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Polyethylene Oxide/Barium Titanate Composites: Optical, Electrical and Thermal Properties

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Our goal is to prepare and study the physical properties (optical, electrical, and thermal) of barium titanate ($BaTiO_3$)/polyethylene oxide (PEO) as ceramic polymer composites (CPCs). CPCs consist of $BaTiO_3$ (0, 2, 4, 7, 10, and 12 by weight percentage concentration) as ferroelectric ceramic fillers and PEO as a host polymer matrix. The composites have been prepared by casting method and their physical properties are investigated and discussed. The optical properties and parameters, such as the optical absorption, optical energy gap, dielectric constant, and refractive index are determined as a function of $BaTiO_3$ concentration and applied UV-wavelength ranges from 300 to 800 nm. The alternating electrical properties have been calculated and studied as a function of $BaTiO_3$ concentrations; applied frequency varies from 50 Hz to 1000 Hz, and temperature is in the range 30-55 °C. Also, the thermal conductivity of the PEO/$BaTiO_3$ CPCs has been calculated as a function of both $BaTiO_3$ concentration and temperature. With the increase in $BaTiO_3$ ceramic content, the optical energy gap is found to decrease from 2.85 eV for 0 wt. % $BaTiO_3$ to 2.26 eV for 12 wt. % $BaTiO_3$. We found that these physical properties depend on the $BaTiO_3$ ceramic content and temperature. This indicates the possibility of manipulation of the physical parameter values by control of the $BaTiO_3$ weight ratio in polymer composites.

Keywords: Polyethylene oxide (PEO), Barium titanate, Optical, Electrical, Thermal, Conductivity.

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1. INTRODUCTION

Barium titanate is considered one of the most important ceramic materials because it has high dielectric constant value [1]. The low dielectric constant value of PEO makes its use in technology limited; here comes the importance to fill it with $BaTiO_3$, which gives new composite materials the desired physical characteristics that are attractive for use in electronic and technological applications [2-4]. These composites can be also widely utilized as ceramic capacitors and piezoelectric transducers [2, 5], in microelectronic packaging [6]. Also, they can be used as high energy storage capacitor dielectrics and sensors in smart systems [7, 8]. Because of simple and convenient process, cheap cost, and excellent physical properties of CPCs, a literature survey shows that many researchers studied different systems of CPCs that have been demonstrated in order to obtain the desirable properties of materials. Louis Lévêque et al. [9] studied the effects of $BaTiO_3$ content on dielectric properties of epoxy-$BaTiO_3$ and epoxy-$SrTiO_3$ composites, and they reported an increase in the real and imaginary parts of the permittivity with increasing $BaTiO_3$ and $SrTiO_3$ contents in the epoxy matrix. Shail K. Gupta et al. [10] studied the PMMA/$BaTiO_3$ nanocomposites and they observed that the frequency dependences of the dielectric constant and loss tangent increase with increasing $BaTiO_3$. Wenhu Yang et al. [11] studied the FeAlSi/epoxy and FeAlSi/$BaTiO_3$/epoxy composites with various volume fractions of FeAlSi and $BaTiO_3$ fillers, and they investigated the complex permittivity, permeability and their relationship with microwave absorption properties. Shu-Hui Xie et al. [5] studied the polyimide/$BaTiO_3$ composites and they reported that the dielectric constants could be controlled by the content of $BaTiO_3$. L. Ramajo et al. [12] studied the dielectric properties and relaxation phenomena of epoxy resin-$BaTiO_3$ composite materials and they reported that the dielectric constant of composites depends on $BaTiO_3$ amount, temperature and frequency. R.K. Goyal et al. [13] studied the dielectric, mechanical and thermal properties of $BaTiO_3$/PMMA composites and they reported that the dc electrical conductivity and dissipation factor increase with increasing $BaTiO_3$ volume fraction.

The present research work aims to investigate the optical, ac electrical, dielectric and thermal properties of PEO/$BaTiO_3$ composites. PEO resin is filled with different concentration of $BaTiO_3$ in the form of particles to dissipate electrostatic charges in the prepared CPCs. Moreover, $BaTiO_3$ has been used because of its ability to impart high dielectric constant to insulating polymers at relatively low filler contents. The obtained optical properties are discussed in the UV-wavelength ranges from 300 to 800 nm that can provide a detailed characterization of the optical dispersion behavior of the composites. AC electrical and dielectric properties are discussed in the applied frequency range (50 to 1000 Hz) that can provide a detailed and quantitative characterization of the dielectric relaxation behavior, and in temperature range (30-55 °C) that can provide more details on the behavior of charges and ability to enhance the electrical and thermal conductivities.

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2. EXPERIMENTAL WORK

The PEO/$BaTiO_3$ CPCs were prepared in the Department of Physics, Materials Physics Laboratory, University of Jordan, Amman-Jordan. Composites with the following $BaTiO_3$ concentrations (0, 2, 4, 7, 10, and 12 wt. %) were prepared by casting method [14]. The sample sheet thickness is 0.2 μm.

The changes in the optical absorption spectra property of PEO/$BaTiO_3$ composites were studied with the UV absorption spectroscopy technique that used to investigate the band structure and electron transition types of these materials. The determined optical quantities in the present paper were calculated from equations and models reported in [15].

The Hewlett Packard (HP) 4192A impedance analyzer was used to measure the impedance and phase angle. The determined ac electrical quantities in the present paper were calculated from equations and models reported in [14].

Thermal conductivities of the tested composites were measured after heating specimens of double discs placed in the sample holder. The procedure and the determined thermal quantities were reported in [14].

3. RESULTS AND DISCUSSION

3.1 Optical Results

In the absorption spectra, as shown in Fig. 1, it is seen that PEO/$BaTiO_3$ CPCs exhibit a strong absorption peak value between 350 and 400 nm. It is shown that the addition of $BaTiO_3$ leads to an increase in the peak intensity and also causes an increase in the density of charge carriers in the solid composite materials which leads to an increase in the absorption spectra [16]. According to the results obtained, tested composites used in the present study have good UV protection especially at high values of filler concentrations.

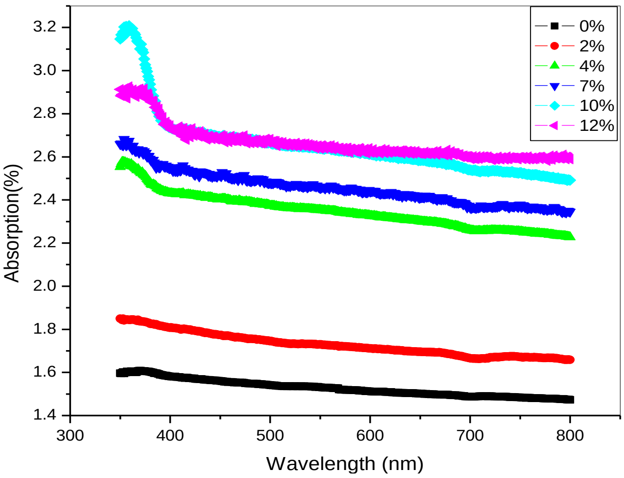

Fig. 1 – The absorption spectra for PEO/$BaTiO_3$ composites

The optical energy gap values were calculated by plotted $(\alpha\hbar\omega)^2$ vs. $\hbar\omega$ as shown in Fig. 2. Extrapolation of the linear part of these curves gives the optical energy gap. The values of optical band gap are found to be 2.85 eV for 0 wt. %, 2.70 eV for 2 wt. %, 2.65 eV for 4 wt. %, 2.51 eV for 7 wt. %, 2.39 eV for 10 wt. %, and 2.26 eV for 12 wt. % $BaTiO_3$ content. The optical energy gap decreases with increasing $BaTiO_3$ content and this may be attributed to the increase in localized states within the forbidden energy gap [17].

Fig. 3 shows the refractive index versus wavelength, and clearly, the refractive index increases as $BaTiO_3$ content increases and decreases as wavelength increases.

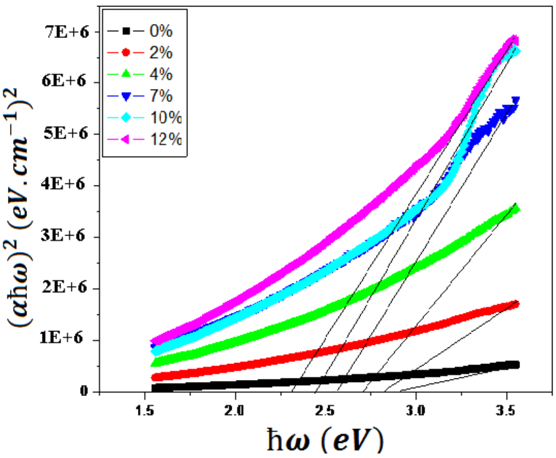

Fig. 2 – Plots of $(\alpha\hbar\omega)^2$ vs. $\hbar\omega$ for PEO/$BaTiO_3$ composites

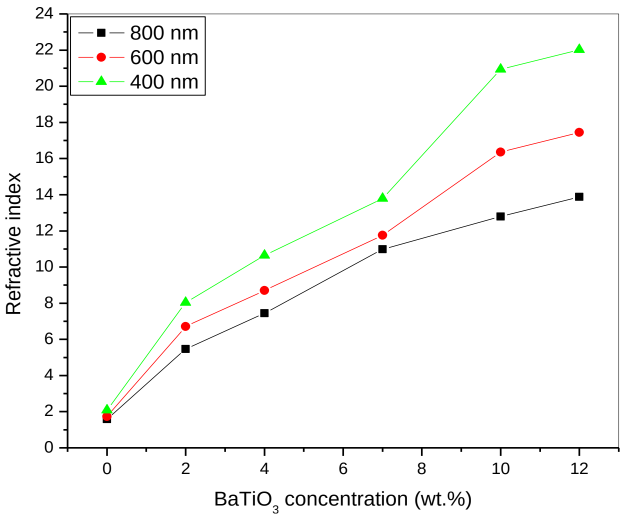

Fig. 3 – The variation of the refractive index with the UV-wavelength for PEO/$BaTiO_3$ composites

Fig. 4 shows the dielectric constant values of PEO/$BaTiO_3$ CPCs. It is clearly observed that as $BaTiO_3$ concentration increases, the dielectric constant increases. This is caused by the interfacial polarization effect created between $BaTiO_3$ particles, since the dipolar polarization effect induced by the permanent dipoles results from the distribution of the charge density between O, Ba and Ti atoms existent in $BaTiO_3$ [2]. Also, it was found that the dielectric constants at low frequencies have high values that results from the effect of space charge, so this increase in dielectric constants values is due to the higher polarization of $BaTiO_3$ than that of the PEO matrix [13].

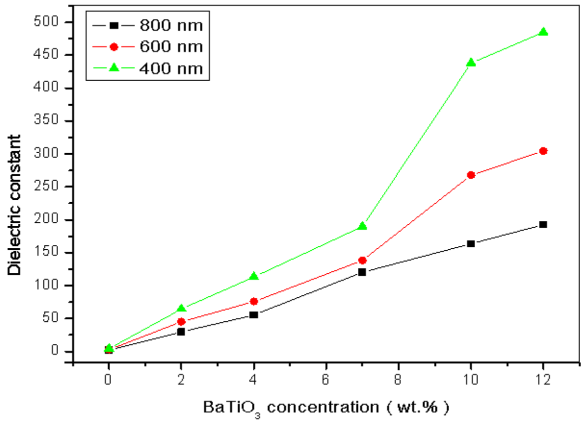

Fig. 4 – The variation of the dielectric constant with $BaTiO_3$ concentration

3.2 AC Electrical Results

Fig. 5 shows the variation of ac electrical conductivity with applied frequency. It was found that the ac electrical conductivity increases with an increase in frequency. This is because the charge hopping process in the polymer composites also increases, thereby increasing the ac electrical conductivity [18].

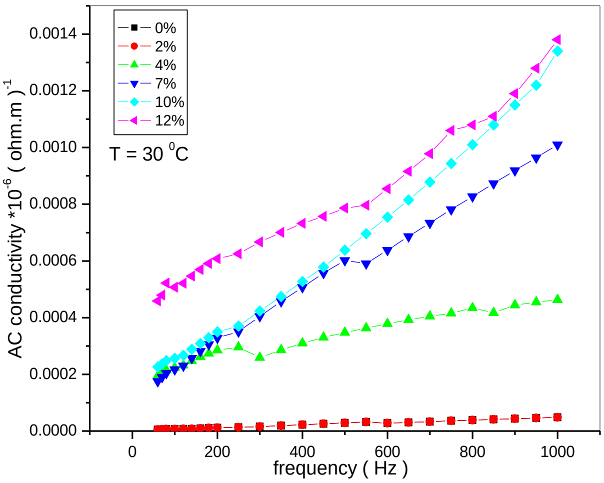

Fig. 5 – The variation of ac conductivity with frequency for $PEO/BaTiO_3$ composites

Fig. 6 shows the variation of the thermal conductivity. It can be seen that the thermal conductivity increases with an increase in both temperature and filler concentration. The $BaTiO_3$ concentration, which makes them major channels for thermal conduction, causes the improvement of the thermal conductivity value [19].

Fig. 7 shows the variation of Lorenz number of the CPCs at absolute temperature 303 K. It can be observed that the Lorenz number increases with increasing $BaTiO_3$ concentration that refers to the difference in the rates of increasing of ac electrical and thermal conductivities with $BaTiO_3$ content [20].

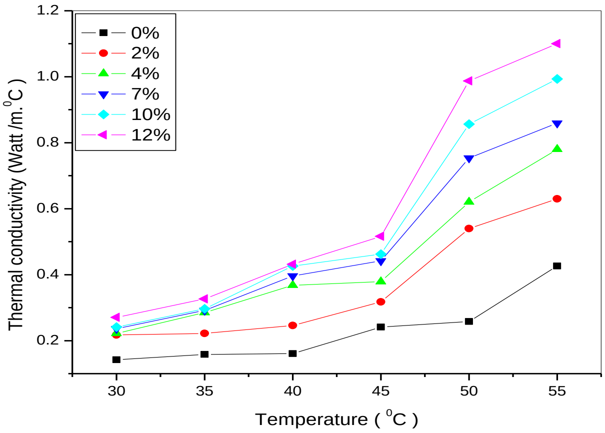

Fig. 6 – The variation of thermal conductivity with temperature for $PEO/BaTiO_3$ composites

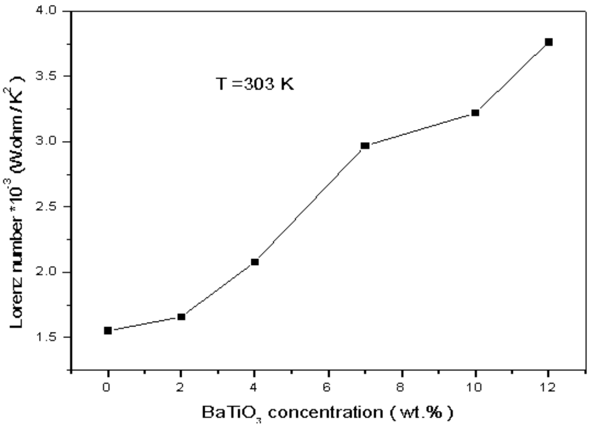

Fig. 7 – The variation of the Lorenz number for $PEO/BaTiO_3$ composites

4. CONCLUSIONS

$PEO/BaTiO_3$ CPCs were successfully prepared and their physical properties were investigated. The optical measurement showed that the CPCs had electronic transition and interaction between $BaTiO_3$ and the host PEO polymer matrix. Measurements confirmed that the dielectric constant and refractive index increase with increasing $BaTiO_3$ content due to the excellent dispersion of its particles throughout the system. Optical energy gap decreases with increasing $BaTiO_3$ concentration which means that the tested CPCs in this study are useful for use in UV protection. With increasing $BaTiO_3$ content, the dielectric constant increases, and it could be concluded that the CPCs obtained in this research paper should be a desirable candidate for high dielectric constant materials. The ac conductivity increases with frequency and $BaTiO_3$ filler concentration. It was found that the thermal conductivity enhanced significantly with increasing $BaTiO_3$ weight fraction and increasing temperature. The highest thermal conductivity value reached 1.1 W/m·°C at 55 °C for 12 wt. % $BaTiO_3$, which is approximately three times higher than that of pure PEO.

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Композити поліетиленоксиду/титанату барію: оптичні, електричні та теплові властивості

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Мета роботи полягає у вивченні фізичних властивостей (оптичних, електричних та теплових) титанату барію ($BaTiO_3$)/поліетиленоксиду (PEO) як керамічних полімерних композитів (CPC). CPC складаються з $BaTiO_3$ (концентрація 0, 2, 4, 7, 10, та 12 вагових відсотків) як сегнетоелектричного керамічного наповнювача та PEO як полімерної матриці-господаря. Композити отримані методом лиття, їх фізичні властивості досліджено та обговорено. Оптичні властивості та параметри, такі як оптичне поглинання, оптична ширина забороненої зони, діелектрична проникність та показник заломлення визначаються як функції концентрації $BaTiO_3$, а використана довжина хвилі УФ випромінювання становить від 300 до 800 нм. Змінні електричні властивості були розраховані та вивчені як функції концентрацій $BaTiO_3$; застосовувана частота коливається від 50 Гц до 1000 Гц, а температура в межах 30-55 °C. Крім того, теплопровідність CPC PEO/$BaTiO_3$ була розрахована як функція концентрації $BaTiO_3$ та температури. Зі збільшенням вмісту $BaTiO_3$ виявляється, що оптична ширина забороненої зони зменшується з 2,85 еВ при 0 мас. % $BaTiO_3$ до 2,26 еВ при 12 мас. % $BaTiO_3$. Ми виявили, що ці фізичні властивості залежать від вмісту $BaTiO_3$ та температури. Це вказує на можливість маніпулювання значеннями фізичних параметрів шляхом контролю вагового співвідношення $BaTiO_3$ у полімерних композитах.

Ключові слова: Поліетиленоксид (PEO), Титанат барію ($BaTiO_3$), Оптичний, Електричний, Тепловий, Провідність.