



Structural, Dielectric and Optical Properties for (Polyvinyl Alcohol–Polyethylene Oxide- Manganese Oxide) Nanocomposites

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DUE to the scientific, industrial, electrical and medical importance, it was necessary to study the properties of overlapping nanocomposites. So it gained the attention and directed scientists and researchers in the study of its structure and electrical characteristics and other characteristics. So, In the present work, (polyvinyl alcohol- polyethylene oxide- manganese oxide nanoparticles) nanocomposites have been prepared with different concentration of blend and MnO₂ nanoparticles. The manganese oxide nanoparticles were added to (PVA-PEO) blend by weight percentages are (2, 4 and 6) wt.%. The optical microscope images indicate that the manganese oxide nanoparticles additive distribution was homogeneous in the (PVA-PEO) blend. FT-IR indicate that no change on chemical structures between the (PVA-PEO) blend and manganese oxide nanoparticles additives. The dielectric properties of nanocomposites were studied. The results showed that the dielectric constant, dielectric loss and electrical conductivity of (PVA-PEO) blend are increased with the increase in concentrations of manganese oxide nanoparticles. The dielectric constant and dielectric loss decrease while the AC electrical conductivity increases with the increase in frequency of applied electric field. Optical measurements are showed the absorbance of (PVA-PEO-MnO₂) nanocomposites is increased with increase of the concentrations of MnO₂NPs. With increasing of the concentrations of manganese oxide nanoparticles, the indirect energy gap (E_g) of (PVA-PEO) blend will be decreased. And by increasing of the weight percentages of manganese oxide nanoparticles, the optical constants as absorption coefficient, extinction coefficient, refractive index, real and imaginary dielectric constants of nanocomposites are varied.

Keywords : MnO₂NPs, Manganese oxide, Energy Gap, Nanoparticles, Polymer blend.

Introduction

In recent years, materials with suitable and sustainable properties have drawn the worldwide community attention. The purpose of doping variety materials to enhance of their properties. The factors the most influencing of these properties depend on the nature and the method of their synthesis. The developing desired devices required cost guide researchers and coaled applications on the choice of materials and the technology. Nanocomposites and polymer blends

are attracting increasing attention from scientists and researchers due to their use in many industrial sectors. Metal oxides like ZnO, TiO₂, SnO₂ and other semiconductors into organic matrices offer more high chemical and physical stability. Organic polymers as matrices and inorganic nanoparticles as filler are endowed the resulting nanocomposites with excellent optical, electrical and mechanical properties. Peroxides or so-called manganese dioxide nanoparticles MnO₂NPs are an inorganic, compound, brown or black-colored compound. It has wide uses as a catalyst, a wastewater processor,

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batteries and capacities. They are molecular weight 228.81 is found in the form of a solid powder whose melting point is 1564C, insoluble in water [1]. The manganese oxide nanoparticles MnO_2 NPs are concentrated due to its differentiated and catalytic adsorption properties. Many of its structural forms have been produced such as β , λ , α , depending on the preparation of MnO_2 nanoparticles. The manganese oxide nanoparticles have been examined through numerous intensive researches that have led to the discovery of their use in many scientific, medical and industrial fields such as antibacterial, biochemical sensors, moisture detection, and dye removal. In addition to : MnO_2 molecules are considered to be potent inhibitors of some bacterial types [2]. The rapidly and suddenly development of nanocomposites has led to interfere in many scientific, industrial, electronic and medical fields. Because they have near-visual and electrical properties which are remarkable, and better in comparison with the physical properties of traditional composites. The addition of very low percentages of nanomaterials, for example, 5% as additives to the polymer leads to improve on the physical properties so they are getting unique physical properties [3].

Nanocomposites exhibit the quality of properties by blending the process ability and flexibility of polymers with nano metal oxides [4]. Owing to flexibility, portability and the miniaturization requirements of electronic products, polymer blend materials that have flexibility and low dielectric constant will be getting more attention in integrated circuit devices as interlayer dielectrics. The heating of the integrated circuit can be reduced efficiently, due to reduce the dielectric constant of the dielectric material used in the integrated circuit. Another important problem about the application of polymer-based low dielectric materials are easy to burn. Polymer-based dielectric layers should have flame retardant properties, so they are used in electronic equipment safely. Compared with unfilled polymer blends, nanocomposites exhibit improved physical properties as dielectric properties, fire resistance and mechanical strength [5]. Poly (ethylene oxide) is a semi-crystalline, biocompatible, biodegradable, nonionic and water-soluble polymer of considerable industrial significance, which finds applications in many different branches of industry. PEO is an organic component in organic-anorganic hybrid material applied in advanced technologies in the field of functional coatings with superior barrier properties

[6]. Recently, some works have been reported the integration of thin films based on polymeric materials such as Polyvinyl alcohol (PVA) as a gate dielectric for the development of organic thin film transistors, due to the high dielectric constant which enhances the gate capacitance, with the advantages of solution processable material, at low cost, non-toxic, with flexible hydrophilic network and low temperature of deposition. PVA is a poor electrical conductor, water soluble, it has carbon chain backbone with OH groups and it is eco-friendly, and its physical properties may be adapted to a specific requirement in conjunction with inorganic materials [7].

Materials and Methods

Films of (polyvinyl alcohol- polyethylene oxide- manganese oxide) nanocomposites were prepared by using casting method. The (PVA-PEO) was prepared by dissolving 1 gm of (PVA and PEO) in 20 ml of distilled water by using magnetic stirrer for 1 hr. The manganese oxide nanoparticles were added to (PVA-PEO) blend with concentrations are (2, 4 and 6) wt.%. The dielectric properties of (PVA-PEO- MnO_2) nanocomposites were measured in frequency ranges from 100 Hz to 5×10^6 Hz by using LCR meter type (HIOKI 3532-50 LCR HI TESTER). The UV/1800/ Shimadzu spectrophotometer device is used to measure the optical properties of (PVA-PEO- MnO_2) nanocomposites in the range of wavelength (200-1100) nm

The dielectric constant (ϵ') of (PVA-PEO- MnO_2) nanocomposites is defined by the following equation [8] :

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

Where, C_p is parallel capacitance and C_o is vacuum capacitor

The dielectric loss (ϵ'') of (PVA-PEO- MnO_2) nanocomposites is given by the equation [9] :

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

Where, D : is dispersion factor.

The A.C electrical conductivity of (PVA-PEO- MnO_2) nanocomposites is defined by the following equation [10] :

$$\sigma_{A.C} = w \epsilon'' \epsilon_o \quad \dots\dots\dots(3)$$

Where, w is the angular frequency.

Absorption coefficient (α) is computed as the following equation :

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

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 [11]. Refractive index (n) is determined by following equation :

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

Where C is the reflectance, can be got from :

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

The extinction coefficient (k) is calculated by the following equation :

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

Where λ is the wavelength of incident light. The dielectric constant is classified into two parts real (ϵ_r), and imaginary (ϵ_{im}) [12]. It can be computed each of the real and imaginary parts of dielectric constant (ϵ_r and ϵ_{im}) as the following equations:

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

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

optical conductivity is the movement of the charge carriers due to alternating electric field of the incident electromagnetic waves produced the electrical conductivity. Optical conductivity (σ_{op}) can be calculated as [13]:

$$\sigma_{op} = \frac{\alpha n c}{4\pi} \quad \dots\dots\dots(11)$$

Results and Discussion

Figure 1 shows the Fourier transform infrared spectra of (PVA-PEO- MnO_2) nanocomposites. For all samples of nanocomposites show broad bands at around 3257.15 cm^{-1} and 2938.87 cm^{-1} are observed because the double bonds of OH groups in the polymer matrix chains. The other bands as (C-O-C) group can be noted in the bands at $(1086.30) \text{ cm}^{-1}$ were attributed to the PVA film was not so transparent. C-H groups are presenting at Peaks $(2888- 2906.1) \text{ cm}^{-1}$ and at peaks

$(1698.57-1698) [14]$. Peaks at $(1086.30) \text{ cm}^{-1}$ were due to the presence of C-O groups. The interaction between MnO_2 NPs and polymer blend causes these changes change in spectral of (PVA-PEO) blend involves a shift in some single and double bonds and change in the intensities due to manganese oxide nanoparticles only [12]. From the FTIR studies note that there are no interactions between (PVA-PEO) blend and MnO_2 NPs. From these figures we noted the transmittance decreases slightly with the increase of MnO_2 NPs concentrations because of the increase in density of nanocomposites [15].

Figures 2 and 3 show the variation of dielectric constant and dielectric loss of (PVA-PEO- MnO_2) nanocomposites films as a function of frequency at different manganese oxide nanoparticles concentrations. The dielectric constant values of (PVA-PEO- MnO_2) nanocomposites films decreases with the increase of frequency. This may be attributed to the tendency of dipoles in polymeric samples to orient themselves in the direction of the applied field. Dielectric loss of (PVA-PEO- MnO_2) nanocomposites drops suddenly with an increase in frequency. This behavior may be due to the fact that at lower frequencies, the dipoles have sufficient time to align with the applied field before it changes its direction. Consequently, the dielectric loss is high [16]. The dielectric constant and dielectric loss decrease with the increase of MnO_2 nanoparticles concentration, this result can be attributed to the increase in conductivity as a result of the increase charge carrier density in polymer matrix [17-25], as shown in Fig. 4. The figure shows that the Fe_2O_3 nanoparticles is aggregated as a clusters at lower concentrations. When increasing the concentrations of Fe_2O_3 nanoparticles, the nanoparticles form a continuous network inside the (PVA-PEO) nanocomposites at concentration 4 wt% for nanocomposites.

Figure 5 shows the variation of A.C electrical conductivity of (PVA-PEO- MnO_2) nanocomposites with frequency for different concentrations of manganese oxide nanoparticles. It is clearly observable from this figure that A.C conductivity increases as the dopant concentration increases. In (PVA-PEO) blend, as the bond rotates with frequency, the existing flexible polar groups with polar bonds cause dielectric α -transition. Thus, there is a change in chemical composition of the polymer repeated unit due to the formation of charge transfer complexes (CTCs) within the

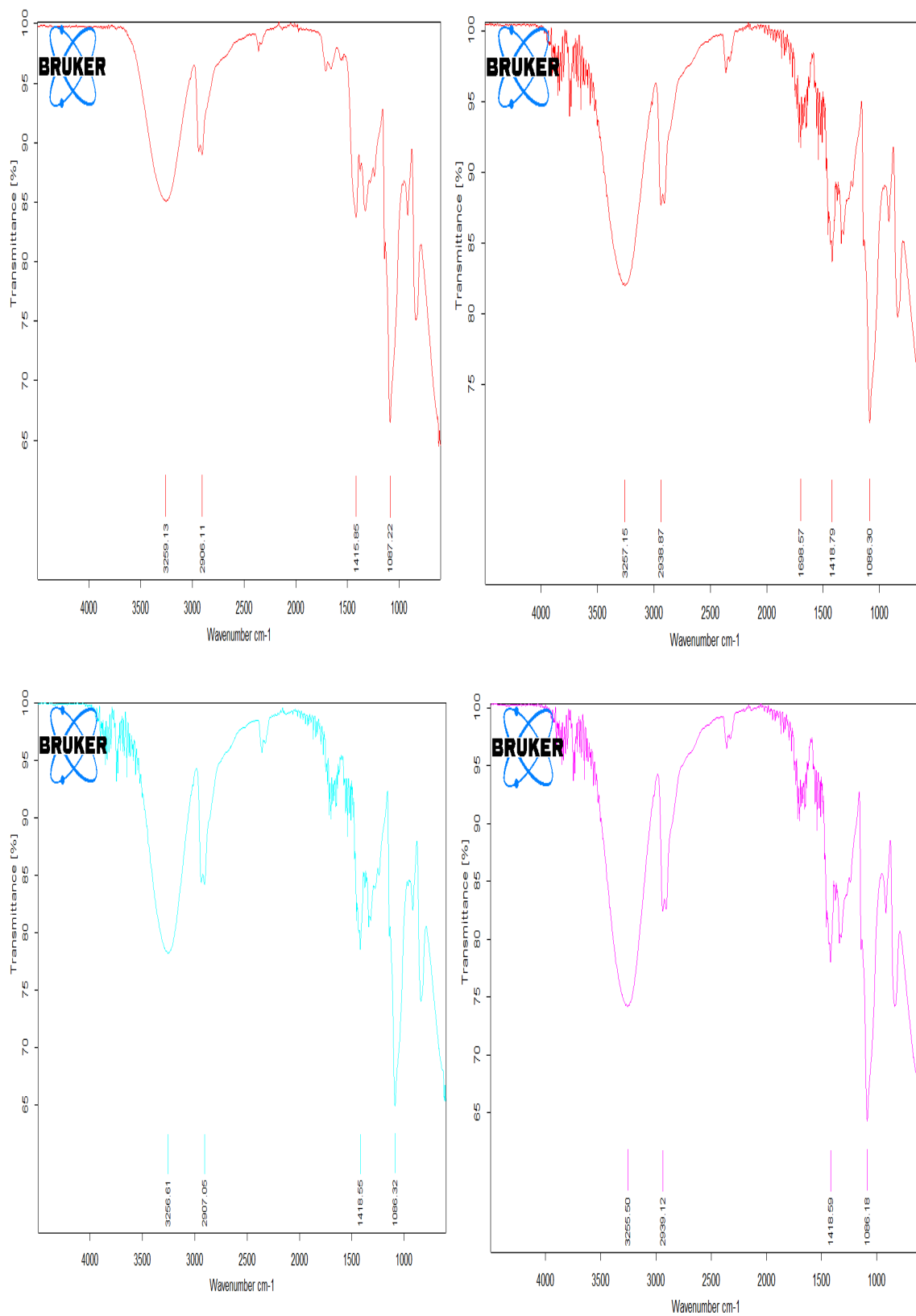


Fig. 1. FTIR spectra for (PVA-PEO-MnO₂) nanocomposites.

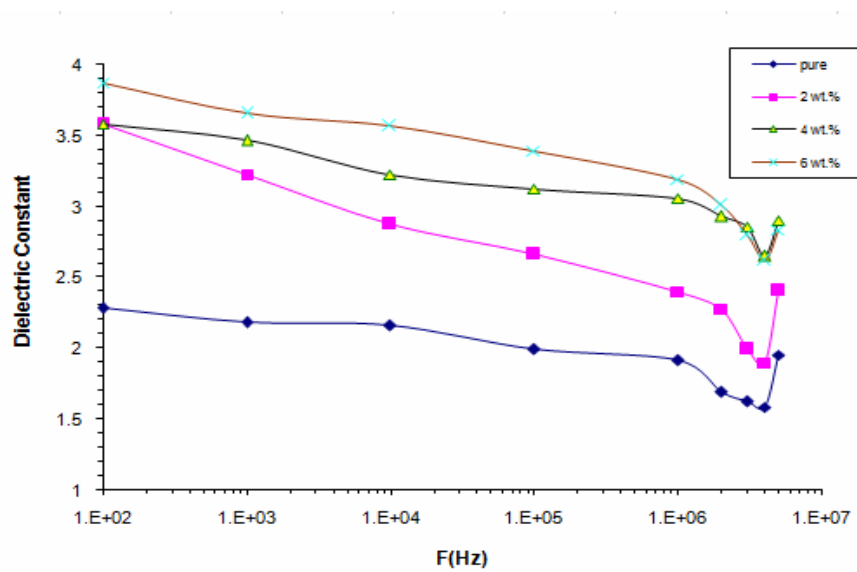


Fig. 2. Variation of dielectric constant for (PVA-PEO-MnO₂) nanocomposites films as a function of frequency at different manganese oxide nanoparticles

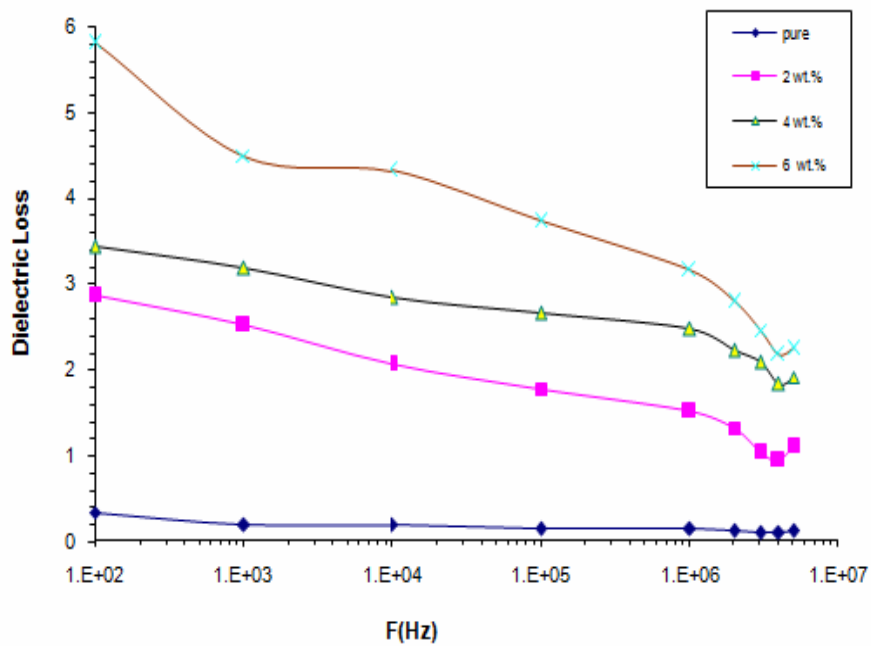


Fig. 3. Variation of dielectric loss for (PVA-PEO-MnO₂) nanocomposites films as a function of frequency at different manganese oxide nanoparticles concentrations.

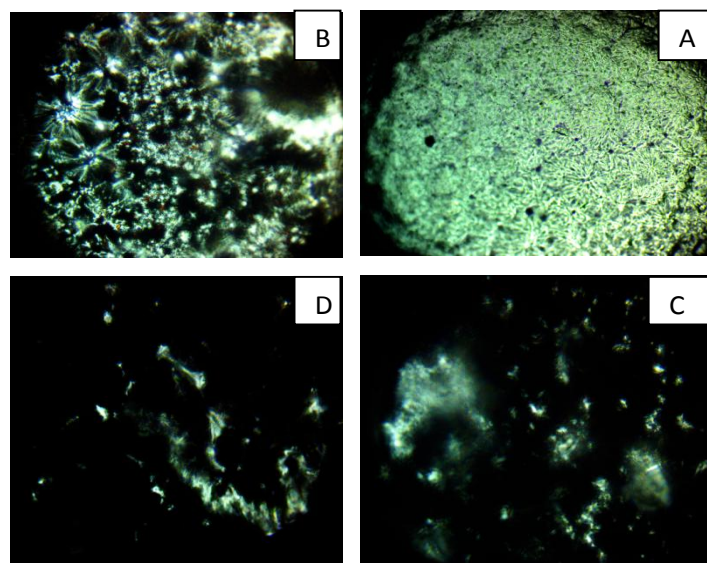


Fig. 4. Photomicrographs (x40) for (PVA-PEO-MnO₂) nanocomposites: (A) for pure (B) for 2 wt.% MnO₂ nanoparticles (C) for 4 wt.% MnO₂ nanoparticles

(PVA-PEO) blend chains, which in turn makes the polymer chains more flexible and hence enhances the A.C electrical conductivity [26]. Also, the increase in conductivity with increase the additive concentration may be attributed to increase the charge carriers numbers in polymer blend [27-35].

To know the effects of manganese oxide filler on the optical properties of (PVA-PEO) blend, UV-visible absorption spectra for (PVA-PEO-MnO₂) nanocomposites was measured.

Figure 6 shows the absorbance for (PVA-PEO-MnO₂) nanocomposites. It's indicated that intensity of the peak increase by increasing manganese oxide additive [9]. Due to specific weight MnO₂NPs, the absorption band shifts toward the longest wavelength. They are giving an idea about the formation of intermolecular hydrogen bonding existing between manganese ions with the adjacent OH groups of the (PVA-PEO) blend chain. Due to the distance between valence and conduction band of (PVA-PEO) blend is higher, its low absorbance. The increase in absorption for (PVA-PEO-MnO₂) nanocomposites, due to increase in manganese oxide nanoparticles which is related to increase the charges carries numbers [7, 36-41].

Figure 7 shows the absorption coefficient against incident photon energy for (PVA-PEO) blend with different percentages of manganese oxide nanoparticles. It shows when energy is low,

the absorption low due to the electron transitions are low. But in the high energy, absorption becomes large due to the higher probability for electron transitions. By increasing of MnO₂NPs additive, absorption coefficient for (PVA-PEO-MnO₂) nanocomposites is increased [10]. The purpose of the absorption coefficient lies in 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 be 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 noted indirect energy band gap due the absorption coefficient for (PVA-PEO-MnO₂) nanocomposites has values are less than (10⁴ cm⁻¹) [11].

Figure 8 shows the variations between absorbance edge $(\alpha h\nu)^{1/2}$ for (PVA-PEO-MnO₂) nanocomposites vs. photon energy. By plotting a line from the upper side of the curve in direction axis (X) in value $((\alpha h\nu)^{1/2} = \text{zero})$ to get indirect forbidden energy gap transition (allowed) [14]. With increasing manganese oxide nanoparticles concentration, the values of forbidden energy gap (allowed) of (PVA-PEO-MnO₂) nanocomposites are decreased. Because of great localized levels in the forbidden energy gap [42-45]. In addition to attribute to oxygen vacancies of MnO₂NPs which due to form non-stoichiometry [11].

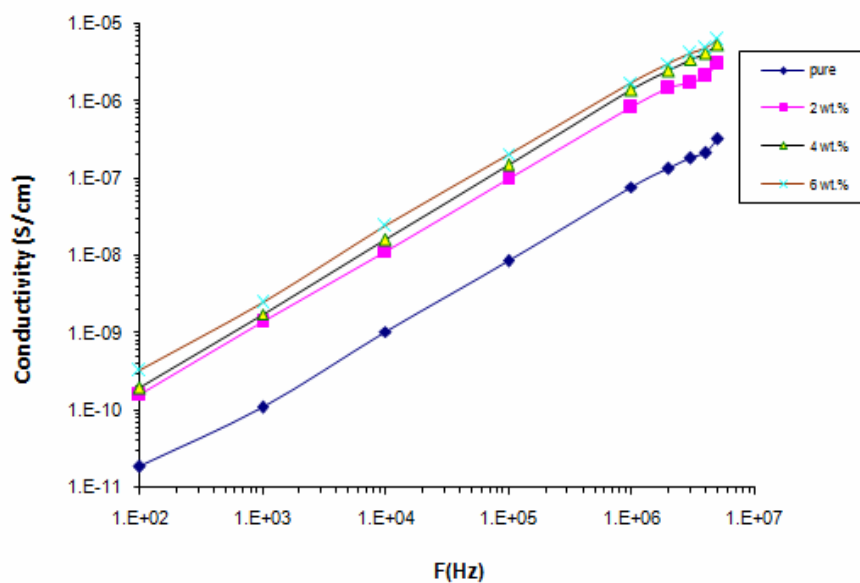


Fig. 5. Variation of A.C electrical conductivity of (PVA-PEO-MnO₂) nanocomposites with frequency for different concentrations of manganese oxide nanoparticles.

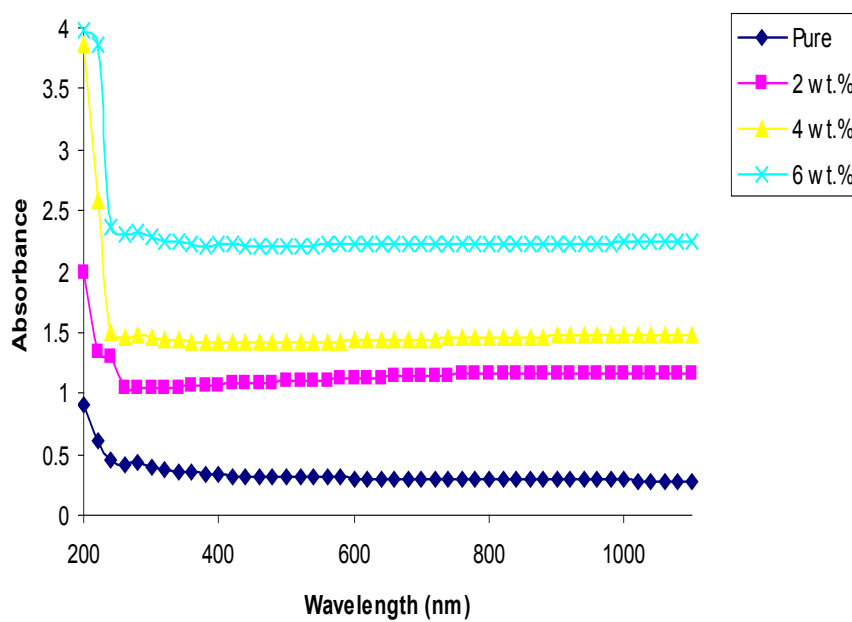


Fig. 6. Variation of absorbance for (PVA-PEO-MnO₂) nanocomposites with wavelength.

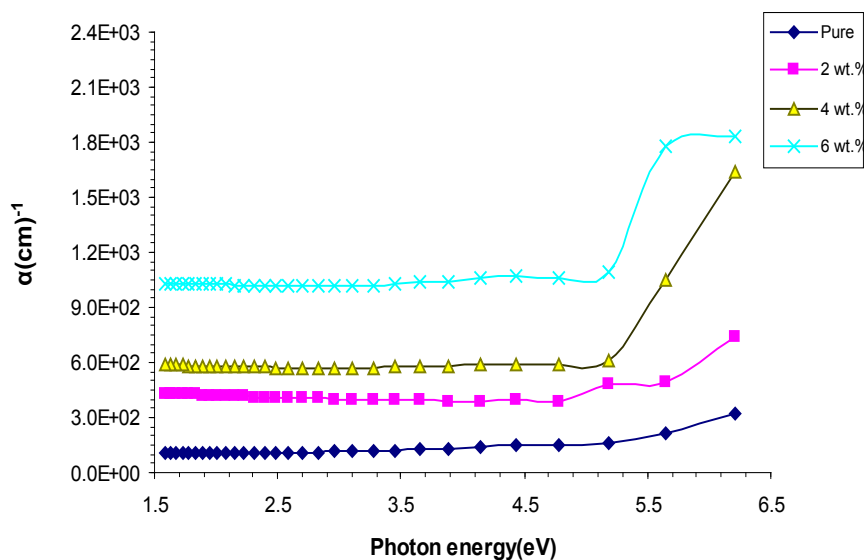


Fig. 7 . Variation of absorption coefficient (α) for(PVA-PEO- MnO_2) nanocomposites with photon energy.

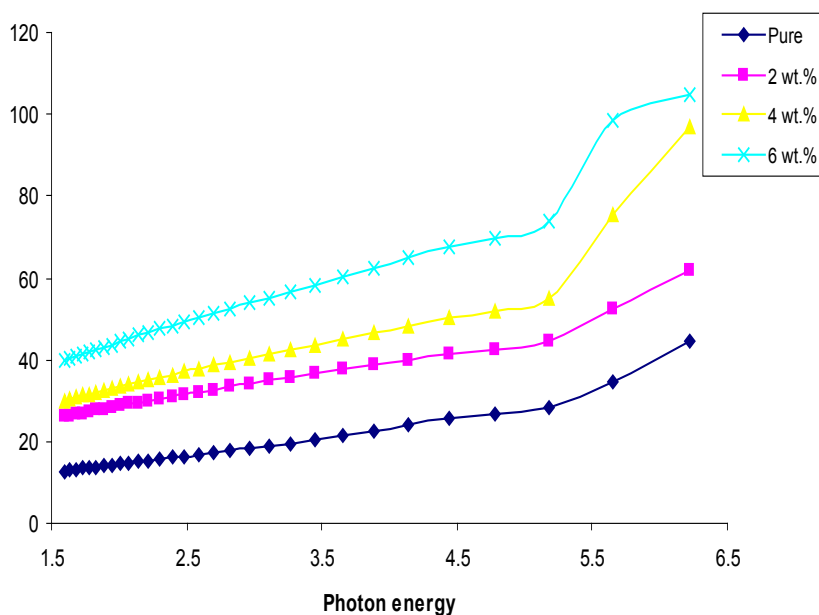


Fig. 8 . Variation of $(\alpha h\nu)^{1/2}$ for (PVA-PEO- MnO_2) nanocomposites with photon energy.

Figure 9 shows the variations between absorbance edge $(\alpha h\nu)^{1/3}$ for (PVA-PEO- MnO_2) nanocomposites vs. photon energy. With increasing manganese oxide nanoparticles concentration the values of forbidden energy gap (prevent) for (PVA-PEO- MnO_2) nanocomposites are decreased, this attribute to forms new levels and the transition of electrons between the tails of localize of the new levels made by the MnO_2 NPs additive [14].

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Figure 10 shows the variations of extinction coefficient as a function of wavelength for (PVA-PEO- MnO_2) nanocomposites. It shows by increasing of manganese oxide nanoparticles for (PVA-PEO) blend the extinction coefficient is increased, due to high absorption coefficient for (PVA-PEO- MnO_2) nanocomposites . Where manganese oxide nanoparticles will modify

the structure of the host (PVA-PEO) blend. An interesting result that when the concentration of MnO_2 NPs increases the absorbance in the visible region increases [13].

To describe the electromagnetic waves in the medium of propagation must be studied refractive index Fig.11 illustrates the plot of the variations of refractive index with wavelength for (PVA-PEO- MnO_2) nanocomposites. There refractive index of (PVA-PEO) blend increases with increasing the manganese oxide nanoparticles which attributed to increase in the intensity of (PVA-PEO- MnO_2) nanocomposites [11]. In addition to increase the scattering of incident photon on surface of samples which causes to increase the reflectance this leads to increase in refractive index for (PVA-PEO- MnO_2) nanocomposites [14].

Figure 12 shows the variations of real part dielectric vs. wavelength for (PVA-PEO- MnO_2) nanocomposites. It can be indicated that real part

dielectric mainly proportional to the square of refractive index. So, it is increased with increasing MnO_2 NPs additive.

Figure 13 shows the variations of imaginary part dielectric vs. wavelength for (PVA-PEO- MnO_2) nanocomposites. Its proportional to the extinction coefficient as shown in equation (11) and its increased with the increasing of the manganese oxide nanoparticles [15].

Figure 14 shows the optical conductivity with wavelength for (PVA-PEO- MnO_2) nanocomposites. The optical conductivity is increased for (PVA-PEO- MnO_2) nanocomposites due to manganese oxide nanoparticles MnO_2 NPs are increased which leads to increase the absorption coefficient [5,46]. 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 [7].

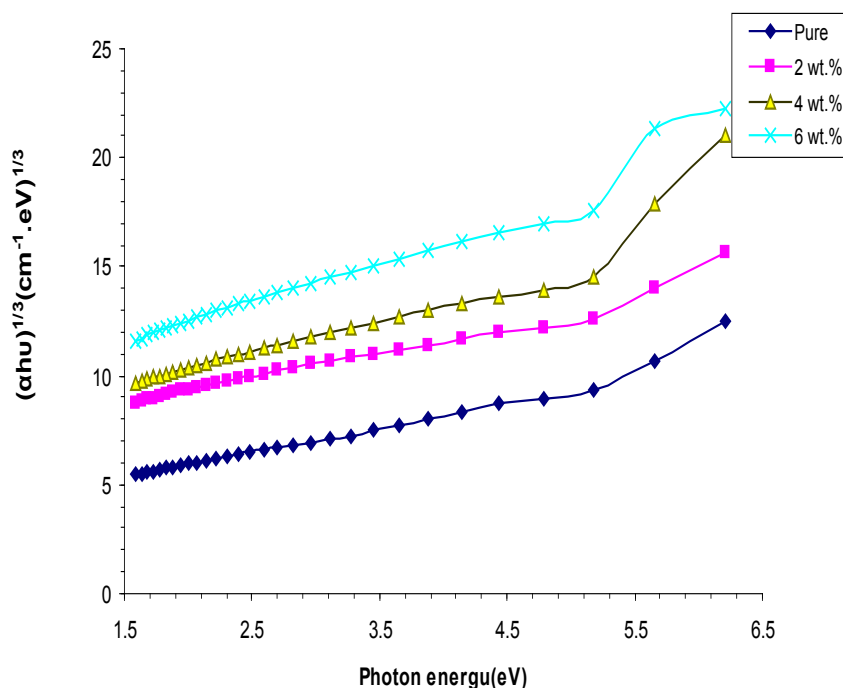


Fig. 9 . Variation of $(\alpha h\nu)^{1/3}$ for (PVA-PEO- MnO_2) nanocomposites with photon energy.

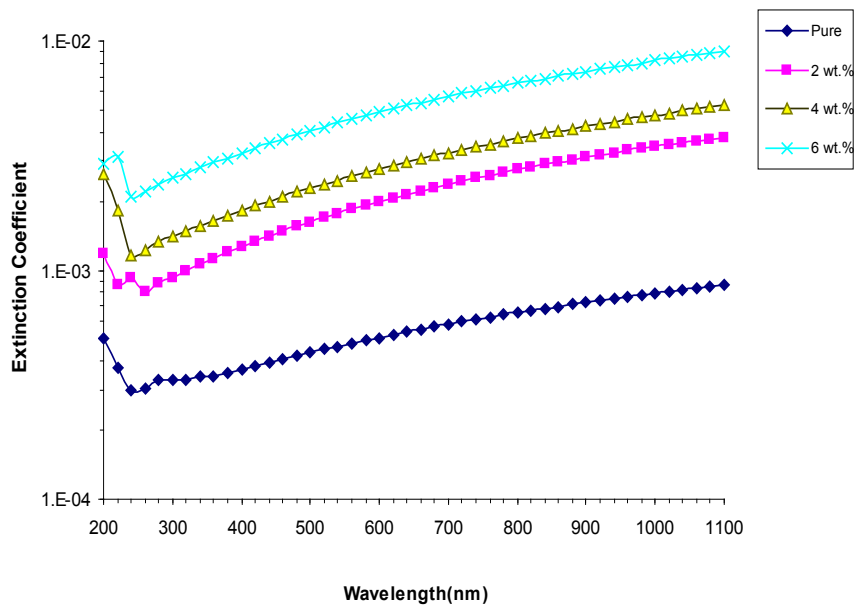


Fig. 10 . Variation of extinction coefficient for (PVA-PEO-MnO₂) nanocomposites with wavelength.

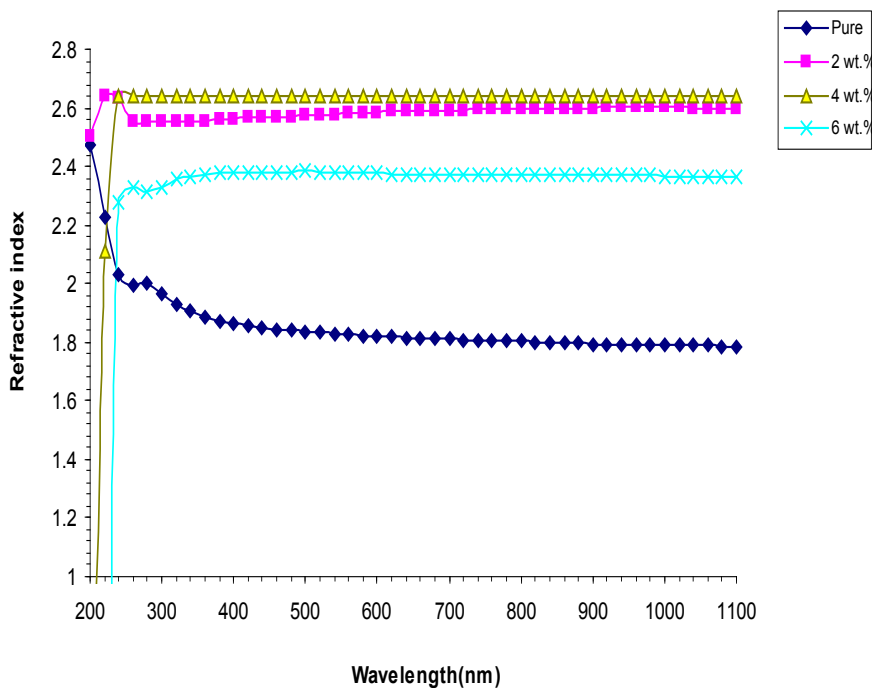


Fig. 11 . Variation of refractive index for (PVA-PEO-MnO₂) nanocomposites with wavelength.

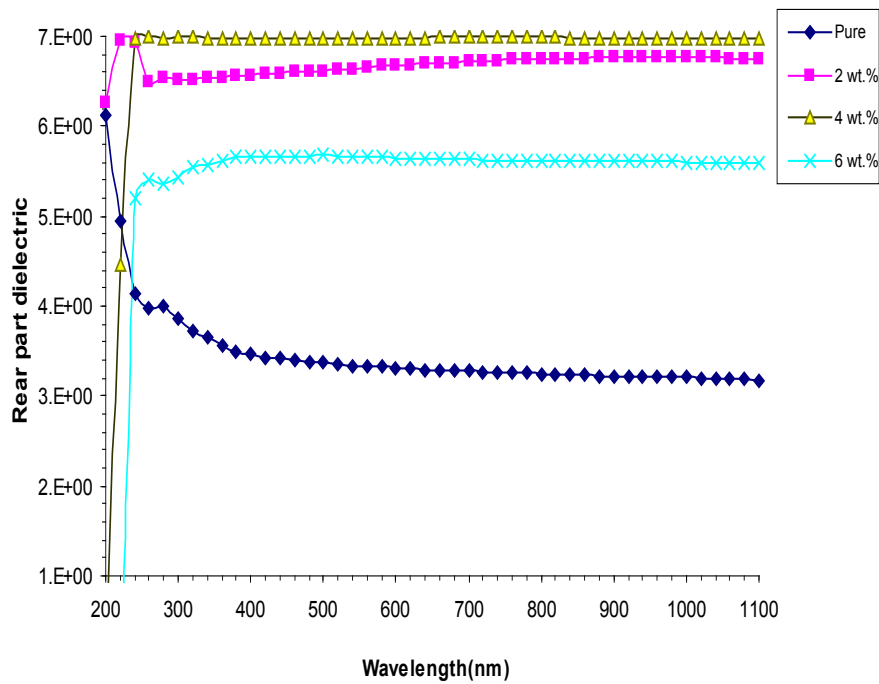


Fig. 12 . Variation of real part of dielectric constant for (PVA-PEO-MnO₂) nanocomposites with wavelength.

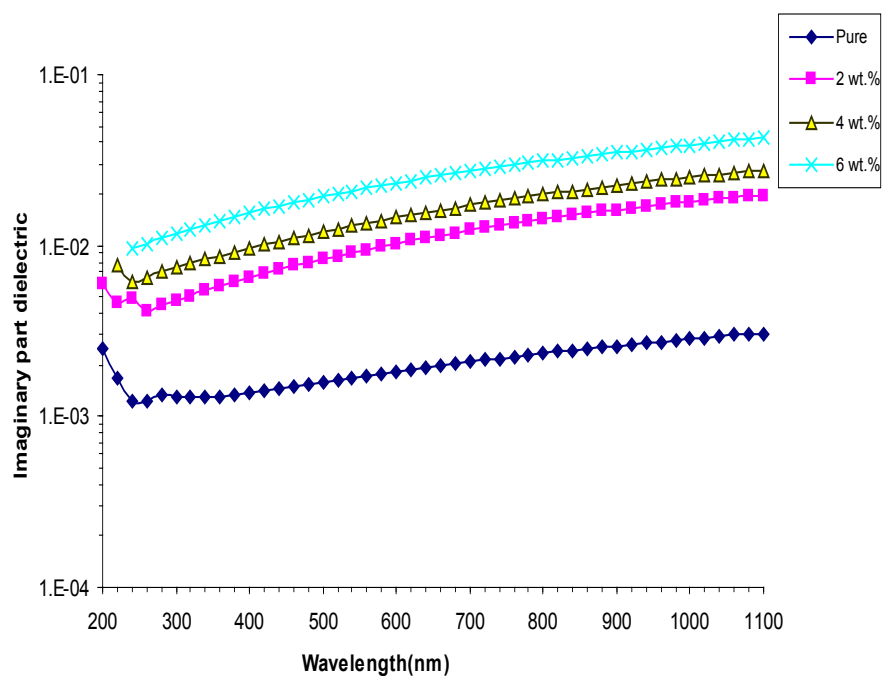


Fig. 13 . Variation of imaginary part of dielectric constant for (PVA-PEO-MnO₂) nanocomposites with wavelength.

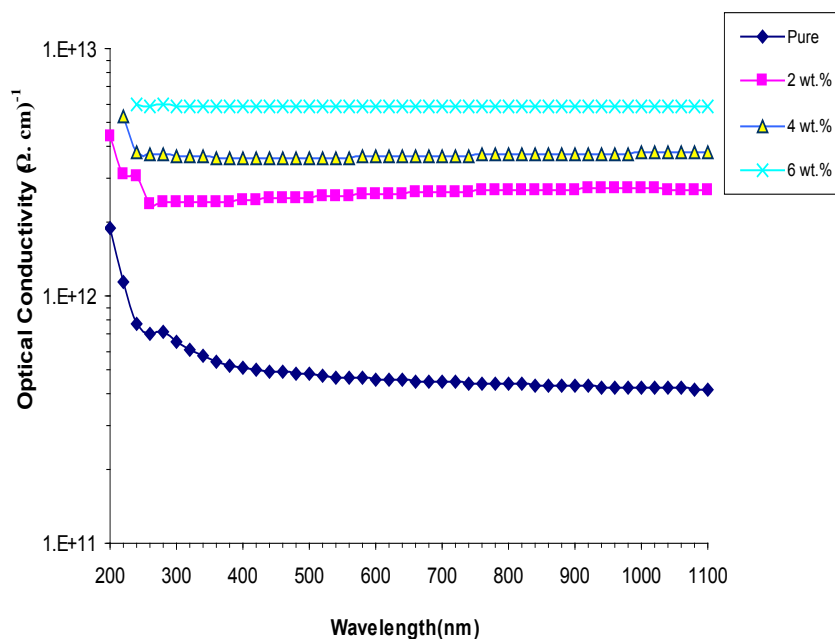


Fig. 14 . Variation of optical conductivity for (PVA-PEO-MnO₂) nanocomposites with wavelength.

Conclusions

- 1- The dielectric constant, dielectric loss and A.C electrical conductivity of (PVA-PEO) blend are increased with an increase the MnO₂ nanoparticles concentrations.
- 2- The dielectric constant and dielectric loss of (PVA-PEO- MnO₂) nanocomposites are decreased while the A.C electrical conductivity increases with increasing of the frequency. The experimental results of dielectric properties showed that the (PVA-PEO- MnO₂) nanocomposites enhancement of dielectric properties for (PVA-PEO) blend by addition the MnO₂ nanoparticles concentrations which is useful for different electronic applications.
- 3- Due to the absorption coefficient of less than (10⁴). The transition electronic of (PVA-PEO-MnO₂) nanocomposites was indirect.
- 4- With increasing weight percentage of manganese oxide nanoparticles for (PVA-PEO-MnO₂) nanocomposites, each of (extinction coefficient, refractive index, real and imaginary part) increases.
- 5- With higher photon energy, optical conductivity for (PVA-PEO-MnO₂) nanocomposites is increased and decrease with low photon energy.

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الخصائص التركيبية، والعزلية والبصرية للمترابكات النانوية (بولي فينيل الكحول- بولي إثيلين أوكسايد- أوكسيد المنغنيز)

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نظراً للأهمية العلمية والصناعية والكهربائية والطبية. كان من الضروري دراسة خصائص المترابكات النانوية. لذا فقد لفتت انتباه العلماء والباحثين في دراسة الخصائص التركيبية والكهربائية وغيرها من الخصائص. لذلك. في هذا العمل. تم تحضير مترابكات نانوية (بولي فينيل الكحول- بولي إثيلين أوكسايد- أوكسيد المنغنيز) بتراكيز مختلفة من الخليط البوليمري وجسيمات أوكسيد المنغنيز النانوية. تمت إضافة جسيمات أوكسيد المنغنيز النانوية إلى خليط (بولي فينيل الكحول- بولي إثيلين أوكسايد) بنسب وزنية (1. ٤. ٢). تشير صور المجهر الضوئي إلى أن توزيع جسيمات أوكسيد المنغنيز النانوية كان متجانساً في خليط (بولي فينيل الكحول- بولي إثيلين أوكسايد). تشير FT-IR إلى عدم حدث أي تغيير في التركيب الكيميائي بين خليط (بولي فينيل الكحول- بولي إثيلين أوكسايد) ومضافات جسيمات أوكسيد المنغنيز النانوية. تمت دراسة الخصائص العزلية للمترابكات النانوية. بينت النتائج أن ثابت العزل الكهربائي والفقداً العزلي والتوصيلية الكهربائية للخليط (بولي فينيل الكحول- بولي إثيلين أوكسايد) يزداد مع زيادة تراكيز جسيمات أوكسيد المنغنيز النانوية. ينخفض ثابت العزل الكهربائي وفقداً العزل الكهربائي بينما تزداد التوصيلية الكهربائية AC مع زيادة تردد المجال الكهربائي المسلط. أظهرت القياسات البصرية زيادة الامتصاصية لمترابكات (PVA-PEO-MnO₂) النانوية مع زيادة تركيز جسيمات أوكسيد المنغنيز النانوية. مع زيادة تركيز جسيمات أوكسيد المنغنيز النانوية فإن فجوة الطاقة غيرالمباشرة (Eg) لخليط (بولي فينيل الكحول- بولي إثيلين أوكسايد) ستقل. وبتزايد النسب الوزنية لجسيمات أوكسيد المنغنيز النانوية. فإن الثوابت البصرية كمعامل الامتصاص. و معامل الخمود. و معامل الانكسار. و ثابت العزل الحقيقي والخيالي للمترابكات النانوية ستتغير.