Investigation of Structural, Photo-luminescence and Self-Cleaning Properties of Thin Layers of GO-ZnO and GO, GO-Ag and Composite Bilayer of GO-ZnO/GO-Ag Prepared by Spray Pyrolysis Method

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Abstract

In this research, the synthesis of graphene oxide-zinc oxide nano-composite, graphene oxidesilver nano-composite, graphene oxide thin layers, and graphene oxide-zinc/graphene oxidesilver composite bilayer was done using spray pyrolysis. Characterization of the synthesized layers was done by XRD, TEM, AFM, PL, FTIR and BET analyses. Based on the TEM images, the given nano-composites are formed and graphene oxide can be a suitable platform for the growth of silver and zinc oxide nano-particles and prevents their accumulation. According to the AFM images, GO-ZnO/GO-Ag sample has the lowest roughness. PL spectrum showed a broad emission peak for GO-Ag thin layer at a wavelength of approximately 550 nm, which is consistent with the reported band gap of 3.6eV for this nanocomposite. From BET results, the surface area was obtained 4 m²g⁻¹ and 14 m²g⁻¹, for GO and GO-Ag respectively which were greater than the similar work. The pore diameter of GO-ZnO was obtained equal to 16.5 nm, indicating the superiority of the meso-holes in GO-ZnO. Also, the surface area of GO-ZnO/GO-Ag bilayer was around 3.6 times larger than the surface area of ZnO. The contact angles of droplet with the surface in GO, GO-Ag, GO-ZnO, GO-ZnO/GO-Ag samples are 55.02, 60.24, 31.28, 56.35, respectively.

Keywords: BET Analysis, Bilayers, GO-Ag Nano-Composite, GO-ZnO Nano-Composite and Self-Cleaning Properties.

1. Introduction

By definition, graphene oxide (GO) consists of a cluster of passive groups of oxygen functional which makes it a strong candidate for using in many applications, through chemical application. GO is made of graphite oxide and has been a desired method for large-scale graphene production [1-3]. However, Graphene has a significant scattering in water and reduces its surface area and its application is thus limited. The limitation is because the density caused by the strong π - π and van der Waals interactions accumulation of sheets of graphene [4]. Graphene oxide (GO), as a single-atom-thick sheet made of oxygenated hydrophilic functional groups in form of ether, hydroxyl, carboxyl, and epoxy, has been considered in the different scientific areas [5-6]. Several intrinsic properties of GO nanoplates such as their smoothness, thickness at the atomic level, high slip length of water, and low cost of mass production through chemical oxidation of graphite provide the new potential applications (programs) in water treatment [7-9]. In addition, special efforts have been made to improve the use of GO such as membrane durability by preventing the connection of hydrophobic hooks or the formation of a protective layer against chlorine attack. Recently, self-cleaning technology has rapidly started, because the windows and cleaning coatings have high commercial call. The materials with self-cleaning capability should have the good photocatalytic activity with optical transparency for the coating surface, high durability, and super-hydrophilic properties [5]. The super-hydrophilic property is a state in which materials are more dependent on water than non-polar air, which is a critical property that is required for a self-cleaning coating [10].

The application of composite materials is widely used in the fields of textile, chemical, energy, defense industry, electronics, and biomedical engineering which is growing and developing on human health. Muflikhun et al. showed that several sizes of nanocomposites can be produced by varying the temperature and baking time by developing a statistical relationship of the diameter (length), temperature and baking time of synthesized Silver-Titanium Dioxide nanocomposites using the Horizontal Vapor Phase Growth (HVPG) technique [11]. Also, nano Silver- Graphene composites were successfully manufactured by HVPG method [12]. GO nano-sheets are used to stabilize the Ag nano-particles and to strengthen the relationship between Ag and bacteria which results in a synergistic effect for these new nano-composites. Although some studies suggest a specific core-shell structure [13] or nano-tube structure [14] based on the different silver salts and chemical reduction methods, most morphologies of GO-Ag nano-composite offer a decoration (arrangement) of good distribution of silver. ZnO is a well-known semiconductor with the band gap energy of 3.2–3.3eV. As an efficient photocatalyst in the degradation of refractory organic pollutants in the environment, it has been extensively studied [15-17]. The transparency properties of graphene and a good photocatalyst of graphene-semiconductor systems can be completely applied in the self-cleaning technique. Most of the works so far reported on graphenesemiconductor photocatalysts have been based on the system of powder composite. The thin layer as a very prominent form for the practical application of self-cleaning is not wellstudied. The self-cleaning applications of Ag and ZnO thin layers based on graphene oxide as well as this layer on each other have not so far been reported [18-20].

In this work, we used the spray pyrolysis deposition method to firstly synthesize the GO-Ag, GO, GO-ZnO thin layers and GO-ZnO/GO-Ag bilayer. Morphological characterizations using TEM and AFM were then performed on them. FTIR analyses were performed to identify the formed bonds and PL to characterize the photo-luminescence. We performed the Brunauer-Emmett-Teller (BET) surface area analysis in order to identify the porosity in the samples. Finally, angle test was performed in the samples for self-cleaning characterizations.

2. Preparation Method

2.1. Preparation of GO-Ag Nano-Composites

Graphene oxide colloid (Borhan Nano Scale Company, Iran) with 1mg/ml concentration was dispersed by the ultrasonic device. We combined the ammonia, deionized water and silver nitrate to prepare 40 M silver diamine hydroxide solution. We mixed the GO solution and Ag (NH₃) OH solution (volume ratio 2/8) and stirred for 30 minutes to prepare GO-Ag nano-composite. We stirred the solution for half an hour in the oil bath at 70-80 ° C, and we then washed the final solution with deionized water to remove the additional materials.

2.2. Preparation of GO-ZnO Nano-Composite

For separation of the layers, GO at a concentration of 1mg/mL was dispersed in the ultrasonic for 30 minutes. 0.2 M hexamethylenetetramine (HMTA) solution and 0.2 M zinc acetate solution were then prepared and added to the GO solution, respectively, and mixed and stirred

for 15 minutes. In order to form nano-composite, we put the final solution in an oil bath at 90 °C for 3 hours, and then washed with deionized water 3 times to remove solvents and prepared for spray.

2.3. Preparation of Thin Layers by Spray Pyrolysis Method

The glass substrates were first washed using acetone and then placed in the ultrasonic for 30 minutes. In order to prepare the thin layers by spray pyrolysis method, 35 ml of each of GO-Ag, GO-ZnO solutions were sprayed using compressed air as carrier gas. Nozzle distance should be set at about 30-40 cm and we selected the deposition rate of 2-2.5. We set the plate temperature of 400 °C and 150 °C for GO-ZnO and GO-Ag thin layers, respectively. For preparation of GO-ZnO/GO-Ag bilayer, at first, GO-ZnO solution was sprayed on glass substrate at 400°C, and then the GO-ZnO thin film was allowed to reach room temperature. Afterward, the GO:Ag solution was sprayed with the same conditions as before on the GO-ZnO thin film at 150°C. Figure 1 shows the schematic diagrams of the spray pyrolysis technique and the GO-ZnO/GO-Ag bilayer.

3. Characterization of Samples

In order to study the morphology and grain size of the samples, TEM images were taken by the EM 208S model of transmission electron microscopy (TEM) device (100KV). We used the Ara-research Company manufactured atomic force microscopes (AFM) to investigate the topology of the films' surface. In order to investigate the chemical bonds in the obtained films, we used the Nicolet AVATAR 370 equipment model of Fourier transform infrared spectrometer within the range of 400-4000 C/m. For measuring the porosity rate and effective surfaces, BET (BEL model BELSORP MINI II) device was used based on the measurement of desorbed by the material surface and the volume of nitrogen gas adsorbed at constant temperature of liquid nitrogen (-77°C). We set the pressure range of our device within the range of 0 to 85kPa. We performed the degassing process of the samples at 200°C for 4 hours. Using the PL device, Perkin Elmer LS 45 model, we examined the PL properties. In order to measure the self-cleaning properties of the samples, we performed the measurement of the angle of contact of water droplets on thin layers under ambient conditions at 25°C. The water droplets were positioned in three different locations for each sample and its mean value was adopted as the contact angle.

4. The results and conclusive discussion 4.1.TEM Images

The results of the X-ray analysis, which confirms the formation of the desired structures, were reported in our previous article [21]. Figure 2a shows the TEM image of pure graphene oxide sample with a magnification of 200 nm. In this Figure, the planar structure of graphene oxide has a broad, wrinkled surface, indicating that GO used in the research is of good quality. Figure 2b shows the TEM image of graphene oxide-silver nano-composite and also shows that Ag nano-particles are well scattered and placed on GO surface. These nano-particles are very small (10-20 nm) and spherical. According to GO image, they can be used as a suitable

platform to grow the Ag nano-particles and prevent their accumulation. Figure 2c shows that the transmission electron microscope image of GO-ZnO nano-composite with a magnification of 100 nm, and provides the direct evidence of the formation of GO-ZnO nano-composites and shows the spherical nano-particles of ZnO scattered on the graphene oxide surface.

4.2. Atomic Force Microscope

Atomic force microscope was used to investigate the topography of GO-Ag, GO-ZnO, GO, GO-ZnO/GO-Ag thin films. 2D and 3D AFM images of thin layers are reported in Figure 3 which shows the graphene oxide plate-shave coated the surface, and nano-particles of Ag and ZnO in the form of prominent peaks on the surface are randomly scattered. GO-Ag, GO-ZnO nano-composites have the heterogeneous morphology but graphene oxide thin layer has the relatively homogeneous morphology. The lateral dimensions of GO plates are about a few micro-meters. By determining the deviation of elevation between peaks and troughs in a particular region, the Root Mean Square Roughness (RMS) is obtained. Based on the results of AFM, values of RMS for GO-ZnO, GO-Ag, GO, GO-ZnO/GO-Ag thin layers were 185.78, 182.8, 131.9 and 115.81 respectively. Thus, the GO-ZnO and GO-ZnO/GO-Ag samples had the highest and the lowest surface roughness, respectively. The data demonstrated here is obtained by Imager software. The numbers seen in this figure are related to the amplitude which, as seen in the 2D and 3D figures, is reported in the direction of the X-axis and the Y-axis based on microns unit. However, in the axis direction Z is reported in mV, which is related to the signals that the device receives from the sample.

4.3. FTIR Analysis

FTIR measurements were used to investigate the interactions of the bonds in GO, GO-Ag, GO-ZnO and GO-ZnO/GO-Ag bilayer and are clarified in Figure 4. As seen from the spectrum of GO, there are bands due to C-O stretching vibrations (1055 cm⁻¹), C=C bond (1619 cm⁻¹), C=O stretching of -COOH groups (1733 cm⁻¹) and O-H stretching vibration (3385 cm⁻¹). The band at 1081 cm⁻¹ confirms the existence of oxidized functional groups after the process of oxidation. The water adsorbed in GO shows the O-H bond of H₂O molecules with a broad peak between 3000 cm⁻¹ to 3700 cm⁻¹ centered at 3385 cm⁻¹ [22]. The fact is confirmed that graphene oxide is a very absorbent material because its ability to become a gel-like solution is confirmed. In the GO-Ag spectrum, the peak at 3390 cm⁻¹ can be assigned to the absorbed water due the stretching vibration of O-H group of carboxyl, alcohol and absorbed water molecules. The broad band between 3000–3700 cm⁻¹ can be attributed to the oxygen groups, mainly to the hydroxyl groups located both on the surface and at the edge of GO sheets. The peaks at 1374, 1602 and 1050 cm⁻¹ can be assigned to the O-H bending vibration, C=C bond and the C–O stretching vibration of alkoxy group, respectively. The peak noticed at 1733 cm⁻¹ in the GO spectrum almost disappeared in the GO-Ag spectrum. Moreover, the intense peak at 1050 cm⁻¹ in the GO-Ag spectrum may indicate a strong interaction between the Ag+ ions and the functional groups on the GO sheets. In GO-ZnO layer spectra, the adsorption peak at 3429 cm⁻¹, which is connected with the tensile vibrations of O-H groups on GO plates and the water adsorbed on GO surface, severely decreased in GO-ZnO composite spectrum, possibly due to the reaction between GO and ZnO nanoparticles. This result may indicate that ZnO particles have been successfully placed on GO sheets. The absorption band at around 1105 cm⁻¹ corresponds to C–O stretching mode. In GO-ZnO spectrum, it shows a sharp peak at about 500 Cm⁻¹ of metallic vibrations. In GO-ZnO/GO-Ag bilayer, the peak centered at 3320 cm⁻¹, which is related to the C-H groups and the peak at 1074 cm⁻¹ corresponds to the stretching vibrations of C=O [23,24].

4.4. Photo-luminescence

The photo-luminescence spectra (PL) for GO, GO-ZnO, GO-Ag, layers and GO-ZnO/GO-AG bilayer thin films at room temperature are displayed in Figure 5. Photo-luminescence spectrum in amorphous carbon stems from the radiative recombination of holes and electrons in the band sequence states made by clusters of sp^2 . However, compared to very disordered sp^3 matrix, clusters of sp^2 in GO are rather small, which may cause fluctuation in the local band gap, severely. Therefore, clusters of sp^2 are embedded with a narrower gap in the matrix sp^3 , which serves as a tunnel and acts as a barrier between clusters, causing the blue shift in photo-luminescence spectrum. In Figure 5, PL spectrum shows a broad emission peak for GO-Ag thin layer at a wavelength of approximately 575 nm. GO thin layer sample exhibits less emission peak intensity than other samples. A spectrum like this blue emission spectrum of chemical graphene oxide has been observed by the previous authors [25,26]. According to PL peak in Figure 5, the intensity of GO-ZnO composite sample is much higher than that of graphene oxide. It has also been shown that incorporation of ZnO in GO can enhance the separation of the optically excited holes and electrons. Therefore, given this intensity, the combination of ZnO and graphene oxide can quench the recombination process significantly. According to the Figure, the peak emission intensity for GO-ZnO/GO-Ag bilayer is higher than all samples.

4.5. BET Analysis

The measurement of surface area, volume and pore distribution have many applications in studying the catalysts, active carbon, pharmaceutical materials. Hence, different methods have been considered to measure the surface area and porosity, which can be referred to the adsorption-based methods. If a solid material is impermeable and has a perfectly stable shape, its total surface area can be approximately measured. But for porous-structured samples, it is difficult to determine the porosity rate as well as the total surface area. The total surface area of porous sample can be measured by BET method which this analysis is one of the most important and accurate method, on the adsorption of some specific molecular species on their surface in the gas state.

The adsorption and desorption isotherms of nitrogen for GO-ZnO, GO-Ag, and GO samples are displayed in Figure 6. The surface area of oxidized graphene sample is significantly lower $(4m^2g^{-1})$ in this work. The mentioned inconsistency can be elaborated using the fact that an increase in BET surface area is connected with the worsening of graphene layer ordering.

The adsorption and desorption isotherm of the adsorbed particles for GO-Ag sample shows in Figure 6, in which gap pores and multilayered structure are seen, and the hysteresis loop of this sample and the graphene oxide sample are approximately similar. BET specific surface area for GO-Ag was calculated compared to graphene oxide, with an increased value of 14m²g¹. According to IUPAC classification, Figure 6 shows the adsorption-desorption isotherm of nitrogen with distinct hysteresis loops that can be attributed to type IV. This behavior indicates that the meso-holes are dominant. In BET analysis, the specific surface area of 10.27m²/g was calculated. The pore diameter shows 16.5 nm with a wide distribution of meso-holes. This behavior also confirms the superiority of the meso-holes in GO-ZnO. The specific surface area of the GO-ZnO/GO-Ag Bilayer nanocomposite was identified by the BET method with N₂ adsorption-desorption, is shown in Figure 6. The measured surface area of GO-ZnO/GO-Ag Bilayer was around 3.6 times bigger than ZnO. By presence of GO-ZnO/GO-Ag in a dark condition, pollutant adsorption is basically contributed by the increased specific surface area (SBET). Graphene, in general, has a very high specific surface area [27]. It could therefore provide a high adsorption capacity. The oxidized form of graphene (i.e., GO), has oxygen functional groups on its surface which can become the sites of adsorption. Therefore, the enhanced degradation capacity under visible light can be attributed to the adsorption power of GO combined as a semiconductor or adsorption substrate [27-29]. In addition, the increased pore size of GO-ZnO/GO-Ag nanocomposite could lead to the increases in adsorption efficiency.

4.6. Contact Angle

The surface is called hydrophile when the angle of contact at the interface of phases of liquid, gas, and solid was less than 90°, the hydrophilic surfaces energy is very high. Albeit, the surface is called hydrophobe, when the angle of contact at the contact location of the solid surface with the liquid droplet is increase more than 90°, the hydrophobic surfaces energy is very low. In this work, the contact angle of GO-ZnO, GO, GO-Ag, GO-ZnO/GO-Ag thin layers was examined using Taiwan, Dino-Lite, AM-7013MZT. Taking image of a 2 µl droplet with a magnification of 50 was done at 20°C and the results are displayed in Figure 7. The contact angles of droplet with the surface in GO, GO-Ag, GO-ZnO, GO-ZnO/GO-Ag samples are 55.02, 60.24, 31.28, 56.35, respectively. The type and amount of impurity, surface homogeneity and roughness of surface are important factors that affect to the contact angle. The surface roughness is one of the highly important factors in the angle of contact, which affects the interaction between the liquid and the surface. It is well understood that by increasing roughness, the hydrophilicity of hydrophilic surfaces generally increases as well as the hydrophobicity of hydrophobic surfaces. For hydrophilic surfaces, when the contact angle is less than 90°, the contact angle gradually decreases as the surface roughness increases, and the hydrophilicity becomes stronger. As graphene oxide is in fact hydrophile, AFM images show that its surface roughness is also relatively large so the angle of contact of its thin layer is equal to 55.02° (i.e., less than 90°), showing the surface hydrophilicity of the layer. According to AFM image, GO-ZnO sample has the minimum contact angle and thus a relatively good hydrophilic surface and the highest roughness. GO-Ag sample has the highest surface roughness and therefore the lowest angle of contact.

5. Conclusion

In this paper, graphene oxide-silver nano-composite, graphene oxide thin layers, graphene oxide-zinc/graphene oxide-silver and graphene oxide-zinc nano-composite bilayer were deposited using the method of spray pyrolysis. According to TEM images, the given nanocomposites are formed and graphene oxide can be a suitable platform for the growth of silver and zinc oxide nano-particles and prevents their accumulation. PL spectrum shows a broad emission peak for GO-Ag thin layer at a wavelength of approximately 550 nm, which is consistent with the reported band gap of 3.6eV for this nano-composite. Analysis of PL results also shows that incorporation of ZnO in GO can enhance the separation of the optically excited holes and electrons. Thus, the intensity of the recombination process can be significantly quenched by combining the ZnO and graphene oxide. The analysis of BET results shows that the graphical structure of the surface area of GO sample is significantly lower $(4m^2g^{-1})$ in this work. BET specific surface area for GO-Ag was calculated compared to graphene oxide, with an increased value of 14m²g¹. BET analysis of GO-ZnO sample shows that the pore diameter is 16.5 nm, with a wide distribution of meso-holes, indicating the superiority of the meso-holes in GO-ZnO sample. The measured surface area of GO-ZnO/GO-Ag Bilayer was around 3.6 times larger than the surface area of ZnO. Based on AFM results, values of RMS for GO-ZnO, GO-Ag, GO, GO- ZnO/GO-Ag thin layers were 185.78, 18.28, 131.9, 115.81, respectively. Thus, the GO-ZnO and GO-ZnO/GO-Ag samples had the highest and the lowest surface roughness, respectively. The investigation of the angle of contact between the surface of the synthesized thin layers and the water droplet shows that GO-ZnO sample has a relatively good hydrophilic surface and the lowest contact angle.

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- Figure 1. The schematic diagram of the: (a) spray pyrolysis technique and (b) GO-ZnO/GO-Ag bilayer
- Figure 2. TEM Images of the sample: (a) Pure GO, (b) GO-Ag Nanocomposite, (c) GO-ZnO Nanocomposite.
- Figure 3. 2D and 3D AFM Images of Thin Layers a) GO-Ag, b) GO-ZnO, c) GO-ZnO/GO-Ag, d) GO Prepared by Spray Pyrolysis Method
- Figure 4. FTIR Spectrum of GO, GO-Ag, GO-ZnO Layers and GO- ZnO /GO-Ag Bilayer
- Figure 5: Photoluminescence (PL) Spectrum for GO-ZnO, GO-Ag Layers, GO-ZnO/GO-Ag Bilayers and GO Thin Layer at Room Temperature
- Figure 6. Adsorption and Desorption Isotherms of Nitrogen for GO, GO-ZnO and GO-Ag Samples and GO-ZnO/GO-Ag Bilayer nanocomposite
- Figure 7: Contact Angle of GO, GO-Ag, GO-ZnO, GO-ZnO/GO-Ag thin Layers Prepared by Spray Pyrolysis Method.





GO-Ag
GO-ZnO
Glass

Figure 1.







Figure 3.



Figure 4.



Figure 5.



Figure 6.



Figure 7.