Improved Physical, thermal, and conductivity strength of ternary nanocomposite films of PVDF/PMMA/GO NPs for electrical applications

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Abstract
Polymer nanocomposites samples polyvinylidene fluoride/Polymethyl methacrylate (PVDF/PMMA) mixed with various concentrations of graphene oxide nanoparticles (GO NPs) were created using solution casting and ultrasonic-assisted solution casting processes. The characteristics of the manufactured polymer samples were studied using XRD, FTIR, TEM, TGA, and DSC. The AC conductivity, dielectric constant, and dielectric loss of PVDF/PMMA-GO nanocomposites were examined as a function of GO concentrations. XRD analysis demonstrates the semicrystalline PVDF/PMMA and shows that the doped PVDF/PMMA films have a lower crystallinity degree (Xc) than the pure blend. The interaction between the GO ions and the PVDF/PMMA matrices is demonstrated by significant fluctuations in the FTIR spectra. TEM revealed that the GO nanoparticles were produced in spherical and cubic shapes, with an average size of 18.6 nm. The energy gap of nanocomposite films reduced as the GO NP’s concentration in the polymer mix increased, decreasing from 4.24 eV to 3.71 eV for the indirect transition. The PVDF/PMMA exhibits a single glass transition temperature (Tg) according to the DSC study, showing that the PVDF and PMMA are miscible. The melting point of the nanocomposite films increased from 244.39 °C to 251.56 °C, increasing their thermal stability. Thermogravimetric analysis (TGA) curves showed that the PVDF/PMMA polymeric matrix had increased thermal stability. Dielectric constant and loss analyses have been performed to gain a better knowledge of charge storage properties and conductivity relaxation. The highest ionic conductivity of PVDF/PMMA blended with 0.08 wt. GO NP’s is $\sim 7.23 \times 10^{-5}$ S/cm. These findings are expected to have a considerable effect on a wide range of applications, such as polymer solar cells, nanoelectronics, polymer organic semiconductors, and energy storage.

Keywords Optical band gap · Activation energy · Thermal stability · XRD · UV–vis · AC conductivity

Introduction
The scientific community has been working for decades to develop composite materials that have unique properties not found in individual components [1, 2]. Polymer composites embedded with metal nanoparticles have been employed as sensors, batteries, and a variety of other industrial applications due to their unique properties [3–5]. Metal nanoparticles are important in influencing polymer composite characteristics [6]. The electrical characterization of the resulting composites is improved by including a conductive nanofiller in the virgin polymer chain [7–9].

Polyvinylidene fluoride (PVDF) is a semicrystalline polymer with a long chain of (CH$_2$-CF$_2$) repeat units. PVDF has a high dielectric constant due to its piezoelectric and pyroelectric capabilities, as well as the presence of fluorine atoms and the C–F bond. When compared to other polymers, it demonstrates tremendous electroactive properties [10, 11]. Some of the physical and electrical properties that make this material better are its excellent dielectric properties, its chemical and thermal stability. It also has a good melting viscosity, making it appropriate for use in a variety of applications without the need for additional stabilizers or additives. A polar organic compound, on the other hand, is the only solvent that can dissolve the PVDF pure polymer [12].

Polymethyl methacrylate (PMMA) is a chemically resistant material with excellent optical transparency. PMMA is
widely used as a transparent cover for a variety of applications, including specific electronic devices. However, its applicability is limited due to its low heat resistance, brittleness, and susceptibility to stress cracking in the majority of organic solvents. PMMA is widely utilized in a variety of industries due to its excellent optical properties, chemical inertness, strong spectroscopic capabilities, thermal and electrical stability, and simplicity of shaping and forming [13].

Using metal nanoparticles in electronics and medicinal research has been a long-standing focus of scientists for more than a century [14]. In recent years, novel synthetic and modification technologies have enabled the creation of fascinating polymers, ligands, medicines, and antibodies that can be mixed with these nanoparticles to create new and exciting applications [15]. Nanoparticles such as graphene oxide nanoparticles (GO NP’s) are one of these nanoparticles that exhibit extraordinary catalytic activity, and their interaction with supramolecular systems has gotten a lot of attention recently [16]. Increasing the functioning of a polymeric chain, on the other hand, necessitates its modification with the inclusion of additional agents [17]. Graphene oxide (GO) is a high-performance reinforcing material for polymers due to its two-dimensional (2D) nanosheet structure [18]. Graphene oxide also has a large specific surface area, excellent chemical stability, and high electrical conductivity [19, 20]. The high surface area to volume ratio of GO, as well as its biocompatibility and corrosion resistance, make it an excellent candidate for incorporation into a nanocomposite [21]. There are two ways in which GO can be toxic: physical disturbance and oxidative stress [22]. Graphene oxide’s mechanical characteristics can withstand stress, which enhances chemical adhesion and interfacial bonding [23, 24]. Our research has shown that metal nanoparticles enhance the polymer’s structural and physical properties [25]. Developing polymeric systems with excellent electrical conductivity is a major goal in polymer research. This is because they might be used in solid-state batteries as both an electrolyte and a separator. Polymers such as PEMA, PS, PVDF, PVP, and PMMA have been used as hosts in this process. PVDF and PMMA were chosen as the polymer hosts in this study to analyze the influence of modifications and verifications in the chemical structure of segments of PMMA on miscibility and compatibility with PVDF, as well as the structure–property relations in these composite materials before and after adding GO NP’s.

The main aim of my article was to present a novel ternary nanocomposite of PVDF/PMMA/GO NPs to improve the electrical properties of polymeric mixture. Graphene oxide nanoparticles/nanosheet were incorporated in PVDF/PMMA blend at various concentrations; (0.0, 0.02, 0.04, 0.06, and 0.08 wt.%). Then investigate the produced PVDF/PMMA/GO NP’s samples by various techniques. Ac conductivity of the obtained PVDF/PMMA/GO NP’s samples was studied using AC as a function of frequency.

**Experimental work**

**Materials**

ACROS Company is selling us PVDF polymer powder with a molecular weight of 350,000 g mol−1. The Sigma-Aldrich Company supplied PMMA with a wt. of 150,000 g mol−1. Graphene oxide NP’s having a surface area of 130 to 160 m2/g were given by Sigma-Aldrich in a 25 nm particle size. Graphene oxide nanoparticles/nanosheet were purchased and were not prepared in our lab because the aim of our study is to enhance some properties of polymer blend for use in different industrial applications. In the preparation of samples, double-distilled water was usually used as a solvent.

**Creation of PVDF/PMMA-GO nanocomposite**

The PVDF/PMMA-GO nanocomposite films were made using a casting solution technique. With the use of a magnetic stirrer, PVDF and PMMA was dissolved in DMF (Dimethyl Formamide) to create PVDF/PMMA samples with a concentration of 50 wt.% PVDF and 50 wt.% PMMA. Ultrasonication was used to disperse GO NP’s nanofiller (0.02, 0.04, 0.06, and 0.08 wt. % GO NP’s) in double-distilled water for 25 min until an aqueous solution was formed. After that, the nanofiller aqueous solution was combined with PVDF/PMMA virgin polymer and ultrasonically processed for another 45 min to complete the process. Until the solution became homogeneous, the solution was vigorously stirred. To dry the prepared films, the Petri dish was placed in a 65 °C oven for four days. The samples were stored at room temperature (RT) after being removed from the Petri plates and stored in a vacuum desiccator. The thickness of the prepared samples ranges from 0.06 mm to 0.08 mm.

**Characterization technique**

A spectrophotometer (Nicolet iS10, USA) was used to measure Fourier transform infrared spectra (FTIR) in the 400 cm−1 to 4000 cm−1 range at room temperature (RT). The X-ray diffraction pattern of the acquired samples was also scanned using XRD (PANalyticalX’PertPRO) with Cu Kα radiation (λ=0.15406 nm) at 5° to 80°. The shape and size of GO NP’s were examined using a transmission electron microscope (JEOL 1200 EX equipment at V = 120 kV Japans). At room temperature, the UV–Vis spectra of polymer nanocomposites samples were recorded with a JASCO UV–VIS Spectrometer (Model V-630, Japan) automated recording double beam spectrophotometer in the wavelength range from 200 to 800 nm.
range 200–1000 nm. DSC was performed on the acquired films using a (SITARAM labrys TG-DSC 16) at a heating rate of 10 °C/min from 30 to 500 degrees Celsius. Thermogravimetric analysis (TGA) was used to assess the films’ thermal stability. During the examination, each design was heated from a coating of gold sputtered on it to avoid the charging effect. Using Broadband dielectric spectroscopy, a Novocontrol Turnkey Concept 40 System (Novo Technologies) is utilized to measure impedance from 0.1 Hz to 20 MHz.

Results and discussion

X-ray diffraction technique (XRD)

The XRD technique has been used to look at the formation of a pure blend and a nanocomposite. It has also been used to look at how different amounts of GO nanoparticles affect the PVDF/PMMA formation. The X-ray diffraction pattern of GO nanoparticles is depicted in Fig. 1. The peaks at 2θ = 26.49°, 27.91, 42.27°, 43.26, 44.53°, 54.77°, and 77.69° are attributed to the Miller’s index of (002), (201), (101), (102), (004), (006), and (102), respectively. The diffraction peaks of GO nanopowders are quite similar to the JCPDS card number 41–1445, which is indexed to the cubic spinel crystal structure of GO NP’s [26]. The Scherrer relation was used to measure the size of the GO crystal, which is 25 nm [27].

\[ D = \frac{K\lambda}{\beta \cos \theta} \]  

where D is the crystal size, K is a dimensionless constant near to unity, \( \lambda \) is the XRD wavelength equal to 1.54 Å, \( \beta \) is the full width at half maximum, and \( \theta \) is the Bragg angle.

Figure 2 depicts the XRD patterns for PVDF/PMMA virgin polymer and PVDF/PMMA/GO nanocomposites samples. The broad peak in Fig. 1 centred on 2θ = 20.38° suggests that the polymer blend (PVDF/PMMA) is semicrystalline and contains both amorphous and crystalline regions. According to Hodge et al., the amorphous broad band is reduced (particularly at 0.08 weight percent GO NP’s), which may be due to the disruption of the PVDF/PMMA polycrystalline structure caused by the GO nanoparticles [28]. In their study, Hodge et al. said that when nanofillers are added, the intensity of the X-ray diffraction film gradually reductions attributed to its amorphous nature. The flexible backbone of amorphous polymers demonstrates that their amorphous nature results in higher ionic diffusivity and enhanced ionic conductivity [29]. In doped samples, we discovered a new sharp peak of 2θ = 26.49°, which is in agreement with XRD peak of GO nanoparticles and was assigned to the (002) plane of cubic spinel structure according to (JSPDS: 41–1445), indicating that the GO NP's

![Fig. 1 The XRD pattern of graphene oxide nanoparticles](image)
interact with the polymeric matrix, most notably with the functional group of the polymer composite C = O, as shown in FT-IR spectrum [30]. The appearance of a 2θ = 26.49° diffraction peak confirms the presence of GO NPs inside the polymer matrix. The other diffraction peaks of GO NP's did not appear in nanocomposite samples due to their low intensity. The crystallinity of the nanocomposite samples was raised by the addition of (0.02, 0.04, 0.06, and 0.08 wt. percent GO NP's) as a result of the complicated creation of the local ordering in the polymer chain. The creation of GO NPs, which fill the interstitial spaces between the amorphous phase polymeric matrix and connect them to the H bond, is due to charge transport activities [31, 32].

**FTIR investigation**

FTIR spectroscopy can be used to investigate intermolecular interactions between groups in a self-polymer or several polymer molecules in a polymeric matrix. Intermolecular interactions like dipoles and hydrogen bonding can cause functional groups to shift their absorbance or frequency [33]. Figure 3 shows FTIR spectra of PVDF/PMMA pure blend and PVDF/PMMA filled with various concentrations of GO NP's in the 4000–1400 cm⁻¹ range. After incorporating GO NP's into a PVDF/PMMA polymer blend, the characteristic transmittance frequencies change. The asymmetric and symmetric C-H stretching (St.) of the methyl groups were attributed to the transmission bands at 2991 cm⁻¹ and 2943 cm⁻¹, respectively. There is a vibrational band at about 1723 cm⁻¹ that is associated to the stretching of the C = O. This double bond is a source of polarons or bipolarons as charged structural defects in the polymeric chains [34]. The band at 1404 cm⁻¹ is thought to be caused by an asymmetrical O-C₂H₅ bending γ(OC₂H₅). It has been determined that the minor peak of 1212 cm⁻¹ observed at the high concentration of GO NP's (0.08) is due to the characteristic band of GO NP's, which would not be present in the PVDF/PMMA composite [34]. It's also important to note that when the concentration of GO NP's increases, the bands at 1000 to 400 cm⁻¹ decrease. It is believed that these bands are due to skeletal stretching, which is associated with C-H deformation (1000 cm⁻¹) and results from intermolecular interactions [35]. The intensity of the stretching frequency of C-H bands at 2991 cm⁻¹ and 2943 cm⁻¹ decreases as the concentration of GO NP's increases. This means that some of the changes that were seen in the FTIR can be explained by hydrogen bonding between GO NP's and some of the functional groups of the polymer blend [36]. Scheme 1a, b depicts a possible interaction between GO nanoparticles and a PVDF/PMMA polymer blend.
TEM image of GO NPs

A transmission electron microscope (TEM), can be used to measure the size of metal nanoparticles, such as graphene oxide nanoparticles. The particles inside the composite could be seen by TEM examination, which was carried out on GO. The TEM micrograph of GO NP’s, as well as their size distribution, are shown in Fig. 4. Graphene oxide nanoparticles/nanosheet have spherical and cubic shapes. The average size of GO nanoparticles is about 11–25 nm, according to the TEM micrograph.

UV/Vis. spectroscopy

The optical characteristics of the PVDF/PMMA polymer blend are investigated using the UV–Visible absorption spectroscopy technique. The ultraviolet–visible spectra of all nanocomposites (PVDF/PMMA/GO NP’s) are shown in Fig. 5 over the wavelength range of 200–1000 nm. As seen in Fig. 5, the absorption band at about 206 nm is related to the n→π* transition [37]. The absorption of polymer blend increases continuously as GO NP’s concentrations increase. A redshift is also observed when the concentration of GO NP’s in the polymer mix increases (the absorption edge move somewhat towards the longer wavelengths), which can be related to an increments in particle diameters produced by the aggregation of nanofiller in the polymeric matrix [38]. The formation of hydrogen bonds among polymer molecules and nanoparticles can also result in changes in the absorption edge of a polymer [32]. The peak absorption intensity rose as the amount of GO NP’s in the sample increased. This is because the nano–size of the graphene increments the surface area of the sample, resulting in high absorption [39]. Due to the polymer-filler complexation, no absorption bands in the visible area were seen for all prepared films. As a result, these samples can be used for a variety of UV shielding applications.

Determination of Optical energy gap (Eg)

The nanocomposite samples’ optical absorption spectra can be used to assess the polymer blend’s optical energy gap. The energy gap can be computed using the Tauc model [40]:

\[ a\nu = C\left(\frac{\nu}{E_{g}^{\text{opt}}}\right)^{r} \] (2)

where C represents for constant, hν refers to photon energy, α represents the absorption coefficient, and E_{g}^{\text{opt}} refers to optical energy gap. The allowed direct and indirect transitions have r values of 1/2 and 2, respectively [41]. Optical bandgap can be estimated by extrapolating the plots of extrapolating the plot of $a\nu^{2}$ and $(a\nu)^{1/2}$ vs $(\nu)$ as shown
in Figs. 6a, b and 7a, b. Table 1 listed the values of the energy gap for the allowable direct and indirect transitions. The values of $E_g^{opt}$ reduced with the addition of GO NP’s, decreasing from 4.81 eV to 3.65 eV for direct transition and from 4.24 eV to 3.71 eV for indirect transition, attributed to unstructured defects that enhance the density of localized states in the bandgap [42]. The optical bandgap decreases slightly because electrons are trapped and released between the bands. According to the FTIR analysis, this decrease is due to the coordination/interaction between the PVDF/PMMA matrices and the GO nanoparticles.

(a): The interaction mechanism for PVDF/PMMA and graphene oxide NPs in 2 dimensions.

(b): The interaction mechanism for PVDF/PMMA and graphene oxide NPs in 3 dimensions.

**Scheme 1**  
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**Thermal analysis**

**DSC measurement**

The DSC technique can be used to study the temperature transitions of polymer nanocomposite. The DSC curves for the nanocomposite samples are depicted in Fig. 8. The DSC curve of PVDF/PMMA blend shows three endothermic peaks and one exothermic peak. The glass temperature ($T_g$) of the PVDF/PMMA blend is attributed to a micro-Brownian motion for the main chain backbone, resulting in
an endothermic peak at 61.24 °C [43]. The α-relaxation in the PVDF crystalline regions causes the single exothermic peak at 416.26 °C. The PVDF and PMMA miscibility can be seen by the presence of a single T_g. The second and third endothermic peaks at 244.39 °C and 403.92 °C, respectively, show the melting point (T_m) and degradation (T_d) temperatures for PVDF/PMMA composite. Table 2 summarizes the impact of GO nanoparticles on the three endothermic peaks.

According to this table, the T_g value for nanocomposite samples has been somewhat enhanced, indicating a significant rise in the segmental mobility of nanocomposite matrices’ amorphous phases. The exothermic peak of the nanocomposites is moved to high-temperature values and largely disappears at high GO NP’s concentrations. This indicates that the crystalline phase content of nanocomposite films has reduced [12]. Also, the T_m position of these samples shifts to higher temperatures in an irregular way, which shows that the crystalline areas of these samples are disrupted and the number of amorphous phases increases because of the addition of GO NP’s to the PVDF/PMMA blend. The XRD method confirms these results. The nanocomposites samples (PVDF/PMMA/GO) have higher T_d values than PVDF/PMMA. This means that the addition of GO NP’s makes PVDF/PMMA more stable at high temperatures because the blend matrix becomes more flexible after the GO NP’s are added. This is because the amount of amorphous phase in the blend increases. Moreover, the increasing T_m and T_d values of nanocomposite samples indicate intermolecular interactions between GO NP’s and PVDF/PMMA chains as shown in FTIR.

**Thermogravimetric analysis (TGA)**

TGA thermograms of residual weight as a function of temperature for PVDF/PMMA filled with different concentrations of GO NP’s at a heating rate of 10 °C/min from 30 °C to 500 °C are shown in Fig. 9. As seen in the figure, the TGA curve degrades in two stages, the first
stage occurs between 232 and 287 degrees Celsius, possibly due to moisture evaporation or adsorbed water. It should be noted that this curve is stable up to 232 °C, and the initial weight loss reaches 30% at 287 °C. Then, without significant weight reduction, there is a plateau that extends up to 399 °C. While the second stage is allocated to the degradation of residual carbon at temperatures ranging from 399 °C to 436 °C. As a result of the addition of graphene oxide to the PVDF/PMMA polymer chain, the

| samples               | $E_g^{in}$ (eV) | $E_g^{out}$ (eV) |
|-----------------------|-----------------|-----------------|
| PVDF/PMMA Blend       | 4.81 ± 0.036    | 4.24 ± 0.032    |
| At 0.02 GO NP's       | 4.35 ± 0.064    | 4.15 ± 0.057    |
| At 0.04 GO NP's       | 4.12 ± 0.023    | 3.89 ± 0.015    |
| At 0.06 GO NP's       | 3.85 ± 0.074    | 3.83 ± 0.063    |
| At 0.08 GO NP's       | 3.65 ± 0.032    | 3.71 ± 0.054    |

Fig. 6 the variation of ($\alpha h\nu$)$^2$ versus $h\nu$ for a PVDF/PMMA blend and b PVDF/PMMA/0.08 GO NP's

Fig. 7 the variation of ($\alpha h\nu$)$^{1/2}$ versus $h\nu$ for a PVDF/PMMA blend and b PVDF/PMMA/0.08 GO NP's
samples are stable up to 500 °C, and the residual weight of the resulting nanocomposite samples has increased. TGA results suggest that the strong hydrogen bonding interactions between GO NP’s and polymer segments inhibited the mobility of both at interfaces, resulting in a delay in polymer degradation. Finally, the nanocomposites have better thermal stability than the pure PVDF/PMMA blend, proving that GO NP’s added to the existing polymer have significant structural modifications.

**Activation energy determination (E)** Two distinct approaches can be used to model the effect of graphene oxide nanoparticles on the thermal degradation rate of PVDF/PMMA. The Coats-Redfern [44] is a useful technique...
for finding activation energy that results in thermal degradation based on the following relationship [44]:

\[
\log \left[ \frac{1 - (1 - \alpha_t)^{1-n}}{T^2} \right] = \log \frac{R}{E} \left[ 1 - \frac{2RT}{E} \right] - 0.434 \frac{E}{RT} \tag{3}
\]

where \( T \) denotes the absolute temperature in Kelvin, \( n \) denotes the reaction order, \( E \) indicates the activation energy in J/mol, \( R \) means the universal gas constant (8.31 J/mol K), and \( \alpha_t \) represents the fractional weight loss related to a specific stage of decomposition. when \( n \neq 1 \), Eq. (3) becomes:

\[
\log \left[ \frac{-\log(1 - \alpha_t)}{T^2} \right] = \log \frac{R}{E} \left[ 1 - \frac{2RT}{E} \right] - 0.434 \frac{E}{RT} \tag{4}
\]

The activation energies (recorded in Table 2) were determined from the slopes of the straight lines produced by plotting \( \log \left[ \frac{-\log(1 - \alpha_t)}{T^2} \right] \) vs \( \frac{1000}{T} \) for each film:

\[
E = 2.303R \times \text{slope} \tag{5}
\]

The second method is the Broido model, which is a simple and accurate approach for computing (E) by the following equation [45]:

\[
\ln \left[ \ln \left( \frac{1 - \alpha_t}{\alpha_t} \right) \right] = -\frac{E}{RT} + \text{const} \tag{6}
\]

Plotting \( \ln \left[ \ln \left( \frac{1 - \alpha_t}{\alpha_t} \right) \right] \) vs \( \frac{1000}{T} \) revealed straight lines that were used to compute the activation energy from their slopes \((-\frac{E}{R})\), and the results were shown in Table 2. The table displays that the activation energy of nanocomposites increases with GO NP's concentrations. This improvement in activation energy indicates that the nanocomposite samples are more thermally stable than the PVDF/PMMA mixture. This data suggests that the current nanocomposite is a promising material for practice in a variety of applications.

**Table 2** The glass transition temperature \( (T_g) \), melting point \( (T_m) \), thermal decomposition \( (T_d) \) temperature, and activation energy \( (E) \) for all prepared samples

| GO NP's wt.% | \( T_g (^\circ C) \) | \( T_m (^\circ C) \) | \( T_d (^\circ C) \) | \( E \) (KJ/mol) Coats-Redfern | \( E \) (KJ/mol) Broido |
|--------------|------------------|------------------|------------------|-----------------|-----------------|
| 0.0          | 61.24            | 244.39           | 403.92           | 13.65           | 20.42           |
| 0.02         | 67.86            | 249.52           | 418.68           | 14.56           | 21.37           |
| 0.04         | 68.79            | 251.66           | 419.46           | 14.88           | 21.69           |
| 0.06         | 68.87            | 257.92           | 420.35           | 15.34           | 22.56           |
| 0.08         | 68.87 s          | 251.56           | 422.89           | 16.84           | 23.71           |

**Electrical investigation**

**AC conductivity**

Figure 10 shows the AC conductivity \( (\sigma_{ac}) \) of PVDF/PMMA virgin polymer and PVDF/PMMA filled with various concentrations of GO NP’s at 30 °C (RT). The conductivity improves in an inaccurate linear manner with frequency in all examined samples. The interfacial polarization effect is responsible for the increase in AC conductivity at lower frequencies, whereas electronic polarizations become active as frequency increases. The inclusion of GO NP’s up to 0.08 loading significantly increases the conductivity, which shows that GO NP’s are important for improving PVDF/PMMA conductivity properties. The addition of nanoparticles increases the number of charge carriers. There is also a gradual buildup of a conducting network in the PVDF/PMMA composite, which permits charge carriers to move through the polymer backbone [46]. The improved conductivity is also due to the space charge defects formed by the GO NP’s in the PVDF/PMMA backbone. The values of \( S \) are clearly inside the range (0.81—0.36). The values of \( s \) are less than 1 and decrease with increasing GO NP’s concentrations, indicating that the conduction mechanism is correlated barrier hopping (CBH).

**Dielectric parameters**

Polymeric materials’ charge storage capability is evaluated using dielectric permittivity analysis. To confirm that increased mobility causes increased ionic conductivity, a dielectric study is performed. The dielectric permittivity of polymer electrolytes is determined using the relation below [47].

\[
\varepsilon \ast = \varepsilon' - j\varepsilon'' \tag{7}
\]

where the \( \varepsilon' \) means the dielectric constant and \( \varepsilon'' \) means the dielectric loss. Figs. 11 and 12 show frequency function plots of the real \( (\varepsilon') \) and imaginary \( (\varepsilon'') \) parts of the dielectric permittivity for virgin polymer (PVDF/PMMA) and GO NP’s loaded virgin polymer samples. The charge carrier refinement density in the space charge accumulation zone, also known as non—Debye type of behaviors, is responsible for the increase in the value of \( \varepsilon' \) and \( \varepsilon'' \) at lower frequencies [48]. The significant cyclic reversal of the field at the interface causes the \( \varepsilon' \) and \( \varepsilon'' \) values to reduce at higher frequencies, and the influence of charge carriers to the \( \varepsilon' \) and \( \varepsilon'' \) reduces as frequency increases. The result reveals that when the frequency increases, the real and imaginary part of dielectric decreases. Another conclusion is that as the concentration of GO NP’s dopant increases, the \( \varepsilon' \) and \( \varepsilon'' \) enhance. As these polymer matrices begin to experience highly rapid chain mobility, the values of \( \varepsilon' \) and \( \varepsilon'' \) of the
Fig. 10 The dependence of log($\sigma_{\text{AC}}$) on log ($f$) for the prepared films at room temperatures

Fig. 11 The variation of $\varepsilon'$ versus log ($f$) for the prepared samples at room temperatures
polymer nanocomposites increases, as observed in XRD measurements.

**Complex modulus study (Argand plot)**

The relaxation process in polymer composites is caused by ion conductivity, as seen by the Argand figure. The Argand plot is shown in Fig. 13 for PVDF/PMMA/GO nanocomposites samples at room temperature. The polymer has a relaxation time distribution that deviates from the semicircular nature. All of the polymer nanocomposites created have a low semicircular arc. The electric relaxation of polymer nanocomposites is represented by the center of this arc, which is located on the M’ axis (X-axis). The electrical conductivity of the prepared films was linked to the length of this depressed semi-circular arc [49]. It can be shown in Fig. 13 that a reduction in the length of the arc is due to an improvement in the concentration of GO
nanoparticles in polymer nanocomposites, which shows an increment in conductivity. This study demonstrates that polymer nanocomposites containing 0.08 GO NP’s have higher conductivity than other compositions and show two relaxation times due to the increase in nanofiller. When the concentration of GO NP’s in polymer nanocomposites rises, the Argand plot attempt to move closer to the center. Moreover, polymer nanocomposites are non-Debye in nature. The non-Debye character results from the contributions of several types of polarization, relaxation mechanisms, and multiple interactions between dipoles and ions.

Conclusion

In this study, several experimental techniques were utilized to analyze the structural, optical, thermal, electrical, and dielectric properties of PVDF/PMMA-GO NP’s complexed polymer nanocomposites films. The crystallinity of pure GO NP’s and the semi crystallinity of PVDF/PMMA nanocomposites samples were shown by XRD patterns. The FTIR analysis was used to confirm the complexation and interaction of GO NP’s with the polymeric matrix. The absorbance spectra of nanocomposite samples increase as the concentrations of GO NP’s in the polymer composite increased, according to the UV–Vis spectrum. Also, the study found that as the nanofiller concentration increased, the expected bandgap values for all nanocomposite samples decreased gradually. The thermal stability of nanocomposite was shown to be greatly increased with the incorporation of GO NP’s to the PVDF/PMMA composite, according to DSC and TGA results. The AC conductivity of all nanocomposite films increases at a higher frequency as a result of the charge transfer through the hopping process. The greatest ionic conductivity was achieved with 0.08 GO NP’s at room temperature, which increased the conductivity from 5.34 $10^{-6}$ S/cm to 7.23 $10^{-5}$ S/cm. finally, it’s possible to use PVDF/PMMA-GO nanocomposites in polymer solar cells, organic semiconductors, and nanoelectronic devices because they have excellent thermal stability, conductivity, and dielectric constant.

Authors’ contribution Investigation, Writing—Review & Editing, Methodology, Formal analysis, Conceptualization. There are no other authors.

Data availability statement The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Declarations

Conflicts of interest The authors declare that they have no conflict of interest.

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