



Graphene oxide on the electrical properties D.C and A.C and dielectric constant of PVP/PVA Nanocomposites

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Graphene oxide (GO) nanofiller were incorporated in PVP/PVA blend films for the preparation of nanocomposite polymer films by the solution cast method. The films were characterized using SEM, D.C and A.C and dielectric properties studies at room temperature. SEM structure shows that the GO exfoliated and uniformly dispersed in PVP/PVA matrix. DC conductivity studies were under taken at (303-336) K, and the conductivity was found to be $3.3 \times 10^{-5} \Omega \cdot \text{cm}^{-1}$ for the polymer film prepared at 336K. The A.C electrical conductivity increase with the increase of frequency. The dielectric properties were measured the frequency range 100Hz-5MHz at room temperature show that the dielectric constant (real and imaginary) decrease with the increase of Graphene oxide nanoparticles concentrations.

INTRODUCTION

Graphene oxide (GO) based materials due to their great in many properties, warm, and mechanical, for example, conductive covering, sensors, microwave engrossing, and vitality stockpiling gadgets. After the revelation of graphene and graphene oxide, a spotlight has been made on the nonvolatile memory gadgets because of their potential applications [1].

Nanocomposite polymer films are set up by scattering the nanocomposites in the polymer framework, for example, graphite, carbon nanotubes, carbon fiber, nanofillers, and uncommon earth-doped particles. By doping the nanocomposites in conductive polymers, to change the electrical conductivity is expanded at a higher rate [2]

Graphene, a monolayer of hexagonally pressed carbon particles. Graphene has a honeycomb-like structure with sp^2 -fortified carbon atoms firmly stuffed with oxygen atoms. Graphene is likewise utilized in numerous applications, for example, batteries, sun oriented cells, power devices, and supercapacitors. Graphene is generally utilized as nanofiller in polymer composites for a few down to earth applications on extensive scale generation and solid association framed between the polar particles of GO is because of the nearness of oxygen-containing utilitarian gatherings to give homogenous scattering [3].

PVA is a standout amongst the most vital polymers because of its great mechanical and warm properties. Its semi crystalline nature permits go around complexities of deciphering properties related with crystallization on expansion of Graphene oxide. PVA is a hydrophilic polymer with hydroxyl ($-OH$) bunches which can interface with $-OH$ or carboxyl gatherings ($-COOH$) present in GO to shape a steady

complex in composites [4].

Polyvinyl pyrrolidone (PVP) has picked on the grounds that it has fantastic qualities, for example, optical, mechanical and electrical properties. PVP has promotion chive natures, it might be utilized in electronic circuit sheets and show gadget applications. PVP has great similarity and can without much of a stretch shape films with extensive inside region. PVP is effortlessly dissolvable in refined water and inorganic solvents, it has nebulous nature. In view of its low diffusing misfortune, it very well may be more valuable for optical applications, bringing about the great scattering and surface development [5].

Electrical conductivity is the most critical property of graphene, when graphene fill into the protecting polymer grid, conductive polymer composites result. Different polymers, including PMMA, PVA, PVC, PP, PE, PA, PS and so on have been utilized as networks to plan electrically conductive graphene/polymer composites [6]. Such composite materials for the most part display a non-straight increment of the electrical conductivity as an element of the filler focus. The two parameters, electrical conductivity and permeation edge are as one related with graphene oxide. At a specific filler stacking division, which is known as the permeation limit (pc), the fillers shape a system prompting a sudden ascent in the electrical conductivity of the composites [7]. Some of the time expansion of a low measure of leading particles can reach to shape successful directing ways and in this way making the entire composite conductive [8].

Graphene is more effective for electrical conductivity improvement than competing nanofillers such as CNTs because of their large specific surface area. An outstanding electrically conductive graphene/polymer composite is expected to have lower percolation threshold and higher conductivity at a lower graphene loading, which can not only decrease

the cost of filler but also preserve the process ability of the composite [9].

In the present investigation, nanocomposite polymer films were prepared by dispersing GO filler with PVP/PVA blend polymers to improve electrical conductivity.

METHODS

Preparation of Nanocomposite Polymer Films

Polymer films were prepared between combination of GO, PVA, and PVP. It takes 100 ml round-bottomed flask and add 20 ml of distilled water with blend polymers PVP/PVA: wt. % (80/20) and Stir all the mixtures till the polymers get dissolved in water. Later, reduced nanoparticles of GO: x% (0.2, 0.4 and 0.6%) Nanocomposites were prepared by casting method.

The dielectric properties of (PVP/PVA/GO) nanocomposites were measured using LCR meter in the frequency range 100Hz-5MHz at room temperature.

Electrical Properties

D.C. Conductivity

The electrical resistance has been measured as a function of temperature in the range (298-336 K) by using the resistivity (ρ) of the films is calculated by using the following equation:

$$\rho = R \cdot AL \dots \dots \dots (1)$$

Where: R is the sample resistance, A is the cross section area of the film and L is the distance between the electrodes. The conductivity of the films was determined from the relation:

$$\sigma_{dc} = 1/\rho \quad (2)$$

The activation energies could be calculated from the plot of $\ln \sigma$ versus $1000/T$.

A.C Electrical

LCR-Meter was used for the A.C measurements. The sample was placed in a holder specially designed to minimize stray capacitance. The frequency range was (100Hz-5MHz) for electric field. The total conductivity σ_{total} at a certain frequency and temperature is defined as

$$\sigma_{tot} = \sigma_{ac}(\omega) + \sigma_{dc} \dots \dots \dots (3)$$

$\sigma_{a.c}$ is the A.C. conductivity, $\sigma_{d.c}$ is the D.C. conductivity, then the empirical relation for the frequency dependence A.C conductivity is given by [10]:

$$\sigma_{a.c}(\omega) = A1 \omega^s \dots \dots \dots (4)$$

A1 is constant parameter, and s is an exponential factor. Its value is $0 < s < 1$ [11]

The exponent (s) is a function of frequency and is determined from the slope of a plot $\ln \sigma_{a.c}(\omega)$ versus $\ln(\omega)$ then,

$$S = \ln \sigma_{a.c}(\omega) / \ln(\omega) \dots \dots \dots (5)$$

And the dielectric constant, ϵ_1 , is calculated from the equation:

$$\epsilon_1 = LC/A\epsilon_0 \dots \dots \dots (6)$$

Where ϵ_0 is the permittivity of free space, L is the thickness, C is the capacitance and A is the cross section area. The dielectric loss, ϵ_2 , is calculated from the equation:

$$\epsilon_2 = \epsilon_1 \tan \delta \dots \dots \dots (7)$$

Where $\delta = 90 - \phi$, ϕ is the phase different angle.

RESULTS AND DISCUSSION

The checking electron microscopy (SEM) is utilized to investigate and look at the structure and morphological surface of the example. SEM of PVP/PVA/GO composites films are appeared in Fig(1) a, b. The pictures uncover the heterogeneous period of nanocomposite polymer frameworks. It is seen that GO lumps are framed which are scattered in polymer film. These GO pieces have crystalline structure installed in the polymer grid with various harshness. The smooth surface back ground relates to the indistinct idea of the host polymer. The larger amount of GO in PVP/PVA will result in preferred scattering of GO over utilizing lower level and it additionally affirms from the SEM pictures. The similarity of the mix PVA/PVP lattice with GO is uniform and homogenous when the GO content is around 0.6 wt % proportion. In this manner it affirms GO was peeled inside the PVP/PVA framework and furthermore uncovers the heterogeneous period of nanocomposite polymer.

D.C Conductivity

Fig (2) shows demonstrates the temperature reliance of D.C conductivity in the temperature go (303-336) K for mix PVP/PVA and PVP/PVA/Go composites, the information uncovered that the conductivity of PVP/PVA: is $3.75 \times 10^{-6} \Omega \cdot \text{cm}^{-1}$ and its esteem expands pointedly to $3.3 \times 10^{-5} \Omega \cdot \text{cm}^{-1}$ at 336 K, demonstrate its higher than that of mix PVP/PVA because of expanding because of free volume and their separate ionic and segmental portability [8].

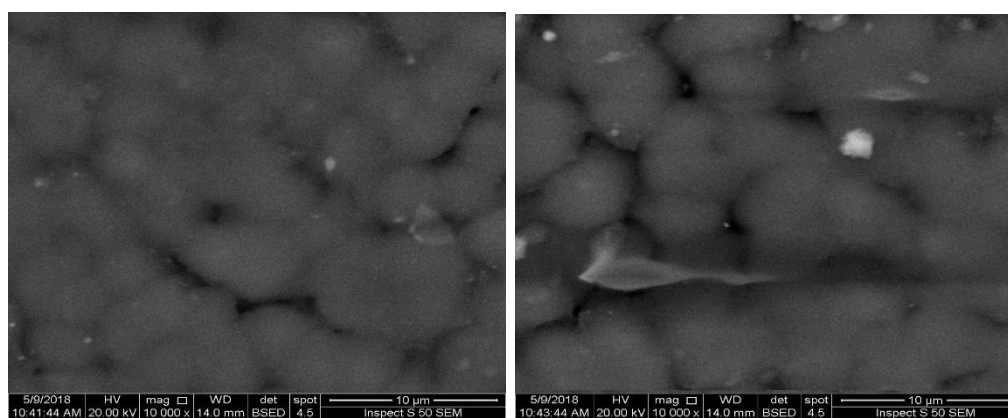
Likewise, clearly the electrical conductivity of the GO doped examples are more prominent than that of unadulterated PVA/PVP mix and increments as expanding the GO doping levels. This could be deciphered as; the decrease of the crystalline stage due to GO doping (announced in SEM) prompts diminishing in the interfacial hindrance and along these lines increment the change versatility of electron trusting over the obstruction. This thusly gives a leading way through the shapeless district of the polymer network bringing about upgraded conductivity.

The plots on the $\ln \sigma_{dc}$ as a function of $1000/T$ are nearly straight lines indicating that the conduction in the samples through an activated process having single activation energy.

The increase in conductivity with electrolyte films also increased. This favors hopping mechanism between the coordinating sides, local structure relaxation, and segmental motion of the polymer. The linear variation of $\ln \sigma_{dc}$ with reciprocal temperature for the present system (Fig.2) suggests an Arrheniuseas relation behavior:

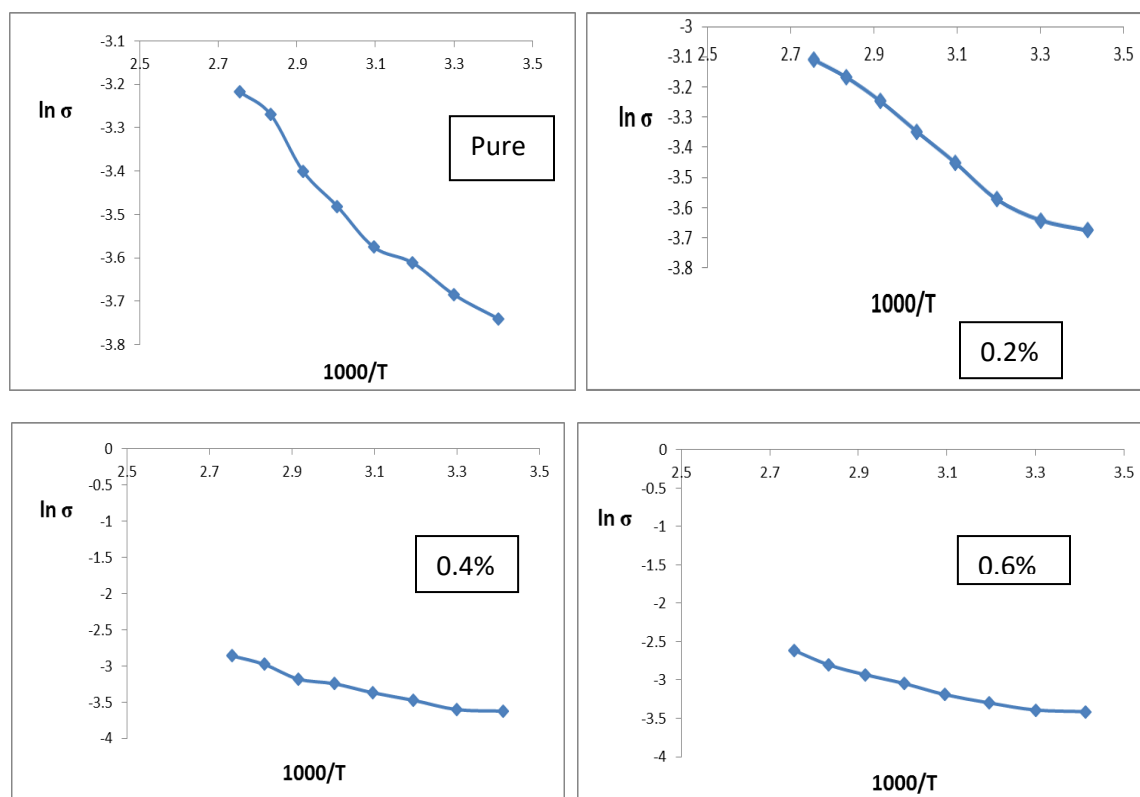
$$\sigma_{dc} = \sigma_0 \exp(-\Delta E_{dc} / k_B T) \dots \dots \dots (8)$$

Where is the proportionality constant, ΔE_{dc} is the activation energy, k_B is the Boltzmann constant and T is the absolute temperature. From this equation the activation energy ΔE_{dc} of the present system can be calculated and is found to be in the range 0.21-0.05 eV. It is also found



PVP/PVA/ 0.2%GO

PVP/PVA/0.6%GO

Figure 1 Scanning Electron Microscopy of samples nanocomposites at (0.2%, 0.6%)**Figure 2** $\ln \sigma$ vs $1000/T$ of blend PVP/PVA and PVP/PVA/Go nanocomposites at (0.2, 0.4, 0.6) wt**Table 1** Values of σ .c and activation energy of nanocomposites

Sample	σ ($\Omega \cdot \text{cm}^{-1}$)	E_g (ev)
PVP/PVA	3.75	0.21
PVP/PVA/0.2%GO	3.53	0.18
PVP/PVA/0.4%GO	3.40	0.09
PVP/PVA/0.6%GO	3.3	0.05

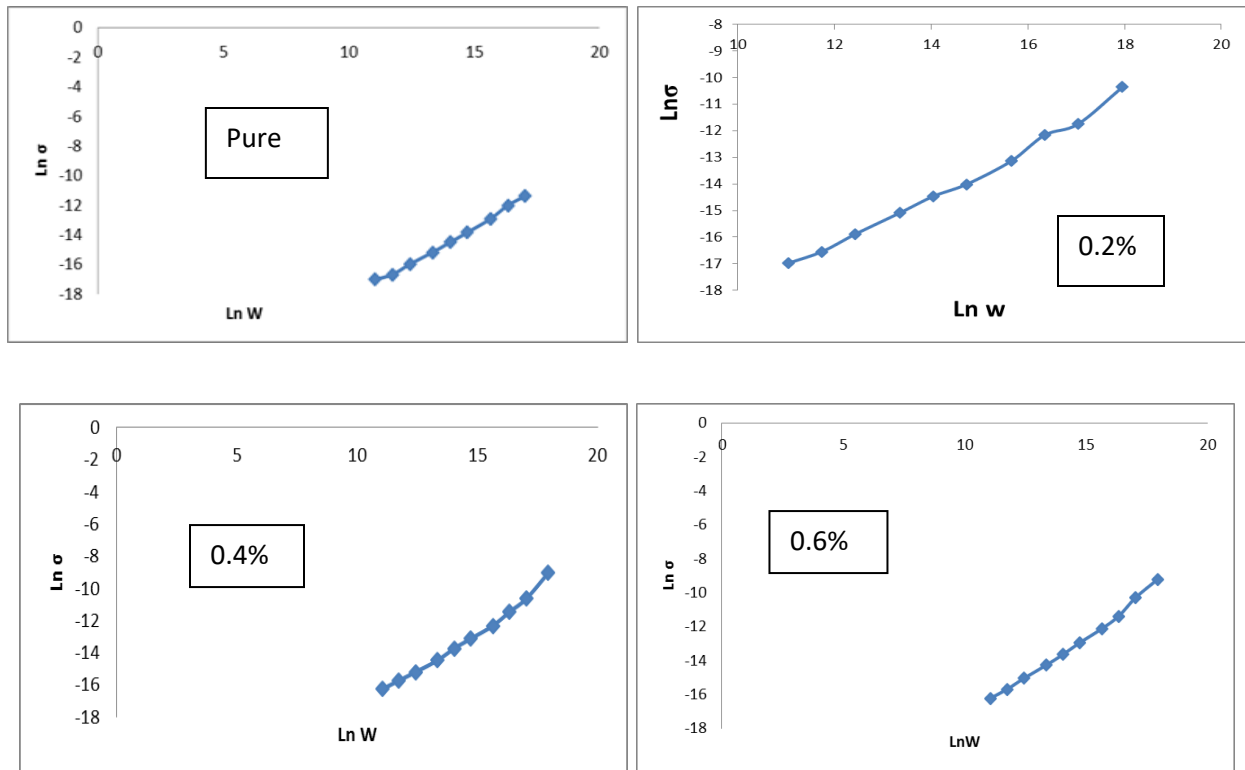
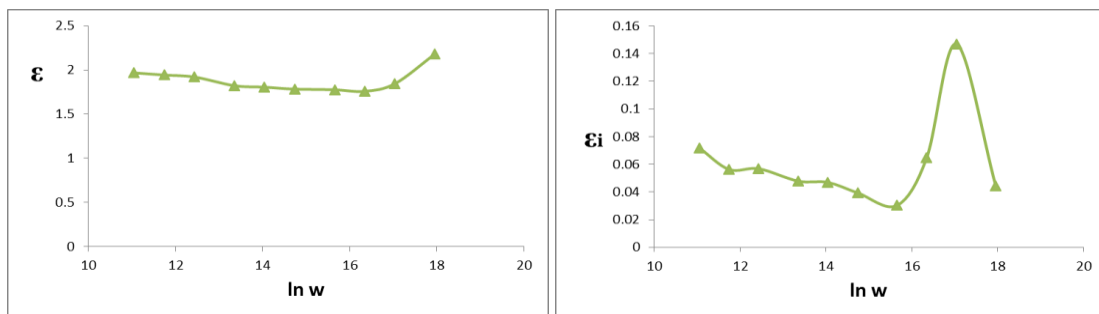


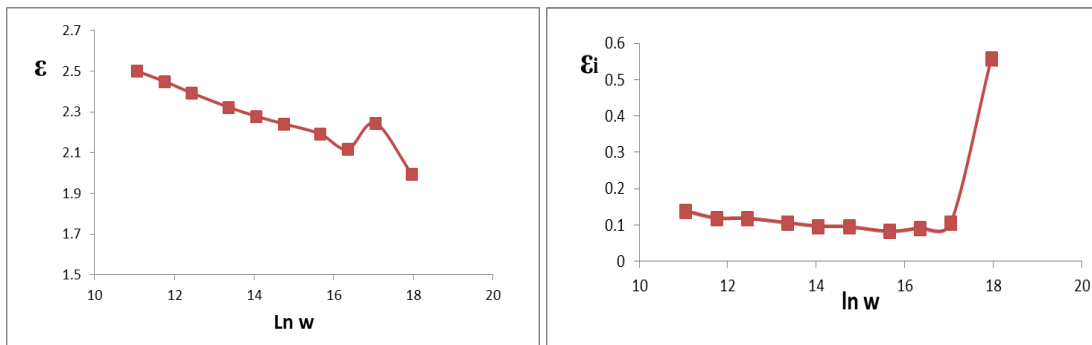
Figure 3 Frequency dependence of the electrical conductivity for PVP/PVA/GO nanocomposites

Table 2 Values of frequency exponent (S) and construction

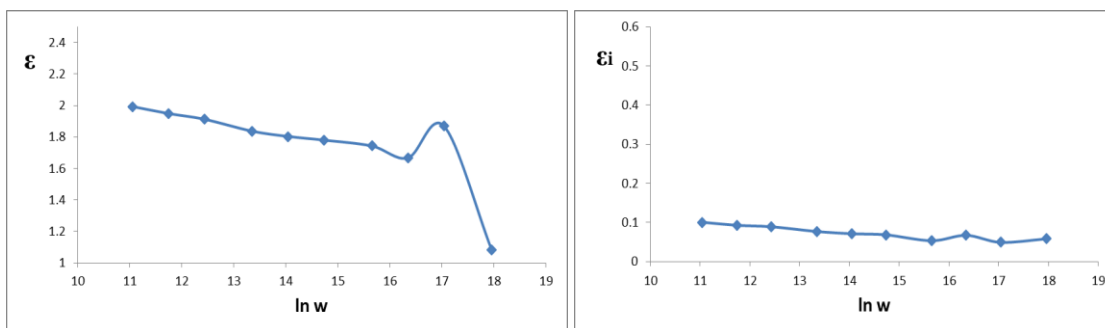
Sample	S
PVP/PVA	0.7
PVP/PVA/0.2%GO	0.5
PVP/PVA/0.4%GO	0.3
PVP/PVA/0.6 %GO	0.2



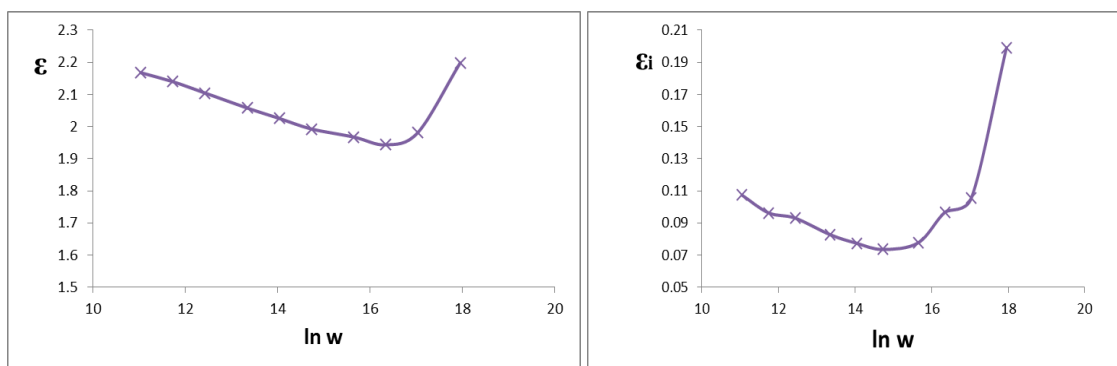
PVP/PVA



PVP/PVA/GO 0.2%



PVP/PVA/GO 0.4%



PVP/PVA/GO 0.6%

Figure 4 Dielectric Properties (real and imaginary) for PVP/PVA/GO nanocomposites

that the values of ΔE_{dc} slightly decrease as the doping level of Graphene oxide increase due to these charge transfer complexes takes place in the host lattice, resulting in the decrease of activation energy.

A.c electrical properties

A.c electrical properties of PVP/PVA/GO nanocomposites were studied as a function of the constriction and frequency, Fig.(3) show the relation between $\ln \sigma_{a.c}$ and $\ln \omega$ for PVP/PVA at room temperature (303) K, and for PVP/PVA/GO nanocomposites. It is clear from the figures that $\sigma_{a.c}(\omega)$ increased with the increase of frequency according to eq.(4) at room temperature. Increase of frequency increased a.c. conductivity by increasing the hopping of conducting electrons present in Graphene oxide. At higher frequencies this hopping frequency could not match the applied field frequency.

According to the eq.(4) ,the value and behaviour of S with temperature can give information about the dominant conduction mechanism. The value of S is the slope of the straight line portion of the relation between $\ln \sigma_{ac}$ and $\ln \omega$ Fig. (3), the dependence of S on the temperature for all the studied samples. It is clear that for pure

PVP/PVA blend the values of S is near unity and are independent on temperature. On the other hand the values of S slightly decrease with increasing Graphene Oxide level in PVA/PVP matrix and remain near unity. Moreover, this behaviour recommends the quantum mechanical tunnelling conduction mechanism to be the dominant mechanism for ac conduction.

The rapidly increasing of $\sigma(\omega)$ with increasing frequency at the frequency greater than 10^3 Hz referred to the electronic polarization effect, and the conductivity is pure a.c conductivity $\sigma_{a.c}(\omega)$ in this region. At low frequencies where the applied electric field forces the charge carriers to drift over large distances. When frequency is raised, the mean displacement of the charge carriers is reduced and the real part of the conductivity, after reaching ascertain critical frequency, f_c , follows the laws a.c. $(\omega) \sim (\omega)^s$ with $0 < s < 1$.

It is observed that the frequency is found to have a pronounced effect on conductivity at lower temperatures. The obtained values of (s) ranged from (0.3 to 0.6) unit that indicate the correlated barrier hopping (CBH). According to the model, a. conductivity $\sigma_{a.c}(\omega)$ can be explained in terms of the hoping of electrons between pairs of localized states at the

Fermi levels [11]. Is the dominant conduction mechanism, and have a tendency to decrease with the increase of frequency and construction as indicated in Table (2).

Dielectric Properties

Fig (4) show the dielectric permittivity as a function of frequency reflects the important effect of the GO content on the properties of pure PVA/PVP blend. The frequency dependence of ϵ_r and ϵ_i at $T = 303$ K for all samples respectively. It is clear that ϵ and ϵ_i decrease with increasing frequency for all samples. The values of ϵ and ϵ_i are high at low frequency and decrease monotonically with increasing frequency until reaching to a constant value at higher frequencies. The high values of ϵ and ϵ_i may be due to the interfacial effects within the bulk of the sample and the electrode effects [38]. Also, it is known that for polar materials, the initial value of ϵ and ϵ_i is high, but as the frequency of the field is increased, the value begins to drop because the dipoles are not able to follow the field variations. At high frequencies, the periodic reversal of the electric field occurs so fast that there is no excess ion diffusion in the direction of the field. The polarization due to the charge accumulation decreases, leading to a decrease in the value of ϵ and ϵ_i [11].

It is noticed that the values of ϵ and ϵ_i for the doped samples are higher than that of pure PVA/PVP and increase with increasing the doping level for all the frequency range. The increase in ϵ and ϵ_i upon increasing MB content can be explained in terms of increasing the accumulated charge because of the polarization of polymer/ dopant interfaces. The polarization makes an additional contribution to the charge quantity. Fig (3) illustrates the increase of both ϵ and ϵ_i with increasing the content of MB at frequency 500 Hz and $T = 333$ K.

CONCLUSION

Samples of blend (PVP/PVA) and sample with different Graphine oxide (PVA/PVP/GO) at concentration of (0.2, 0.4, 0.6) % were prepared and studied. The SEM analysis showed that homogenous phase at low level of Graphine oxide. DC electrical conductivity displayed that the conductivity of the present system increases with increasing of both temperature and the GO nanocomposites. A.C electrical conductivity increase with the increase of Graphine oxide nanoparticles concentrations The dielectric properties were measured the frequency range 100 Hz–5 MHz at 303K Temperature. The results show that the dielectric constant, decrease with the increase of frequency of applied electrical field.

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