



برعاية معالي وزير التعليم العالي والبحث العلمي
الأستاذ علي محمد الحسين الأديب
وبإشراف السيد رئيس جامعة واسط
الأستاذ الدكتور جواد مطر الموسوي

تحت شعار
من تراثنا المعرفي العظيم نستمد العزم لتطوير علومنا ومعارفنا
تعقد جامعة واسط مؤتمرها العلمي الخامس
للمدة ١٣-١٤/١٢/٢٠١١ م
الموافق ١٧-١٨ محرم الحرام ١٤٣٣ هـ



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رئيس الجلسة : أ. م. د. هادي دويج العتابي
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Optical Properties of Polystyrene-Aluminium Phosphate composite films

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Abstract

Optical properties of polystyrene (PS) films, with different levels of Aluminium phosphate ($AlPO_4$), were investigated. UV-VIS spectra of the films were studied using the optical transmittance measurements which taken in the spectral region from 200 – 900 nm. It was found that the optical absorption is due to direct allowed transitions. The energy gaps (E_{opt}) increases from (4.44 to 4.47) eV with increasing $AlPO_4$ concentration, while the width of the tail decreases. The optical constants such as refractive index n , extinction coefficient K , dielectric constant ϵ_r , and dielectric loss ϵ_i have been also calculated. The dielectric constant and refractive index are also affected by $AlPO_4$ content.

Keywords: polymer composite, energy gap, Urbach energy, dielectric constant.

Introduction:

Polymer composites incorporating, semiconductor, carbon black, nano materials, and magnetic materials have been widely used and studied as multifunction and materials with inherent polymer properties [1,2].

The ability of polymers to act as electrical insulators is the basis for their widespread use in the electrical and electronic fields [3]. Non-polar polymers such as PS are especially significant because of their low loss values over the widest frequency range [4]. The dielectric behavior of polymer films is of considerable interest due to their applications for insulation and microelectronics [5]. The dielectric constant and the loss factors are the most convenient and sensitive methods for studying the polymer structure [6].

The measurement of the optical absorption coefficient, particularly near the fundamental absorption edge, provide a standard method for investigation of optically induced electronic transitions and provide some ideas about the structure and energy gap in both crystalline and amorphous materials.

In present work, absorption coefficient (α), optical energy gap E_{opt} , width of band tail E_t , refractive index n , and dielectric constant are determined for Polystyrene-Aluminium Phosphate composite films. The effect of Aluminium Phosphate concentration on these properties are also studied.

Experimental detail:

Films of Polystyrene – Aluminum phosphate composite were prepared by solution caste; 2 gm of pure Polystyrene (PS) supplied from Sigma-Aldrich company has been dissolved in 25ml of Chloroform to obtain a pure PS film; amount of

0.1, 0.2, 0.3, and 0.4 gm of Aluminum phosphate ($AlPO_4$) were added to the pure PS solutions, to prepare the different filling levels (5%, 10%, 15%, and 20%) respectively. The complete dissolution was obtained using a magnetic stirrer at temperature $80^\circ C$ for 2hr. These homogeneous solutions were spread on a glass plate. The whole assembly was placed in a dust free chamber and allowed to evaporate the solvent slowly in air at room temperature for 24h. The thickness of the films was determined using micrometer at different places in each film and an average was taken; and it was found to be (0.298, 0.275, 0.272, 0.275, 0.246)mm for the samples (0, 5, 10, 15, and 20) wt% $AlPO_4$ respectively.

Theoretical:

The most important optical properties are the refractive index n and the extinction coefficient k , which are generally called optical constants, are calculated using the fundamental relations of photon transmittance T and absorbance A . Practically, the optical absorption coefficient α which is a function of wavelength can be calculated from the optical absorbance spectra by using the relation [7,8]:

$$\ln\left(\frac{I_0}{I_t}\right) = \alpha d = 2.303 A \quad (1)$$

where I_0 and I_t are the intensities of the incident and transmitted beams respectively, A the optical absorbance, and d is the film thickness. The extinction coefficient K , is related to the absorption coefficient α through $K = \alpha\lambda/4\pi$, where λ is the wavelength of light.

The refractive index as a function of wavelength can be determined from the reflection coefficient data R and the extinction coefficient K using equation:

$$n = \left(\frac{4R}{(1-R)^2} - k^2 \right)^{\frac{1}{2}} - \frac{(R+1)}{(R-1)} \quad (2)$$

The absorption edge for direct and non-direct transitions can be obtained in view of the models proposed by Tauc et al [9]:

$$\alpha h\nu = C_0 (h\nu - E_{opt})^\gamma \quad (3)$$

where C_0 is a constant related to the properties of the valance and conduction bands [1], $h\nu$ is the photon energy, α is the absorption coefficient, E_{opt} is the optical energy band gap of the material, and $\gamma = 1/2, 3/2, 2, \text{ or } 3$ for direct allowed, direct forbidden, indirect allowed and indirect forbidden transitions respectively [10,11].

A plot of $(\alpha h\nu)^{1/\gamma}$ versus $(h\nu)$ often yields a reasonably good straight line fit to the absorption edge and the extrapolated $(h\nu)$ at which $(\alpha h\nu)^{1/\gamma} = 0$ provides a convenient experimental benchmark for the optical band gap E_{opt} .

The optical absorption coefficient $\alpha(\nu)$ near the band edge for many amorphous and crystalline materials, shows an exponential dependence on photon energy $(h\nu)$ and obeys an empirical relation given by Urbach [12]

$$\alpha(\nu) = \alpha_0 \exp(h\nu/E_t) \quad (4)$$

where α_0 is a constant and E_t is related to width of the band tails of localized states in the normally forbidden band gap that associated with the amorphous nature of

the material. It should be mentioned that this equation is applicable only in the low absorption region $\alpha = (10^3 - 10^4) \text{ cm}^{-1}$.

Finally, the real and imaginary parts of dielectric constant (ϵ_r and ϵ_i respectively) were calculated as follows [11]; The complex refractive index:

$$N^* = n - ik \quad (5)$$

Complex dielectric constant:

$$\epsilon^* = \epsilon_r - i \epsilon_i \quad (6)$$

From the relation $N^* = \sqrt{\epsilon^*}$, there are:

$$\epsilon_r = n^2 - k^2 \quad (7)$$

$$\epsilon_i = 2nk \quad (8)$$

where ϵ_r is the true permittivity, describes the stored energy, and ϵ_i is the imaginary permittivity describes the dissipation energy.

Results and discussion:

The optical absorption spectra for $(PS)_{1-x} - (AlPO_3)_x$ thin films of different compositions ($x = 0.05, 0.10, 0.15$, and 0.20) were recorded at room temperature by UV-VIS double beam spectrometer (Lambda 25) in the wave length range $(190 - 1100) \text{ nm}$. Fig.(1) shows the optical transmittance for different thin films, it could be noted that at longer wavelengths $\lambda > 280 \text{ nm}$ all films become transparent and the value of transmission decreases with increasing $AlPO_4$ contents.

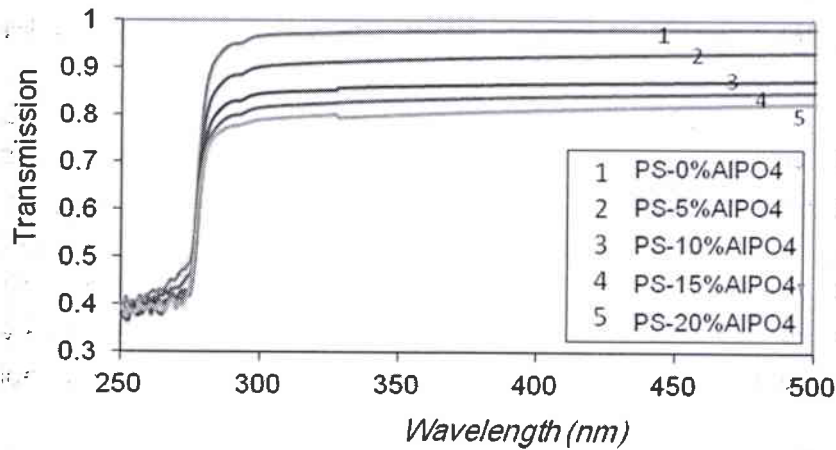


Fig (1): The optical transmittance as a function of wavelength for PS Polymer with different $AlPO_4$ content.

Fig.(2) shows measurements of absorbance (A) against wavelength λ for thin films of different compositions. The absorption coefficient $\alpha(\omega)$ which is a function of λ was calculated from the optical absorbance (A) spectra by using equation (1).

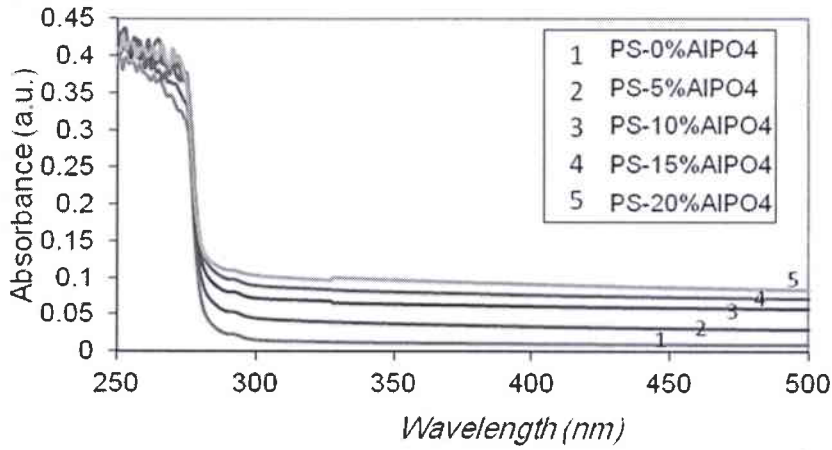
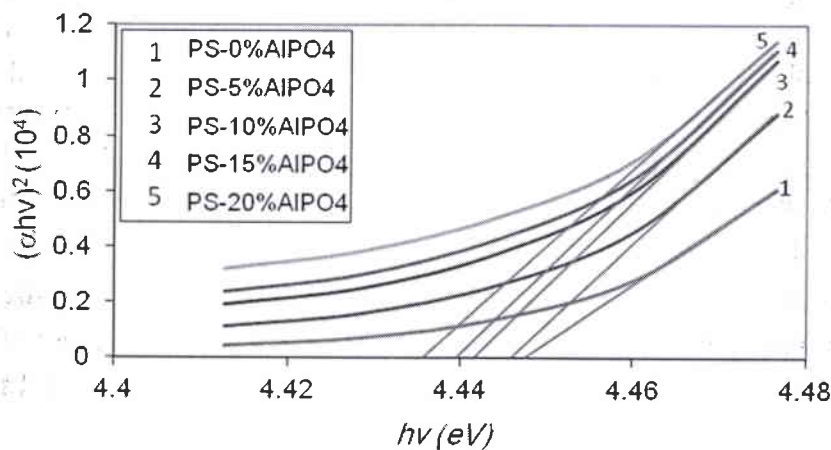


Fig (2): Optical absorption coefficient (α) as a function of wavelength for PS Polymer with different AlPO_4 content.

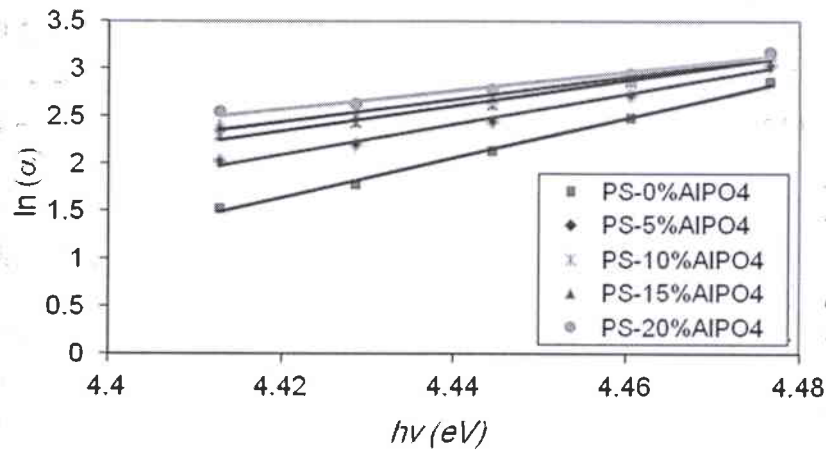
Fig.(3) shows the dependence of $(\alpha h\nu)^2$ on the photon energy ($h\nu$) for direct allowed transitions. It is to be noticed that the curve is characterized by the presence of an exponentially decaying tail at low photon energy. To obtain information about direct or indirect inter-band transitions, the fundamental absorption edge data could be analyzed within the frame work of one electron theory of Bardeen [13]. This theory has been used to analyzed the absorption edge data of molecular solids such as Phthalocyanine thin films [14]. The absorption ($\alpha \geq 10^4 \text{ cm}^{-1}$) is related to direct band transitions [15].

The optical energy gaps (E_{opt}) are estimated from the extrapolation of the linear portion of the graph to the ($h\nu$) axis. It is observed that the E_{opt} increase with increasing AlPO_4 content. The plot shows straight line with some deviation from linear at the lower value of α , which were suggested by Redfield and Afromowitz [16] as possibly due to imperfections in the material.

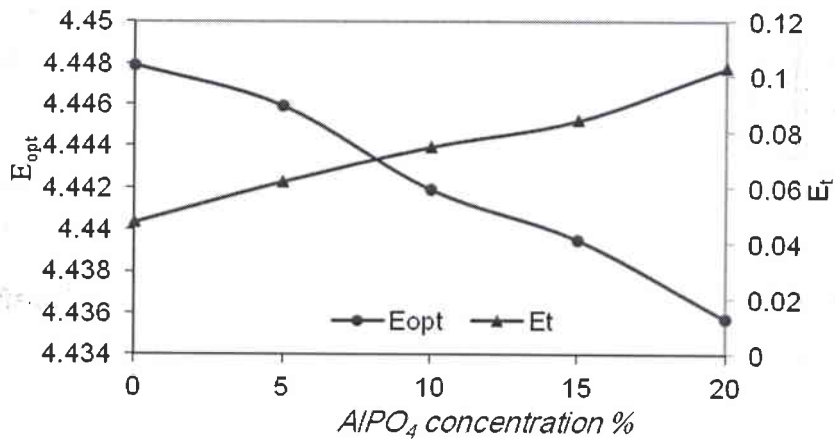


Fig(3): Relation between the $(\alpha h\nu)^2$ and $h\nu$ for PS Polymer with different AlPO_4 content.

The Urbach plot is presented in Fig.(4) in which the absorption coefficient is plotted as a function of $h\nu$. The values of E_t are determined using equation (4). The values of E_{opt} and E_t against $AlPO_4$ content are shown in Fig.(5). It is clear that the E_{opt} increases with increasing $AlPO_4$ concentration, but on the other the width of the band tail of the localized state shift toward lower energies from (0.1 to 0.05) eV. This may be attributed to rearranged of $AlPO_4$ contents as an interstitial atoms in the polymers matrix, which caused the changes in the degree of crystallization, which is known to decrease the width of the localized states, thus increases the value of the E_{opt} . Such a change has been reported by other different polymer compositions [1,17].



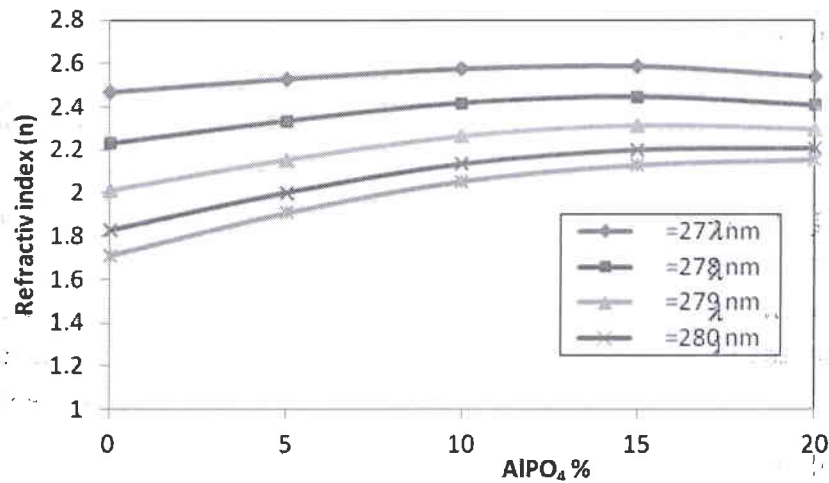
Fig(4): Relation between the $\ln(\alpha)$ and $h\nu$ for PS Polymer with different $AlPO_4$ content.



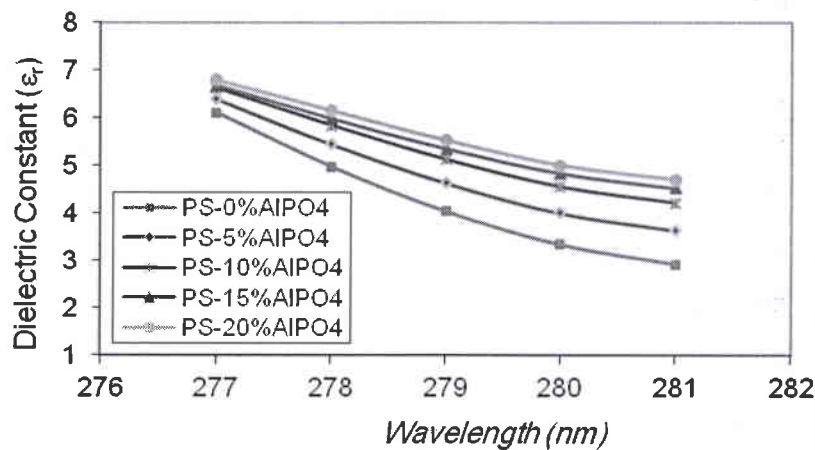
Fig(5): Direct allowed optical energy gap, and band tail for PS doped with Aluminum Phosphate.

Fig.(6) shows the variation of refractive index (n) of all thin films with $AlPO_4$ contents at different wavelength. It is clear that the refractive index (n) increases with increasing $AlPO_3$ contents.

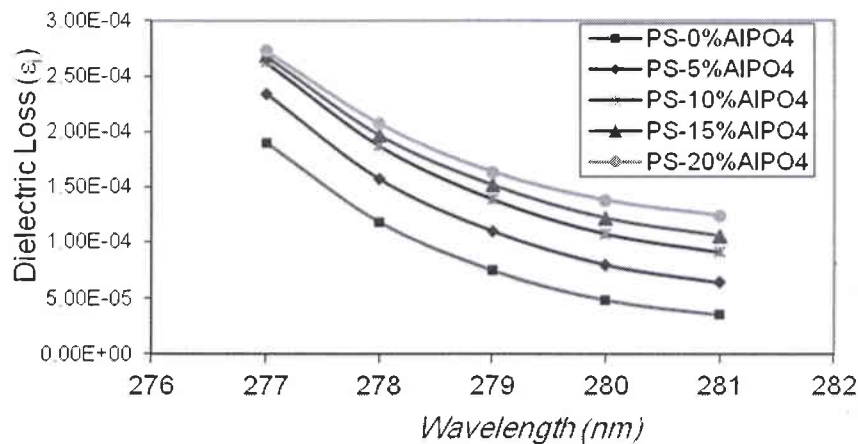
The behavior of the real part of the dielectric constant ϵ_r , and the imaginary part ϵ_i against λ for all samples are displayed in Fig.(7) and Fig(8). It was observed that the dielectric permittivity decrease with increasing λ , and increasing with increases $AlPO_4$ contents, as well as the significant decrease of dielectric constant can be observed in the wave length range (278 – 281)nm.



Fig(6): Refractive index as a function of $AlPO_4$ content for different wavelength in the exponential region.



Fig(7): Dielectric constant as a function of wavelength in the exponential region of the spectrum.



Fig(8): Dielectric loss as a function of wavelength in the exponential region of the spectrum.

Both the electric nature of the polymer and the effect of $AlPO_3$ in the PS polymer can be understood by correlating the dielectric constant values with the refractive index n . If the sample is non-polar insulator, the ϵ_r for long wavelength λ can be expressed by Maxwell's equation $\epsilon_r = n^2$. The difference between the n^2 and ϵ_r is a result of permanent dipoles and the semi-conductive character of the sample [18]. The slightly difference between n^2 and ϵ_r is due to the non polar behavior of PS .

Conclusion:

Optical absorption of $(PS)_{1-x} - (AlPO_3)_x$ ($x = 0.05, 0.10, 0.15, \text{ and } 0.20$) thin films, were recorded in the UV-Visible region at room temperature and various optical parameters such as optical energy gap E_{opt} and Urbach energy E_t were calculated. The optical energy E_{opt} was found to increase with $AlPO_3$ contents. The band tail width E_t obeys' Urbach empirical relation. The optical absorption is due to direct allowed transition. The value of refractive index n , and dielectric constant decreased by increasing the $AlPO_3$ content in PS polymer through the investigated range.

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الخصائص البصرية لأغشية مركبات بوليستيرين-فوسفات الألمنيوم

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الخلاصة

تم في هذا البحث دراسة أغشية البوليستيرين (PS) المتضمنة نسب مختلفة من فوسفات الألمنيوم ($ALPO_4$). تمت دراسة أطيف (UV-VIS) لهذه الأغشية باستخدام قياسات النفوذ البصري ضمن النطاق الطيفي (200-900) نانوميتر. وجد أن الامتصاص البصري ناتجة عن الانتقالات المسموحة المباشرة. وأن فجوات الطاقة (E_{opt}) تزداد من (4.44eV) الى (4.47eV) مع زيادة تركيز ($ALPO_4$) في حين يقل عرض الذيل. تم أيضا حساب ثوابت بصرية أخرى مثل معامل الانكسار (n) ، ومعامل التوهين (K)، ثابت العزل (ϵ_r) وخسائر العزل (ϵ_i)، ووجد أن ثابت العزل ومعامل الانكسار تتأثر أيضا بقدار تركيز ($ALPO_4$).