Graphene-Reinforced the Structure and Mechanical Properties of New PMMA-PVA Hybrid Nanocomposites

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Abstract. Graphene challenges the logic with its behavior and properties because of its multiple uses, so scientists and researchers paid great attention to it. In this study, newly fabricated nanocomposites consist of polymethyl methacrylate and polyvinyl alcohol using Dimethylformamide -co- distilled water and Dimethylformamide solvent with various loading ratios (0.09, 0.18, and 0.27 wt.%) of graphene oxide nanosheets, applying a developed solution casting method. The microstructure, some physical and mechanical properties of the polymethyl methacrylate - polyvinyl alcohol/ graphene oxide nanocomposites, such as density, viscosity, ultrasonic velocity, coefficient of ultrasonic absorption, compressibility and bulk modulus, etc. were measured. The method exhibited the successful synthesis of these nanocomposites for the first time with good homogeneity and good desperation as existing clearly in the images of the optical microscopy. Additionally, strong interfacial interaction was formed between the polymers and nano-graphene sheets as exhibited in Fourier-transform infrared spectra with the most functional group of polymer and graphene oxide in the nanocomposites. The scanning electron microscope images also confirmed this finding. These images illustrated the smooth and homogeneous fracture surface with good dispersion of the nanosheets of graphene oxide into the polymer mixture. The low loading ratio of graphene oxide in the matrix from 0.09 to 0.27 wt.% exhibited significant improvement of the viscosity of ultrasonic velocity, bulk modulus, and coefficient of absorption of the mechanical waves up to 79%, 63 %, 225%, and 127%, respectively, with the blended polymers. These findings could help to realize these new nanocomposites as promising materials for wide applications, such as car glass and backlight and ultraviolet filters applications.
Keywords. PMMA, graphene oxide, PVA, Mechanical properties, Nanocomposites, Structure.

1. Introduction

The use of polymeric materials has permeated every part of our lives and in all areas. It is arduous to imagine the world today with all its prosperity and comfort without the existence of human-made polymeric materials [1]. Polymer science is emerging, fascinating, and easy to process for many applications. Even with these significant characteristics of the polymer, it suffers from mechanical properties weakness [2,3]. Therefore, researchers have been looking every day to produce new methods

and ways to develop polymers with good quality and properties that could be promising for wide and various applications [4,5].

One of the best and successful ways that have been applied is nanotechnology, where polymer-nanocomposite has been involved more and gets more consideration by researchers, engineers, and the industrial sector to bring the high performance of the new nanocomposite with better properties or as required [6,7]. Enhancing is related to many factors, such as used nanofillers, interface interaction, and properties of components, etc. [3,8]. Where the nanofillers effect on the material properties [9,10]. Assuming that the interaction takes place between the dispersed particles and the matrix, Recently, the carbon family has widely been considered as the most significant nanofiller because of its ability to tunable and improve the properties of the materials. Therefore, graphene is realized as the best nanofillers. It has two-dimension sheets, hexagonally arranged a monocular atomic thick layer and unique physical characteristic, etc. [11,12]. Graphene oxide (GO) has the same structure of graphene with the base of oxygen functional groups, where these groups turned GO to hydrophilic and easy to disperse in water and range of solvents, and have a large surface area [13]. This two-dimensional carbon structure opened vast and new potentials for future devices and various applications [14,15].

Polymethylmethacrylate (PMMA) and poly (vinyl alcohol) (PVA) were used in this investigation as model polymers. A thermoplastic polymer PMMA is an amorphous and transparent polymer [9] with very good properties, especially good resistance to harsh cutting conditions and stable to acid and alkalis [10]. It is used in various applications such as lenses, internal and external lighting of cars [10,11]. In comparison, PVA has important hydroxyl groups, which could mix the polymer in the carbon chain backbone [15-18]. PVA is considered as distinguished from other polymers because of its high corrosion resistance, good thermal stability, and mechanical strength [19] as well as it is widely used in various applications, for instance, sensors, fuel cells, electrochromic and bio-medical fields.

Many researchers, such as X. Zhao, et al. 2010 [20], reported that the PMMA or PVA with GO improves the mechanical properties and thermal stability of PVA using graphene oxide. 10 mL of exfoliated GO solution was added to specific PVA powder and blended using stirred at 85 ^{o}C for about 6 hours. An oven with 60 ˚C was used to dry the samples for 24 hours after it is placed on a glass slide. The mechanical properties of the samples showed significant improvements up to 150 % and ten times of tensile strength and Young's modulus, respectively, with a 1.8% vol loading ratio of graphene oxide. In 2015, T. M. Al-Saadi and M. A. K. Jihad [21] reported the synthesis of graphene/ PMMA nanocomposites films using 20 ml chloroform to dissolve the PMMA. Graphene with various loading ratios (0, 0.1, 0.3, 0.5, 1, and 2) % was dispersed in this PMMA matrix under controlled temperature at 80 ^{o}C for 30 minutes; then, the mixture was cast in the mold by hand lay-up method. The results of FT-IR, XRD, and SEM showed that functional groups of graphene, fully dispersed of graphene flakes in the matrix of the PMMA, and the graphene flakes highly agglomerated state and many wrinkles, respectively.

The contribution of the increase in the concentration of graphene flakes presented a vital factor in increasing the absorption of PMMA. Moreover, this exhibited a reduction in the energy gap from 4 eV to 2.8 eV. Additionally, this incorporation of graphene presented notable enhancement in both the mechanical and optical properties of nanocomposites. This finding showed promising materials for various applications, for instance, solar cells, IR- window, and microwave absorbing. S. Kashyap et al. (2016) investigated the effect of adding different amounts of graphene oxide (GO) on the mechanical and electrical properties of PVA. Distilled water (DW) was applied as a solvent for each of the PVA and GO samples. The samples were prepared using the casting method. The results of the study showed that the tensile strength and the modulus of elasticity were improved by 150%. The enhancement in the mechanical properties was qualified for more vital interfacial interaction between the fillers and the mixture of the polymer. The results also showed that the electrical conductivity was improved from -30 to -35. Different measuring devices were used, such as UV-vis, XRD, and SEM [22]. In 2018, K. Sa et al. investigated the mechanical, electrical, and thermal properties of multiwalled carbon nanotubes (MWCNTs) with reducing GO (rGO) and polymethyl methacrylate (PMMA) hybrid nanocomposites with

the contribution of ionic liquid functionalized multiwalled of the carbon nanotubes (IL-MWCNTs). The solvent-casing method was applied to prepare these samples, and the result showed a significant influence of (IL-MWCNTs) on the tensile strength and modulus of elasticity. These mechanical properties were enhanced by up to 119 % and 58 %, respectively. Meanwhile, thermal properties became more stable after the incorporation of IL-MWCNTs in the nanocomposites, where strong interaction were formed between the IL-MWCNTs with RGO for side and with the matrix from another side [23].
All of the reviewed papers [20–23] were investigated one polymer type, either PMMA or PVA alone, with GO nanosheets. However, there is no investigation reported mixing these polymers with GO to the best of our understanding. This study aims to improve the structure and mechanical properties of theses polymers by preparing the nanocomposite (PMMA-PVA/GO). These properties exhibited a significant enhancement as it is characterized by several devices, such as multi frequency-ultrasound measuring, Fourier-transform infrared spectroscopy (FTIR), optical microscope (OM), and scan electron microscope (SEM).

2. Experimental part

2.1. Materials

PMMA (99% purity, 20000-18000 g mol-1, and 213 ºC melting point) and PVA (99% purity, 18000-12000 g mol-1, and 230 ºC melting point) were provided by Tuttlingen Company and Panreac Company, Spain, respectively. Graphite (≤ 39μm), Sodium nitrate ($NaNO_3$), sulfuric acid (H_2SO_4) (99.5% analytical grade), hydrogen peroxide, potassium permanganate ($KMnO_4$), and hydrochloric acid (35%) were purchased to synthesis graphene oxide form Sigma-Aldrich, United Kingdom.

2.2. Graphene oxide nanosheets synthesis

Our group was predicated the graphene oxide nanosheets (GONs), and the full characterization was available in the literature [3,8].

2.3. Syntheses the nanocomposites [15]

Various ratios of PMMA-DMF and PVA-DW/DMF were applied to get the appropriate methods to mix these two polymers with different solvents. The various ratios of PMMA-DMF and PVA-DW/DMF were applied to get the appropriate methods to mix these two polymers with different solvents. Firstly, we reported the methods that accessed not only mixed two of these materials as a fine homogeneous matrix but also with good GO dispersion as briefly in the following steps: PVA (1 g) was dissolved in 50 ml using distilled water (DW), were stirring for one hour at 80 ± 3 ^{o}C [15]; meanwhile, PMMA was dissolved in DMF with 6 g in 50 ml (DMF) using a magnetic stirrer for three hours at 80 ± 3 ^{o}C. Simultaneously, GO nanosheets were dispersed in DMF with 0.09-0.27 wt.% in 50 ml using a magnetic stirrer and sonication bath as an acoustic solution method for good dispersion of GO in the solvent. All these steps were carried out separately before mixing the materials. After the complete dissolving of the PVA in DI, 10 ml of DMF were added to 30 ml of PVA-DI solution then they were mixed using a magnetite stirrer for 30 minutes at 80 ± 3 ^{o}C for better homogeneity. 5 ml PVA-DIS-DMF were added to 25 ml of PMMA-DMF solution and mixed for 4 hours using a magnetic stirrer at 80 ± 3 ^{o}C and the polymers matrix was fine homogenous during this time. Then, the temperature of the PVA-PMMA matrix was reduced to about 35 ± 3 ^{o}C before adding three ratios of GO-DMF to prepare three nanocomposites. After the addition of GO, the mixing-sonication methods were applied. The mixing of the PVA-PMMA/GO matrix was continued for another three hours; then, it was sonicated for 15 minutes. This procedure was repeated four times to get acceptable homogeneity and dispersion of GO in the polymers matrix. Table 1 summarizes the fabrication and preparation methods of (a) GO, (b) PMMA-PVA1, (c) PMMA-PVA/GO2, (d) PMMA-PVA/GO3, and (e) PMMA-PVA/GO4 nanocomposites.

Table 1. The purification methods of (a) GO, (b) PMMA-PVA1, (c) PMMA-PVA/GO2, (d) PMMA-PVA/GO3 and (e) PMMA-PVA/GO4 nanocomposites.

Sample ID	Concentration Wt. %			Total Time h		Drying method
	PVA	PMMA	GO	Sonication	Mixing	
GONs	0	0	100	4	4	Freeze drier
PMMA-PVA1	4	96	0	0	4	40 ± 3 C° under air
PMMA-PVA/GO2	3.91		0.09	4	12	
PMMA-PVA/GO3	3.82		0.18	4	12	
PMMA-PVA/GO4	3.73		0.27	4	12	

2.4. Characterization

Nikon, Olympus model 73346, was used to record the Optical Microscope (OM) images. Fourier transforms infrared **(**FTIR) spectra (Vertex 701) from Bruker, Germany, was utilized to characterize the samples in the region between 4000 - 400 cm^{-1}. The AC electrical conductivity was recorded using an LCR meter (Hi TESTER, HIOKI 3532-50, Japan) with a variable frequencies range of (100Hz-5MHz) that was applied to characterizes the measurement for all samples under the room temperature.

3. Theoretical part

Relaxation time (t) was considered for the nanocomposites using Equation (1) [24,25]:

$$t = \frac{4\eta_s}{3\rho V^2} \tag{1}$$

Where η, p, and V are the viscosity, density, and the ultrasound wave velocity, respectively. Meanwhile, the relaxation amplitude (D) is considered using the following relation .

$$D = \alpha / f^2 \tag{2}$$

The relaxation amplitude is depended on the relaxation frequency (f) and the coefficient of absorption (α). Equation (3) was applied to calculate the impedance audio quality (z).

$$\mathrm{z} = \rho \mathrm{V} \tag{3}$$

Laplace equation was used to measure the compressibility (B) using Equation (4).

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

Whereas, the inverted compressibility represents the elasticity bulk modulus (k) that is exhibited in Equation (5).

$$k = {}^{1}/_{B} = \rho V^2 \tag{5}$$

4. Results and discussion

Density was measured for the polymers and nanocomposites, as revealed in Figure (1). The results displayed an increase in the density, which is due to the increase in the mass of the polymers and the swelling of the polymer particles as a result of their dissolution in distilled water with the addition of the concentration of nanosheets of graphene oxide, which led to the formation of a complex nanostructure with the polymer in the matrix. The contribution of increasing the GO concentrations shows a gradual

improvement in the density values up to 75 % with the rise of GO ratio from 0.09 to 0.27 wt% compared with PM-PV results; these results agreed with the other investigation finding [2].

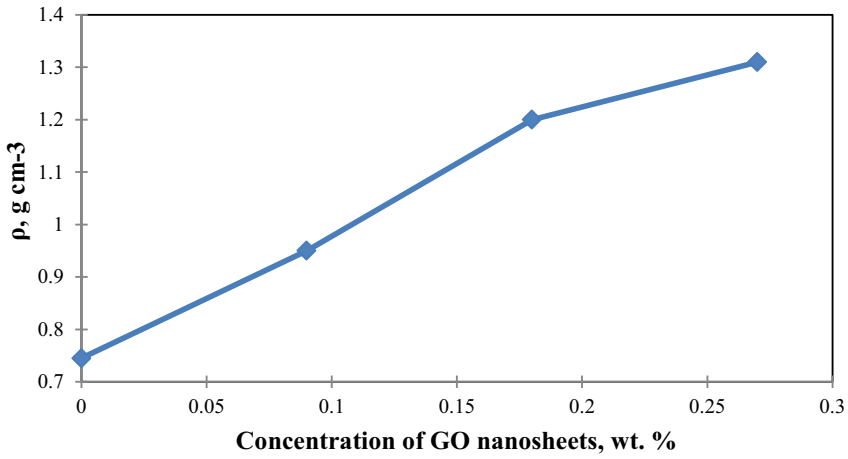

Figure 1. The density of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

Shear viscosity was measured practically by using a viscometer type (NDJ-8S), and Figure (2) displays the change of the shear viscosity with increasing the concentration of GO nanosheets. It is noticed that the viscosity results were improved after increasing the loading ratio of GO because of the transformation to a complex form. This was helped in the formation of large-size molecules of the polymeric chains due to increasing the GO ratio in the matrix. This increase exhibited an increase in the rotation and translational friction forces among the molecules of polymers with the solvent [8] from a side and between the nanosheets of GO from the other side. The reason for this was the increase in the hydrogen bonds formed between the dissolving polymers and its solvent, in addition to the bonding with the nanosheets of graphene oxide. This resulted in the formation of lattice structures and enhanced the structure and all properties [26].

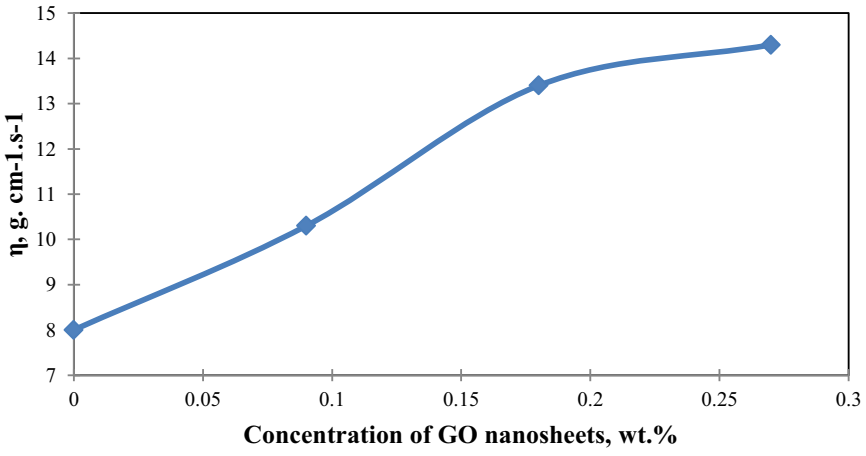

Figure 2. Shear viscosity of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

Ultrasound velocity (V) was measured by dividing the distance value of the traveling waves inside the polymer solution on the delay time. Figure (3) shows the result of the relation between the velocities of ultrasound with the change in the GO concentration. It can be noted that the ultrasound speed increases as the concentration of GO in the polymer matrix increased. The reason was the improvement in the crosslinking due to the interaction which produced the union between the molecules of two types of polymer with its solvent; it resulted in the formation of large (macromolecular) particles within the films. On the other hand, the bonding of carbon groups in both polymers with the carbonyl or carboxyl groups of GO led to the formation of bonds with hydrogen bonds and enhanced the nanocomposite mechanical properties as an outcome of these interactions [27].
It was shown here that the stacking is better with graphene oxide nanoparticles that mechanical waves transmitted from the disturbance source in the form of wave packets. In general, all of the graphene oxide concentrations increase the ultrasound velocity [8,27], and these results are consistent with the other researchers' results [3,24,28].

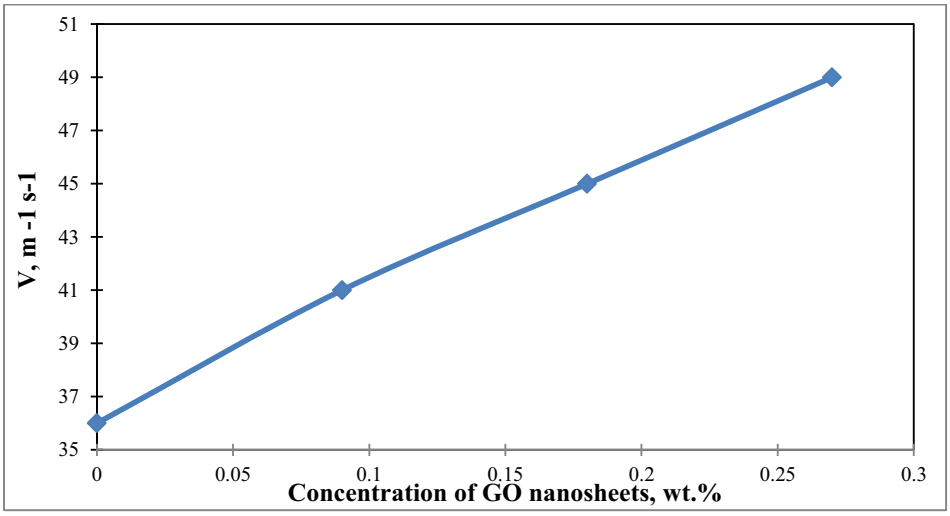

Figure 3. Ultrasound velocity of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

The absorption coefficient of the ultrasound results was calculated by considering the change in the results between the sender and receiver of the ultrasound waves of PM-PV and PMMA-PVA/GO nanocomposites. Figure (4) illustrates the improvement in the absorption coefficient with increasing the ratio of nanosheets of graphene oxide, where the absorption coefficient depends on viscosity, thermal conductivity, and dispersion. The viscosity is considered responsible for enhancing the ultrasound wave absorption. Where any improvement in the viscosity values was associated with increases in the coefficient of absorption as it depends heavily on the ratio in the matrix [3,24].
It was noticed from Figure (4) that an improvement in the coefficient of absorption after adding GO nanosheets, where these are mechanical waves and are transmitted in the medium as compression and vacuum. So, upon addition, the size of the polymeric chains increases and their association with the nanosheets of graphene oxide; thus, increasing their absorption to the incident mechanical waves energy in response to the movement of compression and loosening within the membrane. This behavior is consistent with what the previous researchers obtained [29–31].

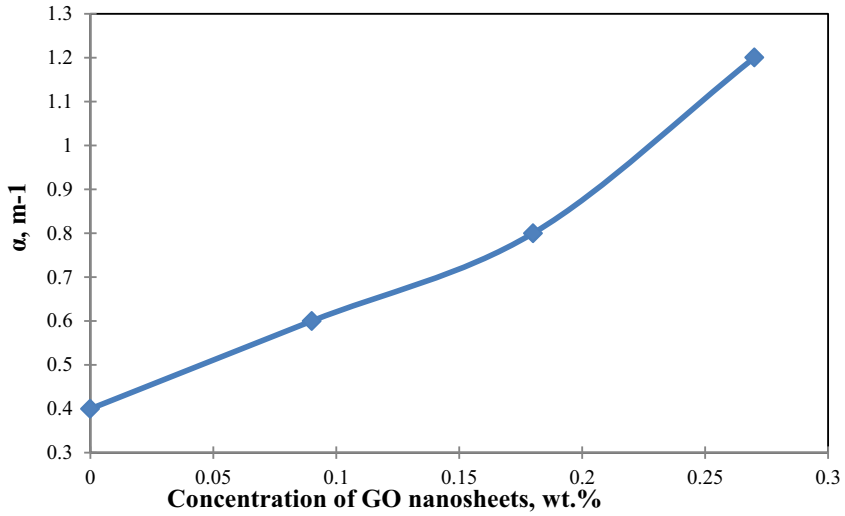

Figure 4. The ultrasonic absorption coefficient of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

The results of relaxation time were measured using the values of the measured density, viscosity, and ultrasound velocity according to the Equation (1). Figure (5) demonstrates the results of the relaxation time with increasing the ratio of GO nanosheets. The relaxation time was reduced with the increase in the loading ratio of GO nanosheets, where due to the increase in the size of the polymeric chains could lead to an increase in the internal friction between the liquid layers resulting from compression and rarefaction as a consequence of the influence of the ultrasound waves. Where the time is essential to return the excited molecule to its first (original) position, and that was reduced with increasing the graphene oxide concentration, which bound with the polymer by hydrogen bonds. This interaction between polymers and GO is associated with more cohesion within the compound. The strength of the mechanical properties of graphene oxide nanoparticles reduces the relaxation capacity. It limits the movement of the polymer molecules, thus reducing the oscillation of the atoms [3].

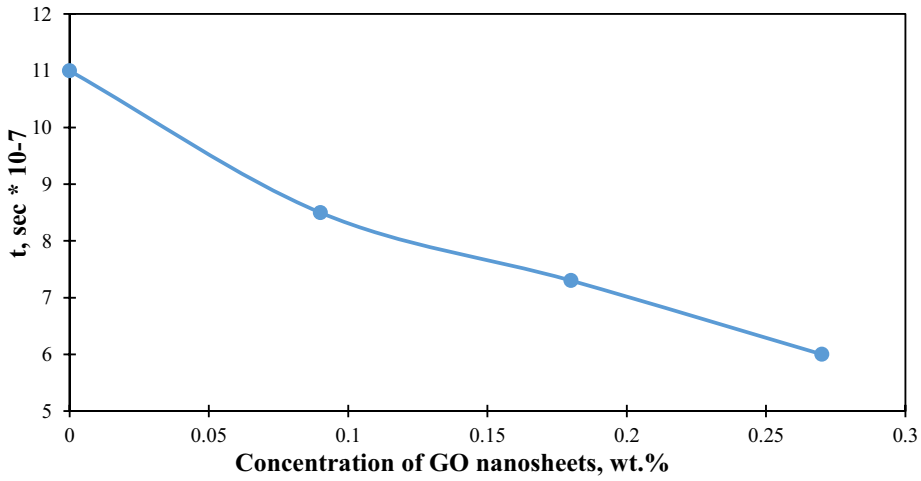

Figure 5. The relaxation time of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

Equation (2) described the relaxation amplitude, and it was used to obtain its value plotted in Figure (6), which illustrates the results of relaxation amplitude with increasing the loading ratio of GO nanosheets in the polymer matrix. It was noticed that the results of the relaxation amplitude were improved with the increase in the loading ratio of the GO nanosheets. The reason was that the considerable traveled distance of the molecule during the excitation process; it could also be related to the big inertia moment of the large molecule [32]. Besides, the values of relaxation amplitude are dependent on the absorption coefficient of the wave energy as directly proportional, which moved faster with rising the ratio of graphene oxide in the superposition, as shown by Equation (2). For both cases, when the frequency is constant, it is estimated to improve the results of the relaxation capacity of the solution particles when the absorption coefficient is increased. These results are consistent with the previous researchers' findings [2,3,29].

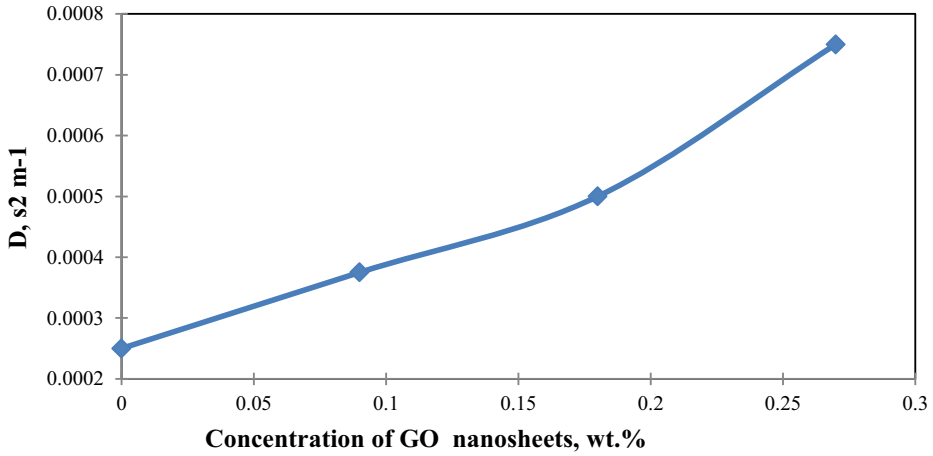

Figure 6. Relaxation amplitude of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

The results of the compressibility were achieved based on Equation (4). The increase in the velocity with concentration reduced the compressibility, where it is proportional to the square velocity. Figure (7) demonstrates the change in compressibility values with increasing the ratio of the GO nanosheets. The reason for this reduction is related to the fusion of two molecules types. This caused a convergence of polymeric chains that are converging each other in addition to the nanosheets of graphene oxide, and this is consistent with what the previous researchers obtained [31,33].

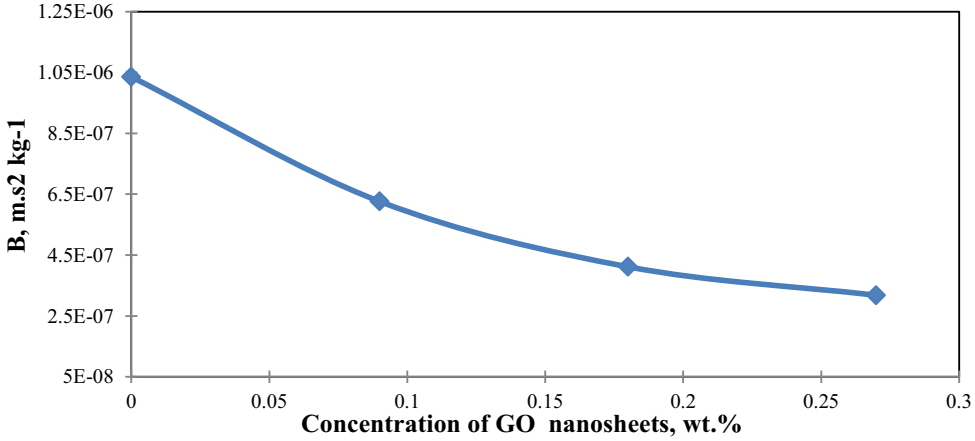

Figure 7. Compressibility of PMMA-PVA1 and PMMA-PVA/GO nanocomposites.

Equations (3) and (5) were used to calculate the values of the modulus of elasticity (K) and the specific acoustic impedance, and the results were summarized in Table 2. As it can be noticed from this Figure, an increase in the values of the mentioned parameters after the addition of the GO, and these results were improved with increasing the GO concentrations in the nanocomposites. The modulus of elasticity is mainly dependent on the velocity of ultrasound; therefore, it is expected that the modulus of elasticity will be increased due to the elastic ability of graphene oxide nanosheets. This result is consistent with the results of the researchers of reference [32].

Table 2. Summarized the values of the elasticity bulk modulus and specific acoustic impedance with increasing the GO concentration of PMMA-PVA1 and PM-PVGO nanocomposites.

Samples	GO ratio (wt. %)	Bulk Modulus (N m^{-2})	Specific Acoustic Impedance (kg sec m^{-2})
PMMA-PVA1	0	965520	26820
PMMA-PVA/GO2	0.09	1596950	38950
PMMA-PVA/GO3	0.18	2430000	54000
PMMA-PVA/GO4	0.27	3145310	64190

Figure 8 shows the FT-IR spectrums range between 4000-500 $cm^{-1}$of GO, PMMA-PVA1, and PM-PVGO nanocomposites. GO FTIR spectrum has revealed that a normal functional peak in agreement with several studies [3,15,34,35], as revealed in Figure (8a). PMMA-PVA blend exhibited peaks as CH_2, C-H stretching, O-H bending at 1646 cm^{-1}, O-H bending and C-O (ester bond) stretching vibration at 1604 cm^{-1}, 1361 $cm^{-1,}$ and between 1260-1000 cm^{-1}, respectively. These are related to the net of both PMMA [33] and PVA [36]. The contribution of the GO in the polymer matrix is associated with the exhibited new strong bands at 2904, 1604, and 887 $cm^{-1,}$ as clearly displayed in Figure 8 (c, d, and e). Increasing the loading ratio to 0.27 wt % caused shifting up to 10 cm^{-1} of the most functional groups of PMMA-PVA/GO4 nanocomposites and increasing the intensity peaks because of the addition of the nucleating agent PMMA-PVA/GO nanocomposites. This is related to the hydrogen bonds formed between the O-H and the C=O in the GO with the functional of PMMA-PVA [15,37].

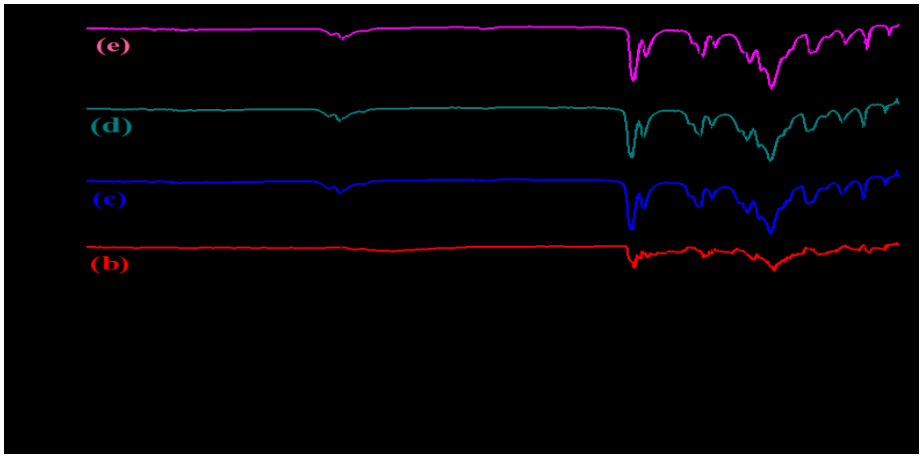

Figure 8. FTIR Spectra of (a) GO, (b) PMMA-PVA1, (c) PMMA-PVA/GO2, (d) PMMA-PVA/GO3 and (e) PMMA-PVA/GO4 nanocomposites [15].

In Figure 9, the XRD patterns of GO and PMMA-PVA/GO4 nanocomposites are illustrated. At 2θ = 11.1°, a sharp peak exists of GONs with 002 diffractions and 0.79 nm an interlayer spacing. This shift

confirmed the synthesis of GO in agreement with other investigations [34,38]. Two sharp broad and peaks at At 2θ = 14.1° and 30° with (111) and (112) diffractions were related to PMMA-PVA1. These peaks are linked to the PMMA in agreement with the studies in the references [38,39]. The GO and PVA peaks are located at 2θ = 11.1° and 2θ = 19.8°, respectively, where both are overlapped with a wide and sharp intensity pattern of PMMA between 7° - 23°; therefore, these peaks were challenging to present.
Otherwise, increasing the loading ratio of GONs from 0 wt.% to 0.09, 0.018 and 0.27 wt % associated to shifting in the PMMA peaks from 2θ = 14.1° to 14.2°, 15° and 16.1° of the PMMA-PVA/GO2, PMMA-PVA/GO3 and PMMA-PVA/GO4, respectively. This could be related to expanding the volume in the macro and the increment of interplanar crystal spacing [40]. These XRD findings support the formulation of strong interfacial interaction between GO and polymers presented in the FTIR spectra and substantial agreement with the finding in the literature [40,41]. In Figure (8 c, d, and e), the new formulation of firm functional peaks at 2904, 1604, and 887 cm^{-1} were exhibited after the contribution of GO. Their intensity also increased with increasing the loading ratio of GO and shifting in up to 10 cm^{-1} of the most functional groups of PM-PV/GO4. This shifting in the position peaks and new firm peaks could be related to the formation of the strong interaction between the polymer and functional group on the surface of GO. This connection is clearly exhibited between the O-H and the C=O in the GO and the PMMA-PVA related to the formulation of the hydrogen bonds between the O-H in the GO with the C=O in the PMMA-PVA, respectively [37].

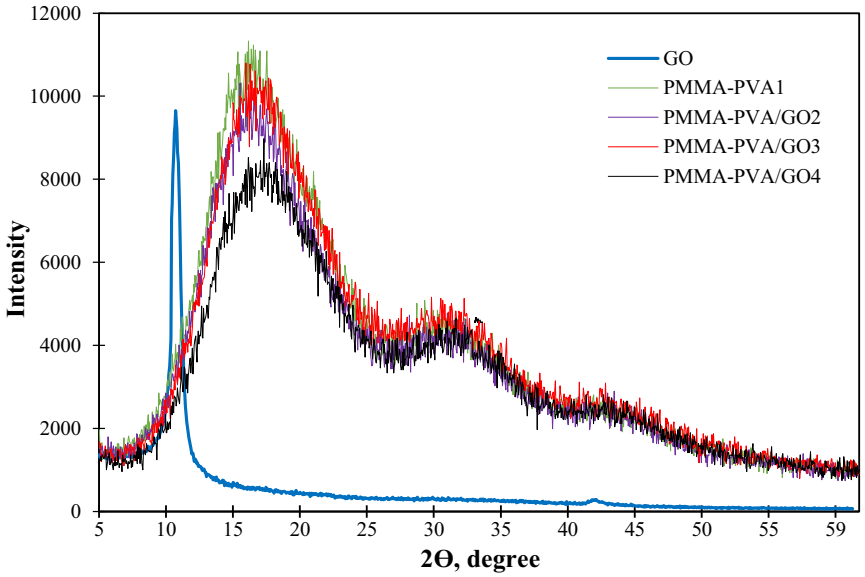

Figure 9. XRD Patterns of GO, PMMA-PVA1, and PM-PVGO nanocomposite.

The optical images for the nanocomposites with magnification power at 40 X have been recorded, as revealed in Figure 10. These images show the high homogeneity between the polymers, as illustrated in Figure (10a). GO was find distribution in the polymer matrix, where increasing the loading ratio of GO was not affecting the distribution without any aggregation. Conversely, the homogeneity was more significant with increasing the concentration of graphene oxide nanosheets, as shown in Figure (10d) related to PMMA-PVA/GO4. Moreover, the contribution of GONs does not influence the transparency of the nanocomposites.

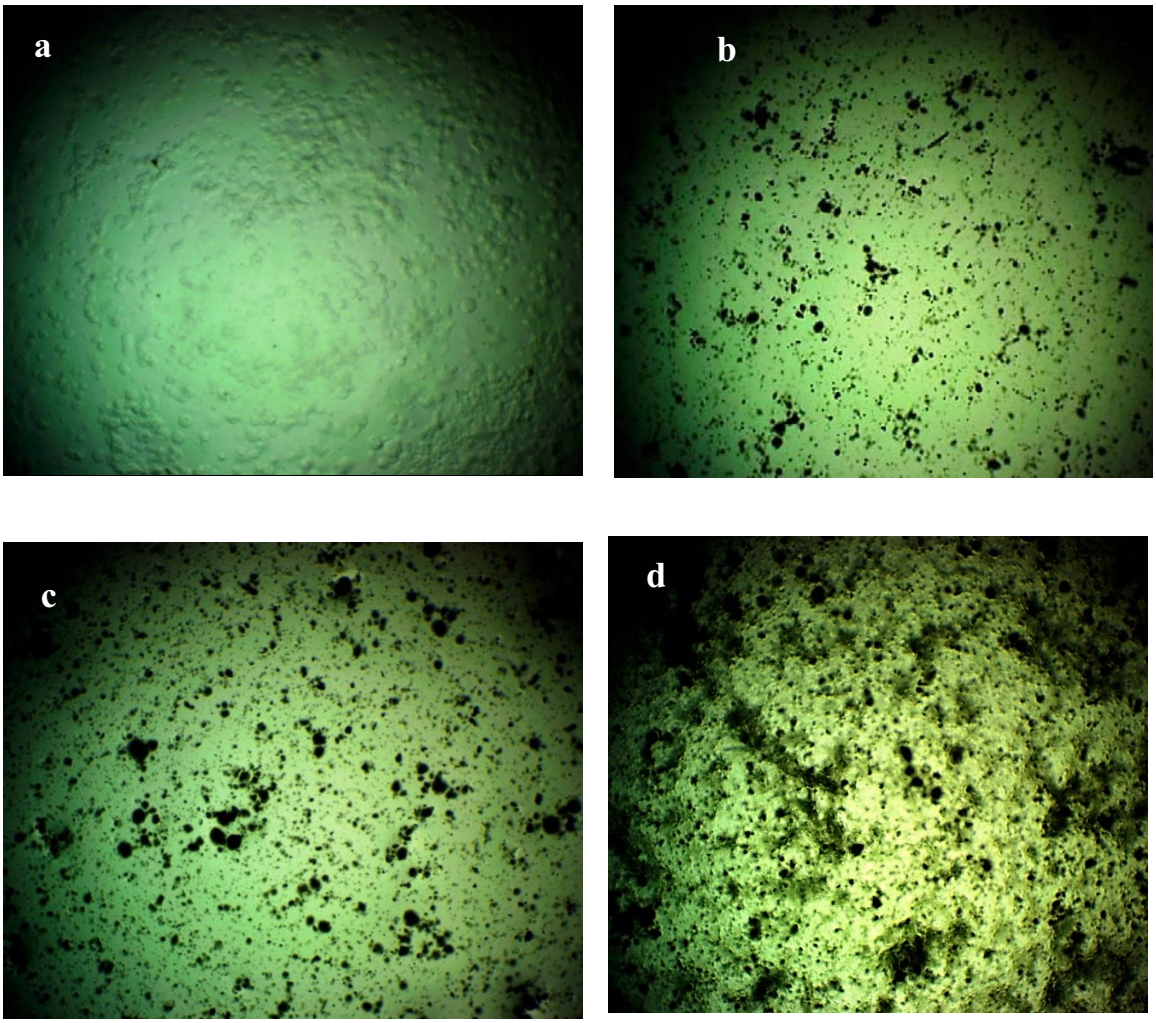

Figure 10. The images of the optical microscopy with a magnification of (40X) of (a) PMMA-PVA1, (b) PMMA-PVA/GO2, (c) PMMA-PVA/GO3, and (d) PMMA-PVA/GO4 nanocomposites.

In Figure (11), the morphology and the surface fraction of the nanocomposites were investigated using the SEM. SEM image in Figure (11a) displays uniform morphology revealing homogeneous polymers surface. The contribution of GO led to changes in the morphology of the surface and presented right distributed and spread densely on the surface; this may be indicating the occurrence of a homogeneous growth mechanism of GO without aggregates, as shown in Figure (11 b, c, and d). The size measurements of GO nanosheets were presented on the surface of nanocomposites of the SEM images. These were between a few nanometers to few microns, as exposed in Figure (11 e, f, and h).

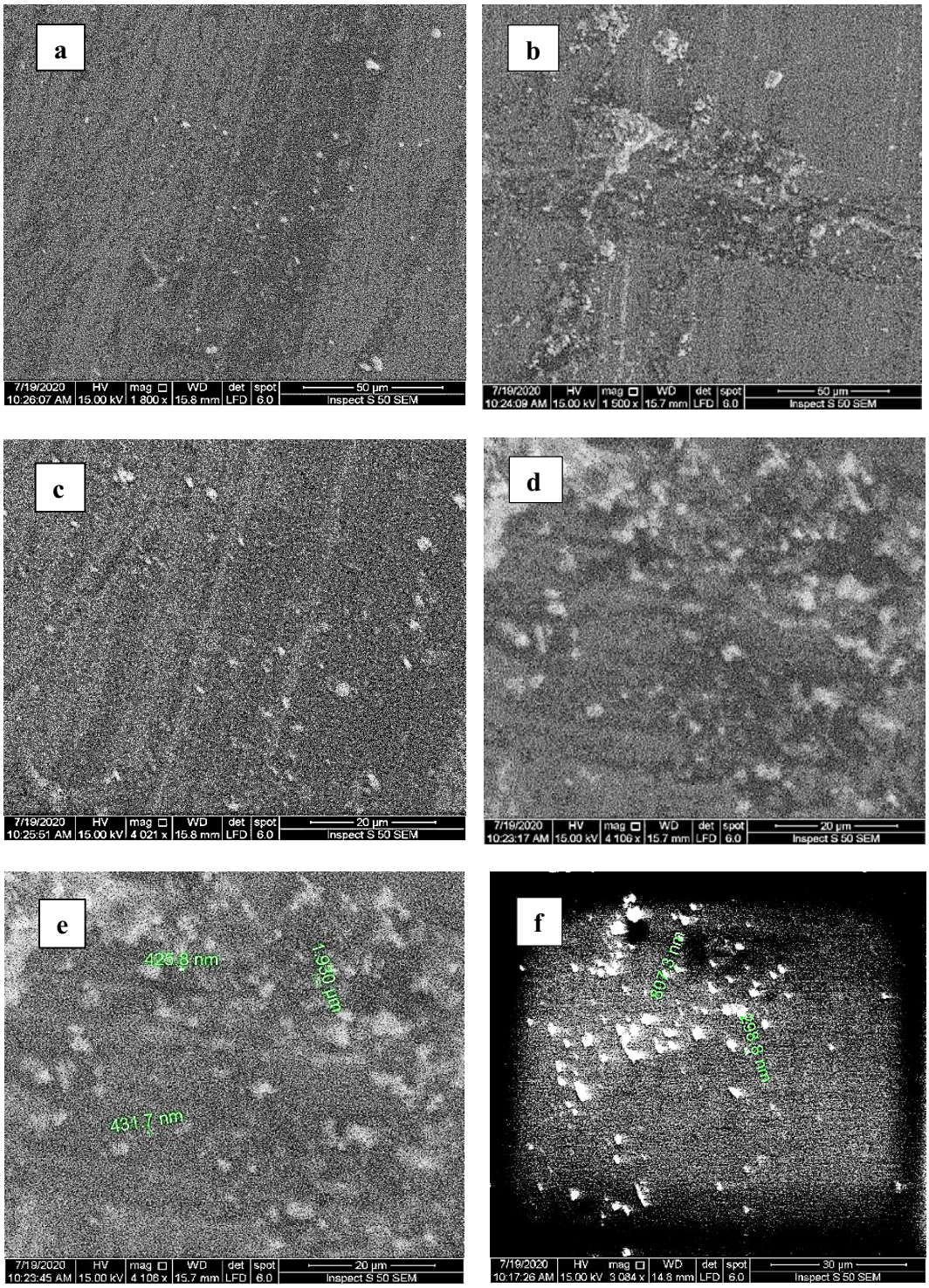

Figure 11. SEM image of (A) PMMA-PVA1, (B) PMMA-PVA/GO2, (C) PMMA-PVA/GO3, (D) PMMA-PVA/GO4 nanocomposite, and (E, F, and H) present the nanosize calculation of GO in nanosheets.

5. Conclusions

The used method was helped to fabricate new nanocomposites from PMMA, PVA, and GO successfully and investigate the structure and mechanical properties using ultrasound. FTIR and XRD exhibited strong interfacial interactions between both polymers and GO with fine homogenous and good dispersion of the GO nanosheets in the nanocomposite, as shown in the OM and SEM images. The contribution of GO and increasing the loading was associated with notable improvement in the most mechanical properties due to the modification of the structure. Where the mechanical properties such as ultrasonic velocity, absorption coefficient and bulk modules were improved by up to 36 %, 127 % and 225 %, respectively. This investigation helped to understand these materials when they are mixed for the first time and, also it reduced the gap of knowledge and discovered promising materials for new and promising applications, such as car glass and backlight, and filters ultraviolet.

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