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NOVEL PRESSURE SENSORS MADE FROM NANOCOMPOSITES (BIODEGRADABLE POLYMERS–METAL OXIDE NANOPARTICLES): FABRICATION AND CHARACTERIZATION

This paper aims to the preparation of novel pressure-sensitive nanocomposites with low cost, light weight, and good sensitivity. The nanocomposites of polyvinyl alcohol, polyacrylic acid, and lead oxide nanoparticles have been investigated. The dielectric properties and dc electrical conductivity of (PVA–PAA–PbO_2) nanocomposites have been studied. The dielectric properties of nanocomposites were measured in the frequency range (100 Hz–5 MHz). The experimental results showed that the dielectric constant and dielectric loss of (PVA–PAA–PbO_2) nanocomposites decrease, as the frequency increases, and they increase with the concentrations of PbO_2 nanoparticles. The ac electrical conductivity of (PVA–PAA–PbO_2) nanocomposites increases with the frequency and the concentrations of PbO_2 nanoparticles. The dc electrical conductivity of (PVA–PAA–PbO_2) nanocomposites also increases with the concentrations of PbO_2 nanoparticles. The application of pressure-sensitive nanocomposites has been examined in the pressure interval (60–200) bar. The results showed that the electrical resistance of (PVA–PAA–PbO_2) pressure-sensitive nanocomposites decreases, as the compressive stress increases. The (PVA–PAA–PbO_2) nanocomposites have high sensitivity to pressure.

K e y w o r d s: sensitivity, piezonanocomposites, resistance, pressure, dielectric properties.

1. Introduction

Flexible pressure-sensitive sensors reveal the advantage over traditional pressure-sensitive sensors and some unique characteristics. They can be used to perceive external forces supplied onto the surface, even bended or folded, and to detect the pressure of any two flexible or soft/rigid contact surfaces. Recently, many flexible pressure-sensitive sensors have been reported. The sensing mechanisms include the capacitive sensing, field-effect transistors, piezoelectric-sensing, and piezoresistive-sensing. Pressure-sensitive materials based on piezoresistive-sensing mechanisms are widely used owing to their attractive advantages,

including feasible preparation, low cost, and easy signal collection. Therefore, they have potentiality to be used to develop flexible pressure-sensitive sensors [1]. The use of polymer piezoelectric materials is certainly desirable, because they have special advantages over ceramics. They are flexible, mechanically more stable, and can be obtained in the form of large-area thin films. Other advantages of polymers are that they can be easily manufactured at much lower temperatures and can be formed more easily with different shapes. Polymers as sensors and actuators offer the advantage of processing flexibility, because they are tough, lightweight, readily manufactured into large areas, and can be cut and formed into complex shapes. Other notable features of poly-

mers are high strength, high impact resistance, low dielectric constant, low elastic stiffness, and low density, which result in a high voltage sensitivity (excellent sensor characteristic), and low acoustic and mechanical impedance (crucial for medical and underwater applications). The purpose of the mixing of piezoelectric ceramics with polymers is to combine the advantages of both materials, which include the mechanical flexibility of polymers and the higher coupling factor [2]. There is the increasing research interest in polymeric nanocomposites owing to improvements in electrical, thermal, optical, and mechanical properties and to their great potential for highly functional materials [3]. Two types of inorganic particles can be used as fillers in polymers for preparing composite materials, i.e., porous and nonporous. Inorganic nonporous nanoparticles have also been introduced into the polymer matrix to prepare a polymer–inorganic nanocomposite [4]. The nanocomposites applications are promising in the industrial and medical approaches of sensors, medical devices, drug delivery, coatings, adhesives, optical integrated circuits, automobiles, microelectronic packaging, injection molded products, packaging materials, aerospace *etc.* [6–9]. Polymers have several advantages such as the low cost, easy processing, high strength, flexibility, and good mechanical properties [10–12]. Nanocomposites on the basis of semiconductor nanoparticles and a polymer matrix are prospective materials for the application in optoelectronics and sensor electronics, for the creation of luminescent materials, *etc.* [13].

2. Materials and Methods

Nanocomposites films of polyvinyl alcohol (PVA), polyacrylic acid (PAA)–lead oxide (PbO_2) nanoparticles have been prepared with different concentrations of the polymer blend and metal oxide nanoparticles. The nanocomposites were fabricated with the following weight percentages of polymers: PVA (83 wt.%), PAA (17 wt.%). PbO_2 nanoparticles were added to the (PVA–PAA) blend with different concentrations (0, 2.5, 5, and 7.5) wt.%. The films of nanocomposites were prepared, by using the casting technique The dc electrical properties of (PVA–PAA–PbO_2) nanocomposites are measured by measuring the dc electrical resistance at room temperature, by using the Keithley electrometer type 2400 source meter. The dielectric properties of (PVA–PAA–PbO_2)
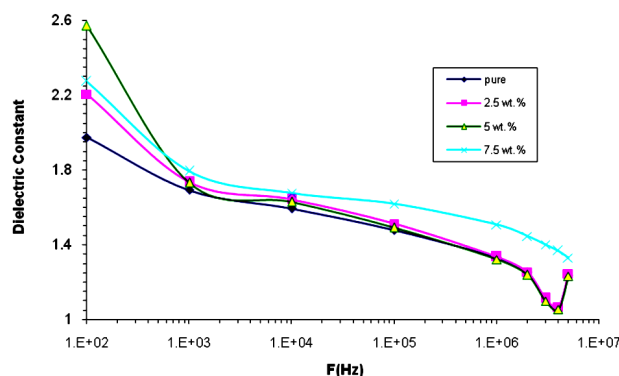
Fig. 1. Variation of the dielectric constant for (PVA–PAA–PbO_2) nanocomposites with the frequency

nanocomposites were examined in the frequency range (from 100 Hz to 5×10^6 Hz), by using an LCR meter HIOKI 3532-50 LCR HI TESTER. The pressure sensor application of (PVA–PAA–PbO_2) nanocomposites was investigated by measuring the resistance between two electrodes on the top and bottom of the samples of (PVA–PAA–PbO_2) nanocomposites for different pressures (80–200) bar. The dc electrical specific conductivity (σ) of (PVA–PAA–PbO_2) nanocomposites in a sample is determined by the formula [14]:

$$\sigma = \frac{d}{RA}, \tag{1}$$

where d is the distance between two plates, A is the area of the sample, and R is the electrical resistance.

3. Results and Discussion

Figure 1: shows a variation of the dielectric constant for (PVA–PAA–PbO_2) nanocomposites with the frequency at room temperature. As shown in the figure, it is clear that the values of the dielectric constant are very high at low frequencies. Such high values of the dielectric constant at low frequencies has been explained by the presence of space charge effects, which is contributed by the accumulation of charge carriers near the electrodes. At higher frequencies, the dielectric constant has been found to be relatively constant with a change in the frequency. This is because the periodical reversal of the field takes place so rapidly that the charge carriers will hardly be able to orient themselves in the field direction, which results in a decrease in the dielectric constant [15]. In addition, the dielectric constant decreases, as the frequency increases, for all concentrations may be due
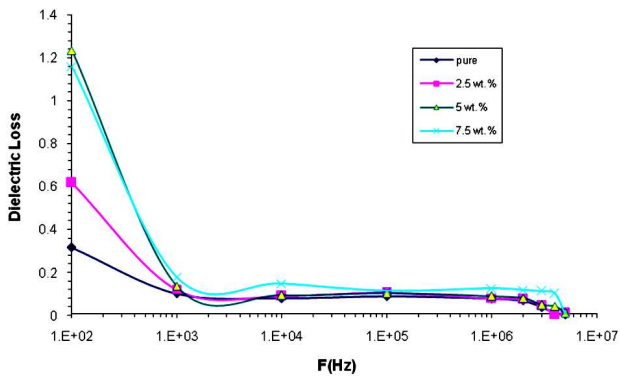
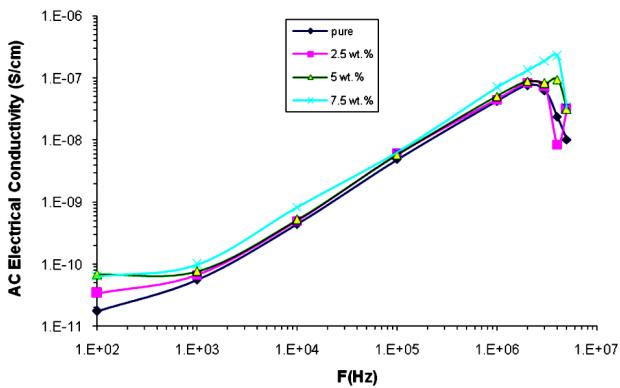**Fig. 2.** Variation of the dielectric losses for (PVA–PAA–PbO_2) nanocomposites with the frequency

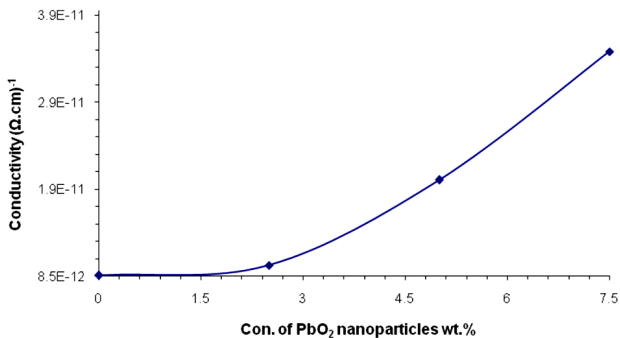**Fig. 3.** Variation of the ac electrical conductivity for (PVA–PAA–PbO_2) nanocomposites with the frequency

Fig. 4. Variation of the dc electrical conductivity for (PVA–PAA–PbO_2) nanocomposites with the concentration of PbO_2 nanoparticles

Fig. 5. Variation of the electrical resistance for (PVA–PAA–PbO_2) nanocomposites with the pressure

Fig. 6. Schematic view of the pressure-sensitive nanocomposites: before the applied pressure (*a*) and after the applied pressure (*b*)

to the relaxation process [16]. A variation of the dielectric losses for (PVA–PAA–PbO_2) nanocomposites with the frequency is shown in Fig. 2. From the figure, it is observed that the dielectric losses decrease, as the frequency increases. This situation has been at-
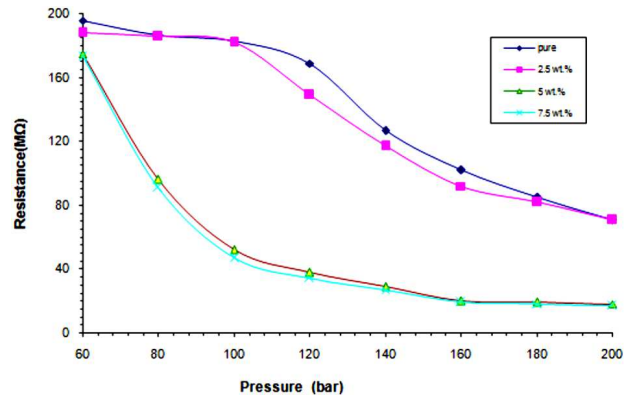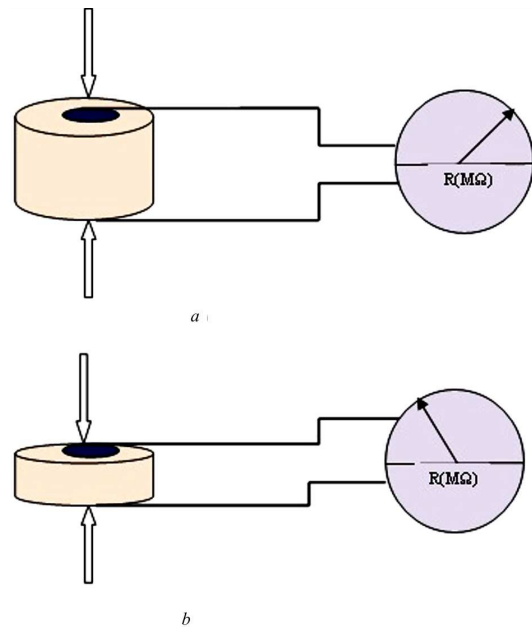
tributed to the fact that, in the low-frequency range, there are all types of polarization. Consequently, the losses due to those types of polarization increase. In the high-frequency range, the types of polarization are decreased, and this is accompanied by a decrease in losses [17].

Figures 2 and 3 show the dielectric constant and dielectric losses of a (PVA–PAA) blend, which increase with the concentration of PbO_2 nanoparticles. This behavior is related to an increase in the polarizations

of free electrons and space charges, while increasing the concentration of PbO$_2$ nanoparticles [17].

Figure 3 shows a variation of the ac electrical conductivity for (PVA–PAA–PbO$_2$) nanocomposites with the frequency at room temperature. The increase of the ac electrical conductivity with frequency is common for polymeric and semiconductor samples. As the filler concentration is increased, the inorganic filler molecules start bridging the gap separating the two localized states and lowering the potential barrier between them, thereby facilitating the charge transfer between two localized states. The frequency-dependent conductivity is caused by the hopping of charge carriers in the localized state. The hopping refers to the sudden displacement of charge carriers from one position to another neighboring site and, in general, includes both jumps over a potential barrier and the quantum mechanical tunneling [18]. As shown in the figure, the ac electrical conductivity of the (PVA–PAA) blend increases with the concentration of PbO$_2$ nanoparticles. This result can be attributed to an increase in the conductivity as a result of the increasing charge carrier density in the polymer matrix [19, 20].

Figure 4 shows a variation of the dc electrical specific conductivity for (PVA–PAA–PbO$_2$) nanocomposites with the concentration of PbO$_2$ nanoparticles at room temperature. From the figure, the dc electrical specific conductivity of the (PVA–PAA) blend increases with theconcentration of PbO$_2$ nanoparticles, which may be attributed to an increase in the number of free charge carriers [21–23].

Figure 5 shows a variation of the electrical resistance for (PVA–PAA–PbO$_2$) nanocomposites with the pressure at different concentrations of PbO$_2$ nanoparticles. As shown in the figure, the electrical resistance of nanocomposites decreases, as the pressure increases. The explanation of the electrical resistance behavior for nanocomposites with the pressure is as follows: the PbO$_2$ nanoparticles are aggregated as clusters in the (PVA–PAA) blend [24], hence, the resistance is high, as shown in Fig. 6, *a*. Applying the pressure causes a decrease in the distance between filler particles inside the matrix and an increase in the number of conductive paths, which leads to a decrease in the resistance of composites [25]. Hence, the resistance is low, as shown in Fig. 6, *b*. The (PVA–PAA–PbO$_2$) nanocomposites combine the advantages of individual materials; for example, the

flexibility and processability of a (PVA–PAA) blend and the selectivity, mechanical properties, electrical properties, and thermal stability of PbO$_2$ nanoparticles. The (PVA–PAA–PbO$_2$) pressure-sensitive nanocomposites have high sensitivity for the pressure, low cost, easy processing, and lightweight. This is similar to the results of researchers in [26–28].

4. Conclusions

1. The dielectric constant, dielectric losses, and ac electrical conductivity of the (PVA–PAA) blend for pressure-sensitive nanocomposites increase with the concentration of PbO$_2$ nanoparticles.

2. The dielectric constant and dielectric losses of (PVA–PAA–PbO$_2$) pressure-sensitive nanocomposites decrease with an increase in the frequency, while the ac electrical conductivity increases with the frequency.

3. The dc electrical specific conductivity of the (PVA–PAA) blend for pressure-sensitive nanocomposites increases with the concentration of PbO$_2$ nanoparticles.

4. The electrical resistance of (PVA–PAA–PbO$_2$) pressure-sensitive nanocomposites decreases with an increase in the pressure in the interval 80–200 bar.

5. The (PVA–PAA–PbO$_2$) pressure-sensitive nanocomposites have high sensitivity for the pressure.

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НОВІ СЕНСОРИ ТИСКУ З НАНОКОМПОЗИТІВ (ПОЛІМЕРИ, ЩО МОЖУТЬ БУТИ БІОРОЗКЛАДЕНІ, ТА НАНОЧАСТИНКИ ОКИСЛУ МЕТАЛУ): ВИГОТОВЛЕННЯ І ХАРАКТЕРИСТИКА

Р е з ю м е

Розроблено та досліджено нові нанокомпозити з низькою вартістю і вагою і хорошою чутливістю до тиску на основі полівінілового спирту, поліакрилової кислоти і наночастинок окису свинцю. Виміряні провідність нанокомпозитів за постійним струмом і діелектричні параметри в частотному діапазоні 100 Гц–5 МГц. Експеримент показує, що діелектрична константа і діелектричні втрати PVA–PAA–PbO₂ нанокомпозитів зменшуються з ростом частоти і ростуть зі збільшенням концентрації наночастинок. Провідність нанокомпозитів за змінним струмом зростає з частотою і концентрацією PbO₂ наночастинок. Провідність за постійним струмом також збільшується з концентрацією PbO₂ наночастинок. Показано, що в інтервалі тисків 60–200 бар опір нанокомпозитів зменшується зі збільшенням тиску за високої чутливості до нього.