Sensors & Transducers, Vol. 229, Issue 1, January 2019, pp. 39-46

www.sensorsportal.com

Sensors & Transducers

Published by IFSA Publishing, S. L., 2019
http://www.sensorsportal.com

Creation of a Humidity Sensor Based on Functionalized Graphene Nanostructures

[1] Tilek K. Kuanyshbekov, [1] Arkady M. Ilyin, [2] Gary W. Beall and [1] Nazim R. Guseinov

[1] al-Farabi Kazakh National University, al-Farabi Avenue, 71, Almaty 050040, Kazakhstan
[2] Texas State University, 601 university Dr, San Marcos, TX 78666-4684, USA
[1] Tel.: 87052817796, 87752028095
E-mail: kuanyshbekov_17@mail.ru, ilyinar@mail.ru, solar_neo@mail.ru, gb11@txstate.edu

Received: 30 November 2018 /Accepted: 31 December 2018 /Published: 31 January 2019

Abstract: In this article, we present the results of GO as a primary material of the humidity sensor and the electro-physical characteristics of this sensitive element. The sensitive humidity sensor created using the GO membrane was obtained via vacuum filtration. Initial material GO was synthesized by the modified Hummers method and characterized by Raman spectroscopy, XRD analysis and SEM. The stability of the electrical resistance, also the rate of recovery and response of a humidity sensor element based on GO was studied as a function of the relative humidity level. The GO sensor was demonstrated to function properly in a wide range of humidity over an extended 10 hour period at room temperature. The deviation of the electrical resistance readings of the device during the experiment did not exceed 2 %.

Keywords: Graphene; Functionalized graphene, Graphene oxide (GO); Hummers method; Humidity sensor, X-ray diffraction.

1. Introduction

Graphene, functionalized graphene, in particular graphene oxide (GO) and other carbon materials are of great interest for the scientific community due to such unique properties as: high level of thermal and electrical conductivity, strength and excellent adsorption characteristics [1-2]. For the first time in 2004, Geim and Novoselov, using the method of mechanical splitting, separated one sheet of graphene from graphite. Since then, due to its unique mechanical, thermal, optical and electrical properties, graphene has attracted more attention in various fields of science and technology. Graphene is a flat layer of the monatomic structure of sp^2 bound carbon atoms, which is tightly packed in a hexagonal structure [3].

One of the main directions in the study of graphene is GO which has another name such as functionalized graphene [4-5]. Compared with pure graphene, functionalized graphene has a wide range of applications in semiconductor electronics for creating biosensors [6], supercapacitors, various gas sensors [7], organic electrodes, light-emitting diodes, etc. [8-9].

GO (functionalized graphene) – is a combination of carbon, hydrogen and oxygen in various ratios, which is formed during processing graphite with strong oxidants [10].

The functionalization of graphene can be carried out by various methods, in particular the creation of radiation defects, hydrogenation, etc., including oxidation [11-12]. In the work [13] being considered that there are two basic categories of functionalization

of graphene: chemical and non-chemical. The chemical functionalization method is understood as chemical modification of the surface by various groups of atoms, as a result of which new covalent bonds between atoms native to RGO/GO and the guest functional groups are formed. In the case of non-chemical functionalization, mainly physical interaction occurs and functionalization occurs on the basis of π interaction between guest molecules and RGO/GO. Both types of functionalization contribute to changes in the properties of graphene, but the most effective is chemical modification [13-15].

Functionalization of graphene with various strong oxidizing agents such as H_2SO_4, HNO_3, $KMnO_4$, $KClO_3$, $NaClO_2$ followed by exfoliation into single sheets facilitates the so-called functionalized graphene or graphene oxide, which in turn, has found widespread application in the field of science and technology due to the modified electronic structure [5].

In our work, the most common Hummers method was used to implement the functionalization of graphene, in which the graphite powder is oxidized with strong acids, then, with subsequent processes, it is exfoliated into few-layer graphene sheets in water or in organic solutions, whereby a GO was obtained [4, 16 and 17].

GO contains hydroxyl, carbonyl and carboxyl functional groups on the basal plane, and this makes it hydrophilic and creates the ability to form stable water suspensions, compared to other carbon materials. This property of graphene oxide makes it sensitive to moisture, which can be used to determine humidity in the atmosphere [18].

In connection with this at present, the role of GO is of great interest in measuring and monitoring the humidity of the environment for industrial, agricultural and human activities [19].

For many years, the creation of highly sensitive sensors for determining the humidity of the environment is relevant, due to the need in such areas of engineering and science as medical applications, textile industry, agriculture, biological products, scientific and technical centers, food industry and other industries. For the creation of highly sensitive sensors, the following physical and electrical characteristics are particularly important: the stability of the sensor operation at various levels of humidity, as well as in aggressive environments; high sensitivity, quick response to moisture and recovery in a short period of time; wide range for determining humidity. In addition to the above characteristics, the most important are the ease of manufacturing technology, low cost. Another important factor in creating a highly sensitive sensor is the selection of a suitable initial material, which can affect many of the above listed sensor properties. One of the promising materials for creating highly sensitive sensors is carbon materials, namely, graphene, graphene oxide, RGO, and carbon nanotubes [19-20].

With the advent of graphene and graphene-like materials, due to their physico-mechanical, as well as the most important electronic properties, much research has been done on the creation of sensitive sensors for determining humidity and various gases.

For the first time, in the work [21] demonstrated the potential of graphene and graphene oxide as an initial material for the creation of nanoscale sensors that can detect the following types of CO_2, NO_2, NH_3 gas.

The creation of highly sensitive sensors based on graphene, graphene oxide, reduced graphene oxide was mainly aimed at determining the following types of gas: NO_2, NH_3, CO_2, which were indicated in the following works: the production of sensitive humidity sensor based on thermal reduced graphene; creation of a sensitive sensor based on epitaxial graphene; creating a flexible sensor for detecting various gases based on carbon nanotubes and graphene; studying the characteristics of the determination of moisture based on thin film of coarse grained graphene oxide; production of a humidity sensor based on bilayer graphene, which was synthesized by the CVD method; the creation of a microscale capacitive humidity sensor, in which a graphene oxide film was used as the initial material; humidity sensor based on high proton conductivity of graphene oxide; using the Electro-spun PVDF / Graphene Membrane to create a capacitive humidity sensor [22-31]. Also, the above works demonstrate the creation of sensitive sensors for determining environment humidity, in which graphene oxide membranes were used as the initial material. In this way, based on all the above works, graphene, graphene oxide and their related structures, due to their physico-mechanical properties, are the most promising materials for creating sensitive humidity sensors.

Therefore, in this work, we examine the humidity sensor based on graphene oxide aimed at studying the potential use of a commercially available product as a sensitive element to a moist environment at room temperature. Compared with other types of sensors based on GO, this sensor has the following advantages: wide range of humidity level, low cost, does not require high technology, and is resistant to aggressive media. Previously, we published an expanded abstract on the topic of the «Sensitive humidity sensor based on functionalized graphene» in which a brief description of the electro-physical characteristics was considered [32].

2. Graphene Oxide Synthesis

In this paper, we studied the humidity sensor based on GO at wide range of humidity for 10 hours at room temperature. We used graphene oxide that was obtained by a modified Hammers method using pure natural graphite as a starting material. The process of synthesis of graphene oxide is a specific sequence, presented below. 23 ml of concentrated sulfuric acid was added to a 250 ml flask with an ice at 0°C, which was filled graphite with weight 1 g, after which 0.5 g sodium nitrate was added. Then, while stirring the

mixture with a mechanical stirrer, 3 g of solid potassium permanganate was gradually added with maintaining the temperature below 20 °C over 2 hours. After raising the temperature of the mixture to 35 °C, it was maintained for 30 minutes at this temperature, then after adding deionized water; the temperature rose to 90 °C and stirred for 30 minutes. At the end, 30 % hydrogen peroxide was added until the color of the mixture changed to bright yellow and until gas is stopped. Then the product was filtered and washed several times in 5 % hydrochloric acid solution to remove metal ions, then washed with deionized water to remove acids. Fig. 1 shows aqueous dispersion of graphene oxide.

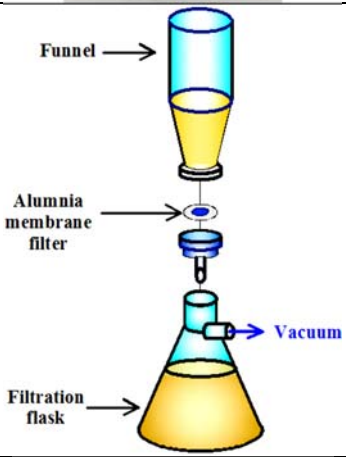
Fig. 1. GO aqueous dispersion.

We used a graphene oxide membrane to create the humidity sensor. For the manufacture of graphene oxide membrane there are the following methods: filtration (vacuum, pressure filtration); casting/coating-based (spinning casting, drop casting, dip-coating, spray-coating); layer-by-layer assembly; evaporation-assembled method; templating method; shear-alignment method and hybrid method [33]. Of all the above methods in our experimental work, we chose vacuum filtration because of the availability, low labor intensity, nano-sized control over the membrane thickness and the possibility of high-scale production of graphene oxide membranes.

The preparation of GO membrane using vacuum filtration was made in the following works: [33-35]. The advantages of the process of obtaining GO membrane using vacuum filtration are that, the graphene oxide nanolayers are connected and located almost in parallel, which is promising as an initial material for creating humidity sensors. One more important advantage of using vacuum filtration is that the physico-chemical property of graphene oxide nanolayers does not change during the manufacture of GO membrane. Synthesis of GO membrane using vacuum filtration consists of the following main processes: selection of an appropriate concentration of GO solution; deposition of GO membrane is carried out by passing a solution of graphene oxide through a porous film, the resulting GO membrane is dried [33, 35]. The result is a uniformly distributed membrane with a relatively flat surface, due to the fluidity of water in the solution filtration process, and controlling the film thickness depends on the concentration chosen.

In this process, we used a 4.0 ml GO solution with concentration 1.5 mg/ml. The GO papers were left overnight to dry at room temperature, and then peeled off from the filter. Table 1 show GO membrane obtained using the vacuum filtration method, in which the dispersion of GO was carried out using a porous alumina membrane filter with a pore size of 800 nm and a diameter of 25 mm.

Table 1. Obtaining GO membrane.

Step 1. Choosing the suitable concentration of GO aqueous dispersion	
Step 2. Vacuum filtration with alumina membrane	
Step 3. Drying and finished GO membrane	

3. Investigation of the GO

The GO obtained by us was studied using Raman spectroscopy, which is intended to study the characteristics of carbon nanostructures, as well as to characterize the quality of graphene in industrial processes and studies, since Raman spectroscopy is not destructive [36-37]. The Raman spectrum of graphene oxide, like graphene, has G and D peaks, the first order of scattering of the E_{2g}-phonon sp^2 carbon atoms G peak is reflected around 1580 cm^{-1}, and also

occurs due to stretching of the C–C bond in the basal plane, which is characteristic of all sp^2 hybridized carbon materials, and the D peak appears in the region of 1200–1400 cm^{-1} and indicates a certain amount of disorder or edges in the carbon structure [38-39]. In the work [40] the ratio D/G, which is about 0.95, which indicates a large number of defects in the crystal lattice, and this ratio increase with chemical reduction. Fig. 2 shows the Raman spectra of graphene oxide, in which the D peak is located in the 1352 cm^{-1} region, the G peak is in the 1584 cm^{-1} region, and the ratio of these peaks corresponds to 0,88 which is in good agreement with the literature data [38-42].

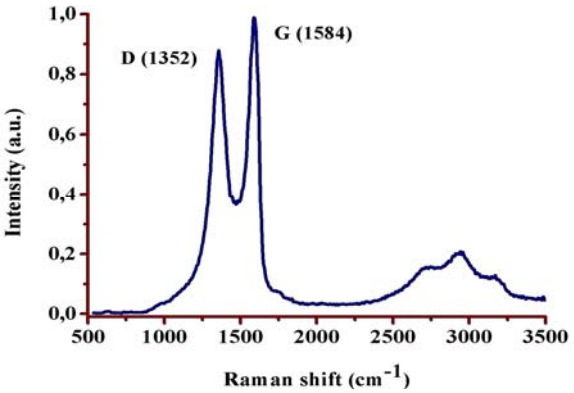
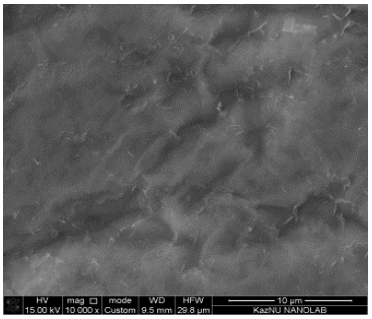
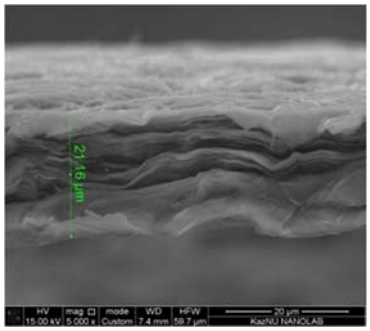


Fig. 2. Raman spectrum of GO.

The SEM of GO micrographs in Fig. 3 (a-surface, b-cross section) clearly show that GO has a two-dimensional sheet-like structure.



a) the surface of GO



b) the cross section of GO

Fig. 3. SEM images of GO.

From the images (Fig. 3(a, b)) of SEM, it can be seen that the layers are stacked one above the other, and there are wrinkled areas on the surface of the GO membrane. According to the Fig. 3(b), the SEM image of the cross section clearly shows that the graphene oxide membrane obtained by us is layered and has a highly ordered lamellar structure, and the membrane thickness is about 20 μm. EDX analysis in the Fig. 4 shows that GO contains about 67 at.% C, 33 at.% O.

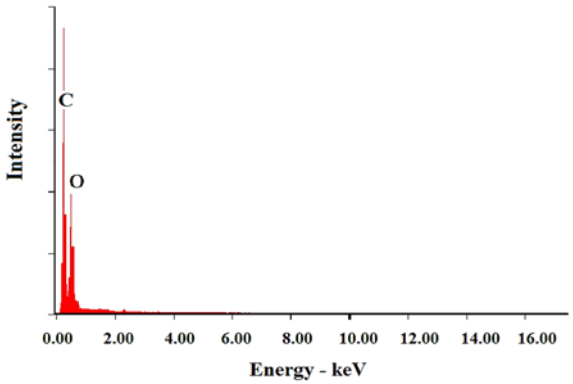


Fig. 4. EDX analysis of GO.

The XRD analysis of graphene oxide and highly-oriented pyrolytic graphite shows in the Fig. 5 that were investigated on the DRON-7 instrument [43].

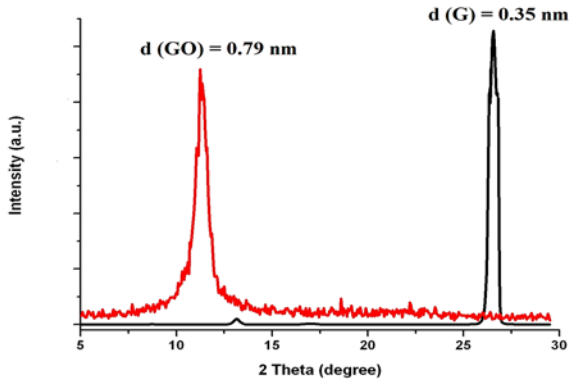


Fig. 5. XRD of graphite and GO.

The interpretation of the XRD allowed to determine the interplanar spacing in the initial GO (d = 0.79 nm) and in the graphite (d = 0.35 nm). The results obtained are in good agreement with the literature data [44].

4. The Structure of the Sensor and Study its Electro-physical Characteristics

The design of the humidity sensor is shown in the Fig. 6, where the GO membrane was mounted on a dielectric substrate and connected at opposite ends by copper wires (diameter 0.15 mm) as electrodes to the GO samples. These electrodes were covered with

conductive silver paint contacts and left to dry over night to insure good electrical contact.

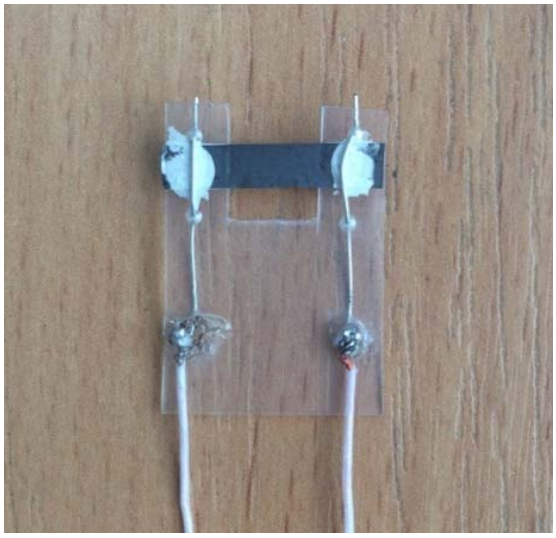


Fig. 6. Structure of the humidity sensor.

The dimensions of the substrate are 2.5×1.8 cm, the thickness of the GO membrane is about 20 μm as shown in the Fig. 3(b), the length is 2.5 cm, and the width is 0.5 cm. The membrane surface is open on both sides, which allows it to react sensitively to the relative humidity of the environment.

As can be seen from the schematic in the Fig. 6, a humidity sensor based on GO and an exemplary DHT 22 sensor by the Arduino platform are placed together in the testing chamber, which was used to control the humidity and DHT 22 sensor has the following technical characteristics: calculated to measure the level of humidity in the range from 0 to 100 % and the measurement accuracy is in the range of 2-5 %.

Schematic representation of an installation for investigating the sensitivity of the sensor to humidity shows in the Fig. 7.

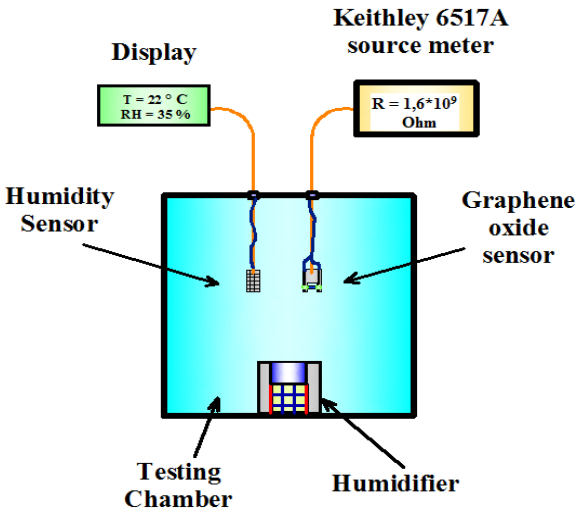


Fig. 7. Schematic view of the installation.

The electro-physical parameters were studied as a function of humidity to determine the operating characteristics of the device. The stability of the electrical resistance was monitored using the Keithley 6517A meter attached to the copper electrodes using alligator clips. The humidity sensor was kept in the sealed chamber with humidity levels controlled at: 5 %, 25 %, 50 %, 75 %, 100 % for 10 hours. The results of testing the sensor for the stability of the electrical resistance under various levels of constant humidity, depending on the time is shown in the Fig. 8. It can be seen from the results that the deviation of the electrical resistance values of the structure for a long time does not exceed 2 %.

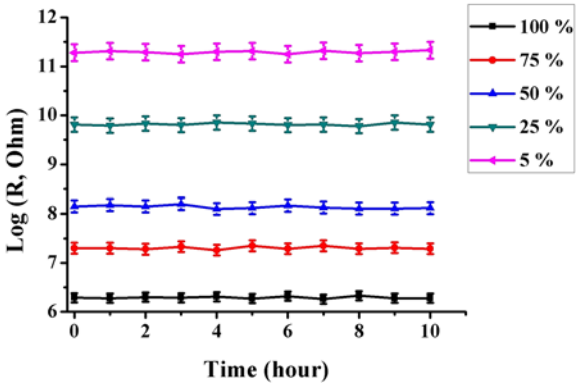


Fig. 8. Testing the humidity sensor on the stability of the electrical resistance readings under different humidity levels

The electrical resistance versus humidity is presented in the Fig. 9. This image shows that the electrical resistance of the sensor decreases from 11,5 to 6,3 Log (R, Ohm) with increasing humidity at the range of 5-100 %.

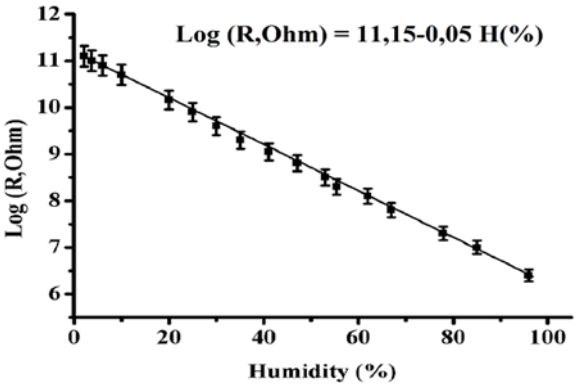


Fig. 9. Dependence of the electrical resistance on humidity.

In the process of raising humidity in the chamber, a drastic decrease in electrical resistance is observed, this would indicate a large amount of water vapor penetrates into the interplanar distance, and is also adsorbed on the surface of the sample altering the resistance.

We also studied the recovery and response of the humidity sensor. The recovery and response time of the sensor were tested at the range from 5 to 100 % humidity. Response dynamics and recovery were measured in the same way as we measured the stability of the electrical resistance.

The dynamics of the response of the humidity sensor as a function of time was studied using the sealed chamber with the hand made humidifier. According to the Fig. 10, a decrease in electrical resistance is observed in the entire range of relative humidity. This is because large amount of water molecules are adsorbed on the GO surface, which significantly increases its conductivity due to proton-electron exchange between graphene oxide and the adsorbed molecules. There are also similar mechanism of changes in electrical conductivity, due to the influence of a water molecule, as a result of which proton-electronic changes occur [28, 30-31, 45]. Fig. 10 shows the dynamics of the humidity sensor response.

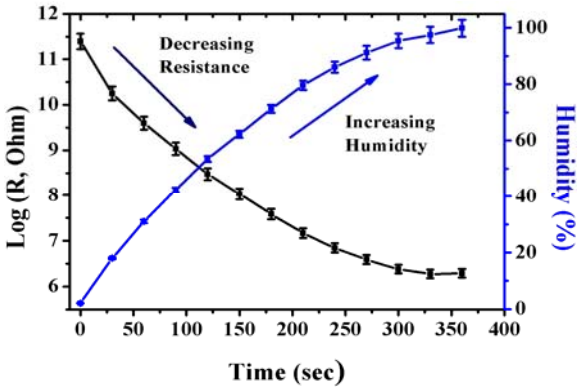


Fig. 10. Dynamics of the response.

Recovery time of the sensor can be seen in the Fig. 11. Increasing electrical resistance of the structure were recorded as a function of time using the Keithley 6517A source meter.

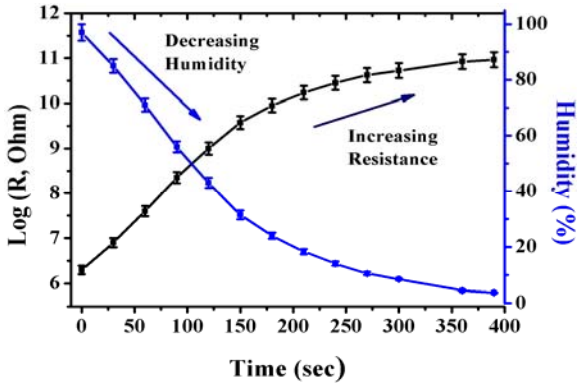


Fig. 11. Recovery dynamics.

The changing in the electrical resistance of the sample mainly depends on the adsorption of the water molecule on the surface of GO therefore the electrical conductivity depends on water molecule on the surface. When the humidity level decreases from 100 to 5 %, adsorbed water molecules are removed, which leads to increasing resistance. Signficantly increases of electrical resistance of the sample observed at the humidity level from 100 to 20 % and resistance changes from 6,4 up to 10 Log (R, Ohm).

5. Conclusions

In this article we created humidity sensor based on GO membrane, which was synthesized by the modified Hummers method using pure natural graphite, then from this GO aqueous dispersion via vacuum filtration GO membrane were produced. The electro-physical characteristics of the created humidity sensor were tested in wide range of humidity level from 5 to 100 %. The humidity sensor was tested for the stability of the electrical resistance at multiple humidity levels of: 5 %, 25 %, 50 %, 75 %, and 100 % for 10 hours at room temperature. The obtained measurement results show that the sensor based on GO is stable (±2 %) in the entire humidity range. Significant changes in the values of electrical resistance are associated with the absorption of a water molecule on the surface and penetration into the interplanar distance of GO. According to the results obtained, the humidity sensor based on GO is able to work stably at wide range of humidity and respond rapidly to changes in humidity, which allows it to be useful as a precise and sensitive device.

Acknowledgements

The article was published with the support of the grant of the Ministry of Education and Science of the Republic of Kazakhstan (IRN: AP05130413).

References

[1]. K. S. Novoselov, A. K. Geim, *et al*., Electric Field Effect in Atomically Thin Carbon Films, *Rev. Sci. Instrum Science,* Vol. 306, Issue 5696, 2004, pp. 666-669.

[2]. A. M. Ilyin, Computer Simulation of Radiation Defects in Graphene and Relative Structures, *"Graphene Simulation"*, Gong, J. R., (Ed), *InTech*, Rijeka, 2011, pp. 39-52.

[3]. A. K. Geim, K. S. Novoselov, The Rise of Graphene, *Nature Materials*, Vol. 6, Issue 3, 2007, pp. 183-191.

[4]. Da Chen, Hongbin Feng, Jinghong Li, Graphene Oxide: Preparation, Functionalization, and Electrochemical Applications, *Chem. Rev*., Vol. 112, Issue 11, 2012, pp. 6027−6053.

[5]. Chernozatonskii L. A., *et al*., Novel graphene-based nanostructures: physicochemical properties and applications, *Russian Chemical Reviews,* Vol. 83, Issue 3, 2014, pp. 251-279.

[6]. Y. Shao, J. Wang, H. Wu, J. Liu, I. Aksay, Y. Lin, Graphene Based Electrochemical Sensors and Biosensors: A Review, *Electroanalysis*, Vol. 22, Issue 10, 2010, pp. 1027-1036.

[7]. K. S. Novoselov, Nobel Lecture: Graphene: Materials in the Flatland, *Rev. Mod. Phys*., Vol. 83, Issue 3, 2011, pp. 837-849.

[8]. R. R. Nair, W. Ren, R. Jalil, I. Riaz, V. G. Kravets, L. Britnell, P. Blake, F. Schedin, A. S. Mayorov, S. Yuan, M. I. Katsnelson, H. M. Cheng, W. Strupinski, L. G. Bulusheva, A. V. Okotrub, I. V. Grigorieva, A. N. Grigorenko, K. S. Novoselov, A. K. Geim, Fluorographene: a two-dimensional counterpart of Teflon, *Small,* Vol. 6, Issue 24, 2010, pp. 2877-2884.

[9]. L. A. Chernozatonskii, P. B. Sorokin, A. A. Artukh, Novel graphene-based nanostructures: physicochemical properties and applications, *Russian Chemical Reviews,* Vol. 83, Issue 3, 2014, pp. 251-279.

[10]. He H., Klinowski J., Forsterb M., Lerf A., A new structural model for graphite oxide, *Chemical Physics Letters,* Vol. 287, Issue 1, 1988, pp. 53-56.

[11]. A. M. Ilyin, G. W. Beall, I. A. Tsyganov, Simulation and study of bridge-like radiation defects in the carbon nanostructures, *Journal of Computational and Theoretical Nanoscience*, Vol. 7, Issue 10, 2010, pp. 2004-2007.

[12]. A. M. Ilyin, N. R. Guseinov, I. A. Tsyganov, *et al*., Computer simulation and experimental study of graphane-like structures formed by electrolytic hydrogenation, *Physica E,* Vol. 43, Issue 6, 2011, pp. 1262-1265.

[13]. Yuhai Hu, Xueliang Sun, Chemically Functionalized Graphene and Their Applications in Electrochemical Energy Conversion and Storage, *InTech*, *Advances in Graphene Science*, Chapter 7, 2013, pp. 162-189. http://dx.doi.org/10.5772/55666

[14]. Sharma R., Baik J., Perera C., Anomalously large reactivity of single graphene layers and edges toward electron transfer chemistries, *Nano Letters,* Vol. 10, Issue 2, 2010, pp. 398-405.

[15]. Lim H., Lee J., Shin H., Spatially resolved spontaneous reactivity of diazonium salt on edge and basal plane of graphene without surfactant and its doping effect, *Langmuir*, Vol. 26, Issue 14, 2010, pp. 12278-12284.

[16]. Ruoff R. S., Graphene: calling all chemists, *Nat. Nanotechnology*, Vol. 3, Issue 1, 2008, pp. 10-11.

[17]. Dreyer D. R., Park S., Bielawski C. W., Ruoff R. S., The chemistry of graphene oxide, *Chem. Soc. Rev.,* Vol. 39, Issue 1, 2010, pp. 228-240.

[18]. G. Naik, S. Krishnaswamy, Room-Temperature Humidity Sensing Using Graphene Oxide Thin Films, *Graphene,* Vol. 5, 2016, pp. 1-13.

[19]. C. Lee & G. Lee, Humidity sensors: A Review, *Sens. Lett.,* Vol. 3, Issue 1, 2005, pp. 1–15.

[20]. A. M. Ilyin, E. A. Daineko & G. W. Beall, Computer Simulation and Study of Radiation Defects in Graphene, *Physica E,* Vol. 42, Issue 1, 2009, pp. 67-69.

[21]. Schedin F., Geim A. K., Morozov S. V., Hill E. W., Blake P., Katsnelson M. I., Novoselov K. S., Detection of Individual Gas Molecules Adsorbed on Graphene, *Nature Materials*, Vol. 6, Issue 9, 2007, pp. 652-655. http://dx.doi.org/10.1038/nmat1967

[22]. Lu G., Ocola L. E., Chen J., Reduced Graphene Oxide for Room-Temperature Gas Sensors, *Nanotechnology,* Vol. 20, Issue 44, 2009, 445502. http://dx.doi.org/10.1088/0957-4484/20/44/445502

[23]. Mansoor Anbia, S. E. Moosavi Fard, A Humidity Sensor Based on Nb-doped Nanoporous TiO_2 Thin Film, *Sensors & Transducers,* Vol. 134, Issue 11, November 2011, pp. 56-64.

[24]. Nomani M., Shishir R., Qazi M., Diwan D., Shields V., Spencer M., Tompa G. S., Sbrockey N. M., Koley G., Highly Sensitive and Selective Detection of NO_2 Using Epitaxial Graphene on 6H-SiC, *Sensors and Actuators B: Chemical*, Vol. 150, Issue 1, 2010, pp. 301-307. http://dx.doi.org/10.1016/j.snb.2010.06.069

[25]. Jeong H. Y., Lee D.-S., Choi H. K., Lee D. H., Kim J.-E., Lee J. Y., Lee W. J., Kim S. O., Choi S.-Y., Flexible Room-Temperature NO_2 Gas Sensors Based on Carbon Nanotubes/Reduced Graphene Hybrid Films, *Physics Letters,* Vol. 96, Issue 21, 2010. http://dx.doi.org/10.1063/1.3432446

[26]. Gautam Naik, Sridhar Krishnaswamy, Room-Temperature Humidity Sensing Using Graphene Oxide Thin Films, *Scientific Research Publishing*, *Graphene,* Vol. 5, Issue 1, 2016, pp. 1-13. http://www.scirp.org/journal/graphene http://dx.doi.org/10.4236/graphene.2016.51001

[27]. Meng-Chu Chen, Cheng-Liang Hsu, Ting-Jen Hsueh, Fabrication of Humidity Sensor Based on Bilayer Graphene, *IEEE Electron Device Letters,* Vol. 35, Issue 5, May 2014, pp. 590-592. https://www.researchgate.net/publication/262224020

[28]. Weng Hong Lim, Yuen Kiat Yap, Wu Yi Chong, Harith Ahmad, All-Optical Graphene Oxide Humidity Sensors, *Sensors,* Vol. 14, 2014, pp. 24329-24337. https://pdfs.semanticscholar.org/bec0/9c6cdbf67fde08d27560cc7a64842cf6089c.pdf

[29]. Hengchang Bi, Kuibo Yin, Xiao Xie, Jing Ji, Shu Wan, Litao Sun, Mauricio Terrones, Mildred S. Dresselhaus, Ultrahigh humidity sensitivity of graphene oxide, *Scientific Reports*, Vol. 3, 2013, pp. 1-7. https://www.nature.com/articles/srep02714

[30]. Sourav Ghosh, Ruma Ghosh, Prasanta Kumar Guha, Tarun Kanti Bhattacharyya, Member, Humidity Sensor Based on High Proton Conductivity of Graphene Oxide, *IEEE Transactions on Nanotechnology*, Vol. 14, Issue 5, 2015, pp. 931-937. https://www.researchgate.net/publication/281521372_Humidity_Sensor_Based_on_High_Proton_Conductivity_of_Graphene_Oxide

[31]. Daniel Hernández-Rivera, Grissel Rodríguez-Roldán, Rodrigo Mora-Martínez, Ernesto Suaste-Gómez, A Capacitive Humidity Sensor Based on an Electrospun PVDF/Graphene Membrane, *Sensors,* Vol. 17, Issue 1009, 2017, pp. 2-11. www.mdpi.com/journal/sensors

[32]. T. K. Kuanyshbekov, A. M. Ilyin, G. W. Beall, N. R. Guseinov, M. A. Tulegenova, Sensitive humidity sensor based on functionalized graphene, in *Proceedings of the 4th International Conference on Sensors Engineering and Electronics Instrumentation Advances (SEIA'18),* Amsterdam, The Netherlands, 2018, pp. 45-46.

[33]. Jinxia Ma, Dan Ping, Xinfa Dong, Recent Developments of Graphene Oxide-Based Membranes: A Review, *Membranes,* 2017, Vol. 7, Issue 3, 52, 2017, P.29. www.mdpi.com/journal/membranes

[34]. Piner R. D., Ruoff R. S., Stankovich S., Zimney E. J., Evmenenko G., Dikin D. A., Dommett G. H. B., Nguyen S. T., Preparation and characterization of

graphene oxide paper, *Nature*, Vol. 448, Issue 7152, 2007, pp. 457–460.
[35]. https://pdfs.semanticscholar.org/2803/0c759adac30dfc0f8f494c0ab815bca18855.pdf
[36]. Ayrat M. Dimiev, Siegfried Eigler, Graphene Oxide: Fundamentals and Applications, *John Wiley & Sons*, 2017, p. 454.
[37]. Bae S., Kim H., Lee Y., *et al*., Roll-to-roll production of 30-inch graphene films for transparent electrodes, *Nature Nanotechnol*, Vol. 5, Issue 8, 2010, pp. 574–578.
[38]. Ni Zh, Y. Wang, T. Yu, Z. Shen, Raman Spectroscopy and Imaging of Graphene, *Journal of Nano Review*, Vol. 1, Issue 4, 2008, pp. 273-291.
[39]. Kumar Challa S. S. R., Raman Spectroscopy for Nanomaterials Characterization, *Springer*, 2012.
[40]. W. Gao, Graphene Oxide, *Springer International Publishing Switzerland*, 2015, p. 154.
[41]. Ferrari A. C., Basko D. M., Raman spectroscopy as a versatile tool for studying the properties of graphene, *Nature Nanotechnol*., Vol. 8, Issue 4, 2013, pp. 235–246.
[42]. Eigler S., Hof F., Enzelberger-Heim M., *et al*., Statistical-Raman-microscopy and atomic-force-microscopy on heterogeneous graphene obtained after reduction of graphene oxide, *J. Phys. Chem. C*, Vol. 118, Issue 14, 2014, pp. 7698–7704.
[43]. https://www.indiamart.com/dinstech/xrd-and-xrf.html
[44]. Thirunavukkarasu Somanathan, Karthika Prasad, Kostya (Ken) Ostrikov, Arumugam Saravanan, Vemula Mohana Krishna, Graphene Oxide Synthesis from Agro Waste, *Nanomaterials*, Vol. 5, Issue 2, 2015, pp. 826-834.
[45]. Anderson J. H., Parks G. A., The Electrical Conductivity of Silica Gel in the Presence of Adsorbed Water, *The Journal of Physical Chemistry*, Vol. 72, Issue 10, 1968, pp. 3662-3668. http://dx.doi.org/10.1021/j100856a051