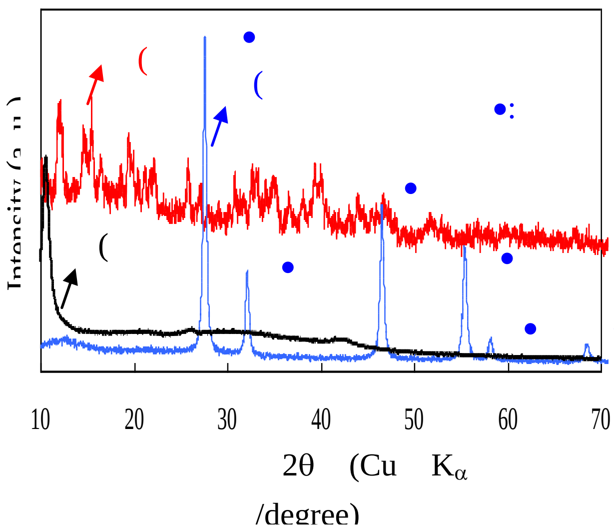

Fig. 3 XRD patterns of (a) GO, (b) pure CeO_2 and (c) RGO/CeO_2 composite that was fabricated by sol-gel.

B. Electrical and humidity-sensing properties of RGO/CeO_2 composite films

Figure 4 presents the effect of the amount of added RGO on the impedance of RGO/CeO_2 composite films as a function of relative humidity. Table 1 summarizes the results concerning sensitivity (defined as the slope of logarithmic impedance (log Z) as a function of %RH), linearity (given by a correlation coefficient that is defined as the R-squared value of the linear fitted curve from 20 to 90% RH). Measurements were made at 25 °C, an AC voltage of 1 V, and a frequency of 1 kHz. The impedance of the RGO/CeO_2 composite films increased with the amount of added partial reduced GO over the range of humidity considered, owing to the low conductive GO embedded in the CeO_2 matrix upon the addition of a large amount of GO. This result suggests that the geometry, dimensions and conductivity of RGO influence the conductance of an RGO/CeO_2 composite film. The impedance of the CeO_2 film changed by a small magnitude as RH increased from 20 to 90%, indicating low sensitivity, and the response was non-linear on a semi-logarithmic scale over the full RH range. The humidity sensor that was made of the 0.1 g RGO/CeO_2 composite film had the highest linearity (R2 = 0.9905) and good sensitivity (0.0388 log Z/%RH).

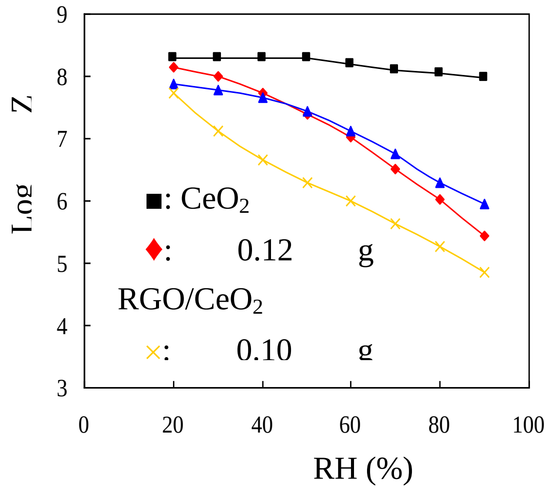

Fig. 4 Impedance versus relative humidity for humidity sensors that were made of CeO_2 film and RGO/CeO_2 composite films with various amounts of added RGO.

Table 1 Effect of amount of RGO on the sensing properties of the RGO/CeO_2-based humidity sensors

Film	Sensitivity (log Z/%RH)[a]	Linearity (R^2)[b]
CeO_2	−0.0043	0.8132
0.08 g RGO/CeO_2	−0.0284	0.9531
0.10 g	−0.0388	0.9905

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RGO/CeO_2		
0.12 g RGO/CeO_2	–0.0392	0.9701

[a]**Sensitivity was defined as the slope of the logarithmic impedance versus relative humidity plot in the range 20 to 90% RH.**

[b]**Linearity was shown as the correlation coefficient of the logarithmic impedance versus relative humidity plot in the range 20 to 90% RH.**

IV. CONCLUSION

The one-pot route was used to fabricate an impedance-type humidity sensor that was based on RGO/CeO_2 composite film. The RGO thus formed effectively dominated the electrical and humidity-sensing properties (sensitivity and linearity) of the RGO/ CeO_2 composite film. The humidity sensor that was based on an RGO/CeO_2 composite film that had been doped with 0.1 g of partial reduced GO exhibited better sensing properties than the CeO_2 film good sensitivity and highest linearity (Y= –0.0388 X + 6.6102; R^2=0.9905) between the logarithm of impedance (log Z) and RH in the range 20 to 90% RH. The improvement in sensing properties was explained in terms of the composition.

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REFERENCES

[1] Y. Sakai, Y. Sadaoka, M. Matsuguchi, Humidity sensors based on polymer thin films, Sens. Actuators B 35-36 (1996) 85-90.

[2] B. Adhikari, S. Majumdar, Polymers in sensor applications, Prog. Polym. Sci. 29 (2004) 699-766.

[3] G. Sberveglieri, R. Murri, N. Pinto, Characterization of porous $A1_2O_3$-SiO_2/Si sensor for low and medium humidity ranges, Sens. Actuators B 23 (1995) 177-180.

[4] T. A. Blank, L. P. Eksperiandova, K. N. Belikov, Recent trends of ceramic humidity sensors development: A review, Sens. Actuators B 228 (2016) 416-442.

[5] C. D. Feng, S. L. Sun, H. Wang, C. U. Segre, J. R. Stetter, Humidity sensing properties of Nation and sol-gel derived SiO_2/Nation composite thin films, Sens. Actuators B 40 (1997) 217-222.

[6] Y. Li, M. J. Yang, Y. She, Humidity sensors using in situ synthesized sodium polystyrenesulfonate/ZnO nanocomposites, Talanta 62 (2004) 707-712.

[7] W. Xie, X. Duan, J. Deng, J. Nie, T. Wang, CeO_2/ionic liquid hybrid materials with enhanced humidity performance, Sens. Actuators B 252 (2017) 870-876.

[8] W. Xie, B. Liu, S. Xiao, H. Li, Y. Wang, D. Cai, D. Wang, L. Wang, Y. Liu, Q. Li, T. Wang, High performance humidity sensors based on CeO_2 nanoparticles, Sens. Actuators B 215 (2015) 125-132.

[9] Hummers, W. S., Offeman, R. E., (1958), Preparation of graphitic oxide, *J. Am. Chem. Soc.* 80, 1339.

[10] Lerf, A., He, H., Forster, M., Kilnowski, J., (1998), Structure of graphite oxide revisited, *J. Phys. Chem. B* 102, 4477-4482.

[11] Lu, G., Ocola, L. E., Chen J., (2009), Gas detection using low-temperature reduced graphene oxide sheets, *Appl. Phys. Lett.* 94, 083111-3.

[12] Pei, S. F., Cheng, H. M., (2012), The reduction of graphene oxide, *Carbon* 50, 3210-3228.

[13] P. G. Su, L. Y. Yang, NH_3 gas sensor based on Pd/SnO_2/RGO ternary composite operated at room-temperature, ens. Actuators B 223 (2016) 202-208.

[14] A. B. Bourlinos, D. Gournis, D. Petridis, T. Szabó, A. Szeri, I. Dékány, Graphite oxide: chemical reduction to graphite and surface modification with primary aliphatic amines and amino acids, Langmuir 19 (2003) 6050–6055.