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Important Factors Affecting the Microstructure and Mechanical Properties of PEG/ GO-Based Nanographene Composites Fabricated Applying Assembly-Acoustic Method

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Abstract. Graphene-based nanocomposites are one of the most important materials attracted the attention of researchers and engineers. This investigation focused on the effect of two considerable factors using the three molecular weights (Mw) 4k, 8k and 20k (g.mol⁻¹) of polyethylene glycol (PEG) and reinforcing with low loading ratio of graphene oxide nanosheets (GONSs) on the mechanical properties of the polymer matrix. New developed sonication-mixing-aquatic methods were applied to synthesize the different MW of PEGs and PEGs/ GONSs nanocomposites then cast as films. Range of characterization was considered such as structural, rheological and mechanical properties. These were characterized using density and viscosity tolls, Fourier transforms infrared (FTIR), X-ray diffraction (XRD), optical microscopic (OM) and ultrasonic. The FTIR results exhibited the most functional group of PEGs and GONSs, also confirmed the significant strong connection between PEGs with GONSs. Crystalline peaks of the PEGs were presented clearly in the nanocomposites using XRD, whereas GONSs peak was not clear showed that related to the full desparation of the GONSs in the samples as presented by OM. Most of the mechanical properties presented notable improvements with the increase of the molecular weights of net PEGs in the matrix and significant enhancements with the reinforcement of the GONSs, where the most characterizations such as, ultrasonic velocity, absorption coefficient and bulk modules appeared a significant enhancements up to 50%, 700% and 126%, whereas the compressibility and relaxation time results were reduced to 75% and 54%. Moreover, the contribution of GONS with increase Mw of PEGs demonstrated an outstanding improvement of the mechanical properties up to 100%, 800%, and 307%, meanwhile the compressibility and relaxation time was reduced to 57% and 18%. Generally, increasing of the PEGs molecular weights and reinforcing using GONSs led to notable enhancement in the properties of nanocomposites, where the sample contain higher molecular weight in both samples PEGs and PEG/ GONSs nanocomposites. From the obtained results the nanocomposite specimens perform good medium for transfer the ultrasound waves particularly in high molecular weights (20k), that means it presented good stability against and absorbance mechanical waves, therefore it could be applied for a coated material for objects that wanted to be invisible under sonar or in storage energy applications.

INTRODUCTION

Polyethylene glycol (PEG): HO-(CH₂CH₂O)_n-H, is available in a variety of molecular weights from 200 to tens of thousands [1]. It is commonly used as solid-liquid phase change materials because of the suitable melting point, chemical stability and high phase change enthalpy. This behaviour depends on the molecular weight [2]. Moreover, it is considered soluble-water polyether compounds. Where the oxyethylene groups are the repeated subunits as well as the terminal hydroxyl groups, which rise the polarity. Additionally, this turns the PEG to become more inter-active towards polar analytics and surfaces. Therefore, PEG is commonly wide and extensively used in research and range of application [3–5]. Despite all the advantages of this polymer, it suffers from a weakness in mechanical properties.

Nanotechnology and develop science have been used as an important method to overcome these drawbacks. Modifying the polymer structure in the matrix is an important factor to enhance and developed new moderate composite [6]. Where polymeric is the most film that is currently used and its production is extremely developed applying these methods [7]. Therefore, polymer-based nanocomposites have been attracted more attention because of the high performance of the polymer. This attribute to the drastic improved of the properties of the polymers and materials through the impact of the nanoparticles. Consequently, studies are carried out to design the

mechanical properties of these type of materials [8]. Polymer-nanocomposite has been widely applied in numerous applications [6].

Recently, carbon family are an important filler to enhance the properties and reinforcement the materials, meanwhile, graphene is one of the best nanofillers, it is stronger and strength a two-dimension sheet of a single atomic thick layer of a hexagonally arranged carbon atom with a range of extraordinary unique physical properties [9]. These make graphene an ideal candidate for a wide range of applications such as supercapacitors [10], hydrogen storage [11,12] and nanocomposites [13]. Graphene oxide nanosheets (GONSs) is one of the most interesting nanofillers that have the same feature of graphene with additional advantages, which are the functional groups on the surface of the GONSs. Due to the existence of various oxygenated functional groups, GONSs is derivative from graphite using oxidized methods that reduced a highly hydrophilic surface of GO with numerous containing of oxygen functional groups and unique properties of graphene that make it attracted a lot of attention as nanostructured material [14,15]. GONSs can be synthesized via various ways and Hummers [16] that is the most famous one, also, GONSs has a large surface area and significant potential to fabricate micro-electrical devices for various applications [17]. GONSs can basically interact with various materials, polymers, etc to reinforce or/and improve the properties such as bioactivity, thermal, mechanical electrical, conductivity, and so on [18]. To achieve high performance and improvements in the properties of the materials, GONSs must have a good homogeneous and fine dispersion in the matrix [19]. Whereas, the large surface area and strong van der Waals forces of the GONSs could lead to strong aggregation in the composite.

Polymer- graphene oxide based nanocomposites are developing a novel class of materials. This display significant enhancement in the properties of materials and do not achieve by conventional composites or pristine polymers and hold promise for applications. The improvements can be obtained even at a very low loading ratio of the nanofillers in the polymer matrix. That depends on many factors, for instance, the components, morphological, dispersion and interface interaction of the GONSs with the polymer in the matrix [12,15].

[20] studied a series of GO-PEG nanocomposites that were syntheses in an aqueous medium applying ultrasonic dispersion. They investigated the influence the different PEG molecular weights on the orientation structure and intercalation behaviour of the samples, where XRD result presented the increasing in the PEG molecular weights increased the interlayer spacing of nanocomposites and that reached the maximum values with 20k PEG of the GO/PEG nanocomposites, whereas after this MW, the results were decreased. Moreover, FT-IR spectra exhibited a significant shift of O-H functional absorption peak of GO sample. This shifting a significant increased for GO/PEG20k nanocomposites. This indicating strong interfacial interactions of the hydrogen bonding that made between GO nanosheets and PEG20k molecules. Also, the electrical conductivity of PEG was improved used GO. [21] Studied the effect of molecular weight on the properties of the films. Also, the effect of nano-clay and graphene oxide (GO) on the PEG plasticised film properties using sonicating methods and casting on a petri dish. They find a strong influence of the molecular weight of PEG on the water vapour permeability and mechanical properties, whereas there is no clear effect on the contact angle or initial degradation temperature of the samples. Additionally, plasticise shows more affected by the lower MW, 400-1000 g.mol⁻¹ of PEG than a higher molecular weight > 1000 g.mol⁻¹. [11] Reported using GO as reinforcement materials using a simple and effective one-step hydrothermal method to fix and fabricate PEG PCMs. They reported enhancing in the thermal conductivity and achieving high energy efficiency after using the GO compared with PEG. This is related to strong hydrogen bonding that widely exists in the GO/PEG composites [11].

In spite of all the above various studies and reports [11,15,18,20,21] the effect of increasing molecular weights as well as with addition GONSs reinforcement as nanofillers on the mechanical properties need more investigated [22,23]. This study was focused to reduce the gap of knowledge in this filed. Therefore, a new method was developed to prepare polymers and polymer-GONSs hybrid nanocomposite films to investigate the effect of increasing the molecular weights of the PEGs and reinforcement with low loading ratio of graphene oxide nanosheets on the mechanical properties. Three different molecular weights (4k, 8k and 20k) g mole⁻¹ of Polyethylene glycol (PEG) with were selected as polymer model. Range of devices were used to characterize the samples such as density and viscosity tolls, Fourier transforms infrared (FTIR), X-ray diffraction (XRD), optical microscopic (OM) and ultrasonic.

EXPERIMENTAL PART

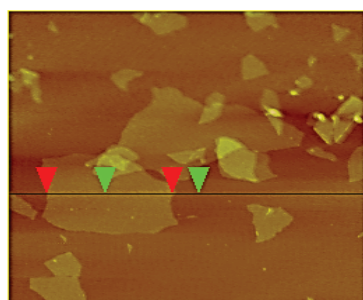
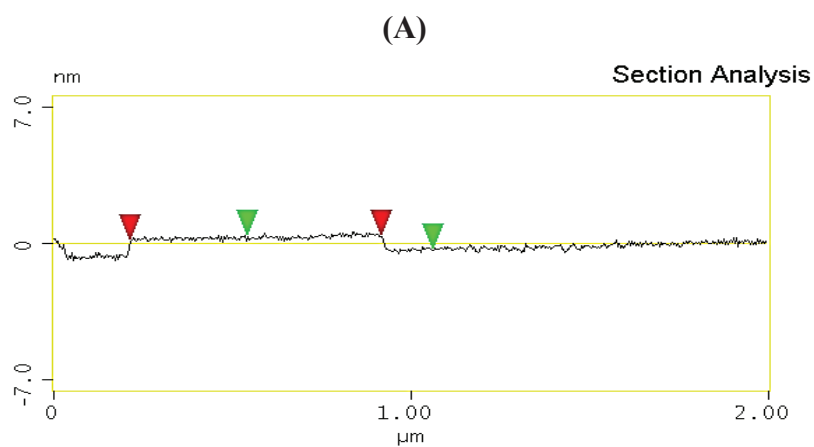
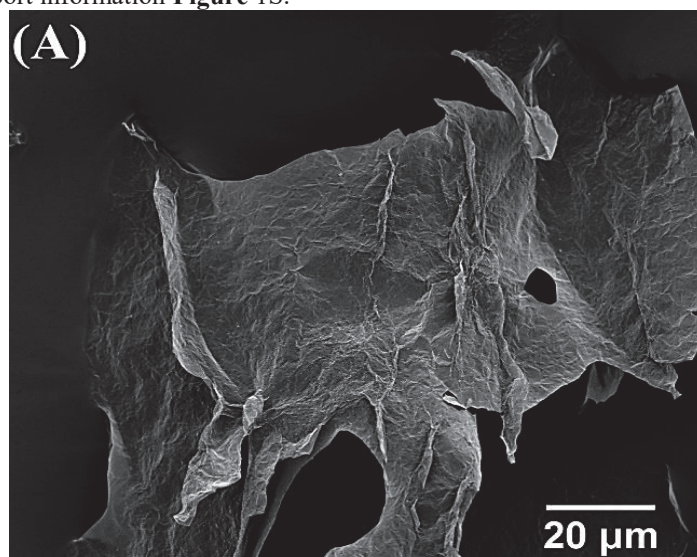
Materials

PEG with three different molecular weights (4k, 8k and 20k) g- mole⁻¹ were supplied from (Central Drug House, Ltd, Company), India. Graphite powder ($\leq 40\mu\text{m}$), hydrochloric acid (35%), potassium permanganate, sulfuric acid (99.8%), hydrogen peroxide and sodium nitrate were obtained from Sigma-Aldrich Company, UK.

METHODS

Synthesis of the Graphene Oxide Nanosheets

GONs were synthesis by our group and full characterizations were available in our previous literature [24], as explained in the support information **Figure 1S**.



Surface distance	703.41 nm
Horiz distance(L)	703.13 nm
Vert distance	0.238 nm
Angle	0.019 $^{\circ}$
Surface distance	519.73 nm
Horiz distance	519.53 nm
Vert distance	0.700 nm
Angle	0.077 $^{\circ}$
Surface distance	
Horiz distance	
Vert distance	
Angle	
Spectral period	DC
Spectral freq	0 / μm
Spectral RMS amp	0 nm

(B)

FIGURE 1. A) SEM and B) AFM images of GO nanosheets (24).

Purifications of Nanocomposites

GONSs was dispersed in distilled water with 1 wt% using sonication bath then stirred for 24 h of GO in the distilled water and sonicated for 20 minutes for fine dispersion. PEGs were dissolved in distilled water with 99 wt% using magnetic stirrer, then GONSs-H₂O solution was added to PEGs to synthesis the nanocomposites applying mixing-sonication-acoustic methods as shown in Table 1.

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

Sample ID	PEG MW	Ratio wt%		Total Mixing time during synthesis	Sonication time during synthesis	Final mixing time	Dry method
		PEG	GONS				
GONSs	0	0	100	-	0.5 h	-	Freeze drier
PEG1	4k	100	0	-	-	3 h	At room temp. 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-GONSs	20k	99	1	6 h	1.5 h	12 h	

briefly, after added the solution of GONSs to the PEGs, the samples were mixed using a magnetite stirrer for 60 minutes then sonicated for 0.5 hour, followed with another mixing time for 120 minutes then another sonication for 0.5 hour, then mixture also was mixed and stirred for 180 minutes and sonication for 30 minutes with monitoring to get fine homogeneous and dispersion of the GONSs in the polymer matrix. Finally, these nanocomposites mixtures were mixed for 12 h, then the samples were pleased in petri dish applying casting methods, which was casting the mixture in the plastic petri dish under air until to dray to get films. PEGs with different MWs were prepared to flow the same procedure to prepare PEGs films with different three different MWs. Figure 1 summarizes the preparation steps of PEGs and nanocomposites. The thicknesses of the sample films were between 240 – 290 μm.

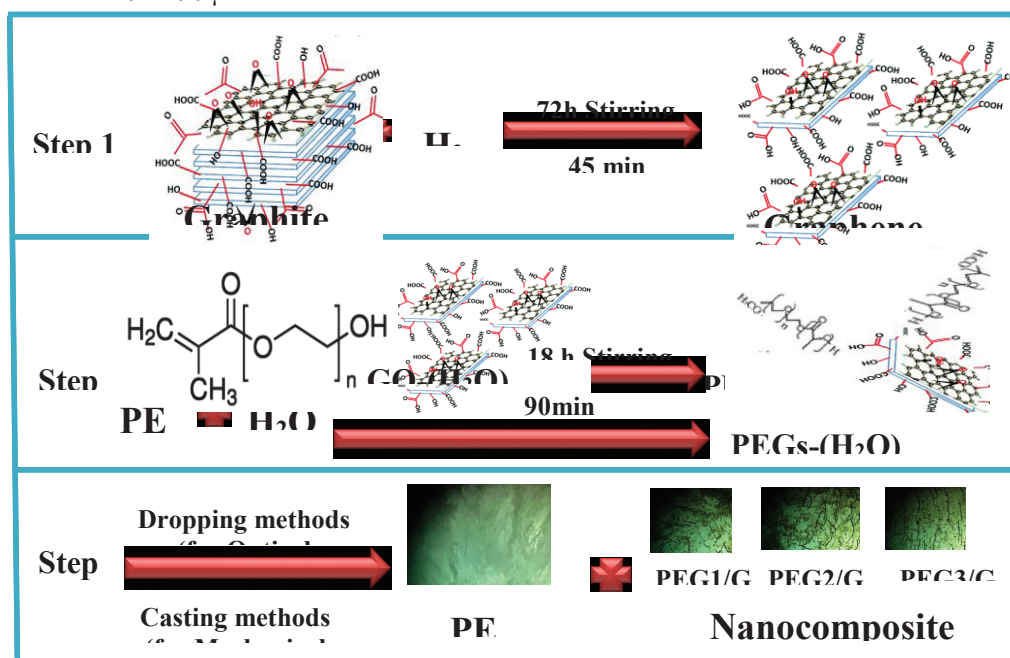


FIGURE 2. Schematic of purification the PEGs and Nanocomposites.

CHARACTERIZATION

Density was measured using a density bottle with a capacity of 25 ml and the sensitive balance (0.0001) (Switzerland Company). The viscosity of the aqueous solutions is measured using Ostwald's viscometer. This was calibrated using distilled water. The water bath was used during the experimental using the Ostwald's Viscometer in room temperature. Fourier transforms infrared (FTIR) spectra were recorded by FTIR (type vertex 70, Bruker Company), Germany. FTIR is the preferred technique to obtain an infrared spectrum of absorption or emission of

a gas or solid and liquid. “Infrared” refers to any electromagnetic radiation, the most attractive for chemical analysis are located in the region between 4000 - 400 cm^{-1} . X-ray diffraction (XRD) analysis was used to recognize the crystal structure of PEGs/ GONSs nanocomposite films. The X-ray diffraction instrument type (Shimadzu 6000) was made in Japan. The surface morphology of PEGs/ GONSs nanocomposites was examined using the optical microscope (Nikon-73346) that was supplied from Olympus Company. Mechanical properties were measured using ultrasonic (SV-DH-7A/SVX-7) with a pulse technique of sender-receiver type with multiple frequencies 5 Hz -50k Hz and all measurement were at 30 kHz.

THEORETICAL PART

The viscosity of the experimental liquid η_s was calculated using equation [25]:

$$\frac{\eta_s}{\eta_0} = \frac{t_s \rho_s}{t_0 \rho_0} \quad (1)$$

η_0 means the viscosity of water, t_0 means the time of flow of water, ρ_0 means the density of water, t_s means the time of flow of the experimental liquid and ρ_s means its density. The ultrasonic velocity (V) was measured by applying the following equation [26]:

$$V = \frac{X}{t} \quad (2)$$

X means the sample thickness and t means the time of wave across the sample. The absorption coefficient (α) was calculated from the Lambert-Beer law [27]:

$$A = A_0 \exp(-\alpha X) \quad (3)$$

A_0 means the initial amplitude of ultrasonic waves and A means the amplitude of the wave after absorption. Compressibility (B) was calculated using the Laplace equation [28,29]:

$$B = (\rho V^2)^{-1} \quad (4)$$

The Bulk Modulus of elasticity (k) was calculated by applying the equation:

$$k = B^{-1} = \rho V^2 \quad (5)$$

Where D means the relaxation amplitude of an ultrasonic wave that was calculated using the equation:

$$D = \alpha / f^2 \quad (6)$$

where α means the absorption coefficient. The relaxation time (τ) was calculated from the equation.

$$\tau = \frac{4\eta_s}{3\rho V^2} \quad (7)$$

The specific acoustic impedance (Z) of a medium that was calculated applying equation:

$$Z = \rho V \quad (8)$$

RESULTS AND DISCUSSION

The FTIR spectra of PEGs, nanocomposites were presented in Figure 2. PEGs with three different molecular weights exhibited different functional groups such as, $-\text{CH}_2$ that was asymmetrical stretching at 2881 cm^{-1} , $-\text{CH}_2$ and $-\text{CH}_3$ were the bending vibrations at 1466 cm^{-1} and 1340 cm^{-1} , respectively. At 1279 cm^{-1} , this peak was attributed to the bending vibration of the ether O-H groups and the $-\text{C}-\text{O}-\text{C}-$ symmetrical and asymmetrical stretching were located at 1098 cm^{-1} and 958 cm^{-1} , respectively. these peaks of PEGs showed a good agreement with the behaviour reported by another author [24,30] of PEGs as shown in Figure 2.

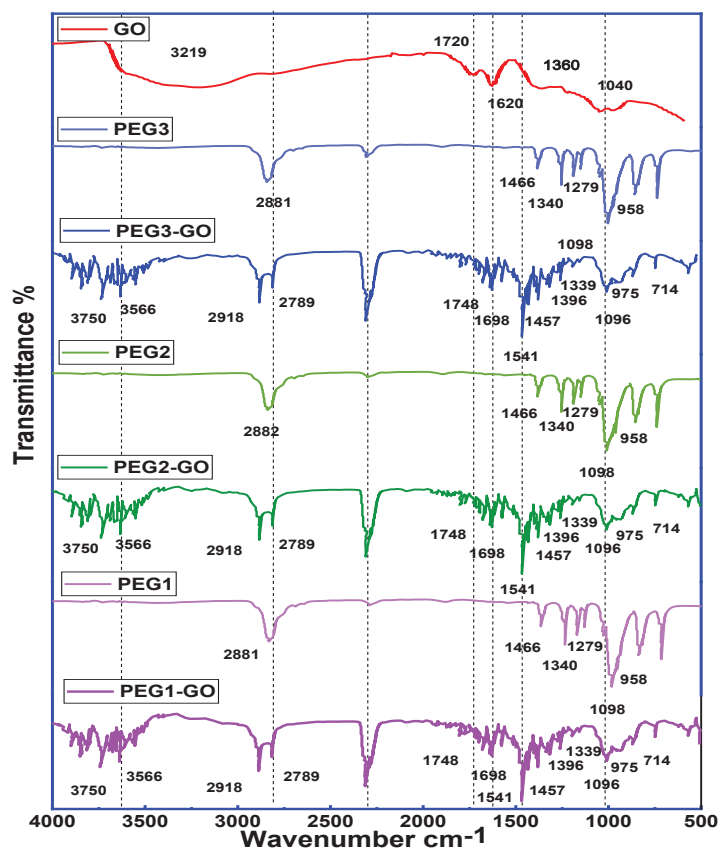


FIGURE 3. FTIR spectra of PEGs, GO and PEGs/GO nanocomposite films.

The FTIR spectrum of GONS showed the broadband at 3219 cm^{-1} that was related to the stretching vibrations of hydroxyl groups (O–H). Around 1720 cm^{-1} , this characteristic stretching adsorption bands that were corresponded to the carbonyl groups (C=O) at the edges of the GONS [13]. the peak at 1620 cm^{-1} can be attributed to the C=C skeletal ring stretching vibration of un-oxidized sp^2 carbon domain [20]. Meanwhile, the peaks at 1360 cm^{-1} and 1040 cm^{-1} were linked to C-O or epoxy groups (C–O–C), as shown in Figure 2.

Another peak presented in the nanocomposites that related to PEGs that were presented at 2918 cm^{-1} and 2789 cm^{-1} which were attributed to CH_3 and CH_2 the symmetrical stretching vibration, respectively. Additionally, CH_3 and CH_2 at 1457 cm^{-1} and 1396 cm^{-1} were asymmetrical stretchings. The band at 1541 cm^{-1} could be assigned to the aromatic C=C stretching, the bending vibration of the O–H at 1339 cm^{-1} and the C–O–C stretching vibration at 1096 cm^{-1} . The deformation vibration of the C–H bond at 975 cm^{-1} aromatic C-H in-plane bend (several), while the bond C–H at (714 cm^{-1}) is aromatic C-H out-of-plane bend (several).

In addition, the PEGs/ GONSs nanocomposites showed a slight shift in the peak positions and change of relative intensity in the C=O, O–H, C-H, and C=C peaks, that suggest the correlation and strong interaction between both PEG and GO which may related to the hydrogen bonding between PEGs and GONSs [20], This was confirmed the synthesis of the nanocomposites [11,20].

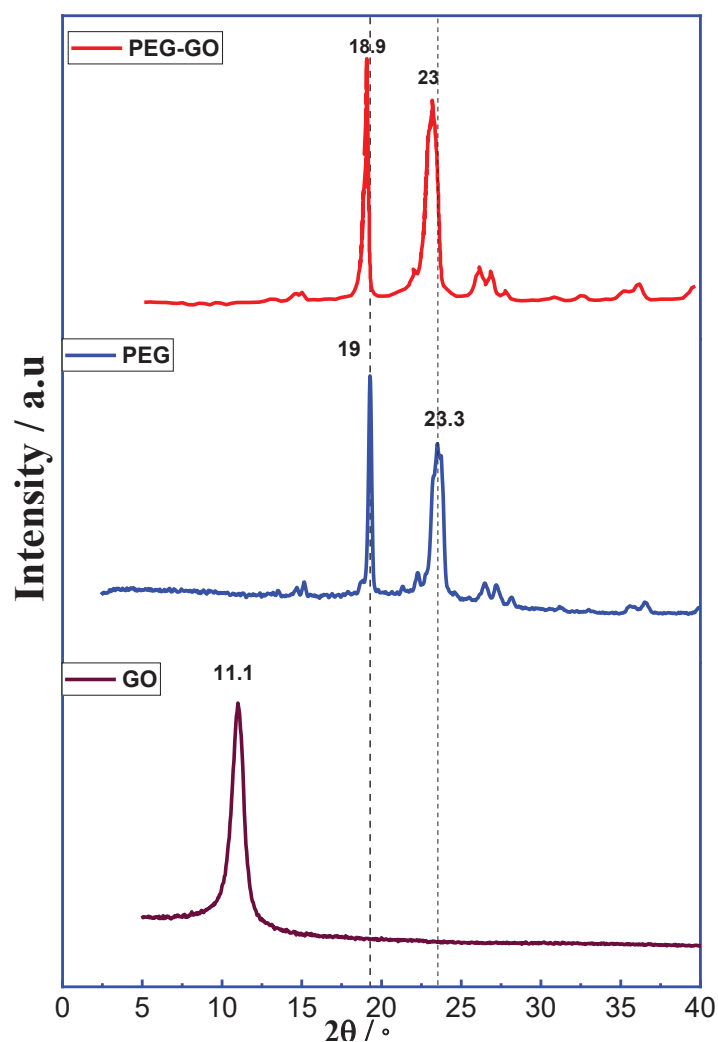


FIGURE 4. XRD patterns of the PEG3 and PEG3/ GONSs nanocomposites.

Fig. 3 shows the XRD patterns of GONSs, PEG3 and PEG3/ GONS nanocomposites. GONSs spectra illustrated a peak at $2\theta = 11.1^\circ$ that is related to 002 diffractions of GONSs flakes demonstrating a 0.79 nm as interlayer spacing, in agreement with the literature [16]. PEG pattern demonstrated two peaks at $2\theta = 19^\circ$ and 23.3° as usually confirmed by literature reported [31]. These PEG peaks were clearly presented in the PEG3/ GONSs nanocomposites, it presented a slight left shift in the position of PEG1 peak in to 18.9° that related to the increment of interplanar crystal spacing. This led to the observable in the volume expansion in the macro [11]. Whereas the peak of GONSs was not clearly presented that may be related to many reasons reported by many research [24,32]. They may be related to overlapping by higher intensity PEG peaks or to the full desperation of GONSs in the PEGs/ GONSs nanocomposites that presented clearly in the OM images or because of orientation. However, this finding indicated the reinforcing using GONSs did not influence on the crystal structure of PEG in the nanocomposite in strong agreement with the literature [11].

The results of the density and viscosity measurement values of the different molecular weights PEG1, PEG2, and PEG3 g mole^{-1} of polyethylene glycol (PEG) and its graphene-based nanocomposites solution were tabulated in Table (2). The viscosity values increased with the increasing the molecular weights of PEGs as well as after the addition of graphene oxide nanosheets (GONSs). The result of the all prepared PEGs/ GONSs nanocomposites was significantly presented better and higher than the net PEGs. That was due to the increase of the rotational and transition friction forces between the polymer and solvent molecules from the side and between the graphene oxide nanosheets from the other side [23]. That could be related to the formation of hydrogen bonds between the disintegrating polymer and the solvent in agreement with FTIR results and literature [24].

TABLE 2. The viscosity measurements values of the based-polymeric solution.

Sample ID	Density (g.cm-3)	Flow Time (s)	Water Density (g.cm-3)	Water Flow Time (s)	Viscosity (g.cm-1.s-1)
PEG1	1.008	38.7			2.191
PEG2	1.01	51.9			2.944
PEG3	1.012	69.3			3.939
GO	1.007	20.3	1	17.8	1.148
PEG1/GO	1.011	41.25			2.342
PEG2/GO	1.015	59.3			3.381
PEG3/ GO	1.025	73.2			4.215

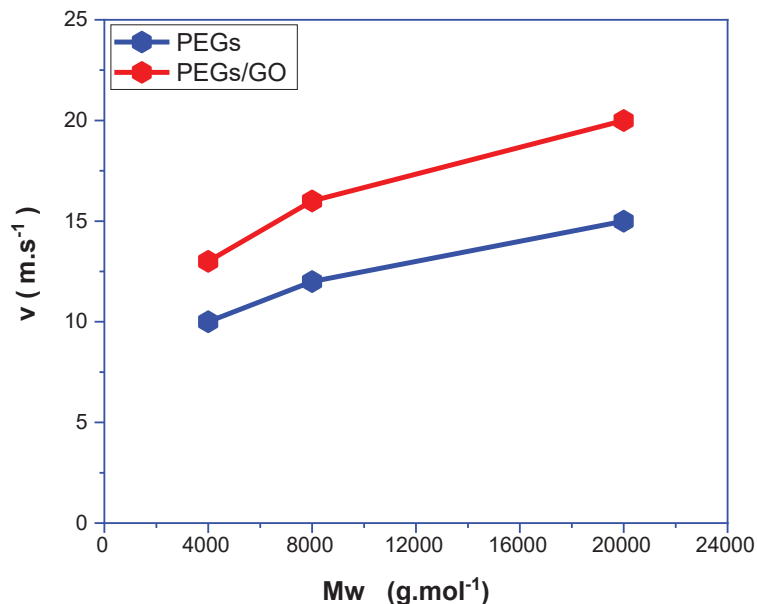


FIGURE 5. The Ultrasonic velocity with the molecular weights of the PEGs and nanocomposites.

Fig. 5 shows the Ultrasonic velocity of the PEGs and PEGs /GONSs nanocomposites. The results displayed increasing in the velocity results with increasing molecular weights of the PEGs from PEG1 to PEG2 then from PEG2 to PEG3 and up to 20% and 25%, respectively. In comparison, the velocity was increased up to 35% after increasing the MWs of PEGs from PEG1 to PEG3. Furthermore, the contribution of GONSs with increase Mw of PEGs led to an increase of up to 19%, 20% and 35%, respectively. This recognized to the interaction or (increasing) resulting in the connections between the types of molecules; polymer and GONSs. The values of 'V' indicates that the presence of solute-solvent interactions because of hydrogen bonding [20,24], Also, the molecular weight of the polymer can influence the magnitude and nature of intermolecular interactions [28,33]. The presented of GONSs in the PEGs nanocomposite can be filled the spaces between polymer chains and interact with ether group of PEGs in more than one part as presented in the FTIR spectra, where when comparing the improvement in the transferring ultrasonic wave through the sample that presented 100% enhancement in the result when compared between pure polymer PEG1 and PEG3-GONSs. These entanglements enhanced the prepared composite to be a good medium for transferring the ultrasonic wave.

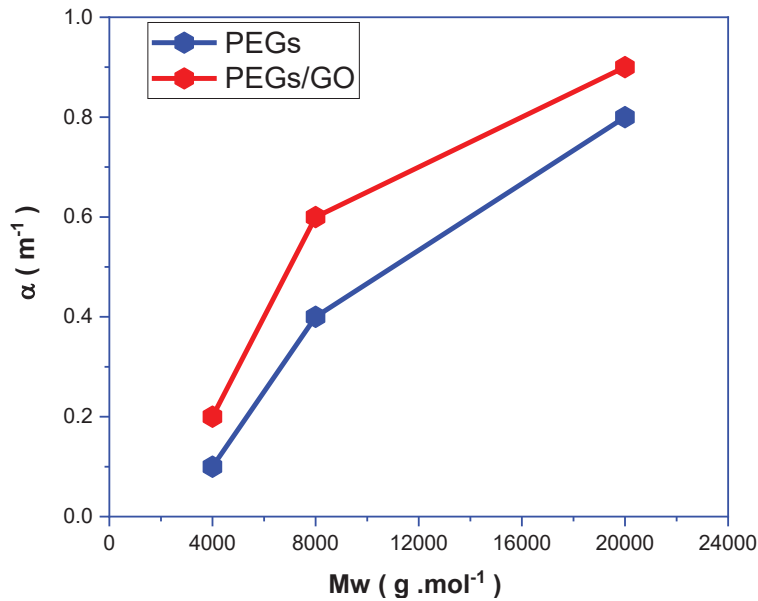


FIGURE 6. Absorption coefficient with the molecular weights of the PEGs and nanocomposites.

Fig. 6 shows the absorption coefficient (α) with the different molecular weights of PEGs and PEGs/GONSs nanocomposites. Ultrasonic absorption coefficient values were measured of the PEGs and PEGs/GONSs nanocomposite from the difference in value between sender and receiver ultrasound absorption waves as presented in the oscilloscope screen. The α values were increased with the increasing up to 300%, 100% and 700% when increased the PEGs MWs from PEG1 to PEG2 then PEG2 to PEG3 and between PEG1 to PEG3, whereas the addition of GONSs with increasing the molecular weights of PEGs caused an improved in the absorption coefficient of the ultrasonic wave up to 200%, 50% and 350%. Whereas, it presented a notable enhancement of the absorption coefficients of the ultrasonic wave up to 800% during the comparison between the results between PEG1 and PEG3-GONSs.

This attributed to the attenuation of ultrasonic energy depends on many factors such as nature of components, intermolecular interaction, density and shear viscosity [34,35], Where increasing the strong interfacial interactions between the polymer molecules, solvent and nanofillers was the main cause of increasing shear viscosity as presented in Table 1. As a sequence, this led to increasing the absorption coefficient as a result of these interactions which were clearly presented in the FTIR results. Increasing the molecular weights attributed to the mechanism of hydrogen bonding attached to oxygen sites [11,20,24], which led to increasing the entanglement between filler molecules (GONSs) and polymer chains. This caused randomly coiled and strong interfacial interactions and led to an increase in absorbed of the ultrasound waves the nanocomposites compared to the molecule of the polymer, as shown in Figure 6.

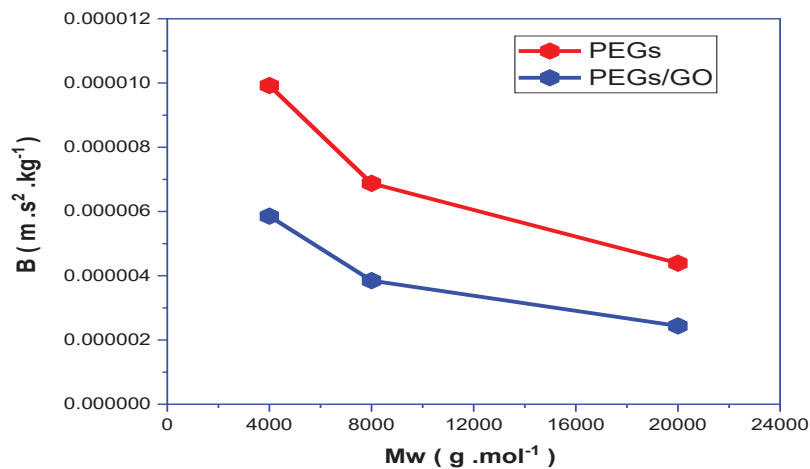


FIGURE 7. Compressibility with the molecular weights of the PEGs and nanocomposites.

In Fig. 7, the compressibility (B) was presented of the PEGs and nanocomposites with increasing the MWs of PEGs. The B values were decreased with the increasing the Mw of the PEGs when increased the PEGs MWs from

PEG1 to PEG2 then PEG2 to PEG3 to 31% and 36%, meanwhile, the contribution of the GONSs with increasing the MWs of the PEGs reduced up to 36% 39%. In comparison, whereas B reduced to 59% in the comparison between PEG1 and PEG3, whereas the higher MWs that PEG3 with addition GONSs PEG3/ GO nanocomposites exhibited stronger compared with all samples as compressibility reduced up to 75%. This concludes that increasing the MW was an important factor to improve the compressibility in both PEGs and hybrid nanocomposites, as shown in Fig. 7.

The compressibility values exhibited decreasing in the values with both increasing the Mw and increased the decreasing of the B after the contribution of the GONSs with increasing the Mw. This attribute to the fact that the polymer molecules come close to the solvent molecules, leaving a sufficient space around them [36], also the reason for the fusion of two types of molecules, which in turn led to the clustering of polymeric chains converging with each other due to their strong interfacial interaction with the GONSs nanosheets as presented in the FTIR results that decreasing the spacing and built strong interaction between both nanofillers and polymer matrix. These helped to prepare stronger materials showed resistance against the compressibility also that presented clear with the velocity behaviour. Additionally, this behaviour of the B is supported by the results of the density and velocity that are considered important and effective factors on the compressibility, which exhibited opposite behaviour as shown in equation (4).

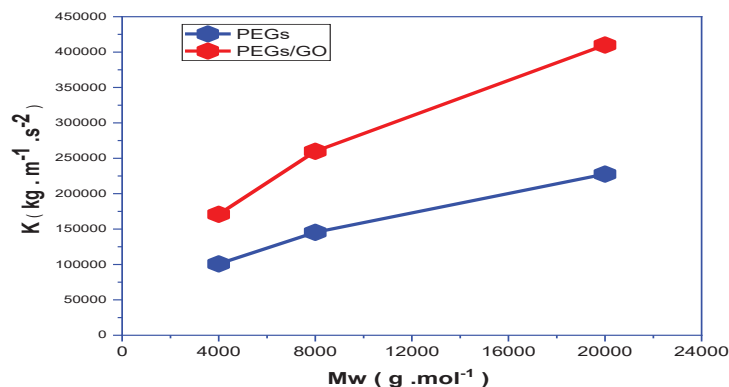


FIGURE 8. Bulk modulus with the molecular weights of the PEGs and nanocomposites.

Bulk modulus (K) of the PEGs and nanocomposites results were shown in Figure 6 that illustrated the effect of the various and increasing in the molecular weights (PEG1, PEG2 and PEG3) g.mole⁻¹ of the PEGs. The results showed enhancement the K results with increasing polymer MWs from PEG1 to PEG2 then PEG2 to PEG3 and between PEG1 to PEG3 up to 44%, 57% and 126% respectively, whereas the contribution of GONSs with increasing the Mw enhanced up to 52%, 59%, and 140%, respectively. The increase in bulk modulus for samples suggested a strong connection between polymers molecular weights with GONSs nanosheets, as presented in the FTIR results. Also, this finding was supported with a significant increase in the ultrasonic velocity that is considered an essential factor effect on the behaviour of the bulk modulus (K), as presented in the relations [4 and 5]. Generally, the bulk modulus (K) of the nanocomposite films behaviour demonstrated a significant improvement especially of the higher MWs of the PEGs, moreover increasing of the molecular weights the transferring ultrasonic waves within the spacemen and enhanced the K in all samples, as shown in Fig. 8.

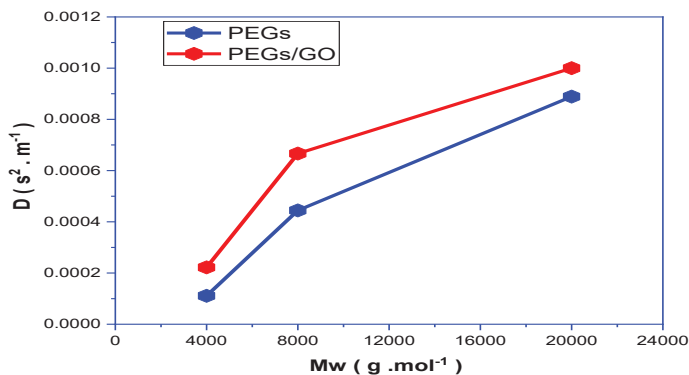


FIGURE 9. Relaxation amplitude with the molecular weights of the PEGs and nanocomposites.

Figure 9 shows the relationship between the relaxation amplitude and increasing the molecular weights of PEGs and PEGs/GONS nanocomposites. The results of PEGs displayed that the relaxation amplitude increased with the increasing the molecular weights of the PEGs from the PEG1 to PEG2 and from PEG2 to PEG3 then

between PEG1 and PEG3 improved up to 300%, 102%, and 709%, respectively. Whereas, the addition of the GONSs in the polymer matrixes improved the relaxation amplitude up to 205%, 67%, and 400%, respectively. Interestingly, comparing the addition of GONSs with increase molecule weights of the PEGs of PEG3/ GONSs nanosheets with PEG3 demonstrated an improving up to 900%. This related to an increase in the distance travelled of the molecules during the excitation process. Moreover, the determination of the inertia of the large molecule was significant due to its interrelationships with the GONSs nanosheets [15,37]. In addition, the relaxation amplitude was directly proportional to the absorption coefficient, as a form in the equations (6). When the frequency is constant for both cases, the relaxation amplitude of the solvent molecules is expected to increase when the absorption coefficient is increased.

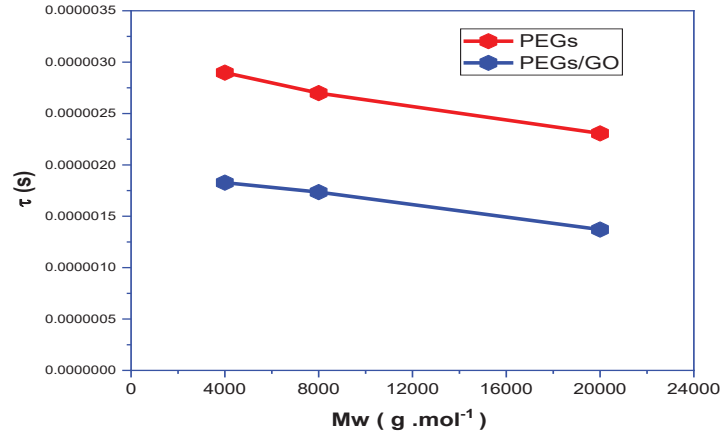


FIGURE 10. Relaxation time with the molecular weights of the PEGs and nanocomposites.

Fig. 10 illustrates the relationship between relaxation time and Mw of PEGs with PEGs/GONSs nanocomposites. The results showed reducing of the relaxation time with the increasing of the molecular weights up to 7% and 15% respectively, whereas, it reduction up to 21% increasing the MWs from PEG1 to PEG3. The contribution of the GONSs with increasing the molecular weights led to increasing the reduction of the relaxation time up to 6% 18% and 28%, respectively. Interestingly, the nanocomposites results exhibited notable improvement due to the contribution of GONSs compared with the net of the PEGs samples, where the reinforcement of GONSs enhanced the interfacial interaction and reduced the relaxation time. In comparison, the addition of GONSs with increasing the MW of PEG to PEG3 exhibited a reduction in up to 54% of the PEG3/ GONSs nanocomposites compared with PEG3 that contained PEG3. The decrease in the relaxation time is explained because increasing the volume of the polymer chains. This led to increased internal friction between the layers of films resulting from compressing and deactivation as a result of ultrasound effect and the strength of interfacial interaction with GONSs nanosheets [31], which restricted the movement of the polymer molecules. Moreover, based on the dominance of the inverse relationship between the relaxation time (τ) and the ultrasonic velocity (V) according to equation [7], the relaxation time decreases with the molecular weight before and after the reinforcement using the GONSs, regardless of its relationship to viscosity. This results exhibited a decreasing in the time that required to restore the excited molecule back to its original state [24].

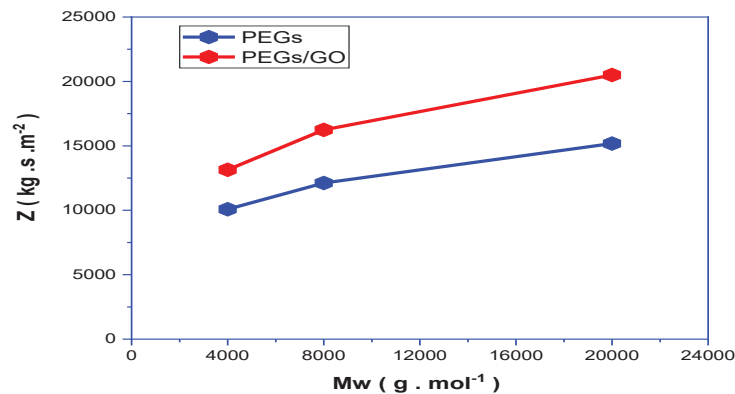


FIGURE 11. Specific acoustic impedance with the molecular weights of the PEGs and nanocomposites.

Fig.11 demonstrates the specific acoustic impedance curves of PEGs and PEGs/GONSs nanocomposites. The results of specific acoustic exhibited an improve with increasing of the molecular weights of PEGs from PEG1 to PEG2 then from PEG2 to PEG3 up to 20% and 25%, respectively. Whereas, 51% was the total enhancement of increasing the Mw of the PEGs from PEG1 to PEG3 as shown in Figure (9). In addition, the contribution of GONSs

with increasing the molecular weights of PEGs from PEG1 to PEG2 then from PEG2 to PEG3 that was improved the result of up to 24% and 26%, respectively. Whereas the impact of GONSs with higher MW illustrated a total enhanced up to 56% of PEG3-GONSs compared to PEG1/GONSs. Interestingly, the reinforcement of the GONSs showed a notable increase in the specific acoustic impedance results compared to the net PEGs samples. This increasing reached up to in compassion between the PEG3-GONSs to net PEG3. Generally, the specific acoustic impedance of the nanocomposites the exhibited higher that PEGs samples, moreover a notable improvement up to 103% was displayed with the reinforcement of the nanoparticle (GONSs) and increasing the MW of the PEG1 to PEG3 in the comparison between PEG1 and PEG3-GONSs nanocomposites. Where increases the intermolecular becoming the main cause of impedance in the propagation of ultrasound waves [38].

This significant enhanced could relate to the specific strong conation and strong interfacial interaction between the GONSs with polymer matrix as presented in the stronger functional groups between ether groups at the end of net PEGs and the C-O bond in the surface of the GONSs. This resulted in stacking the polymer molecules with nanofillers material and led to an increase in the mechanical properties. This led to reduce the space and restrict the movement of the most particle in the polymer matrix that supports the previous with the finding of the reduction in the results of compressibility with relaxation time and in agreements with the literature [28]. Additionally, from the relation [8], it presented that the velocity coefficient is the effective variable coefficient, whereas the density has a very small impact with respect to velocity variation. This explains the behaviour of the specific acoustic impedance t was similar to the ultrasonic velocity behaviour.

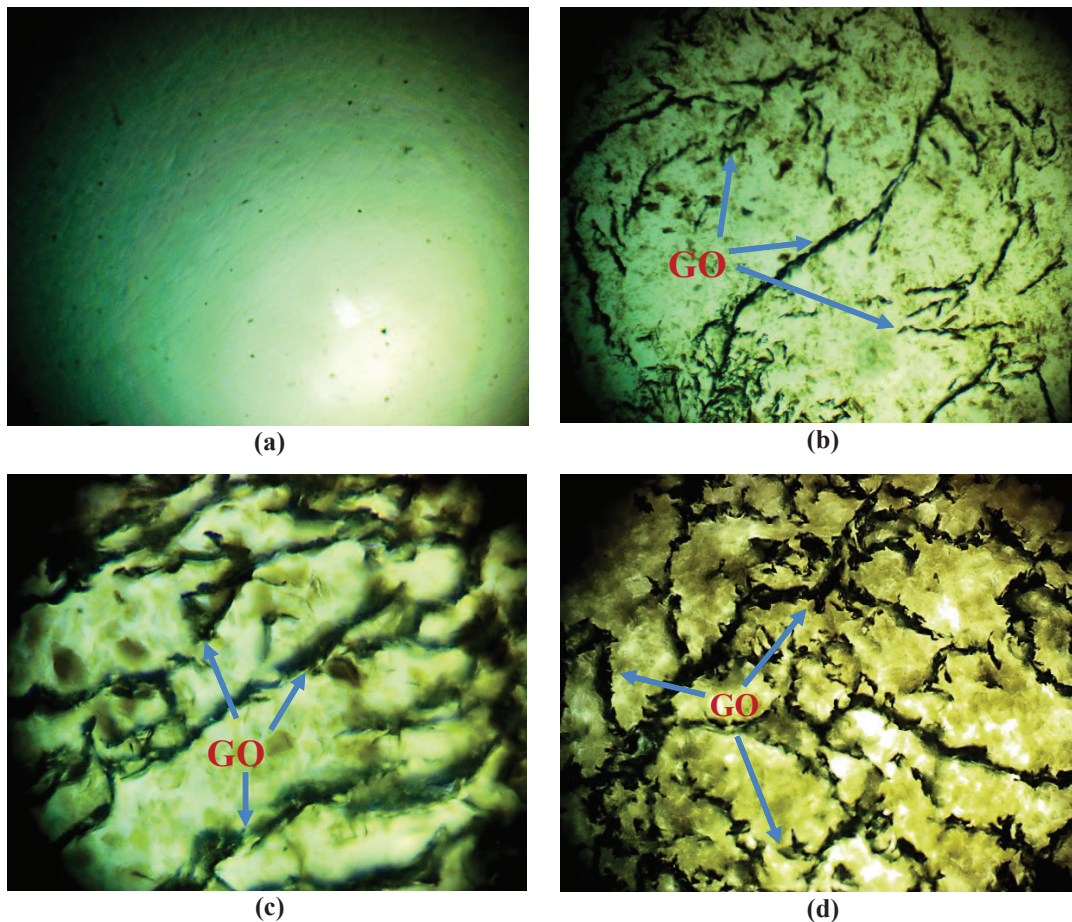


FIGURE 12. Optical microscopy images at magnification power 40X of (a) PEG, (b) PEG1/ GO, (c) PEG2/ GO and (d) PEG3/ GO nanocomposites.

Fig. 12 exhibits the OM images of the PEG and nanocomposites at 40X magnification power. These images showed the change in the dispersion of the GONSs in the polymer matrix. It is clear that the homogeneity and fine distribution of GONSs increased with increasing MW of the PEG. Where, Figure (11b) exhibits area of PEG without GONSs and not fully homogeneous of the PEG1/ GO nanocomposites, where GONSs attracted each other, whereas this phenomenon reduced more in the PEG2/ GO and more in the PEG2/ GO nanocomposites. Increasing the PEG molecular weight display better network and homogeneous without any sign of aggregation which is one and important key to enhance the properties of the nanocomposites without the effect of the transparency in agreement with the literature [15,39,40].

CONCLUSIONS

The new developed sonication-mixing-aquatic method was succeeded to get the fine dispersion and good homogenous of the samples as presented in the OM images. Strong interaction and interfacial connection were displayed to syntheses the nanocomposites in the characterizations as presented in the FTIR, XRD and mechanical. Increasing the molecular weight of PEG showed better mechanical properties and the significant strong linking with GONSs, where the mechanical properties results showed notable enhancement with increasing the molecular weights of PEG from PEG1 to PEG3, as well as the addition of low loading ratio of graphene oxide as nanosheets showed more improvement, especially with increasing the Mw of PEG to PEG3. The nanocomposite specimens prepared from GONSs addition is a good medium for transfer ultrasound waves particularly in high molecular weights (PEG3), that is mean it has good stability against mechanical waves, so these nanomaterials could be applied as storage energy or a coated material to cover objects that unwanted detection by sonar.

REFERENCES

1. J. Chen, SK. Spear, JG. Huddleston, RD. *Green Chemistry*. **7**,64–82(2005).
2. C. Wang, W. Wang, G. Xin, G. Li, J. Zheng, W. Tian, P. *Eur. Polym. J.* **74**, 43–50 (2016).
3. Q. Dai, C. Walkey, W. C. W. P. Chan. *Angew Chem Int Ed.* **53**,5093 –5096 (2014).
4. E. Kianpour, S. *FUEL*.; **137**,36–40(2014).
5. C. Cruje, DB. P. Chithrani. *Reviews in Nanoscience and Nanotechnology*. **3**, 20–30 (2014).
6. Kononova S V, Gubanova GN, Korytkova EN, Sapegin DA. *Polymer Nanocomposite Membranes. Appl Sci*.; **8**,1181(2018).
7. B. D. Freeman, N.. *Macromolecules*, **32**, 375–80(1999).
8. E. Sharifzadeh, I. Ghasemi, M. Karrabi, H. Azizi, *Iran. Polym. J.* **23**,835–45(2014).
9. V.C. Tung, M. J. Allen, Y. Yang, R. B. Kaner., *Nat. Nanotechnol.* **4**, 25–9(2009).
10. N. Mahmood , *J. Mater. Chem.* **2**,15–32(2014).
11. Y. Fu, W. Xiong, J. Wang, J. Li, T. Mei, X. Wang., *J. Nanosci. Nanotechnol.* **18**, 3341–7(2017).
12. P. Chem, C. Phys, V. Tozzini, V. Pellegrini, *Phys Chem Chem Phys.* **15**, 80–9 (2013).
13. H. Chang, H. Wu, *Energy & Environmental Science.* **6**, 3483–3507 (2016).
14. DR. Dreyer, S. Park, W. Bielawski, RS. Ruoff, *Chemical Society Reviews The.* **39**,228–40 (2010).
15. E. Al-bermany, D. Qais, S. Al-rubaye "Graphene Effect on the Mechanical Properties of Poly (Ethylene Oxide)/ Graphene Oxide Nanocomposites Using Ultrasound Technique Graphene Effect on the Mechanical Properties of Poly (Ethylene Oxide)/ Graphene Oxide Nanocomposites Using Ultrasound Techn". *Journal of Physics: Conference Series* 2019), pp.1234.
16. W.S. Hummers, R. E. Offeman. *J Am Chem Soc.* **80**,1339(1958).
17. I. V. LIGHTCAP., P. V.KAMAT, S. Storage, and Sensing. *Account of Chemical Research.* **10**,102(2012).
18. AM. Díez-Pascual, Díez-Vicente AL. *ACS Applied Materials and Interfaces.*, **8**, 17902–14(2016).
19. A.M. Díez-pascual, M.A. Gómez-fatou, Ania F. *Progress in Materials Science.* **67**,1–94(2015).
20. Li C, Xiang M, Ye L., *RSC Advances [Internet]*., **6**,72193–200(2016).
21. RF. Faradilla, G. Lee, P. Sivakumar, M. Stenzel, Arcot J., *Carbohydrate Polymers* .**205**, 330–9(2019).
22. R. Padmanaban, K. Venkatramanan, S. Girivel, K. Kasthuri, A. Usharani, R. Vellaichamy. *In Recent Trends in Materials Science and Applications* 723–30 (2017).
23. K. Venkatramanan, R. Padmanaban, V. Arumugam, *Acoustic.. Physics Procedia.* **70**, 1052–6 (2005).
24. E. Al-Bermany, B. Chen, *Surface Adsorption and Interfaces. Doctoral dissertation, University of Sheffield;*(2017).
25. H. Baharvand *Iran. Polym. J.*, **13**, 479–84 (2004).
26. P. Boutouyrie, M. Briet, S. Vermeersch, B. Pannier *Assessment of pulse wave velocity.* 3–8(2009).
27. Z. Wu, DC. Liu, Wu. Zongfang and C. Dong Lin. *Method of improved scatterer size estimation without attenuation known a priori.* 10–3 (2010).
28. A-KJ. Rashid, E. D. Jawad, B. Y. Kadem, A-KJ. Al-Bermany, E. Al-Bermany, B. Y. Kadem. *European Journal of Scientific Research.* **61**, (2016).
29. A-KJ. Al-Bermany, F. S. Hashim, E. D. Al-Bermany, *Chemistry and Materials Research.* **2**, 2225–956 (2012).
30. C. Zhang, L. Wang, T. Zhai, X. Wang, Y. Dan, L. S. Turng. *J. Mech. Behav. Biomed. Mater.* **53**, 403–13(2016).
31. M. Pulst, M. H. Samiullah, U. Baumeister, M. Prehm, J. Balko, T. Thurn-Albrecht, et al. *Crystallization of Poly(ethylene oxide) with a Well-Defined Point Defect in the middle of the Polymer Chain.* *Macromolecules.* **17**, 6609–6620(2016).
32. Y. Han, T. Wang, X. Gao, T. Li, Q. Zhang. *Applied Science and Manufacturing.* **84**, 336–43 (2016).
33. R. Joshi, ND. *Der Pharmacia Lettre.* **10**,126–33 (2015).
34. M. Bhattacharya *Materials.* **9**,1–35 (2016).
35. T. Hornowski, A. Józefczak, A. Skumiel, M. Łabowski, *International Journal of Thermophysics.*, **31**,70–6(2010).
36. N. M. Saeed, *Iraqi journal of science* **53**, 88–95(2012).
37. S. B. Jagtap, R. K. Kushwaha, D. Ratna, *RSC Adv.* **39**, 30555–63 (2015).
38. A. Jain, S. K. Upadhyaya, *Portugaliae Electrochimica Acta.* **4**, 241–8 (2013).
39. Y. Feng, X. Zhang, Y. Shen, K. Yoshino, W. Fengab, Y. Feng., *Carbohydrate Polymers* **87**,644–9 (2012).
40. N. R. Wilson, N. R. Wilson, P. A. Pandey, P. A. Pandey, R. Beanland, R. Beanland, *ACS nano.* **3**, 2547–56 (2009).