



## Structural, A.C electrical and Optical properties of (Polyvinyl alcohol–Polyethylene Oxide–Aluminum Oxide) Nanocomposites for Piezoelectric Devices



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**T**HERE needs to study of structural, electrical and optical properties of overlapping materials to determine their suitability for electronic, industrial and solar cell applications. Polymer blends and nanocomposites gained considerable importance for researchers and scientists. In addition to; they have entered into a wide range of electronic and industrial applications. So in this paper, it will make preparation a new kind of (polyvinyl alcohol–polyethylene oxide– aluminum oxide) nanocomposites with different concentrations of aluminum oxide nanoparticles. From the results, it's noted that the dielectric constant of (PVA-PEO) blend is increased by increasing concentrations of Al<sub>2</sub>O<sub>3</sub>NPs and applied load at 100Hz. The dielectric loss of (PVA-PEO) blend is increased by increasing concentrations of Al<sub>2</sub>O<sub>3</sub>NPs as well as with low percentage with increasing mechanical stress at 100Hz. AC electrical conductivity of (PVA-PEO) blend is increased with increasing of Al<sub>2</sub>O<sub>3</sub>NPs concentrations. The optical measurements are shown that the absorbance of (PVA-PEO-Al<sub>2</sub>O<sub>3</sub>) nanocomposites is increased by increasing the percentage weight of Al<sub>2</sub>O<sub>3</sub>NPs. The indirect energy gap (E<sub>g</sub>) of (PVA-PEO) blend decreases with increasing of concentrations of Al<sub>2</sub>O<sub>3</sub>NPs. The optical constants as absorption coefficient, extinction coefficient, refractive index, real and imaginary dielectric constants of nanocomposites are a variation with increasing of Al<sub>2</sub>O<sub>3</sub>NPs concentrations.

**Keywords:** Polymer blends, Mechanical stress, Aluminum oxide, Energy gap, Applied load.

### Introduction

In general; Piezoelectric effect or direct piezoelectric effect is a phenomenon that generates electrical energy as when the material is subjected to pressure, applied force or mechanical stress. Where the charges are separated due to apply force. In short; energy conversion phenomena [1]. The Converse piezoelectric effect is called when materials are generated a mechanical strain when an electric field is applied [2]. First discovered in the phenomenon of piezoelectric effect are the two brothers Jacques Curie and Pierre Curie in 1880. They are observed when some crystals naturally

occurring as quartz, cane sugar, Rochelle salt and topaz or Man-made crystals as a quartz analogue crystal, Gallium orthophosphate (GaPO<sub>4</sub>), and Langasite (La<sub>3</sub>Ga<sub>5</sub>SiO<sub>14</sub>), nanocomposites when exposed to pressure gave electric effect. In addition, lead zirconate titanate, which was the first piezoelectric ceramic, was discovered when it was exposed to mechanical stress and generated an electric effect [3].

Piezoelectric materials have played a major role in research and development of many fields as pressure sensors and actuators. So, they are defined as a materials have the ability to convert

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mechanical energy into electrical energy when exposed to mechanical stress. The molecules of these materials are poly or monocrystalline [4]. Piezoelectric materials have many applications in many fields, for examples, sensors, as pressure sensor, force sensor, and strain gauge; actuators as loudspeaker, piezoelectric motors, nanopositioning in AFM and STM, and acoustooptic modulators; high voltage and power source such as cigarette lighter, energy harvesting, and AC voltage multiplier; transducers; surface acoustic wave devices; and frequency control [5].

Nanocomposites are defined as materials are resulting from the mixing of two substances; polymer as a matrix and the additive such as nanometals, nanofibers and nanometals oxide or any other materials. To obtain of piezoelectric nanocomposites must be selected polymers should be semi crystalline polymer, fibers, biocompatibility, biologically derived polymers, high ionic conductivity, biodegradability, brunch linking, lightweight nanocomposites and high specific strength. Both poly vinyl alcohol (PVA) and polyethylene Oxide (PEO) are characterized by these properties mentioned above [6].

### Theoretical part

The dielectric constant ( $\epsilon'$ ) of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites is calculated by the eq.:

$$\epsilon' = \frac{C_p}{C_o} \quad \dots\dots\dots(1)$$

Where,  $C_p$  is parallel capacitance and  $C_o$  is vacuum capacitor

The dielectric loss ( $\epsilon''$ ) of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites is given by the equation [7]:

$$\epsilon'' = \epsilon' D \quad \dots\dots\dots(2)$$

Where,  $D$ : is dispersion factor.

The AC electrical conductivity of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites is determined by the following eq. [8]:

$$\sigma_{AC} = w \epsilon'' \epsilon_o \quad \dots\dots\dots(3)$$

Where,  $w$  is the angular frequency.

Absorption coefficient ( $\alpha$ ) is calculated as the following eq.:

$$\alpha = 2.303 (A/t) \quad \dots\dots\dots(4)$$

$A$ : is the absorbance. For amorphous polymers, the indirect transition model can be computed as:

$$\alpha h\nu = L(h\nu - E_g)^x \quad \dots\dots\dots(5)$$

Where  $L$  is a constant,  $h\nu$  is the photon energy,  $E_g$  is the optical energy band gap,  $x = 3$  for forbidden indirect transition and  $x = 2$  for allowing indirect transition [9]. Refractive index ( $n$ ) is determined by following eq.:

$$n = \frac{1 + \sqrt{C}}{1 - \sqrt{C}} \quad \dots\dots\dots(6)$$

Where  $C$  is the reflectance, can be got from:

$$C = 1 - A - T \quad \dots\dots\dots(7)$$

The extinction coefficient ( $k$ ) is determined by the following eq.:

$$k = \frac{\alpha \lambda}{4\pi} \quad \dots\dots\dots(8)$$

Where  $\lambda$  is the wavelength of incident light. The dielectric constant is classified into two parts real ( $\epsilon_r$ ), and imaginary ( $\epsilon_{im}$ ). It can be computed each of the real and imaginary parts of the dielectric constant ( $\epsilon_r$  and  $\epsilon_{im}$ ) as the following eqs.:

$$\epsilon_r = n^2 - k^2 \quad \dots\dots\dots(9)$$

$$\epsilon_{im} = 2nk \quad \dots\dots\dots(10)$$

Optical conductivity can be defined as the movement of the charge carriers due to alternating electric field of the incident electromagnetic waves produced the electrical conductivity. Optical conductivity ( $\sigma_{op}$ ) can be determined as: [10].

$$\sigma_{op} = \frac{\alpha n c}{\pi} \quad \dots\dots\dots(11)$$

### Materials and Methods

In this paper the materials used are polymers (Poly vinyl alcohol (85 wt. %)), (Poly ethylene oxide (15 wt. %)) as a polymer blend and the nanoparticles as magnesium oxide as additive. To get a homogeneous solution to (PVA-PEO) blend has been used magnetic stirrer in mixing process when each of the (PVA and PEO) polymers are dissolved in distill water. Aluminum oxide NPs were added to solution with concentrations is (0, 2, 4, and 6) wt. %. To preparation the (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites, the casting method has been used. The dielectric properties of nanocomposites were measured in frequency ranges from  $10^2$  Hz to 1MHz by using LCR meter type (HIOKI 3532-50 LCR HI TESTER). The films of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites were prepared by dissolving (4g) of polymers in (60

ml) of distilled water with different concentrations are 85 wt.% PVA and 15 wt.% PEO by using a magnetic stirrer to blend the polymers for one hour to get a more homogeneous solution. Aluminum oxide nanoparticles are added to (PVA-PEO) blend with different concentrations are (0, 2, 4 and 6) wt. %. The sample was placed between copper plate and applied load on two plates where sample between them. The capacitance for different applied load range (60-300) bar was measured by using LCR meter type (HIOKI 3532-50 LCR HI TESTER at 100Hz, which locally manufactured.

To measure the optical properties of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites in the range of wavelength (200-1100) nm, The UV/1800/ Shimadzu spectrophotometer device has been used.

### Results and Discussion

Figure 1 show the photomicro graphs for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. From these shapes are noted homogenous distribution of  $\text{Al}_2\text{O}_3$  NPs through (PVA-PEO) blend and they are form a path network in the (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites when the concentration of nanoparticles exceeds 6 wt. %.

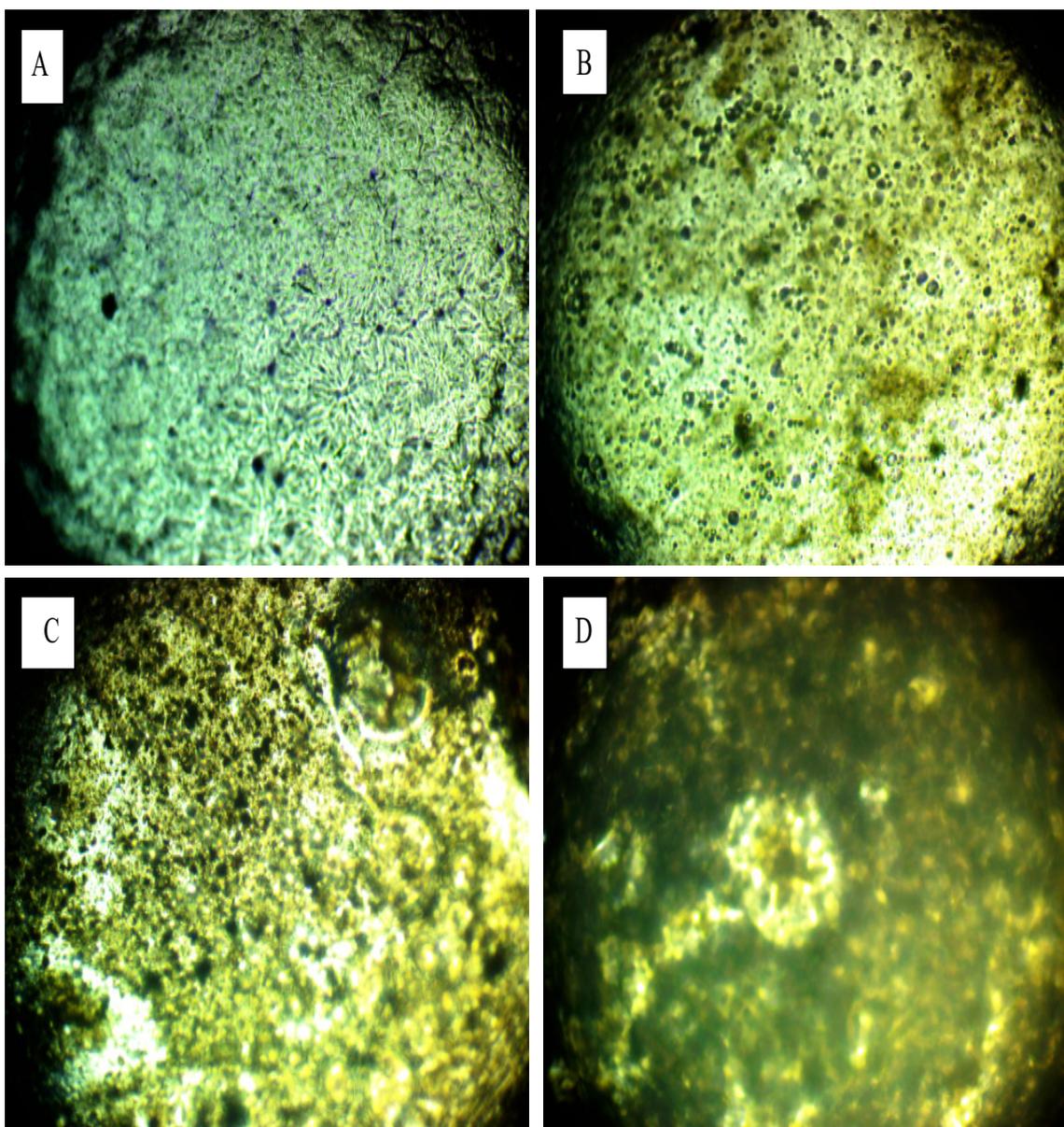


Fig. 1. Photomicrographs (x40) for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites :  
(A) for pure (B) for 2 wt.%  $\text{Al}_2\text{O}_3$  NPs (C) for 4 wt.%  $\text{Al}_2\text{O}_3$  NPs

Figure 2 shows the FTIR spectra of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. Double bonds of OH groups in the polymer matrix chain are observed for all samples of nanocomposites at around  $3255.75\text{ cm}^{-1}$  and  $3648.99\text{ cm}^{-1}$ . At  $(1088.71)\text{ cm}^{-1}$  the other bands as (C-O-C) group are noted due to the PVA film was not so transparent. Bonds of C-H groups are presented at  $(2955-2918)\text{ cm}^{-1}$ . The C=C stretching mode is appeared in the bands at  $(1706-1698.48)\text{ cm}^{-1}$ . The peak at  $(1091.02-1420.46)\text{ cm}^{-1}$  the band is strong for all samples of nanocomposites this is

due to the stretching mode of C-O group. The two strong bands observed at around  $(1418\text{ cm}^{-1}$  and  $842\text{ cm}^{-1})$  are due to the stretching and bending modes of  $\text{CH}_2$  group respectively [11].

The interaction between polymer blend and  $\text{Al}_2\text{O}_3$  NPs leads to change in spectral of (PVA-PEO) blend. From the FTIR studies note that there are no interactions between (PVA-PEO) blend and  $\text{Al}_2\text{O}_3$  NPs. Due to the increase in  $\text{Al}_2\text{O}_3$  NPs concentrations, the transmittance is decreased slightly as figures show [12].

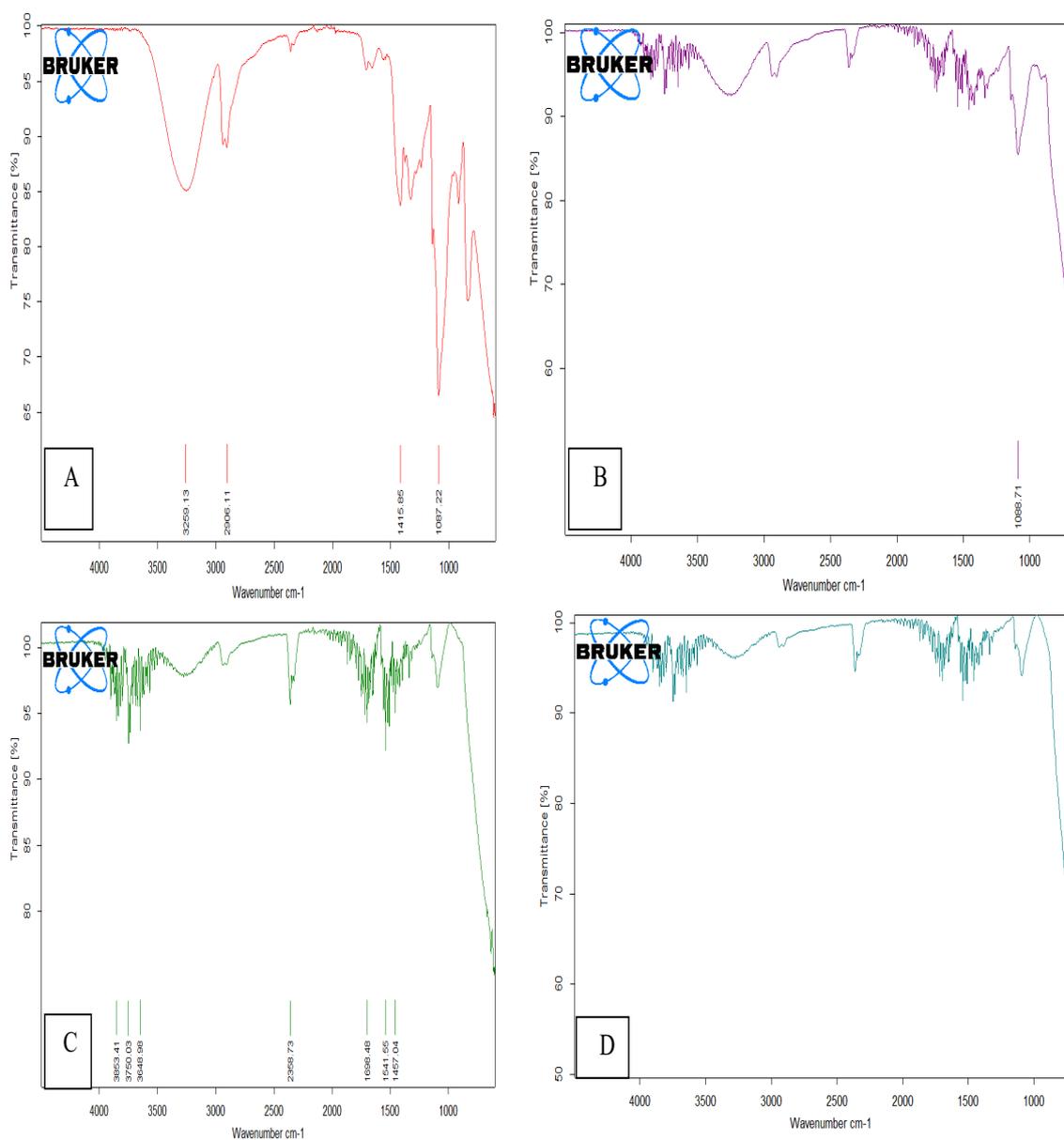


Fig. 2. FTIR spectra for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites A- (PVA-PEO) blend B- 2 wt.%  $\text{Al}_2\text{O}_3$  NPs C- 4 wt.%  $\text{Al}_2\text{O}_3$  NPs D- 6 wt.%  $\text{Al}_2\text{O}_3$  NPs.

Figures 3 and 4 show the plot between dielectric parameters against the frequency for different concentrations of aluminum oxide nanoparticles at room temperature. At low frequencies; each of dielectric constant and dielectric loss of nanocomposites are high, but they are decreasing rapidly when frequency increases due to the electric dipoles as interfacial polarization have sufficient time to adjust with the electric field before the field changes its

direction. On the other hand, at high frequencies, the dielectric constant and dielectric loss of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites are decreased due to the limited time available for electric dipoles compatibility. As a result, they are increased with increasing of aluminum oxide ( $\text{Al}_2\text{O}_3$ ) nanoparticles concentrations, due to the increase in charge carrier density in (PVA-PEO) matrix [13-21].

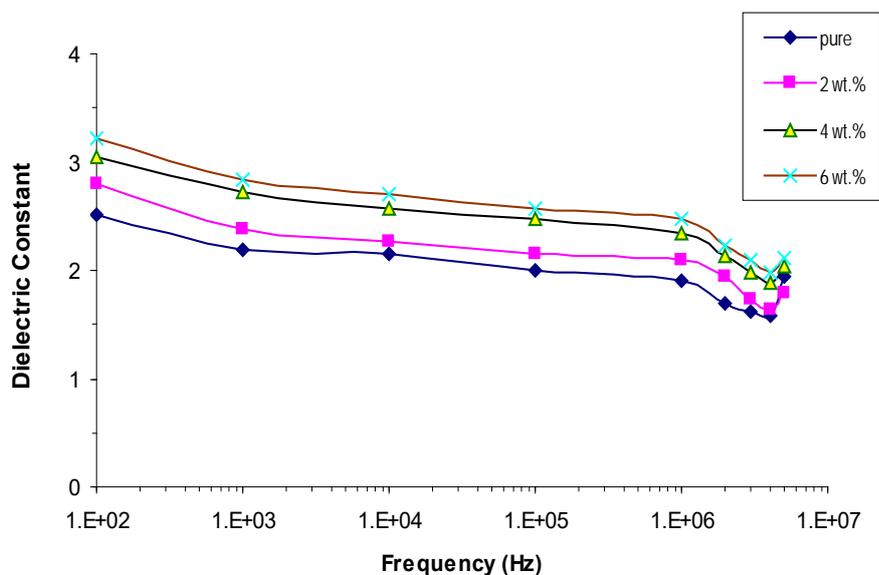


Fig. 3. variation of dielectric constant with the frequency for different concentrations of aluminum oxide nanoparticles at room temperature.

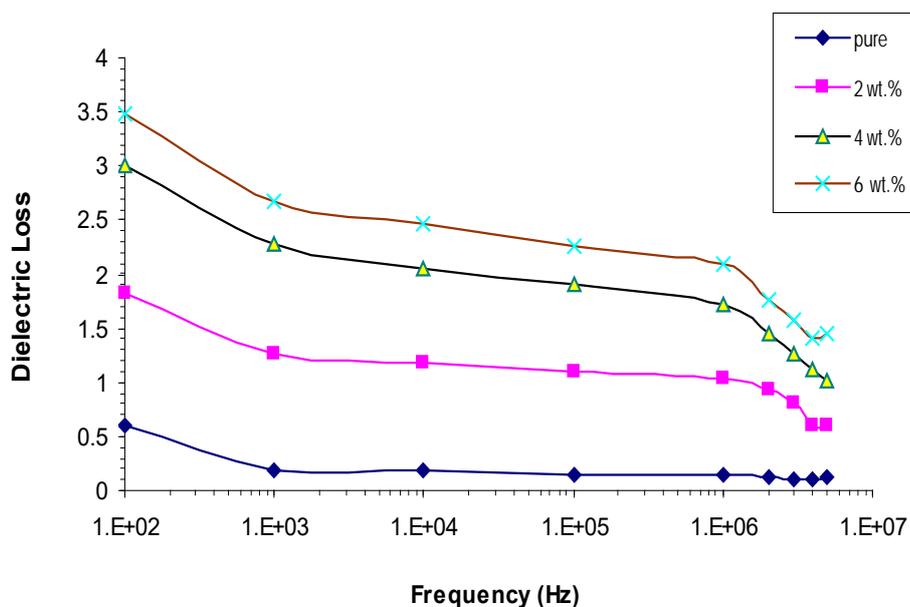


Fig. 4. variation of dielectric loss with the frequency for different concentrations of aluminum oxide nanoparticles at room temperature.

Figure 5 shows the variation between A.C conductivity and frequency for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. It is observed that conductivity increases with increasing frequency due to interfacial polarization. In addition to; the increasing of conductivity for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with increase in  $\text{Al}_2\text{O}_3$  NPs. The increase of electrical conductivity with increase in  $\text{Al}_2\text{O}_3$  NPs attributed to increase the charges carries in (PVA-PEO) blend [22-30].

Figure 6 shows the plot between dielectric constant for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with different concentrations of  $\text{Al}_2\text{O}_3$  NPs versus applied load. It's observed that dielectric constant increases with increasing applied load of (PVA-

PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites due to the samples are exhibited high polarization and ability to orient the molecular dipoles. Where both of space charge and orientation polarization are responsible for piezoelectricity in these nanocomposites. In addition to; (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites are more flexible [9]. On the other hand the increase in dielectric constant may be attributed to the increase of aluminum oxide nanoparticles additive because of only each oxygen atom has the same distance to the aluminum atoms. This is because of the formation of net dipole moments owing to the change in the position of the atoms owing to mechanical stress, which led to creation of polarization and an electric field,, respectively [3].

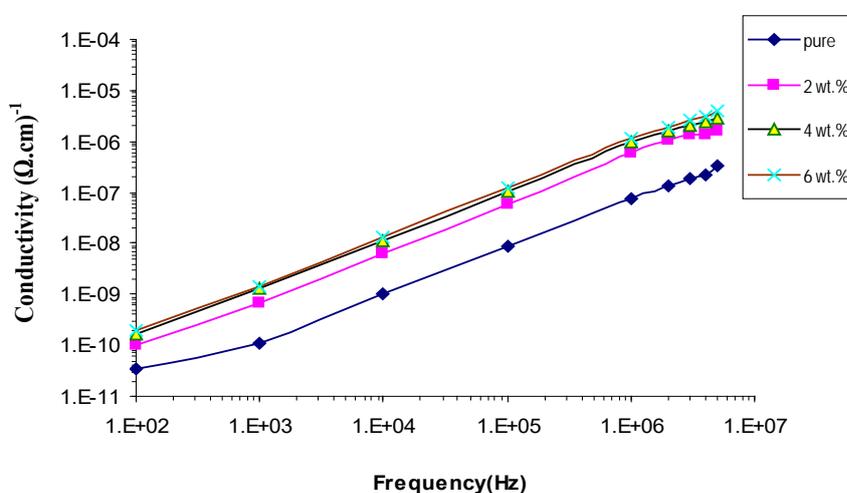


Fig. 5. variation of A.C conductivity with the frequency for different concentrations of aluminum oxide nanoparticles at room temperature.

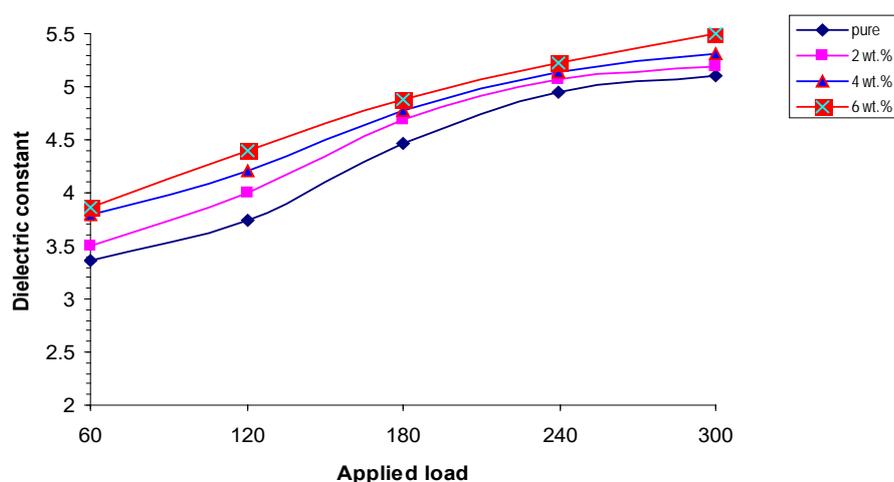


Fig. 6. variation of dielectric constant for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with applied load at 100Hz.

To know the effects of aluminum oxide filler on the optical properties of (PVA-PEO) blend, UV-visible absorption spectra for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites was measured. Fig. 7 shows the absorbance for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. It's indicated the intensity of the peak increase with increases aluminum oxide filler. All samples have higher absorbance at UV region. The absorption band shifts due to specific weight  $\text{Al}_2\text{O}_3$  NPs and the formation of intermolecular hydrogen bonding existing between aluminum ions with the adjacent OH groups of the PVA and PEO main chain. Its note that (PVA-PEO) blend has low absorbance because of the distance between valence and conduction band is higher. The increase in aluminum oxide particles leads to the increase in absorption which is related to the increase the number of carries charge inside nanocomposite [31-44].

Figure 8 shows the plot of absorption coefficient vs. incident photon energy for (PVA-PEO) blend with different percentages of aluminum oxide nanoparticles  $\text{Al}_2\text{O}_3$  NPs. It shows the absorption low when energy is low, this means the electron transitions are low. In the high energy, absorption becomes large this indicate the highest probability for electron transitions. The absorption coefficient for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites is increased by increase of  $\text{Al}_2\text{O}_3$  NPs additive [45]. The advantage of the absorption coefficient to conclude the transition electrons nature, when the value of absorption

coefficient is high in the higher energy expected direct transition of electrons. The energy and momentum conservation can by electrons and photons, when the value of absorption coefficient low expected indirect transition of electrons. The momentum conservation is by phonons only. From the results it's indicated absorption coefficient for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites has values are less than ( $10^4 \text{ cm}^{-1}$ ) which mean it's have indirect energy band gap [46].

Figure 9 shows the variations between absorbance edge  $(\alpha h\nu)^{1/2}$  for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites as a function of photon energy. It can be indicated that the values of forbidden energy gap (allowed) of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites are decreased with increasing aluminum oxide nanoparticles concentration this is attributed to great localize levels in the forbidden energy gap, in addition to oxygen vacancies of  $\text{Al}_2\text{O}_3$  NPs which form non-stoichiometry [45-47].

Figure 10 shows the variations between absorbance edge  $(\alpha h\nu)^{1/3}$  for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites as a function of photon energy. Its indicate that the values of forbidden energy gap (prevent) for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites decrease with increase aluminum oxide nanoparticles concentration this attribute to forms new levels and the transition of electrons between the tails of localize of the new levels made by the  $\text{Al}_2\text{O}_3$  NPs additive [31,49].

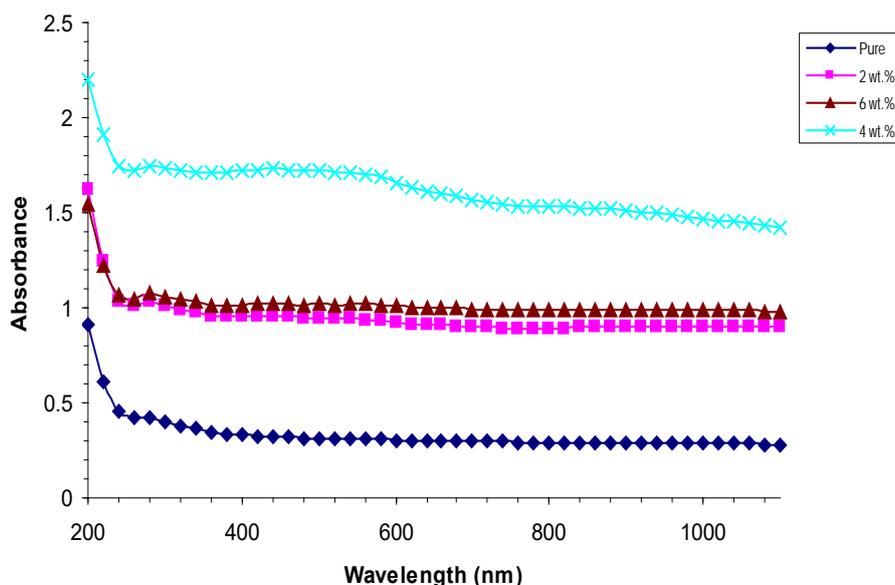


Fig. 7. variation of absorbance for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with wavelength.

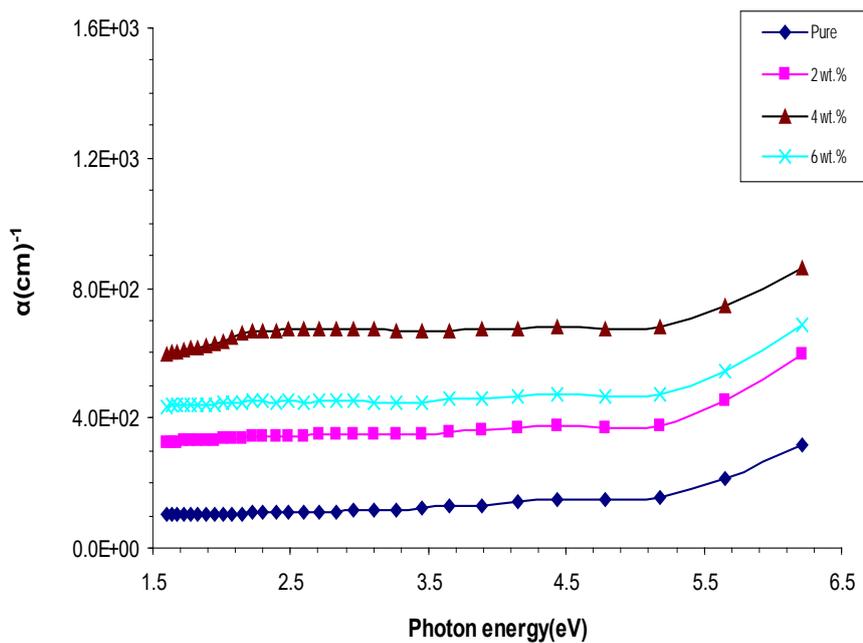


Fig. 8 . variation of absorption coefficient ( $\alpha$ ) for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with photon energy.

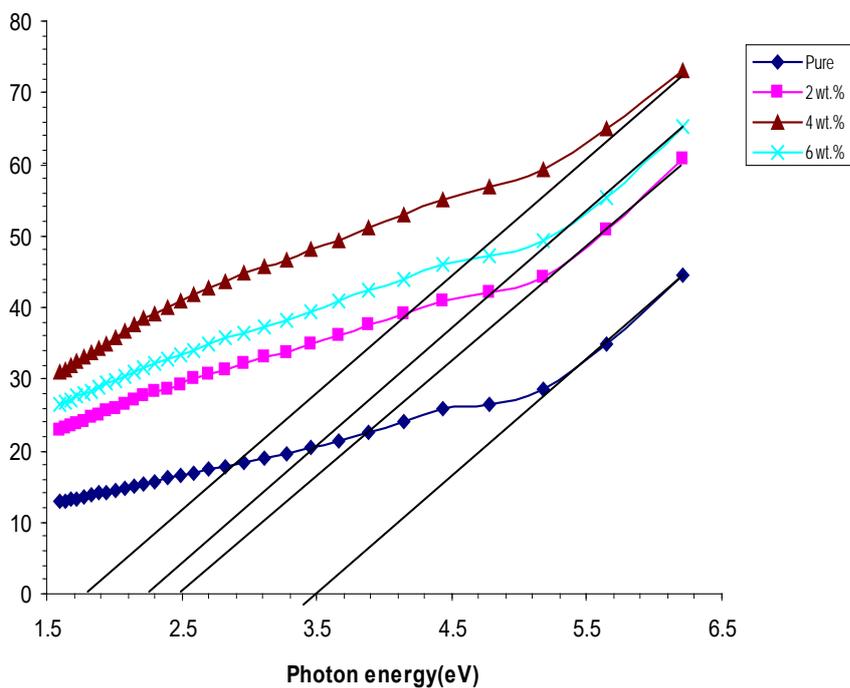


Fig. 9 . Variation of  $(\alpha h\nu)^{1/2}$  for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with photon energy.

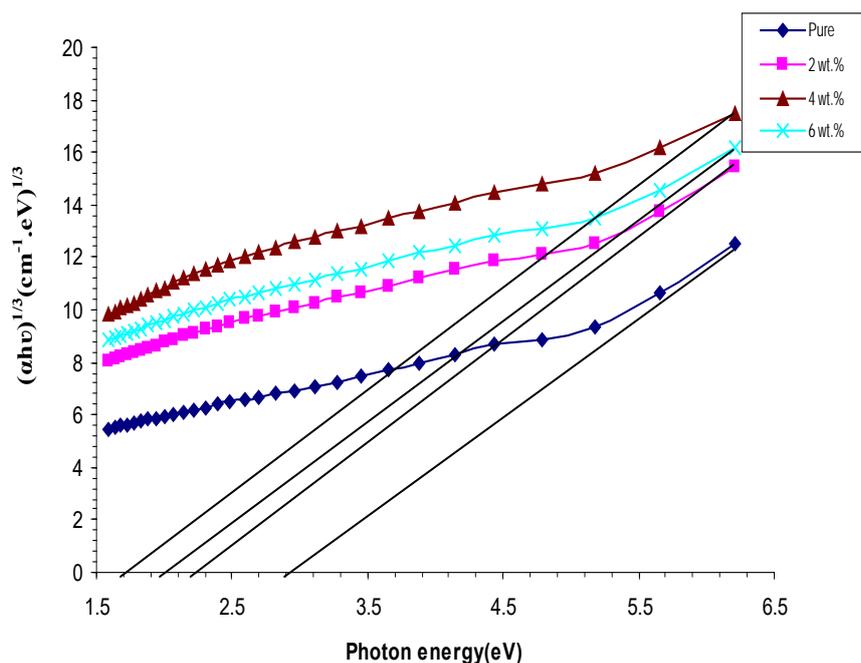


Fig. 10 . variation of  $(\alpha h\nu)^{1/3}$  for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with photon energy.

To describe the attenuation of the waves must be studying the extinction coefficient, Fig. 11 shows the variations of extinction coefficient against wavelength for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. It shows an increase in extinction coefficient with increasing of aluminum oxide nanoparticles for (PVA-PEO) blend. The increasing of extinction coefficient is attributed to high absorption coefficient. Where aluminum oxide nanoparticles will modify the structure of the host (PVA-PEO) blend. An interesting result that when the concentration of  $\text{Al}_2\text{O}_3$  NPs increases the absorbance in the visible region increases [50].

To describe the electromagnetic waves in the medium of propagation must be studied refractive index Fig. 12 illustrates the plot of the variations of refractive index with wavelength for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. There refractive index of (PVA-PEO) blend increases with increasing the aluminum oxide nanoparticles, which attributed to increase the scattering of incident photon which causes to increase the reflectance. In addition to; further addition of aluminum oxide nanoparticles causes increasing the intensity for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites [45].

Figure 13 shows the variations of real part dielectric vs. wavelength for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. It can be indicated that real part dielectric mainly proportional to the square of refractive index. So, it is increased with increasing  $\text{Al}_2\text{O}_3$  NPs additive

Figure 14 show the variations of imaginary part dielectric vs. wavelength for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. Its proportional to the extinction coefficient as shown in equation (11) and its increased with the increasing of the aluminum oxide nanoparticles [9].

Figure 15 shows the optical conductivity with wavelength for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites. The optical conductivity is increased for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites due to aluminum oxide nanoparticles  $\text{Al}_2\text{O}_3$  NPs are increased which leads to increase the absorption coefficient [12]. In addition to; This increase due to the creation of new levels in the band gap, lead to ease of passage carrier charges from the valence to the conduction band, as a result decreasing in the band gap and the conductivity increase [50-52].

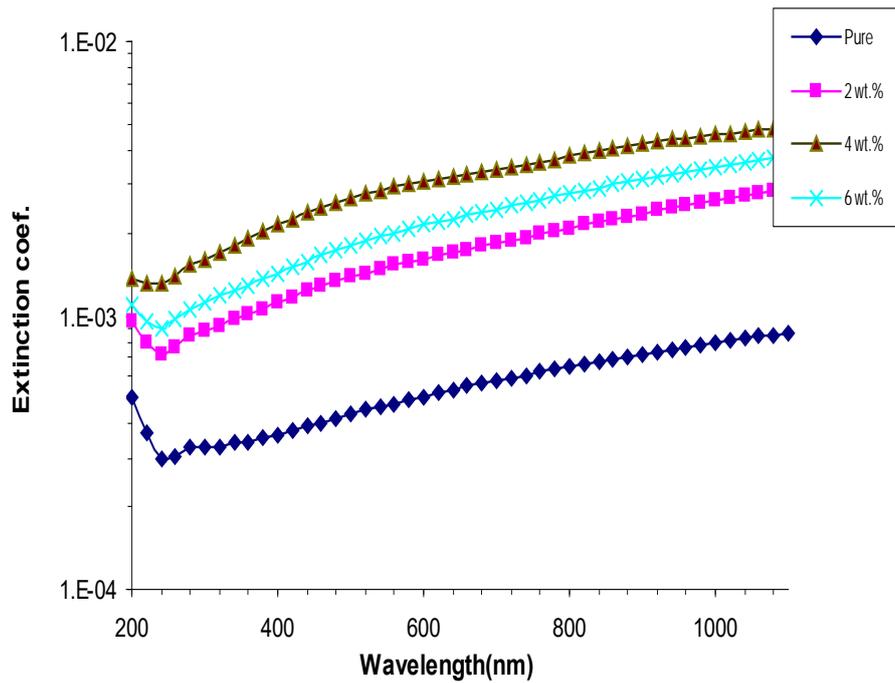


Fig. 11. variation of extinction coefficient for (PVA-PEO-Al<sub>2</sub>O<sub>3</sub>) nanocomposites with wavelength.

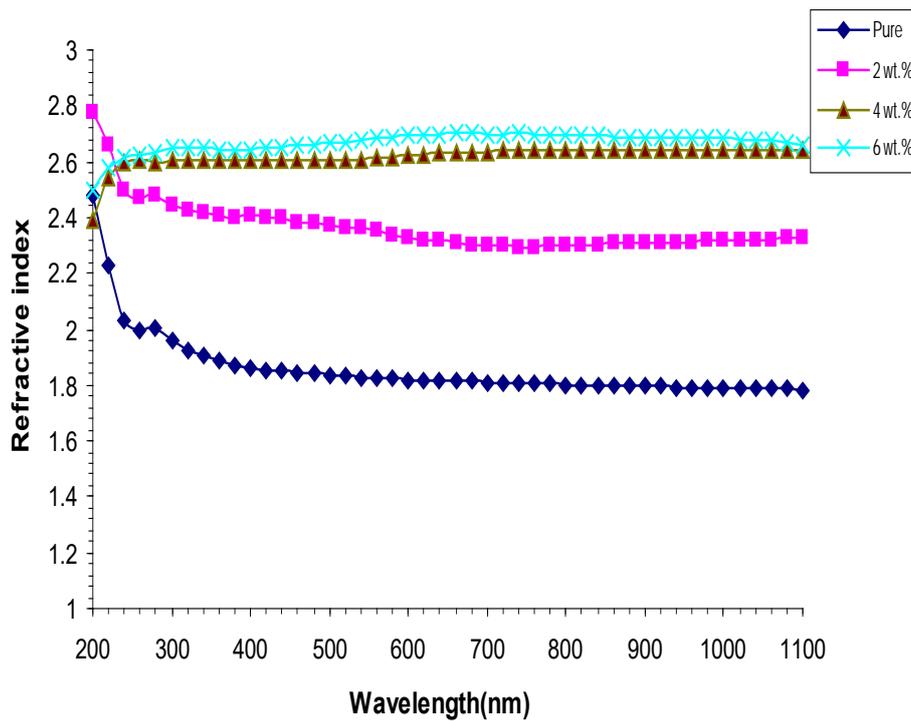


Fig. 12 . Variation of refractive index for (PVA-PEO-Al<sub>2</sub>O<sub>3</sub>) nanocomposites with wavelength.

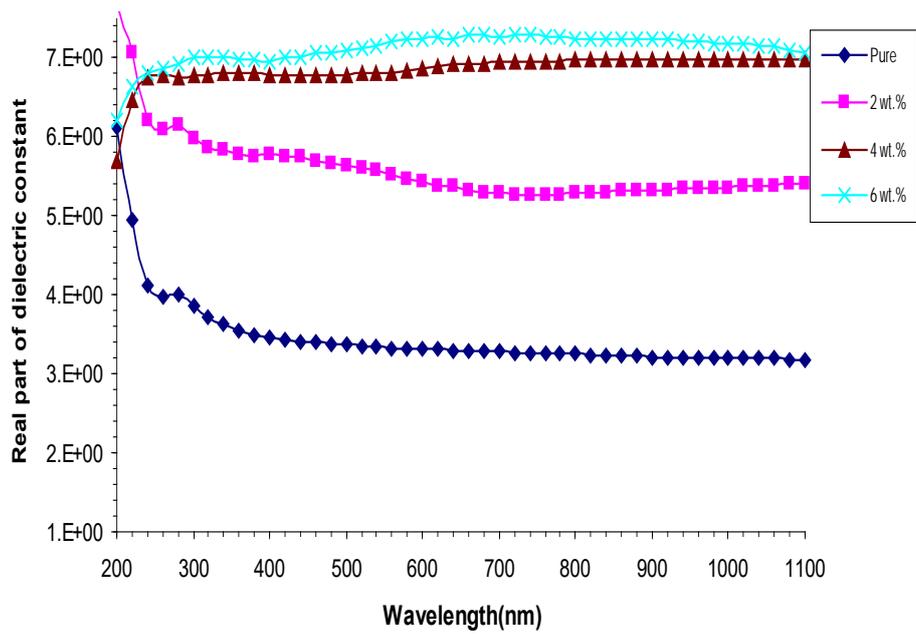


Fig. 13 . variation of real part of dielectric constant for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with wavelength.

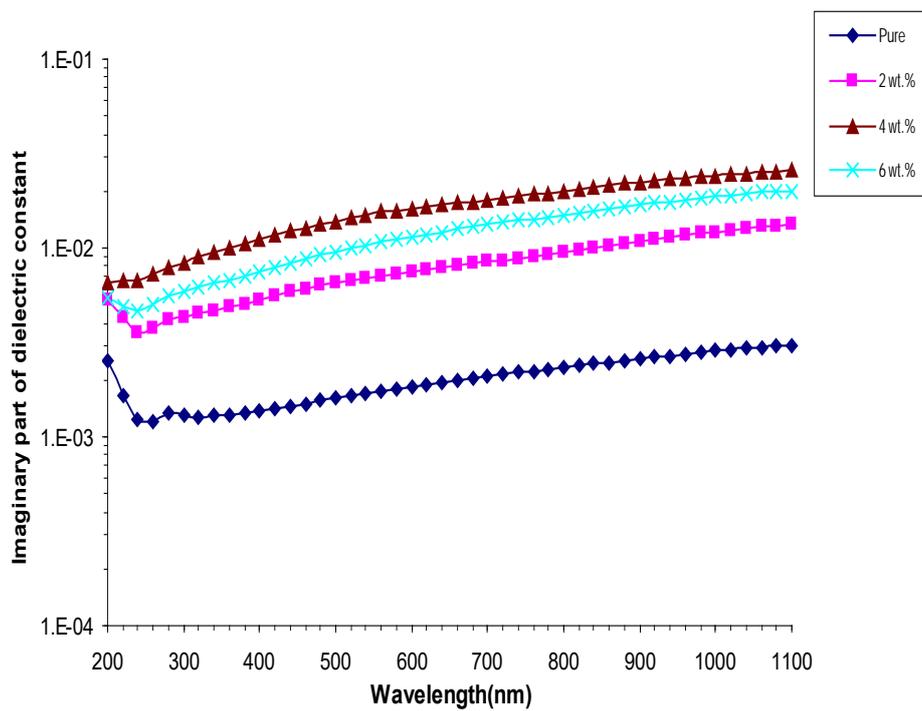


Fig. 14 . variation of imaginary part of dielectric constant for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with wavelength.

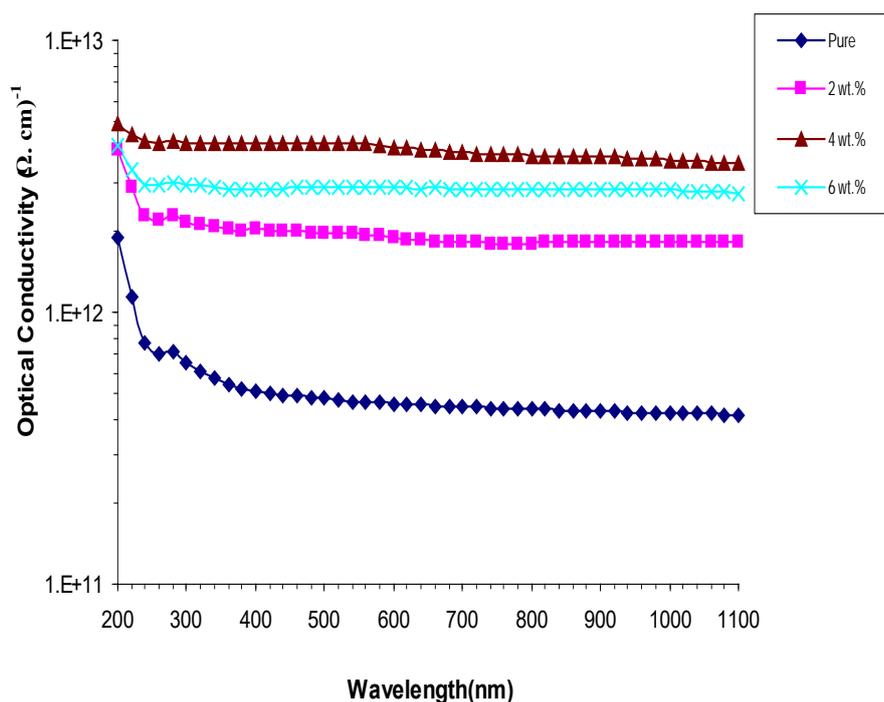


Fig.15. Variation of optical conductivity for (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites with wavelength.

### Conclusions

1. The dielectric constant of PVA-PEO blend increases about 77% at 100Hz by increasing the concentrations of  $\text{Al}_2\text{O}_3$  NPs to 6 wt.%. Moreover, it is increased to about 86% when the applied load is 60 bars and increase in the concentrations of  $\text{Al}_2\text{O}_3$  nanoparticles is 6 wt.%. So, (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites can be used as piezoelectric sensors.
2. The dielectric loss of (PVA-PEO) blend has increased about 17.6 % at 100 Hz by increasing concentrations of  $\text{Al}_2\text{O}_3$  nanoparticles at 6 wt.%. Moreover, it increased to about 13.7% when mechanical stress is 60 bars and increase in the percentage weight of  $\text{Al}_2\text{O}_3$  nanoparticles at 6 wt.%.
3. The optical measurements are shown that in infrared region the absorbance of (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites has increased about 59% by increasing the percentage weight of  $\text{Al}_2\text{O}_3$  nanoparticles at 6 wt.%. So, (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites can be used as shielding systems.
4. The indirect energy gap ( $E_g$ ) of (PVA-PEO) blend decreases about 48% with increasing of concentrations of  $\text{Al}_2\text{O}_3$  nanoparticles at 6wt.%. So, (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites can be used as opt electric devices.

5. The results showed that the (PVA-PEO- $\text{Al}_2\text{O}_3$ ) nanocomposites can be used for different applications such as: sensors, solar cells, and medical fields.

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## الخصائص التركيبية، و الكهربائية المتناوبة والبصرية للمترابكات النانوية (بولي فينيل الكحول- بولي اثيلين اوكسايد- اوكسيد الالمنيوم) للأجهزة الكهروضغطية

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هناك حاجة لدراسة الخصائص التركيبية والكهربائية والبصرية للمواد المتداخلة لتحديد مدى ملاءمتها للتطبيقات الإلكترونية والصناعية والخلايا الشمسية. اكتسبت الخلائط البوليمرية والمترابكات النانوية أهمية كبيرة لدى الباحثين والعلماء. بالإضافة الى ان هذه المواد دخلت في تطبيقات الكترونية وصناعية واسعة. في هذا البحث , مترابكات نانوية من بولي فينيل الكحول- بولي اثيلين اوكسايد- جسيمات اوكسيد الالمنيوم النانوية تم تحضيرها بتراكيز وزنية مختلفة من اوكسيد الالمنيوم النانوية. بينت النتائج ان ثابت العزل الكهربائي للخليط (PVA-PEO) يزداد مع زيادة تراكيز جسيمات اوكسيد الالمنيوم النانوية وزيادة الحمل المطبق عند تردد ١٠٠ هيرتز. الفقدان العزلي لخليط (PVA-PEO) يزداد بزيادة التراكيز الوزنية لجسيمات اوكسيد الالمنيوم ويزداد الجهد المسلط عند التراكيز الواطئة. تزداد التوصيلية الكهربائية لخليط (PVA-PEO) مع زيادة تراكيز جسيمات اوكسيد الالمنيوم النانوية. القياسات البصرية بينت ان الامتصاصية للمترابكات النانوية (PVA-PEO-Al<sub>2</sub>O<sub>3</sub>) تزداد مع زيادة تراكيز جسيمات اوكسيد الالمنيوم النانوية. فجوة الطاقة غير المباشرة (E<sub>g</sub>) تقل مع زيادة تراكيز جسيمات اوكسيد الالمنيوم النانوية. الثوابت البصرية (معامل الامتصاص، معامل الخمود، معامل الانكسار، ثوابت العزل الحقيقي والخيالي، و التوصيلة البصرية للخليط (PVA-PEO) تتغير مع زيادة النسب الوزنية لجسيمات اوكسيد الالمنيوم النانوية.