DIELECTRIC PROPERTIES OF (GO-MGO-POPDA- PVA) NANOCOMPOSITE FILMS

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ABSTRACT

In this work, a pure (PVA) polymer film reinforced with magnesium oxide, graphene oxide poly (o-phenylene diamine) (GO-MgO-PoPDA) was created utilizing the solution casting process in various weight ratios (0, 2, 4, 6, 8, 10 wt%). It was investigated how varied weight ratios of the nanoparticles magnesium oxide (MgO) and graphene oxide (GO) affected the Dielectric Properties of nano composite films. FTIR, SEM,X-RAY were used to characterized the nanocomposite. The results of the dielectric properties showed that the values of the alternating electrical conductivity of the prepared films increase when adding (GO-MgO-PoPDA) nanoparticles and with the increase of the frequency of the applied electric field and the increase of the particle content, while the values of the dielectric constant increase with the increase of each of the content of (GO-MgO- PoPDA) nanoparticles, but it decreases with increasing frequency. Whereas, the dielectric loss coefficient of the prepared films decreases when nanoparticles are added and with increasing frequency.

KEYWORDS

Dielectric Properties, Nanoparticles, Triple hybrid, Nanocomposite, Dielectric Constant, dielectric loss, Electrical Conductivity.

PAPER INDEX

1.   INTRODUCTION

The research on polymer dielectrics (PD) For the past two decades, research has focused mostly on materials with high energy-storage density for dielectric applications due to their affordability, biocompatibility, flexibility, straightforward design, and ease of processing[1-6].This method of expansion employed insulating ferroelectric polymers, such as polyvinylidene fluoride and polyvinylidene fluoride tetrafluoroethylene. Polymer nanocomposites have recently attracted interest as energy storage materials [1-19], uses for electromagnetic interference shielding [20, 21] different kinds of nanofillers, such as CNT, CB, graphite, etc., are incorporated into the ferroelectric polymer matrices during the construction of these PNC [1-26]. These PNC fillers use a variety of fillers with various conductivities, sizes, forms, and co-functionalization [1-26]. These PD are made using either the classic mixed technique, which involves solution casting followed by heat molding, While most recently, our team created the cold pressing method, which safeguards the ferroelectric polymers' spherulites [9-12]. Since they have more interfaces in the PNC, the spherulites also store electrical charge, this causes increased interfacial polarization and raises the effective dielectric constant value. For enforcement in a more general scenario and the associated commercialization, we were competent to demonstrate the value of ferroelectric polymer matrices, the influence of filler surface area, the high value of (eff) of 2400 in these PNC based on nanocrystalline nickel (n-Ni) filler, and the value of cold pressing in the development of PD. [9-12]. Polymer/metal composites, polymer/conductor composites, and polymer/ceramic composites are three examples of PNCs, are being developed as part of the development of these PD [7-26]. Interesting percolation behavior is displayed in one class [27].

2.   EXPERIMENTAL PART

2.1. SYNTHESIS OF BINARY NANOCOMPOSITE

The (GO) prepared by the modified Hummer method [28].The(MgO) prepared through (sol-jel) method. The binary nanocomposite (GO-MgO) was synthesized by dispersing (0.5 g) of (GO) in 100ml of non-ionic water using an ultrasonic water bath at 25ºC for 1 hour to form a GO solution.(0.5g) of MgO added to the GO solution. Moving for two hours at 25ºC and then dispersing for another one hour, the precipitate was separated by centrifugation and dried at 80ºC for two hours.[29].

2.2. SYNTHESIS OF THE TERNARY NANOCOMPOSITES

The triplet nanocomplex was prepared by dispersing (0.5g) of the GO-MgO binary complex in 50ml of non-ionic water by an ultrasonic bath at 25ºC for 1 hour and then mixing with (1.62g) of o-PDA dissolved in 100ml of 0.1M (HCI) acid with constant stirring for 1 hour in an ice bath. The oxidizing agent solution was synthesized by dissolving (6g) of(APS) in 100 ml (HCI) at a concentration of 0.1M. It was gently added dropwise to the main mixture with vigorous moving. Then the new mixture was

kept in an ice bath under stirring for 4 hours and then removed from the ice bath. Then, moderate stirring for 20 hours at 25ºC then the precipitate filtered washed with acetone and distilled water several times and dried in the oven at 80ºC for 2 hours[29].

2.3. SYNTHESIS OF THE QUATERNARY COMPOSITES

The following six concentrations are used to create pure PVA polymer membranes that are reinforcement by the triple nanolayer (GO-MgO-POPDA):

GO-MgO-PoPDA	PVA	The ratio
Zero	1	0 %
0.02	0.98	2 %
0.04	0.96	4 %
0.06	0.94	6 %
0.08	0.92	8 %
0.10	0.90	10 %

The triple composite was dispersed in 3 ml of non-ionic water and gently added to PVA solution after the PVA had been thoroughly dissolved in (15 ml) of non-ionic water with constant stirring at (60 °C). The mixture heated to a temperature of (50°C) while being agitated for an additional hour and a half to create a homogenous solution. Transferred to a petridish and dried at (25°C). Finally, the nanocomposites' films were peeled off in order to examine their physical characteristics.

3.   RESULTS AND DISCUSSION

3.1. INFRARED SPECTRUM OF THE TRIPLE COMPOSITES (GO-MGO-POPDA)

The infrared spectrum of the triple compound is depicted in the figure. It reveals the presence of several distinct bands for the triple hybrid compound, with the absorption bands at the region (3440.8 cm^{-1} - 3110 cm^{-1}) belonging to the group (NH-,NH2) and the appearance of the band (1680.7cm-1) connected to the stretching vibrations of the (C=O) group (which is connected to graphene oxide). Two bands that are centered at (1617.7 cm^{-1} - 1531 cm^{-1}) are a result of the group's stretching vibrations. (C=C) Regarding the absorption bands, they are (C-N) in the quinoid and benzoide groups, together with the bundles (1137.3 cm^{-1}-1393 cm^{-1}) that belong to the alkoxy and epoxy groups, and the bundle at (436.29 cm^{-1}) that belongs to the magnesium oxide group MgO.
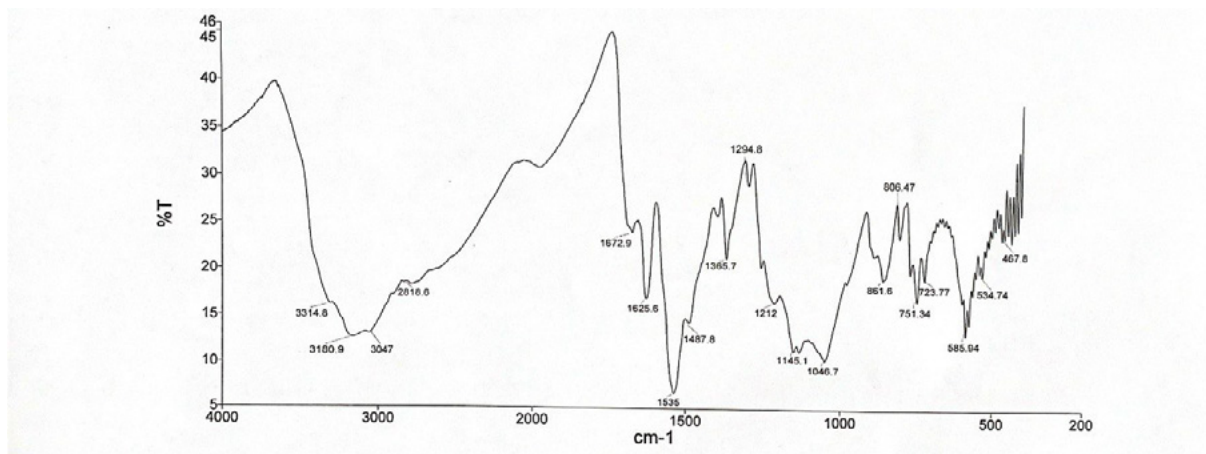
Figure 1. Infrared spectrum of the triple composite (GO-MgO-PoPDA).

3.2. SCANNING ELECTRON MICROSCOPE (SEM) OF TRIPLE COMPOSITE (GO-MGO-POPDA)

Scanning electron microscope with three composites (GO-MgO-PoPDA) SEM images of (GO-MgO-PoPDA) with magnification powers of (1 mμ), (500nm), (200nm) and are shown in Figure (2).The investigation reveals that all of the photos contain asymmetrical structures, both in terms of shape and size. It should be highlighted that the polymerization of the monomer oPDA on the surface of the binary compound is the reason why graphene oxide and magnesium oxide did not appear clearly in this test (GO-MgO) and that these asymmetrical forms are the result of the monomer's haphazard diffusion across the oxide surfaces in the first step, which is followed by polymerization when APS is added to it in the second step.
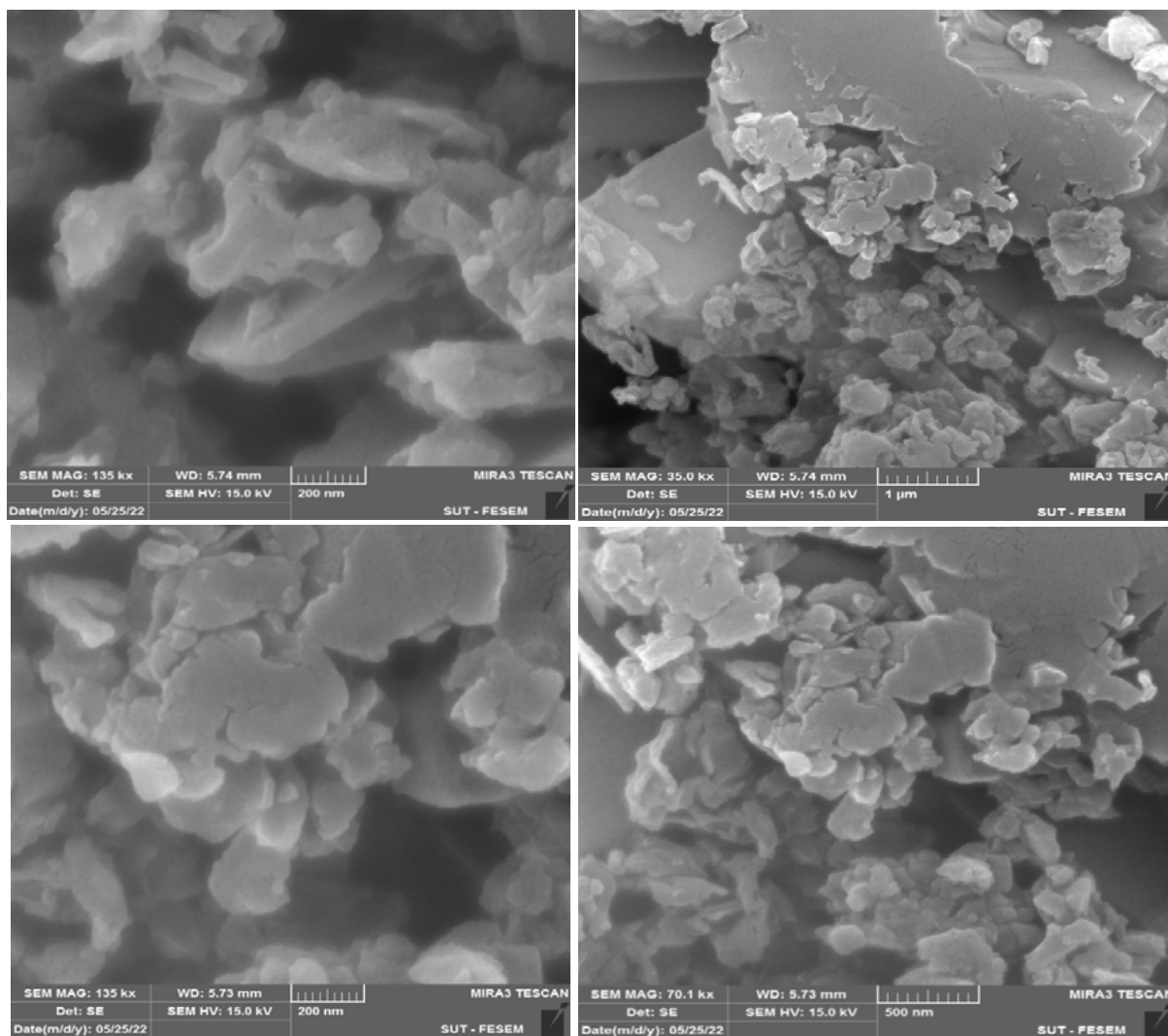
Figure 2. SEM of Triple Composites (GO-MgO-PoPDA).

3.3. X-RAY DIFFRACTION OF THE TRIPLE COMPOSITES (GO-MGO-POPDA).

The X-ray of the (GO-MgO-PoPDA) nanocomposite is depicted in Figure (3). The image below shows that the created triple nanocomposite can differentiate the peaks of magnesium oxide, and it is noteworthy that the maximum values of diffraction angles were obtained ($2\Theta=26.7460$, 27.9421, 19.7250). The diffraction angles clearly overlap the PoPDA polymer, and no graphene oxide peaks are seen as a result of the overlap of the polymer and graphene oxide peaks.
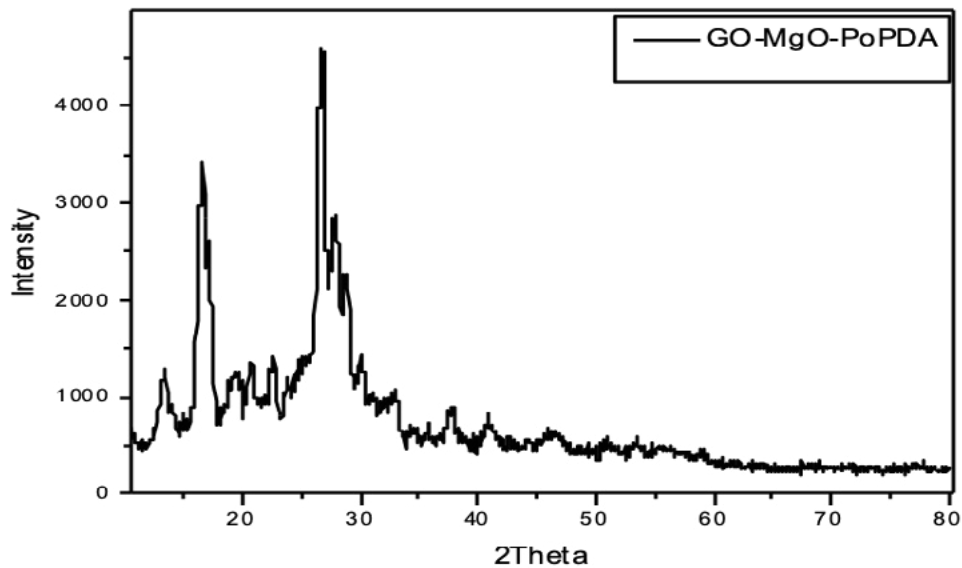
Figure 3. X-ray diffraction of the Triple Composites (GO-MgO-PoPDA).

4. DIELECTRIC PROPERTIES:

4.1. DIELECTRIC CONSTANT ($\dot{\varepsilon}$)

The dielectric constant was calculated for the pure (PVA) polymer film and the (PVA:PoPDA-GO-MgO) nanocomposite films varying the weight ratios (2,4,6,8, 10wt%) at room temperature (25 °C) as shown in figure (4), when it can be seen from the graph that the dielectric constant drops as the frequency increases and for the films of all hybrid nanocomposites [30]. Dipolar groups in insulating polymers can arrange themselves in the direction of the electric field at low frequencies, but it is difficult for this group (Dipoles) when it is large to arrange itself towards the fast and time-varying electric field at high frequencies because the time period is short less than the time it takes for the molecules to be able to arrange themselves in the direction of the outgoing electric field, the electronic polarization occurs within a very short but longer period of time than the ionic polarization, while the dipole polarization takes a relatively long time compared to all polarizations, therefore the dielectric constant of non-polar polymers remains almost constant at high frequency and therefore ($\dot{\varepsilon}$) values decrease dramatically and sharply with increasing frequency in low frequency regions [31], this may be the reason for the decrease in ($\dot{\varepsilon}$) values with increasing frequency. Another reason for this decrease may be attributed to the decrease in the polarization of the space charges to the total polarization [32]. We also note from figure (4) that the dielectric constant ($\dot{\varepsilon}$) of the hybrid composite (PVA:PoDA-GO-MgO) at the same frequency range rises with increasing weight ratios of the nano-oxides (Magnesium Oxide and Graphene Oxide) added. The insulation was recorded at the weight ratio (2 wt%) and its lowest value at the weight ratio (8 wt%), and all of these dielectric constant values are higher than the value of () for pure

(PVA), and generally speaking, this rise in the dielectric constant value An increase in polarity and an increase in charge carriers are thought to be the causes of electricity [32].

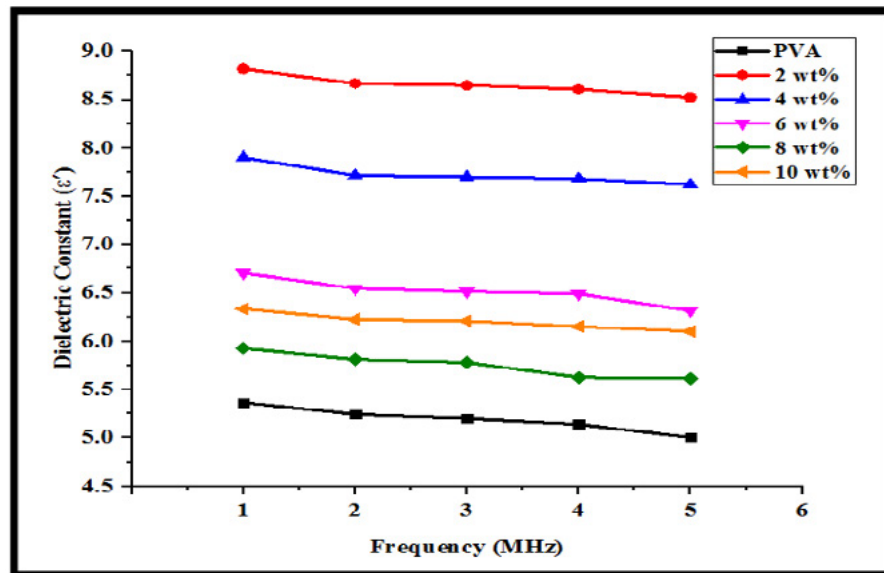**Table 1.** Dielectric constant of hybrid nanocomposite with different weight ratios of (GO, MgO).

Figure 4. Dielectric constant as a function of hybrid nanocomposite films with different weight ratios of (GO, MgO).

4.2. DIELECTRIC LOSS FACTOR ("ε)

The energy dissipation in insulating materials is directly proportional to the dielectric loss factor, hence, understanding the importance of this component offers huge advantages in the use of composite materials. So that the behavior of polar polymers in an alternating electric field depends on the position of the dipoles, whether they are within the polymeric chain or in its side groups. The insulating loss factor was calculated for the prepared samples pure PVA polymer film and hybrid nanocomposites films (PVA:PoPDA-GO-MgO) varying the weight ratios (2, 4, 6, 8, 10 wt%)) at room temperature (25 ^oC) and within the frequency range (1MHz-5MHz), from figure (5), It is evident that for all prepared samples, the dielectric loss factor decreases with increasing frequency. Additionally, we observe that as applied frequency rises, the dielectric loss first starts to decline at low frequencies, due to the decrease in the polarization of the vacuum charges (Space Charge Polarization) [32]. There is another reason for this decrease in (ε") values with increasing frequency, which is because of the decrease of dipoles in nanocomposites [33]. The dielectric loss factor is characterized by a high level of polarization of space charges for the nanoparticles of oxides (Magnesium Oxide and Graphene Oxide), which leads to a decrease in the values of (ε") with increasing frequency [34]. There is another reason for (ε") to change with frequency because the dipoles absorb energy from the electric field in the system in order to overcome the resistance of the viscous materials that surround them during rotation, and this absorbed energy reduces the charge carriers

moving between the limits in the widening with the increase in the frequency of the applied field Thus, these dipoles require high energy that is higher than the system to obtain relaxation and thus decreases (ε'') with increasing frequency [31]. Figure (5) note that the value of the dielectric loss factor (ε'') at the same frequency range increases when the weight ratios of nano-oxides (GO, MgO) increase, and that the highest value of (ε'') for the weight ratio (6 wt%) and the lowest value of the weight ratio (2 wt%) and the cause for this is due to the increase in the number of electrons in the nano-oxides used, and thus the dipole charge increases, which leads to an increase in electrical conductivity, thus increasing the value of (ε'') [32]. Table (2) shows the dielectric loss factor values for hybrid compounds with different weight ratios.

Table 2. Dielectric loss factor of hybrid with different weight ratios of (GO, MgO).

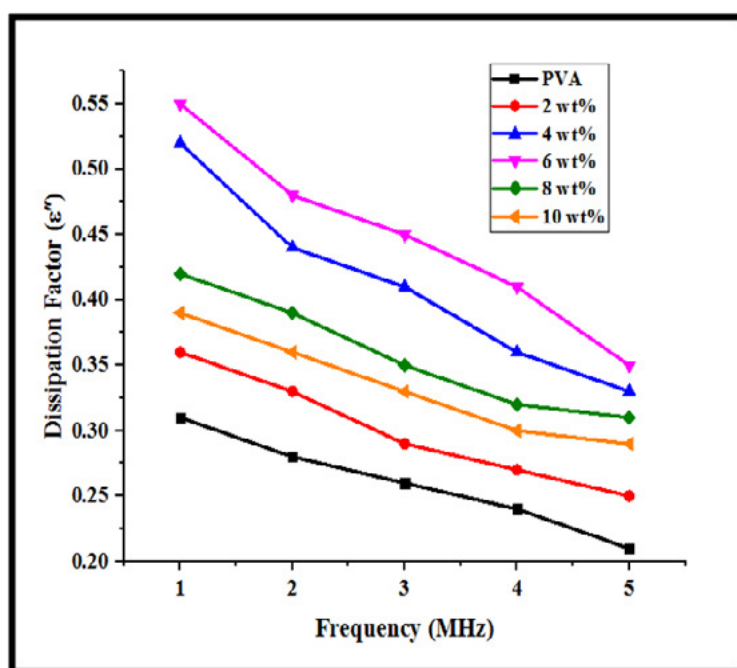| Frequency (MHz)	Dissipation Factor (ε'')					
	PVA	2wt%	4wt%	6wt%	8wt%	10wt%
1	0.31	0.36	0.52	0.55	0.42	0.39
2	0.28	0.33	0.44	0.48	0.39	0.36
3	0.26	0.29	0.41	0.45	0.35	0.33
4	0.24	0.27	0.36	0.41	0.32	0.3
5	0.21	0.25	0.33	0.35	0.31	0.29

Figure 5. Dielectric loss factor as a function of frequency of hybrid nanocomposite films with different weight ratios of (GO, MgO).

4.3. ELECTRICAL CONDUCTIVITY (σa.c)

Electrical conductivity is defined as the process of moving electric charge from one place to another through a medium when an electric field is applied. Alternating electrical conductivity (σa.c) was calculated for the membrane of pure PVA polymer and hybrid nanocomposites films (PVA: PoPDA-GO-MgO) at room temperature (25 ºC) and varying the weight ratios (2,4,6,8,10wt %) and in the frequency range (1MHz -

5MHz). As shown in figure (6), where we notice from the figure that the AC electrical conductivity (σa.c) increases with the rise in frequency and for all the prepared compounds, this increase in (σa.c) is attributed to the increase in the applied frequency to the polarization and the charge carriers (Carriers Charge) that It is transmitted by hopping process between electronic levels, and these hoppings of charge carriers across the voltage barrier and quantum tunneling increase the value of AC electrical conductivity [32]. From the figure, we also notice the absence of relaxation peaks, which indicates that the increase in (σa.c) is due to the rise in the density of ions (Ionic density) rather than the structural relaxation process in order to rearrange the dipoles in the system or molecular relaxation for polymer chains [35]. Alternating electrical conductivity (a.c.) in insulating materials is determined by power lost as a result of applying an alternating electric field, which appears as heat and rotates dipoles in their positions or causes charges to vibrate by changing the direction of the alternating electric field. For this reason, the amount of power lost determines the frequency of the applied field [36]. By increasing the weight ratios of the reinforcement with nano-oxides (MgO, GO), it is noted that the alternating electrical conductivity is high compared to the alternating electrical conductivity of the pure PVA membrane, which is few, as well as the formation of a continuous network in the form of agglomerates when the weight ratio of nano-oxides is increased, which it leads to the formation of a continuous path inside the hybrid nanocomposite films material as the weight ratio increases, Because of the formation of a continuous conductive path, this leads to an increase in the AC electrical conductivity, and its value is determined by the effect of polarization and electron movement in the substrate [32]. Table (3) shows the AC electrical conductivity values for all the prepared hybrid nanocomposites films.

Table 3. Electrical Conductivity values of the hybrid nanocomposites films with different weight ratios of (GO, MgO).

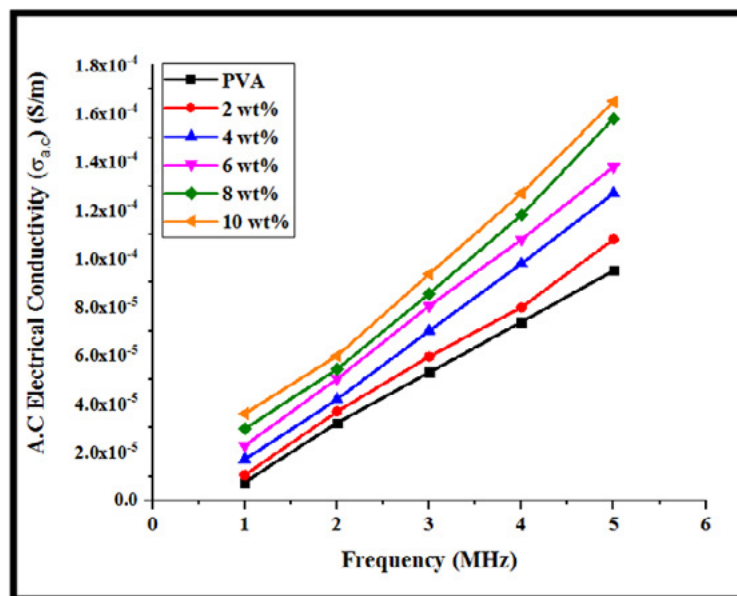| Frequency (MHz)	(A.C) Electrical Conductivity (σa.c (S/m)					
	PVA	2 wt%	4 wt%	6 wt%	8 wt%	10 wt%
1	7.65×10^{-6}	1.06×10^{-5}	1.7×10^{-5}	2.27×10^{-5}	2.97×10^{-5}	3.59×10^{-5}
2	3.21×10^{-5}	3.67×10^{-5}	4.18×10^{-5}	5.03×10^{-5}	5.43×10^{-5}	5.98×10^{-5}
3	5.29×10^{-5}	5.95×10^{-5}	7.02×10^{-5}	8.05×10^{-5}	8.55×10^{-5}	9.36×10^{-5}
4	7.38×10^{-5}	7.98×10^{-5}	9.79×10^{-5}	1.08×10^{-4}	1.18×10^{-4}	1.27×10^{-4}
5	9.5×10^{-5}	1.08×10^{-4}	1.27×10^{-4}	1.38×10^{-4}	1.58×10^{-4}	1.65×10^{-4}

Figure 6. Electrical Conductivity as a function of frequency of hybrid nanocomposite films with different weight ratios of (GO, MgO).

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