Synthesis of Novel (Polymer Blend-Titanium Carbide) Nanocomposites and Studying their Characterizations for Piezoelectric Applications

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Abstract

Piezoelectric nanocomposites are very important for many applications as a pressure sensors. Fabrication of (polyvinyl alcohol - polyvinyl pyrrolidinone -titanium carbide) nanocompos- ites and study their structural, electrical, dielectric and optical properties have been in- vestigated. The effect of adding the TiC nanoparticles on structural, electrical, dielectric and optical properties of polymeric blend has been studied. The results showed that the electrical conductivity of (PVA-PVP-TiC) nanocomposites is increasing with the increase of TiC nanoparticles concentrations at room temperature. The FTIR analysis showed there is no interactions between (PVA- PVP) polymer blend and TiC nanoparticles. The dielectric studies showed the dielectric constant and dielectric loss of nanocomposites increase with the increase of TiC nanoparticles concentrations and they decrease as frequency increased. The A.C electrical conductivity increases with the increase of TiC nanoparticles concentra- tions and frequency. The results of optical properties showed that the optical absorbance of (PVA- PVP) polymer blend increases with the increase of TiC nanoparticles concentrations. The optical constants change with increase in TiC nanoparticles concentrations. The piezo- electric application results of (PVA-PVP-TiC) nanocomposites showed that the electrical resistance of (PVA-PVP-TiC) nanocomposites decreases with an increase of the pressure which make it is suitable for piezoelectric applications or pressure sensors.

Keywords: piezoelectric, titanium carbide, dielectric properties, nanocomposites, pres- sure

1. Introduction

Piezoelectricity refers to the change of the electric polarization with stress; this vary results in a alter in voltage across the material in the direction of the electric polarization [1]. Piezoelectric transducers are used for determining mechanical properties (acceleration, weight, effort, pressure, deformations, moments, angular speed, etc.), thermal sizes (expense, temperature, electric parameters, vacuum, etc.) and for gas concentration, humidity, structure control, and micro weights. For accuracy, these devices in many applications sur- pass transducers based on other physical resolution principles. Piezoelectric sensors can be divided into two large groups depending on their fundamental physical effects: 1. Sensors in the first group use a straight-line piezoeffect. They are used for determining dynamic and quasistatic pressure, linear and vibrating accelerations, as well as parameters of sound and ultrasonic fields, etc. 2. A second but no less general class of sensors concerns the so-called resonant piezotransducers, which use the return piezoeffect. They

are resonant sensors from piezoelectric resonators, and they can also make straight-line piezoeffects. (These are resonant piezoelectric transformer sensors.) In addition, other physical effects can be used, e.g., thermosensitivity, acoustosensitivity, tensosensitivity, etc [2]. Piezoelectric composites have a several range of applications in actuators, energy harvesting, and sensors. There are many kinds of piezoelectric materials as quartz, $Pb(Zr, Ti)O_3$ and TiO_2 oxide. They concluded that electricity was produced if a mechanical stress was applied to a crystal material and they found the electricity was proportional with the mechanical stress. Composite mate- rials show good piezoelectric properties can be used in medical synthesis because of some composites materials are flexible. Polymers have flexible and light property like poly vinyl carbonate, polyvinylidene fluoride and Nylon 11. Some polymers possess specific a crystalline region or called semicrystalline polymers as PVDF film and PVA can be considered good piezoelectric polymers due to that have an internal dipole moment [3]. Composite materials, developed in the automotive, aerospace, railway, aeronautic and nautical fields for high velocity vehicles are necessary to be premeditated to be the best with deference to the technical concert and weight decrease, together with the made to order price for the targeted market [4]. Composites are usually used in our day to day living. Relate to their ability and low weight to be customized for specific end use they have gained a significant ground in the high concert applications, such as automobile industry and aerospace. The idea of linking two or more different constituents into on material gives almost unlimited possibilities to generate new engineering materials propertied by variety of different proper- ties. Composites because of these varied properties are successfully used in approximately all areas of science and industry [5] [6] [7] [8] . Single advantage of nanoparticles, as polymer fillers appear to have is that compared to conventional fillers, loading necessities are rather low. Micro-sized particles used as reinforcing agents dispersion light, thus decreasing optical clarity and light transmittance. Efficient nanoparticle scattering combined with excellent polymer material particle interfacial adhesion eliminates dispersion and allows the exciting possibility of rising strong yet transparent films, coatings and membranes [9]. Optical characteristics of polymers comprise an important aspects in the study of electronic conversion and the possibility of their application as a cover in solar collection, optical filters, green house and selection surfaces [10]. The present work deals with preparation of (polyvinyl alcohol - polyvinyl pyrrolidinone - titanium carbide) nanocomposites and study their structural, electrical, dielectric and optical properties for piezoelectric and pressure sensors applications.

2. Theoretical part

The volumetric electrical conductivity (σ_V) of (polyvinyl alcohol - polyvinyl pyrrolidi- none -titanium carbide)nanocomposites can be calculated for a regular body with a section has along the length (L), a constant area (N) and electrical resistance (R) by using the relation [11]

$$\sigma_V = \frac{L}{RN} \qquad \dots\dots\dots\dots\dots\dots(1)$$

The dielectric constant ε of (polyvinyl alcohol-polyvinyl pyrrolidinone-titanium carbide) nanocomposites was calculated by using the equation [12] :

$$\varepsilon = \frac{C_p}{C_\circ} \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(2)$$

Where C_p is parallel capacitance and C_o is vacuum capacitor which be calculated by [13]:

$$C_\circ = \varepsilon_o M/t \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(3)$$

Where is ε_o vacuum permittivity. M: is the area of capacitance plate. t: is the distance between two plates. Dielectric loss ε'' can written by following equation [6] :

$$\varepsilon'' = \varepsilon\, D \qquad \ldots\ldots\ldots\ldots\ldots\ldots..(4)$$

D: is dispersion factor. The alternating conductivity of (PVA-PVP-TiC) nanocomposites is given by [8] :

$$\sigma_{A.C} = w\, \varepsilon'' \varepsilon_\circ \qquad \ldots\ldots\ldots\ldots\ldots..(5)$$

Where w is the angular frequency.

The absorption coefficient (α) can be calculated by following equation [14]:

$$\alpha = 2.303\ (A/t) \qquad \ldots\ldots\ldots\ldots\ldots.\ldots\ldots(6)$$

Where A: is the absorbance. For amorphous semiconductors, the transitions model is [14]:

$$\alpha h\upsilon = B(h\upsilon - E_g)^m \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(7)$$

Where B is a constant, $h\upsilon$ is the photon energy, h is Planck's constant, E_g is the optical energy band gap, m = 2 for allowed indirect transition and m = 3 for forbidden indirect transition.

The refractive index (n) of (PVA-PVP-TiC) nanocomposites calculated by using follow- ing equation [15] :

$$n = (1+R^{1/2})/ (1-R^{1/2}) \qquad \ldots\ldots\ldots (8)$$

The extinction coefficient (k) is given by using the equation [15]:

$$K = \alpha\lambda/4\pi \qquad \ldots\ldots\ldots\ldots (9)$$

The real and imaginary parts of dielectric constant (real part ε_1 and imaginary part ε_2) for (PVA-PVP-TiC) nanocomposites can be calculated by using equations [16] :

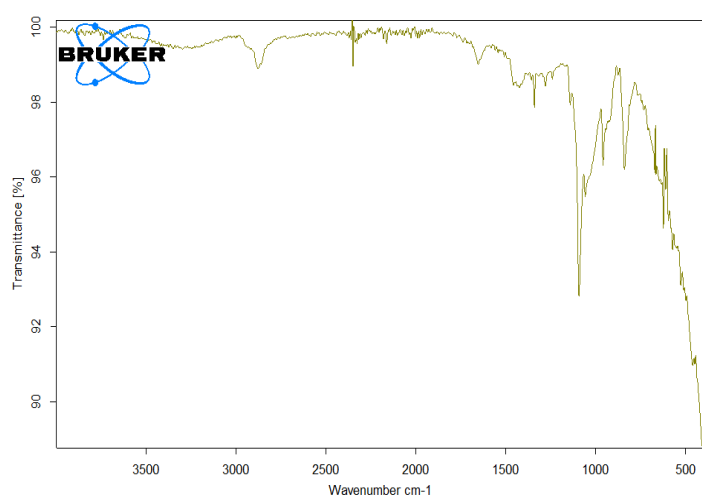$$\varepsilon_1 = n^2 - k^2 \qquad \text{........ (10)}$$

$$\varepsilon_2 = 2nk \qquad \text{...... (11)}$$
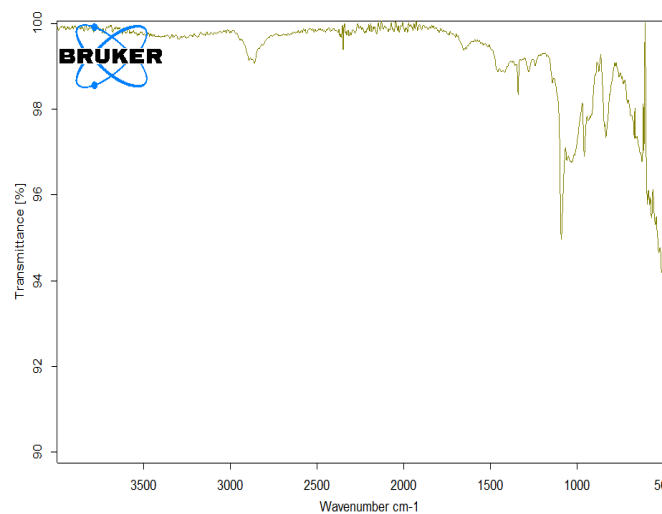
3. Experimental Part:

The (polyvinyl alcohol-polyvinyl pyrrolidinone-titanium carbide) nanocomposites films were prepared with different concentrations of polymer blend and titanium carbide nanoparticles by using casting method. The solutions of nanocomposites are prepared by dissolving 1 gm of polymers (86 wt.% PVA, 14wt.% PVP) in 30 ml of distilled water by using magnetic stirrer to mix the polymers to obtain more homogeneous solution. The TiC nanoparticles was added to (PVA-PVP) polymeric blend with different weight percentages are (0,3,6 and 9) wt.%. FTIR spectra (Fourier transform infrared spectrometer) were examined in wavenumber range $(500 - 4000) cm^{-1}$ by FTIR (Bruker company, German origin, type vertex -70). The D.C electrical properties of (PVA-PVP-TiC) nanocomposites were measured by determining the D.C electrical resistance at 25^0C by using the Keithley electrometer type 2400 source mater. The dielectric properties (dielectric constant; dielectric loss and A.C. electrical conductivity) of (polyvinyl alcohol-polyvinyl pyrrolidinone -titanium carbide) nanocomposites were tested for different frequency range $(10^2-5 \ 10^6)$ Hz by using LCR meter type (HIOKI 3532-50 LCR HI TESTER). The optical properties of (PVA-PVP-TiC) nanocomposites were measured by using UV/1800/ Shimadzu in range of wavelength (220-800) nm. The piezoelectric application of (polyvinyl alcohol-polyvinyl pyrrolidinonetitanium carbide) nanocomposites has tested by measuring the electrical resistance between two electrodes on the top and bottom of the nanocomposites films for different pressure range (60-200)bar.
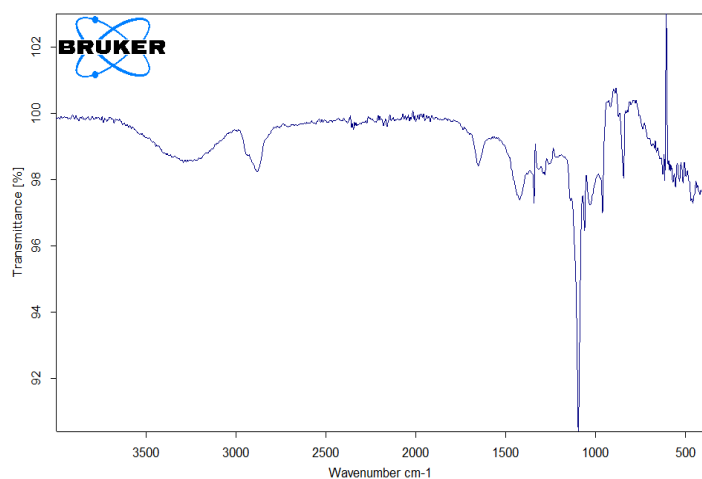
4. Results

Fig. 1: shows the FTIR analysis of (PVA-PVP-TiC) nanocomposites. As shown in figure, the broad bands at around $3260 \ cm^{-1}$ are observed due to -OH groups in the PVA chain. The bands at around $1200 \ cm^{-1}$ were attributed to the ether bonds (C-O-C). The peaks at around $1650 \ cm^{-1}$ suggests the presence of such free C=O groups. C−N group of PVP due to the overlapping by C=O group, could not visibility. The peaks at around $1400 \ cm^{-1}$ were assigned to the C-O groups of PVA based films. The band at around $2800 \ cm^{-1}$ was attributed to the C-H groups [17]
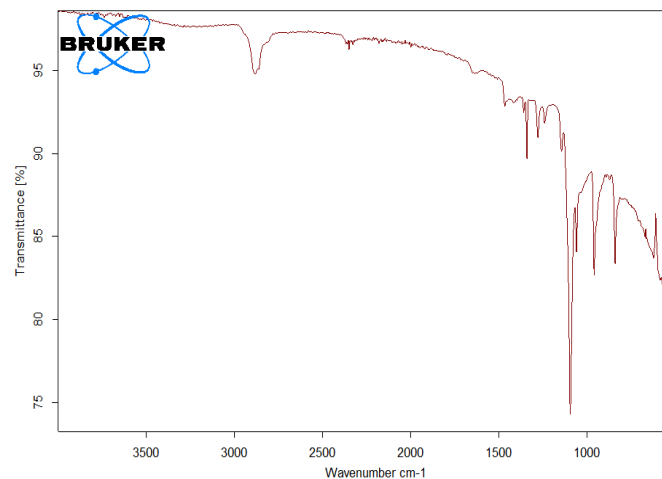
Fig. 1: FTIR spectra for (PVA-PVP-TiC) nanocomposites
A- Pure (PVA-PVP)   B- 3 wt.% TiC nanoparticles

C- 6 wt.% TiC nanoparticles   D- 9 wt.% TiC nanoparticles

Fig.2: shows the D.C electrical conductivity of (PVA-PVP) blend with different weight percentages of titanium carbide nanoparticles at room temperature. From the figure the conductivity of (PVA-PVP) blend increases with the increase in titanium carbide nanopar- ticles concentrations. This behavior may be attributed to the some different mechanism or effect that have a major position in the conduction process. At low TiC weight percentages, the nano-particles of titanium carbide aggregate diameter is small, and therefore the Coulomb blockade influence is not obvious. At high TiC weight percentages, a large num- ber of titanium carbide nanoparticles aggregated to form larger sized particles and reach diameters which are large sufficient to display the Coulomb blockade influence apparently. The Coulomb blockade influence will thus result in a minimum in electrical conductivity of TiC/PVA-PVP nanocomposites. At higher TiC weight percentages, the TiC nanoparti- cles aggregates diameter and TiC nanoparticles concentration increase to the degree that nanoparticles become too big and show the way to the increase in electrical conductivity. The increase of TiC weight percentages and the associated increase of nanoparticles aggregates diameter to a large size which may help to create new conductive paths in nanocomposite leading to the increase of electrical conductivity [18] .
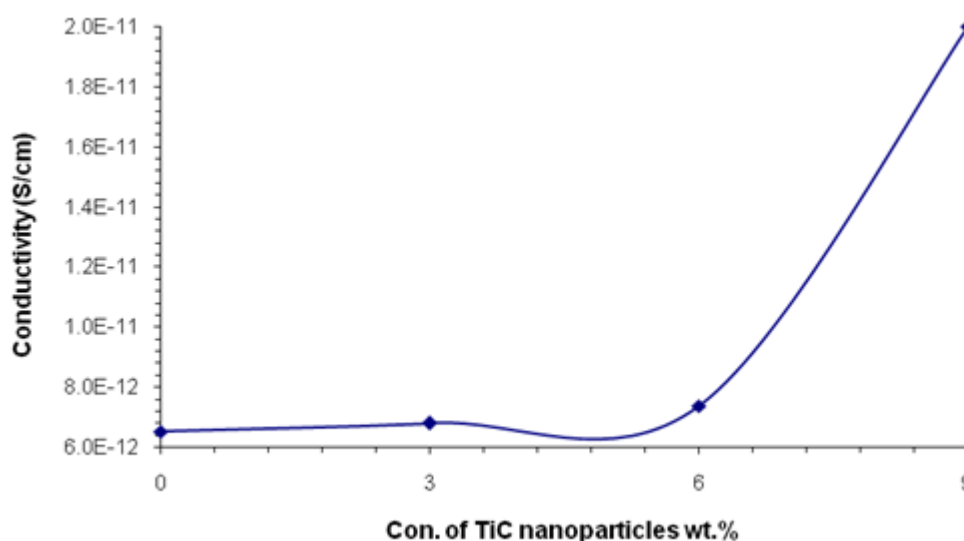

Fig.2 the D.C electrical conductivity of (PVA-PVP) blend with different weight percentages of titanium carbide nanoparticles at room temperature.

Fig.3 and Fig.4 show the dielectric constant and dielectric loss of nanocomposites films as a function of frequency at different TiC nanoparticles concentrations. The figures show that the dielectric constant (ε') values of nanocomposites films decrease with increase in 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 (ε'') 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 [Chandrakala*et al.*, 2014]. Also, the values of dielectric constant and dielectric loss of (PVA- PVP-TiC) nanocomposites increase with the increase of the titanium carbide nanoparticles concentrations. Enhanced values of dielectric constant and dielectric loss, mainly at low frequencies, can be attributed to increased conductivity, and/or interfacial polarization, and/or electrode polarization. Electrode polarization is related to the build up of space charges at the sample-electrode interfaces and is propertied by very high values of both real and imaginary parts of dielectric constant. Interfacial polarization results from the accumulation of unbounded charges, at the interfaces of the constituents, where they form large dipoles [19].
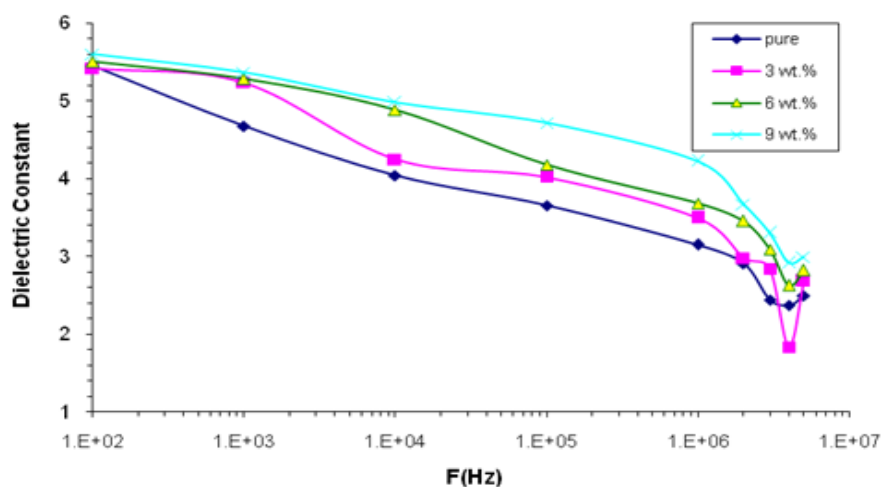

Fig.3: the dielectric constant of nanocomposites films as a function of frequency at different TiC nanoparticles concentrations.
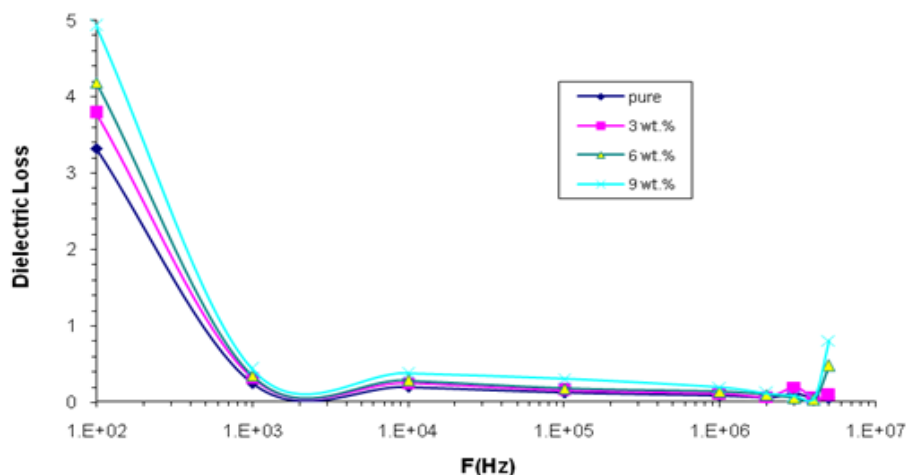

Fig.4: the dielectric loss of nanocomposites films as a function of frequency at different TiC nanoparticles concentrations.

Fig.5   shows   the   variation   of   A.C   electrical   conductivity   of

nanocomposites films with frequency. From the figure, The electrical conductivity spectrum consists of two different behaviors: the low frequency dispersion region which followed by the frequency independent plateau region. Low frequency dispersion was relate to space charge polarization effects. At low frequency region, more charge accumulation occurred at the electrode of sample leading to a decrease the number of mobile ions and electrical conductivity [20].
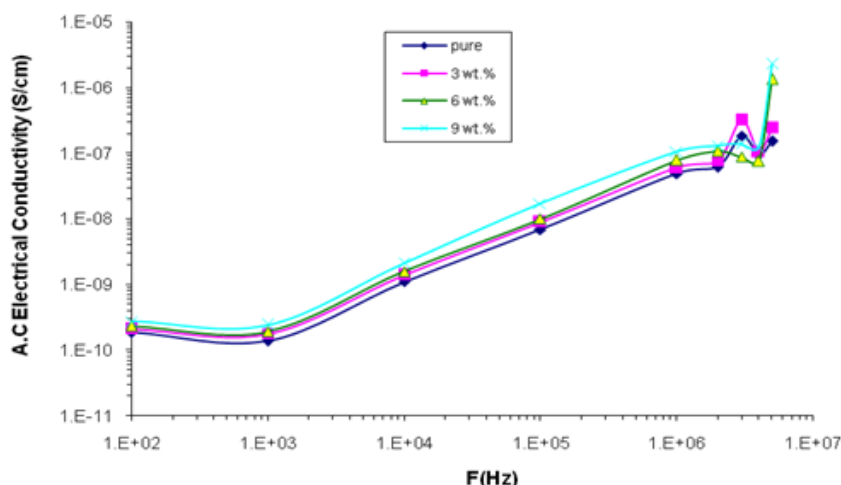

Fig.5: the variation of A.C electrical conductivity of nanocomposites films with frequency.

The charge carriers mobility was higher in the high-frequency; hence the electrical con- ductivity increases with frequency. As shown in figure, the A.C conductivity of (PVA-PVP) blend increases with increase in TiC concentrations which is attributed to the enhance- ment of the charge ions mobility and the larger number of charge carriers in polymer bend system [20].

The absorbance spectra of pure and TiC nanoparticles dispersed PVA-PVP blend are presented as given in Fig.6. It is well known that PVA-PVP blend absorbs the radiation strongly in the wavelength region 200–400 nm. Fig.6 signifies the shift in both absorption bands and the band edges towards the higher wavelengths with different absorption inten- sities for different concentrations of doped PVA-PVP blend samples. This can be more enlightened by using Beer's law which states that the absorption of radiation is directly proportional to the number of absorbing molecules in the sample. The shift witnessed in the absorption edge of the doped PVA-PVP blend composites are essentially due to the variation in crystalline parameters which in turn changes the energy band gap [21] [22] [23] . The values of energy band gap for indirect transition calculated for different titanium carbide nanoparticles concentrations of the nanocomposites samples are presented in Fig.7.
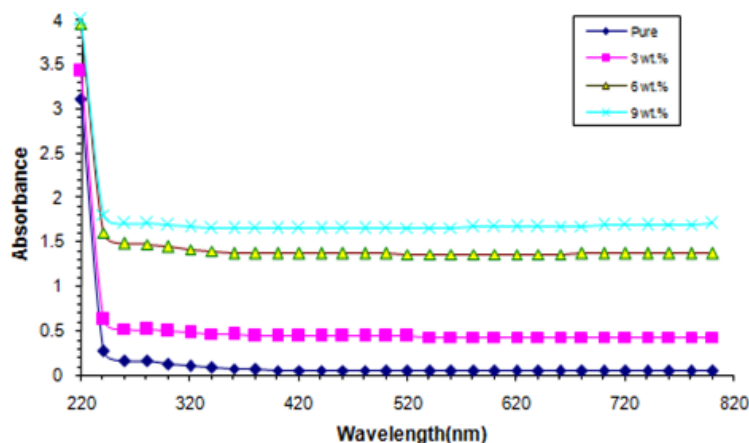
Fig.6: the absorbance spectra of PVA-PVP-TiC nanocomposites

It is very clear from Fig.7 that as the TiC concentration increases the band gap value decreases. The decrease in the optical energy gap (E_g) values are due to the presence of local cross-linking that occurred within the noncrystalline phase of the composite polymer. This increase in the local cross-linking further increases the degree of ordering in PVA-PVP blend
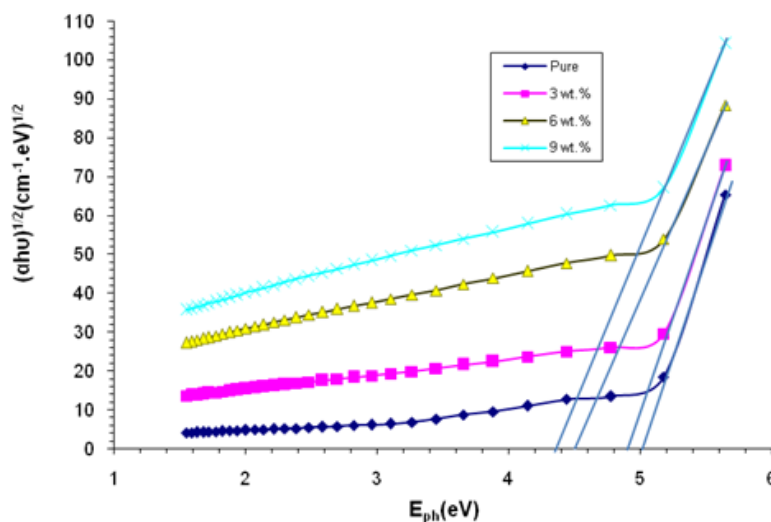

Fig.7: the values of energy band gap for allowed indirect transition calculated for different

titanium carbide nanoparticles concentrations of the nanocomposites

Fig.8: shows the refractive index distributions of the (PVA-PVP-TiC) nanocomposites films. The refractive index of PVA-PVP blend increases with increasing of TiC concentra- tions. Also, the refractive index increases with increasing of the photon energy. It has been concluded that when the incident light interacts with a material has a large amount of par- ticles, the refraction will be high and hence the refractivity of the films will be increased [24].
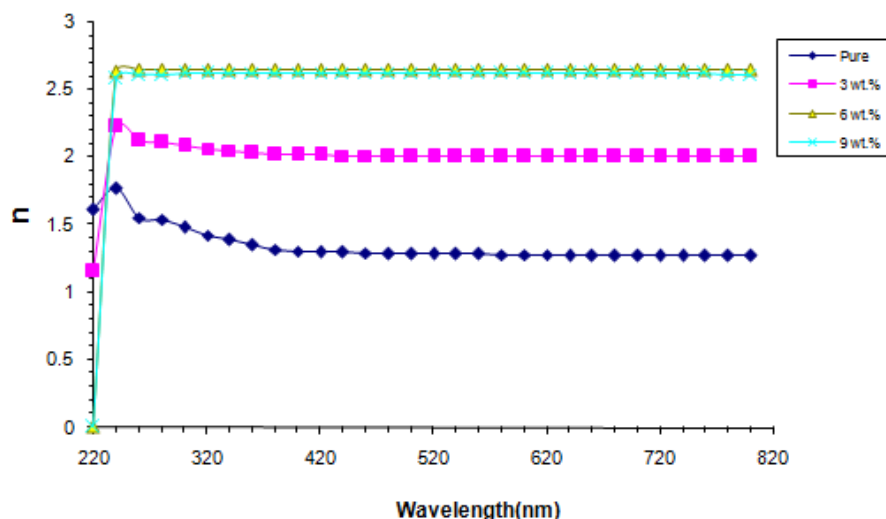
Fig. 8: the refractive index of (PVA-PVP-TiC) nanocomposites with wavelength

The variation of extinction coefficient (k) with wavelength for (PVA-PVP-TiC) nanocom- posites is as shown in Fig.9. The extinction coefficient increases with increasing of TiC NPs concentration. This behaviour of extinction coefficient can be ascribed to high absorption coefficient. The extinction coefficient is high at the longest wavelengths and high concentration. The variation of real and imaginary parts of dielectric constants (PVA-PVP-TiC) nanocomposites with wavelengths are shown in Figs. (10 and 11). As shown in figures, the real and imaginary parts are changed with TiC NPs concentration and wavelength. It is concluded that the variation of ε_1 mainly depends on (n^2) because of small values of (k^2), while ε_2 mainly depends on the (k) values which are related to the variation of absorptioncoefficients. The values of the real dielectric constant (PVA-PVP-TiC) nanocomposites are high with respect to the imaginary dielectric constant [25] [26] .
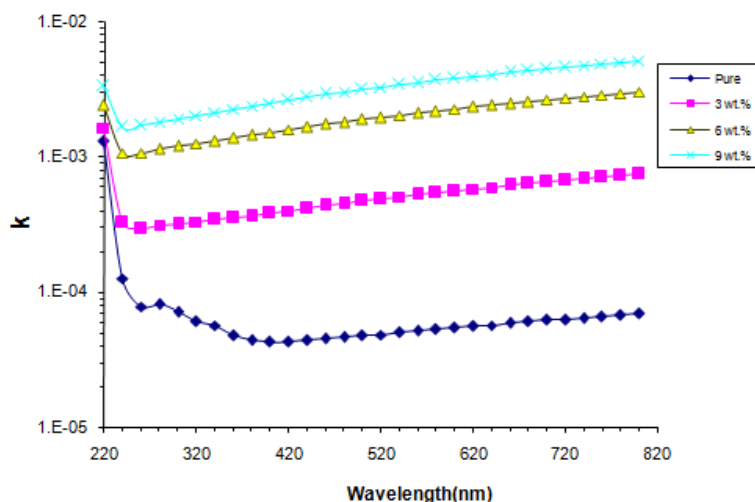
Fig.9: the variation of extinction coefficient with wavelength for (PVA-PVP-TiC) nanocomposites
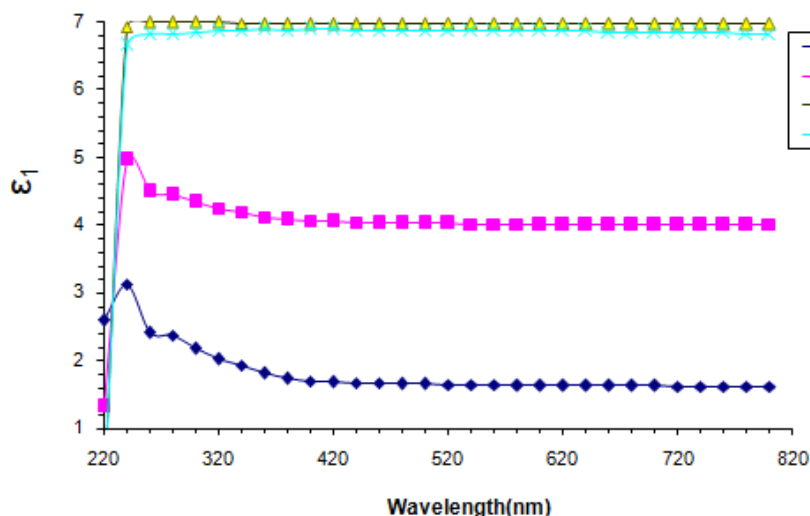

Fig.10: the variation of real part of dielectric constants (PVA-PVP-TiC) nanocomposites with wavelength
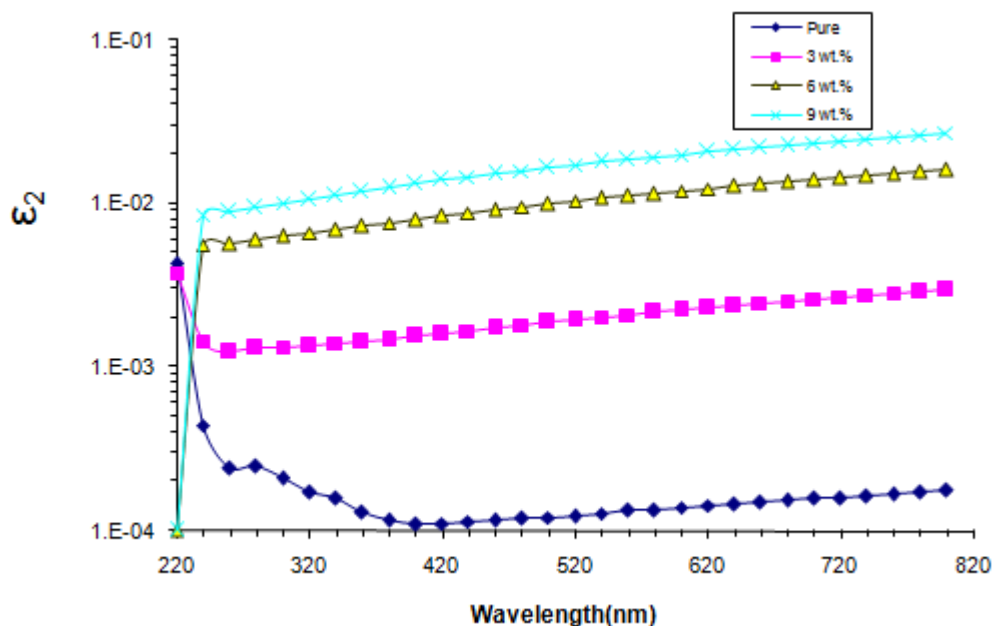

Fig. 11: the variation of imaginary part of dielectric constants (PVA-PVP-TiC) nanocomposites with wavelength

Fig.12: shows the variation of electrical resistance for (PVA-PVP-TiC) nanocomposites with pressure. As shown in figure, the electrical resistance decreases with an increase of the pressure, this behavior can be explained: The piezoelectric property is dictated by the manner in which the atoms of the crystal align themselves. The crystal consists of multiple interlocking domains which have positive and negative charges. These domains are symmetrical within the crystal, with the result that the crystal has a net charge of zero. When a stress is applied to the crystal this symmetry is

broken, and in order to restore the symmetry these domains realign themselves, and through the realignment, generate a current and the resistance will be decreased. When such a crystal is deformed, the positive charge centres and the negative charge centres are displaced with respect to one another. Overall the crystal remains electrically neutral, but the difference in the charge centres results in an electric polarization inside the crystal. The converse piezoelectric effect works on the opposite principal. The crystal atoms are aligned and have a zero charge. When a voltage is applied over the crystal, the symmetry is broken because of the electricity that flows through the crystal. In order to restore the zero charge, the crystal deforms. Piezoelectricity is used in sensors, actuators, clocks, lighters and transducers [27].
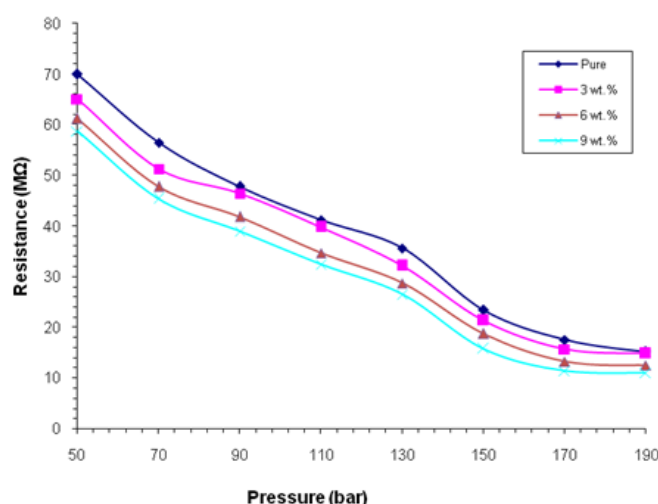

Fig. 12: the variation of electrical resistance for (PVA-PVP-TiC) nanocomposites with pressure.

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الخلاصة

ان المواد المتراكبة الكهروضغطية مهمة جدا" لتطبيقات عديدة منها: متحسسات الضغط. لذلك تم تصنيع المتراكبات النانوية (بولي فينيل الكحول- بولي فينيل بيروليدون- كاربيد التيتانيوم) ودراسة خواصها التركيبة، الكهربائية، العزلية والبصرية. درس تاثير اضافة جسيمات TiC النانوية على الخواص التركيبية، الكهربائية، العزلية والبصرية للخليط البوليمري. بينت النتائج ان التوصيلية الكهربائية للمتراكبات النانوية (PVA–PVP–TiC) تزداد مع زيادة تركيز جسيمات TiC النانوية عند درجة حرارة الغرفة. اختبار FTIR اوضح عدم وجود تفاعلات بين الخليط البوليمري (PVA–PVP) و جسيمات TiC النانوية. بينت الدراسات العزلية ان ثابت العزل والفقدان العزلي للمتراكبات النانوية يزدادان مع زيادة تراكيز جسيمات TiC النانوية ويقلان مع زيادة التردد. وان التوصيلية الكهربائية المتناوبة تزداد مع زيادة تراكيز جسيمات TiC النانوية والتردد. اوضحت نتائج الخواص البصرية ان الامتصاصية البصرية للخليط البوليمري (PVA–PVP) تزداد مع زيادة تركيز جسيمات TiC النانوية. وان الثوابت البصرية تغيرت مع زيادة تركيز جسيمات TiC النانوية. بينت نتائج تطبيق الكهروضغطية للمتراكبات النانوية (PVA–PVP–TiC) ان المقاومة الكهربائية للمتراكبات النانوية (PVA–PVP–TiC) تقل مع زيادة الضغط مما يجعلها مناسبة لتطبيقات الكهروضغطية او متحسسات الضغط.

الكلمات المفتاحية: كهروضغطية، كاربيد التيتانيوم، الخواص العزلية، المتراكبات النانوية، الضغط.