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Enhance the Optical Properties of the Synthesis PEG/Graphene- Based Nanocomposite films using GO nanosheets

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Abstract. Graphene is one of the most interesting and attracting nanofillers. The investigation focused on the effect of two significant factors using graphene nanosheets and polymer molecular weights (Mw) on the optical properties of polymer graphene-based nanocomposites. New sonication-mixing-aquatic methods were applied using the three Mw, 4k, 8k and 20k of polyethylene glycol (PEG), as a polymer model, with low loading ratio graphene oxide nanosheets (GONSs) to synthesise the nanocomposites. Fine distribution and good homogeneity of GONSs were successfully presented in the PEG matrix as examined applying the optical microscope (OM). The results presented an enhance in the most optical properties, which shows significantly in the ultraviolet region (~300 nm in wavelength), such as, absorbance, absorption coefficient, real and imaginary dielectric constants up to 71%, 355%, 37% and 41% after increasing the Mw, except the allowed and forbidden indirect optical energy gap were reduced to 18% and 29%, respectively. Moreover, the contribution of GO with Mw of PEG exhibited a notable improvement of the optical properties up to 100%, 440%, 48% and 61%, whereas the allowed and forbidden indirect optical energy gap were reduced to 43% and 86%. These results illustrated significant roles of the effect of MW and GO in the optical properties that give rise to better photovoltaic performances of heterojunction solar cells and may use as filters and antireflection coating in the substantial applications.

Keywords: graphene, PEG, nanocomposite, optical properties, Molecular weight, films.

1. Introduction

Polymer nanocomposites consist of a polymeric substance and a nanoscale material. These materials show considerable improvements in different properties; mechanical, optical, thermal stability and chemical resistance etc. The enhancement of the properties with the addition of nanoparticles can be achieved due to different factors, one of the most important of them is a respectable interfacial interaction between the nanoparticles and the matrix with good dispersion of particles within the matrix (1,2). Poly (ethylene glycol) (PEG) has a flexible structure of C-O-C bonds and an essential type with of thermoplastic polymer, moreover PEG is considered a solubility in water and organic solvents and

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hydrophilicity high crystallinity with self-lubricating property. Therefore, PEG is one of the most prefer polymer for the growing and development of various significant applications as a solid polymer electrolytes such as, sensors and batteries (3,4).

Many of previous studies focused on using the nanoparticle, clay, polymer, graphene and other to improve the properties of the polymers, especially, the reinforcement the mechanical properties literature (4–8) these studies were barely studied. However, the effect of molecular weight and substantial band gap alteration of GO on the optical properties for instance (the energy band gap) of the PEG as nanocomposites films has not been reported to date (9,10).

Graphene oxide is considered as one of the best a prospective nanofillers because of their feature in many significant optoelectronics applications for instance, organic solar cells(11), luminance organic light-emitting diodes (12), flexible transparent electronics (13) and chemical sensors (14). Additionally, GO have high mechanical strength (15–17), significant conductivity (18) and still have high transparency (19). GO was synthesized by Hummers methods (20) since the early nineteenth century. This method is considered the most interest one among other purification methods to prepared GO due to the more reliable, much produced of GO and lower cost than other methods (21). GO surface and structure are abundant with various functional groups such as hydroxyl, carbonyl, carboxyl and epoxy groups. The presence of these groups turn the GO nanosheets to be hydrophilic (22), able to distribute with more homogenous in the polymer nanocomposites (4,23) and strongly interact in the matrix with the polymer (24). GO has been chosen manipulate the structural to improve the optical properties of PEG polymer and use in the significant applications (21,25).

Although it is displayed fluorescence and water-soluble, from all these properties of GO that present it as better attractive nanosheets compared to graphene for different application such as, optoelectronics (26,27). These fabrication methods result in a range of optical and electronic properties of GO due to the procedure of synthetic. Meanwhile, other feature of these abundant function group on the surface of GO give promise for modification of GO especially, their optical properties (10). The band gap of GO is able to tune that is strongly desired in devices that use in the energy storage/conversion technique (10). Where controlling the performance of solar cells in devices using the electronic gap varying of GO-based active nanosheets that give high power conversion efficiencies and mainly over the short circuit current as well as the open circuit voltage (28). Moreover, these features of GO may use ultrafast lasers as a saturable absorber (26) and in mid-Infrared irradiation range photodetectors (29).

Researchers (10) utilized and tuned the band gap of GO as an aqueous solution that significantly enhance its applications, such as ozone treatment. Accordingly, investigating the optical absorption and energy band gap (Eg), as the optical properties in particular, of polymer-graphene based nanocomposites bring a huge interest for researchers those try to understand how these properties can be changed with the polymer composition changes to open new way for novel practical applications (10,28). In spite of the previously reported methods, it is still difficult to control because of their complexity (10).

According to available literature, this work was characterized the process of preparing nanocomposite of PEGs-GONS in the form of a film for the first time. GO could utilize to control and tune the band gap of GO, which may significantly enhance its applications such as, optoelectronics. Therefore, in this study, to reduce the gape of knowledge in this area and improve the optical absorption (in particular, the energy band gap) of PEG polymer were the main purpose to identify the effect of different molecular weight and the addition of low loading ration GONSs on the optical properties of PEG4000, PEG8000 and PEG20000 films prepared using sonication-mixing acoustic methods. The pure and nanocomposites films thicknesses were between 5-10 μ m.

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2. Experimental part

2.1. Materials

PEG with three different molecular weights (4k, 8k and 20k) g- mole⁻¹ were supplied from (Central Drug House, Ltd, India) Company. graphite powder (40μ m), sulfuric acid (H₂SO₄) (analytical grade, 99.8%), potassium permanganate (KMnO₄), sodium nitrate (NaNO₃), hydrogen peroxide and hydrochloric acid (HCl, 35%) were obtained from Sigma-Aldrich Company, UK.

2.2. Method

2.2.1. Purification of Graphene Oxide

GONSs were synthesis by our group and full characterizations were available in our previous literature (4).

2.2.2 Purification of the nanocomposites

GONSs was dispersed in distilled water with 1 wt% using stirring-sonication bath method to get fine dispersion of GO in the distilled water. PEGs were dissolved in distilled water with 99 wt% using magnetic stirrer, then GONSs was added to PEGs to synthesis the nanocomposites using mixing-sonication methods, where the samples were mixed using magnetite stirrer for 60 minutes then sonicated for 30 minutes, followed by another mixing for 120 minutes then sonicated for 30 minutes, then also mixture was mixed for 180 minutes and final sonication was 30 minutes for fine dispersion and homogeneous of GONSs in the polymer matrix. Finally, these nanocomposites mixtures were mixed for 12 h, then the samples were pleased on a glass slide and petri dish. PEGs without GONSs were fellow the same procedure to prepare PEGs films. The method summarized in Table 1.

Sample ID	PEG MW	PEG con. wt%	GO con. wt%	Total Mixing time during synthesis	Total Sonication time during synthesis	Final mixing time	Dry method
GONSs	0	0	100	-	0.5 h	3 h	Freeze drier
PEG1	4k	100	0	-	-	3 h	Under air
PEG1-GO	4k	99	1	6 h	1.5 h	12 h	
PEG2	8k	100	0	-	-	3 h	
PEG2-GO	8k	99	1	6 h	1.5 h	12 h	
PEG3	20k	100	0	-	-	3 h	
PEG3-GO	20k	99	1	6 h	1.5 h	12 h	

Table 1. Summarized the purification of GONSs, PEGs and nanocomposites films.

2.2.3 Characterization

The surface images of the polymers and its nanocomposite films procured by Optical Microscope (OM), supplied from Olympus (Nikon - 73346) with light intensity automatic controlled camera under magnification. The UV-Visible spectrophotometer (Shimadzu UV- 1650 PC) made by Phillips, (Japanese company), that used to record the transmission and absorption spectra in the wavelength range (300-1100) nm at room temperature.

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The transmittance spectra (T) was calculated from the following relation:

$$T = \log A \tag{1}$$

The absorption coefficient () (cm)⁻¹ was considered from Lambert formula:

$$= \ln (1/T) / t$$
 (2)

Where t means the thickness. Both the allowed and forbidden indirect optical energy gap for PEG and PEG/GO nanocomposite films are estimated from the intercept of the extrapolated linear part of the curve against the photon energy (h) at $(h)^n = 0$, as shown in Figures (5), and (6) respectively, using the following equation:

$$h\upsilon = B (h\upsilon - E_g^{opt} \pm Eph.)^r$$
(3)

Eph. means the energy of phonon, (-) means when phonon absorption and (+) means when phonon mission, r means the exponential constant; its value depends on the type of transition, r=2 for the allowed indirect transition, meanwhile r=3 for the forbidden indirect transition.

The refractive index (n) and extinction coefficient (k) of the film are calculated from the equations(31):

$$n = \frac{1+R}{1-R} + \left[\frac{4R}{(1-R)^2} - K^2\right]^{1/2}$$
(4)
$$K = \frac{\Gamma}{4f}$$
(5)

where R means the reflectance, and is the wavelength of Cu K line (1.5406).

Real and imaginary dielectric constant (r and i) for pure PEG and PEG-GO nanocomposite films are calculated from the equations (31):

$$_{r} = (n^{2} - k_{\emptyset}^{2}) \tag{6}$$

 $_{i} = (2nk_{o}) \tag{7}$

4. Results and discussion

The OM images of the surface of PEG1, PEG2 and PEG3 with the nanocomposite films at magnification power 40X were shown in Figure (1). It demented good homogeneity and fine distribution of GONSs inside the PEGs matrix, which is evident with increased molecular weight. This presented a successful preparation method that forms suitable conditions and used to prepare these PEG-GO nanocomposites. Compared PEGs films with nanocomposite films that noted a notable change with lower addition ratio of the GONSs led to many difference among these films without aggregations or loss of transparency. Where, PEGs exhibited clear pristine PEGs of all samples even with a different molecular weight as shown in Figure (1- a, c and e). Meanwhile, the nanocomposites presented clear difference behaviour after the contribution of GO. The PEG1-GO showed less dispersion of GO in the polymer matrix with a bright area for PEG as shown in Figure (1 b), whereas, in figure (1 d), this issue was overcome with increasing the PEG that exhibited a good network of the

PEG2 with GO. This network was considerably improved with increasing the PEG molecular weight especially the higher molecular weight of PEG3, where the sample Figure (1 f) illustrated significant and great network and line as that may present as significant tunnels to move the electrons through the polymer matrix and without loss of transparency in agreement with the literature (5,25).





(b)

(a)



(c)

(**d**)



(e)

(**f**)

Figure (1): Optical microscopy images with (40X) of (a) PEG1, (b) PEG1-GO, (c) PEG2, (d) PEG2-GO, (e) PEG3 and (f) PEG3-GO.

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The absorbance spectra for PEG1, PEG2, PEG3 and PEG-GO nanocomposite films in the wavelength range 300-1100 nm were recorded at room temperature using a double-beam spectrophotometer. Figure (2) shows the variation of optical absorbance spectrum versus wavelength of incident light for PEGs and PEG-GO nanocomposite films. It clear noticed from the figure that the absorbance of all films had a high values at a wavelength in the ultraviolet region of the electromagnetic spectrum (About 300 nm in wavelength), then the absorbance decreases with the increase of wavelength. The absorbance values increased with the increase of molecular weight that showed an important role to effect on the absorption. The highest absorption value for PEG3 appears in the ultraviolet region at 0.63, meanwhile, the contribution of the GONSs presented another significant enhancement up to 0.80for PEG3-GONSs nanocomposite film. Generally, the result presented enhance in the absorbance value up to 71% after increasing the Mw, and 100% with the contribution of GONSs. The nanocomposite samples exhibited a notable increase in the results compared to all the PEGs films because the electrons outside orbits can absorb the electromagnetic energy of the incident light and travel to higher energy levels (32). This procedure is not accompanied by the emission of radiation because the travelled electron to higher levels had occupied vacant positions of energy bands, thus part of the incident light was absorbed by the substance and does not penetrate through it. Indeed, higher molecular weights are expected to increase charge carrier mobility's and therefore give rise to better photovoltaic performances of heterojunction solar cells, and may use as filters and antireflection coating (33,34). These finding match the other finding in the literature (10), which found that the highest absorbance value appears in the ultraviolet region (About 228 nm in wavelength) of the electromagnetic. This is to be achieved through our work with PEG3-GONS nanocomposite film. Meanwhile, the ability to tune the band gap of GO is highly desired in energy storage-conversion devices for instance, in solar cells, varying the electronic gap of GO-based active material that require high power conversion efficiencies will allow controlling the performance of the device, mostly over the open circuit voltage and short circuit current (10).



Figure (2): The absorbance spectra versus wavelength of PEGs and PEGs/GO nanocomposite films.

Figure (3) illustrations the variation of optical transmittance spectra versus wavelength of incident light for three molecular weight of PEG polymers and its nanocomposite films with GONSs. For all samples in the lower wavelengths region (ultraviolet region), the transmittance strongly increased decreased and then slowing gradually with the increase of wavelength, except PEG1 and its nanocomposite, it stays

constant at the highest wavelengths. The optimum value of transmittance is about 98.5% for PEG1 film and about 96% for its nanocomposite PEG1-GO at high wavelengths (VIS-NIR region). The decreased in the optical transmittance was caused by the increase of the molecular weight and GONSs as nanofillers.



Figure (3): The transmittance spectrum versus wavelength of PEGs and PEGs/GO nanocomposite films.

The obtained results of absorption coefficient $(cm)^{-1}$ versus photon energy for PEGs and its nanocomposite films were presented in Figure (4). It clear to see that was the smallest at low energy, which means the little possibility of electron transition. Because of the energy of the incident photon was not satisfactory to transfer the electron from the valence to the conduction band, whereas at high energies the absorption of the electron is excellent. The results of the absorption coefficient were less than (10^4 cm^{-1}) which means there was a large probability of the indirect transition.



Figure (4): The absorption coefficient (cm)⁻¹ versus photon energy for PEGs and PEGs/GO nanocomposite films.

Both the allowed and forbidden indirect optical energy gap for PEGs and PEGs-GO nanocomposite films were estimated from the intercept of the extrapolated linear part of the curve against the photon

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energy (h) at (h) n = 0, as shown in Figures (5) and (6), respectively. The obtained values of the optical energy gap decreased with the increase in the molecular weights of PEGs and GONS additive as shown in Table 2. This could be explained as follows: Increasing molecular weight lead to an increasing in the disorder degree. Furthermore, the selective absorption of photon energies of the incident light indicates which such energy was dedicated to breaking up and hence deforming the partially crystalline structure of the polymers. The rising in the disorder degree of polymer leads to a reduction in the estimated optical gap (35), as well as the contribution of GO with the ability to tuneable the band gap showed that significant of a reduction in the energy gap values in agreement with the literature (10).



Figure (5): Optical energy gap for the allowed indirect transition $(h)^{1/2}$ versus photon energy of PEGs and PEGs-GO nanocomposite films.



Figure (6): Optical energy gap for the forbidden indirect transition $(h)^{1/3}$ versus photon energy of PEGs and PEGs/GO nanocomposite films.

Table (2): Optical energy gap values for the allowed and forbidden indirect transition of PEGs and PEG/GO nanocomposite films.

Sample ID	Allowed Indirect Transition (eV)	Forbidden Indirect Transition (eV)
PEG1	3.68	3.5
PEG1-GO	3.59	3.35
PEG2	3.4	3.1
PEG2-GO	3.21	2.8
PEG3	3.1	2.71
PEG3-GO	2.5	1.8

Figure (7) shows the change of the refractive index for PEGs and PEG/GO nanocomposite films versus wavelength. It can be noted that the refractive index increased up to 2.25 with the increase of molecular weights of PEGs, meanwhile the contribution of the GONSs presented another significant enhancing up to 2.4. This result corresponds to the research done by J.D. Ingham and D.D. Lawson (31), they proved the linear relationships between the refractive index and molecular weights for PEGs, in a wide wavelength region from ultraviolet to infrared, which is the reason for the increase refractive index.



Figure (7): The refractive index (n) with wavelength for PEGs and PEGs/GO nanocomposite films.

Figure (8) demonstrations the change of the extinction coefficient (K) for PEGs and PEG/GO nanocomposite films as a function of wavelength. The result showed that the extinction coefficient increased with the increase of molecular weight of PEGs and additional GONSs in all wavelength regions. This is because of the close relationship between the absorption coefficient and extinction coefficient as shown in equation 5. The increasing of extinction coefficient in the lower wavelengths (ultraviolet region) is due to the high absorbance of films in that region.



Figure (8): Graphically relation between extinction coefficient and wavelength for PEGs and PEGs/GO nanocomposite films.

The variation of the real ($_r$) and imaginary ($_i$) parts of the dielectric constant results with wavelength in the range (300-1100) nm for PEG1, PEG2, PEG3 and PEG-GO nanocomposite films were illustrated in Figures (9) and (10), respectively. The real part represents, how much slower down the speed of light in the material, and the imaginary part represents how a dielectric absorbs energy from the electric field due to dipole motion. The behaviour of r was similar to the refractive index because of the smaller value of k compared with n according to equation (6), while i mainly depended on the extinction coefficient (k) value. Generally, the results presented increasing in both r and i with increasing both effect factors that molecular weight of PEGs and GONPs additive in all wavelength regions. Figures implying that the values of the real part were higher than those of the imaginary part.



Figure (9): Graphically relation between r and wavelength for PEGs and PEGs/GO nanocomposite films.



Figure (10): Graphically relation between _i and wavelength for PEGs and PEGs/GO nanocomposite films.

5. Conclusions

The fine dispersion and good homogenous nanocomposites were achieved using sonication-mixingacoustic methods. The results presented the highest absorbance value appears in the ultraviolet region (About 300 nm in wavelength) of the electromagnetic at PEG3-GO nanocomposite film. Meanwhile, the optical energy gap was decreased with the increase of molecular weight of PEGs and addition of GONSs. All the other results showed notable enhancement in the optical properties that indicate the influence of the Mw of the polymer and addition of GONSs playing important roles to enhance the optical properties of the polymer, where improve the absorbance of ultraviolet waves allows using in the significant applications.

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