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Effect of 1,4-dihydroxyanthraquinone dye on the dispersion parameters of poly(methyl methacrylate) polymer composites

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ABSTRACT

The casting approach was used to make films of poly methyl-methacrylate (PMMA) doped with varied weight percentages of 1,4-dihydroxyanthraquinone dye. These films were made on glass substrates at room temperature. Two different methods, including elemental analysis, FT-IR, and UV-Visible spectroscopies, were used in order to characterize the films that were created. The spectrum properties and the dispersion parameters of the dye-doped polymers have been examined with the help of spectrophotometric measurements. These experiments were performed in the wavelength range of (300-900) nm. According to the Wemple-Didomenico single oscillator model, many significant optical dispersion characteristics of the spectrum absorption have been identified. These include the dispersion energy, E_d the oscillator energy, E_o the wavelength of the single oscillator, λ_o and the average value of oscillator strength, S_0. It was discovered that dye doping had an influence on each of these different factors.

***Keywords*:** PMMA, Dispersion parameters, oscillator strength, Spectroscopic measurements

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1. INTRODUCTION

As a result of ground-breaking research and technological advancements in the area of polymer science and engineering, the human way of life has undergone a significant transformation over the course of the last century [1-3]. There has a long history of prosperous growth, which was made possible by the significant efforts of a large number of individuals. Motivated by the potentially lucrative commercial applications of polymeric materials in light emitting diodes, solar cells, magnetic storage media, light-stable color filters, optical sensors, optical data communication and optical data storage [4-6] field effect thermal and optics transistor (FET) and photonic and photo-electronic devices, polymeric materials have emerged as an area of interest [7, 8]. It is possible for the optical characteristics of polymers to be adjusted in an appropriate manner by the addition of dopants. The kind of doping, concentration, and their reactivity with the host matrix, which refers to the manner in which it penetrates and interacts with the chains of the polymer, are all factors that play a role in this modification. It is reasonable to anticipate that a polymeric system will improve both the electrical, mechanical and optical performance [9-11]. In-depth research into doped polymers, in which the same dopant is added in varying amounts, opens the door to the option of choosing the qualities one wants. The investigation of the optical characteristics of polymeric materials has gained greater significance as a result of the fact that it enables one to get a knowledge of the band structure and optical transitions of these materials [12, 13]. In spite of the fact that a significant amount of outstanding work has been published on the optical behavior of polymeric materials [14-17], it is still significant to expand the study of these polymers.

For the most part, inorganic materials, such as dye, gallium arsenide (GaAs), and cadmium- telluride (CdTe), have been utilized in the production of solar cells during the course of the previous several decades. Over 85 percent of all solar cells are azo or organic dye-based, so-called dye-based azo solar cells [18]. Synthesis and design of a few crucial low band gap polymers with higher absorption capacities have been described only recently, and scientists have taken a significant step forward in the process of constructing polymer solar cells [19].

The power conversion efficiency of polymer photovoltaics is lower than that of inorganic solar cells [20, 21], but polymer photovoltaics have the benefits of being mechanically flexible, having a cheap cost, and being able to be fabricated on a wide scale [22, 23].

To put this into perspective, in this paper, we have made an attempt to prepare 1,4 Hydroxyanthraquinone dye incorporated into Poly(methyl methacrylate) (PMMA) polymer films with low concentrations of 1,4-dihydroxyanthraquinone dye by using a simple solution casting technique. Our goal is to find new polymer composites with tuned properties that can be used for linear optical properties and optoelectronic applications. In addition, an effort is made to investigate the impact that dispersing 1,4-dihydroxyanthraquinone dye has on the optical properties of the PMMA system in order to investigate the electrical properties of the composite.

2. EXPERIMENTAL DETAILS

2. 1. Materials

The Poly(methyl methacrylate) PMMA in the form of granules with chemical formula $(C_5H_8O_2)_n$ is used in this paper. 1,4-Hydroxyanthraquinone dye was purchased from Sigma Aldrich (St. Louis, MO, USA) was used as a dopant, which is an organic compound is known since ancient times an important red pigment. Other compounds were utilized without additional purification since they were of analytical grade or above. 1,4-dihydroxyanthraquinone is a solid appearing reddish-orange as crystals and brownish-yellow as powder, with chemical formula ($C_{14}H_8O_4$). It is used chiefly in the synthesis of other dyes [24, 25]. Chloroform ($CHCl_3$) was used as a solvent for PMMA, which is an organic compound with the chemical formula ($CHCl_3$), and is a versatile solvent [26].

2. 2. Preparation of the Samples

Casting method technique from solution has been carried out to prepare films of 1,4-dihydroxyanthraquinone dye doped PMMA. The solution of PMMA was prepared by dissolving PMMA granules of weight 0.2 gm in 5 ml of $CHCl_3$ to obtain solution of 6% wt by using magnetic stirrer in mixing process for more than an hour until the polymer granules completely dissolved. On the other hand however 1,4-dihydroxyanthraquinone dye was dissolved easily in CHC to obtain the solution of dye with the same proportion as above of 6% wt. The solution of 1,4-Hydroxyanthraquinone dye was added to the dissolved polymer by choosing weight percentages of dye relative to pure PMMA (1, 2 and 3 wt %) and mixed for 30 min to get homogeneous solution. The mixture was poured into clean flat glass strate that were placed on a platform that could be leveled with the aid of a spirit level that was fixed on the platform. The dishes were then kept inside a dry-oven at a temperature of 40 °C for at least 24 hours to ensure that all traces of the solvent were removed, after which they were allowed to slowly cool down to the temperature of room temperature, which was 25 °C. In order to confirm that the dried samples are free of bubbles and are not damaged by heat, many films that are identical in appearance have been made. The prepared films' thickness was measured using a digital micrometer, and the results showed that the thickness was in the range of 143.2, 160.4 and 188.6 µm, respectively. These films were homogenous, free of any notable imperfections, and had a light brownish tint.

2. 3. Measurement

The measurement of the amount of energy that is absorbed by an electromagnetic wave in accordance with its frequency or wavelength is the purpose of the method known as absorption spectroscopy. The process of absorption causes an interaction between the sample and electromagnetic forces, and these changes in the absorption spectra may be interpreted to reveal the nature of the interaction. A molecule's or polymer material's unique fingerprint may be found in its absorption spectra. The study of the interactions that occur between electrons and radiation often makes use of the analytical instrument known as UV-Vis absorption. On the other hand, infrared absorption is a technique that is often used for the purpose of analyzing the interactions between the vibrational energy of bonds and electromagnetic waves. The most significant aspects of an object's optical qualities are its ability to respond to light, absorb light, reflect light, and scatter light. Because every kind of polymer has its own unique absorption band in the infrared portion of the spectrum, the infrared spectrum is one of the most useful tools for analyzing different types of polymers. Several different approaches were used in order to investigate the characteristics of the finished films. The Fourier Transform Infra-Red (FTIR) studies were recorded to the produced samples, and the infrared spectrum was viewed using an FTIR spectrophotometer model 8400S by Shimadzu in the range 4000-400 cm^{-1}. All of the tests were performed at room temperature, and optical absorption spectra were collected across the wavelength range of 190 to 900 nm using a twin beam UV/Vis-7200 (England) spectrophotometer.

3. RESULTS AND DISCUSSION

3. 1. FT-IR spectral analysis

Research using infrared spectroscopy was carried out in order to learn more about the nature of the chemical bonds. In Fig. 1, several typical infrared spectra for the 1,4-dihydroxyanthraquinone dye are shown. It is evident from Fig. 2 that aromatic C-H bending is responsible for the formation of many varied bands in the range of 867-711 cm^{-1}. The C=O stretch yields a band at 1672 cm^{-1}, while the C-H aliphatic offers a band at 2970 cm^{-1}. At 3084 cm^{-1}, an additional aromatic C-H emerged. There is a shift from 1697 cm^{-1} to 1681 cm^{-1}, which can be seen when previewing the Figure, this shift may be ascribed to the overlapping of the carbonyl esters C=O band of the PMMA polymer with the carbonyl esters C=O band of the dye. This shift can be observed when previewing the Figure. The stretching of aliphatic C-H bands has caused the appearance of a new band at 2953 cm^{-1}.

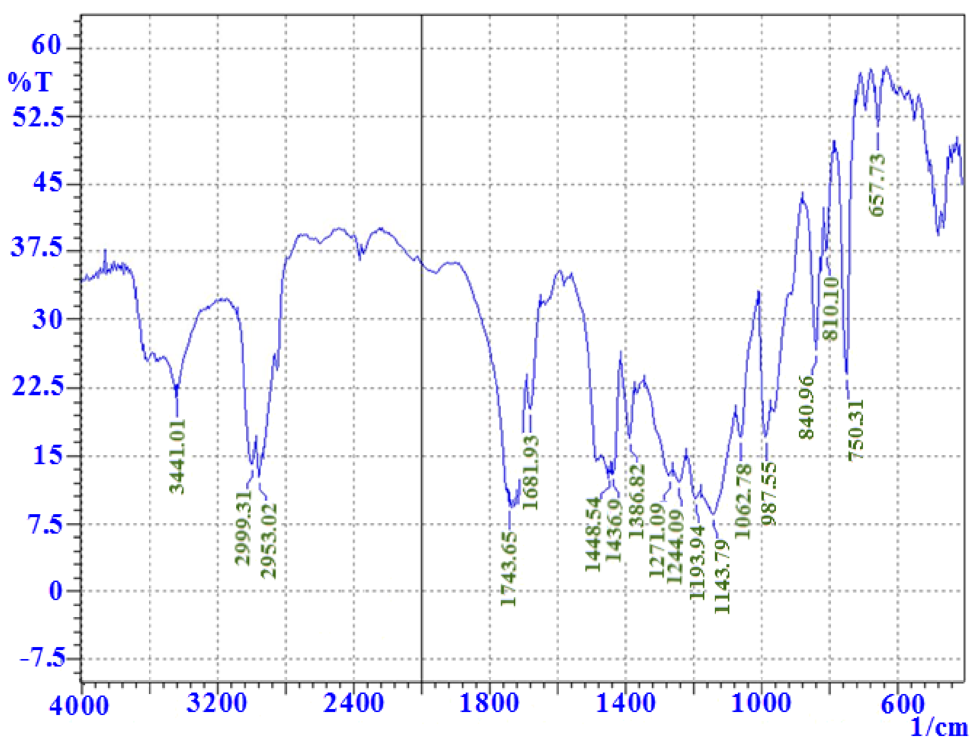

Fig. 1. FTIR spectrum of the 1,4-dihydroxyanthraquinone dye-doped PMMA

3. 2. Optical absorption studies

Figure 2 depicts the spectrum that is produced by polymer films with varying concentrations. There is a correlation between the concentration of 1,4-dihydroxyanthraquinone and a rise in the absorption values. This correlation may be explained by the presence of localized states of doping materials in the energy gap. The localized states lead to a narrowing of the energy gap, which in turn leads to an increase in absorption value [27, 28].

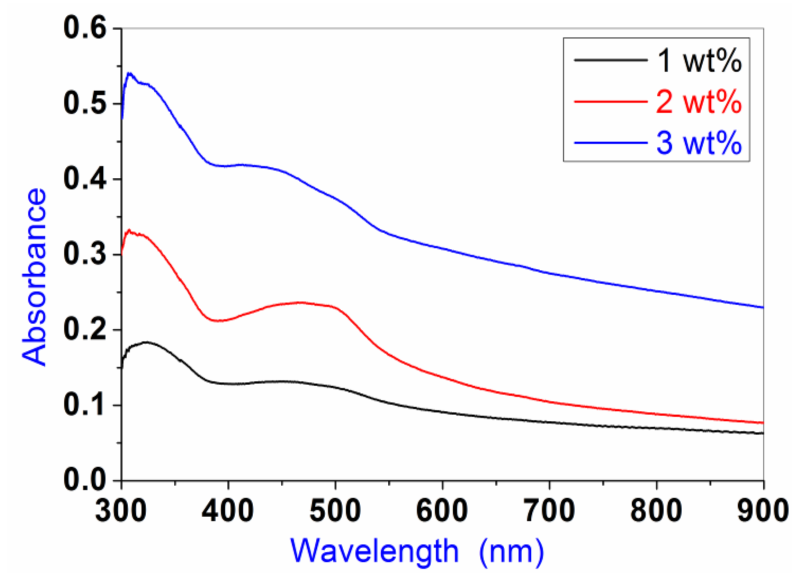

Fig. 2. UV-Vis absorption spectrum of Polymer films at different weight percentages.

The spectrum that may be obtained via the process of electromagnetic radiation absorption is referred to as the "absorption spectrum," and this is the phrase that is used to refer to the spectrum. The optical absorption spectra of dye film at room temperature is given in Figure 3, and as can be seen from the spectrum, the optical absorption excitonic peak is shifted to the lower wavelength side in contrast to that of 1,4-dihydroxyanthraquinone (520 nm).The optical absorption spectrum of dye film at room temperature. The conclusions of the several pieces of study that have been presented [29, 30] are in line with this conclusion, therefore it is not surprising. In the computation, the Tauc relation plays a role in determining the energy band gap of the material [31, 32].

$$(\alpha h\nu)^{\frac{1}{n'}} = B(h\nu - E_g) \qquad (1)$$

where B is a constant $(B = 4\pi\sigma_{\circ}/ncE_c)$, c is the speed of light, $\sigma_{\circ}$ is the extrapolated dc conductivity at T=∞, E_c is a measure of the extent band-tailing, E_g is the band gap of the materials, and n' is an exponent that varies according to the type of transition. In this equation, is a measure of the extent band-tailing, is the band gap of the materials, and is an exponent that varies according to the kind of transition. When n' is equal to 1/2 for transitions that are directly allowed, when n' is equal to 2 for transitions that are indirectly approved, and when n' is equal to 3 for transitions that are directly forbidden. When calculating the likely transitions, the plot of $(\alpha h\nu)^2$ versus $h\nu$ that is shown in Figure 4 is referred to as an important resource. It is possible, by applying the formula, to obtain the value of from the connection [33, 34].

$$\alpha = \frac{2.303}{t_{thk}} \log\left(\frac{I_i}{I_t}\right) = \left(\frac{2.303}{t_{thk}}\right) A_{bso} \tag{2}$$

where A_{bso} is the amount of absorption and t_{thk} represents the sample's thickness. The value of the optical fundamental gap of the produced polymer sample may be obtained by extrapolating the straight line to the point where $(\alpha hv)^2$ equals 0. It was discovered that the direct band gap values of the 1, 4-Hydroxyanthraquinone films are, respectively, 2.44, 2.51 and 2.62 eV.

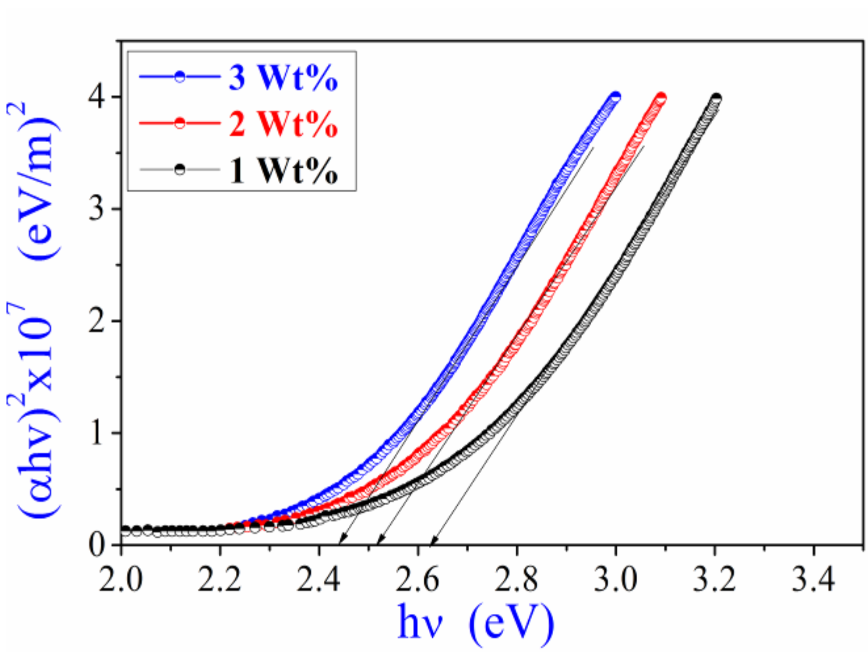

Fig. 3. $(\alpha hv)^2$ vs. photon energy *of* 1,4-dihydroxyanthraquinone at different weight percentages.

3. 3. Refractive index

Because it is directly tied to the electronic polarization of ions and the local field that exists inside materials, the refractive index (n) is one of the most basic features that an optical material may possess. Using the following equation [35], one can determine the refractive index of both the pure and dye-doped films based on the reflectance values of the samples that were studied.

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \tag{3}$$

$$n = \frac{(1 + R^{\frac{1}{2}})}{(1 - R^{\frac{1}{2}})} \tag{4}$$

where R is the amount of light that is reflected. Figure 4 illustrates how the refractive index (n) of clean and doped films varies with wavelength depending on the weight percentage of 1,4-dihydroxyanthraquinone dye used 1, 2 and 3 wt%. As can be seen from the figure, the refractive index falls with an increase in wavelength, and more rapid fluctuations in value may be seen when the wavelength is less than 300 nm.

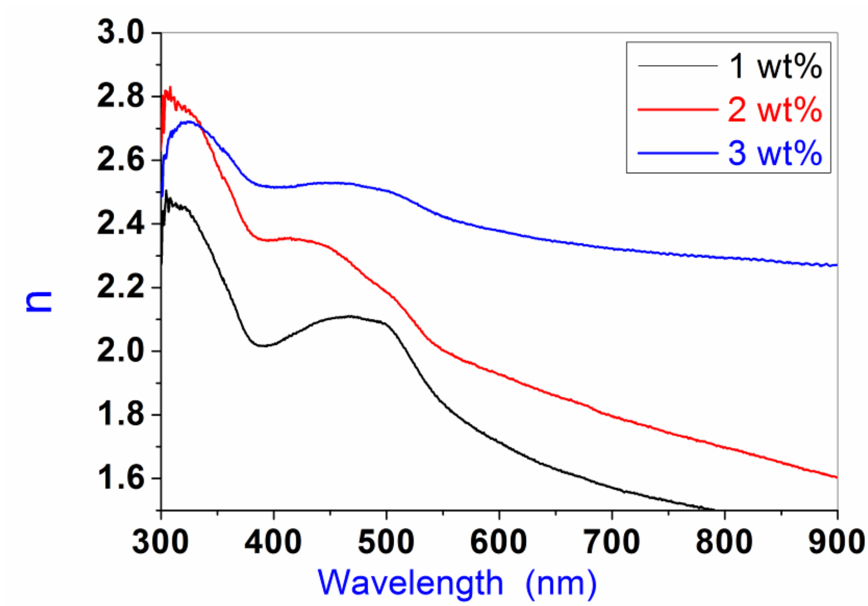

Fig. 4. The variation of the refractive index with wavelength at different weight percentages

3. 4. Optical dispersion parameters

According to the single oscillator model that Wemple and Di-Domenico suggested, the data on the dispersion of n were analyzed and assessed. They came to the conclusion that the following formula could, to a very good approximation, be used to explain all of the data [36].

$$n^2 = 1 + \frac{E_d E_o}{E_o^2 - (h\nu)^2} \tag{5}$$

where hν represents the energy of a photon, E_o represents the energy of an oscillator, and E_d represents the oscillator strength or dispersion energy, which is a measure of the intensity of inter-band optical transitions. The graphs in Figure 5 show the relationship between $1/(n^2 - 1)$ and $(h\nu)^2$ for doped PMMA films with dye at a variety of concentrations. Both E_o and E_d may be found by calculating the intercept of the y-axis, which is written as $E_\circ / E_d$, and the slope, which is written as $1/(E_\circ / E_d)$. The values of the static refractive index n and the static dielectric constant ε_∞ are shown in Table 1, together with the influence of single effective oscillator parameters on 1,4-Hydroxyanthraquinone. After determining the value of N based on equation 5, which states that $n_\circ^{\,2} = 1 + (E_d / E_\circ)$, the value of ε_∞ was computed using the formula $\varepsilon_\infty = n_\circ^{\,2}$, respectively.

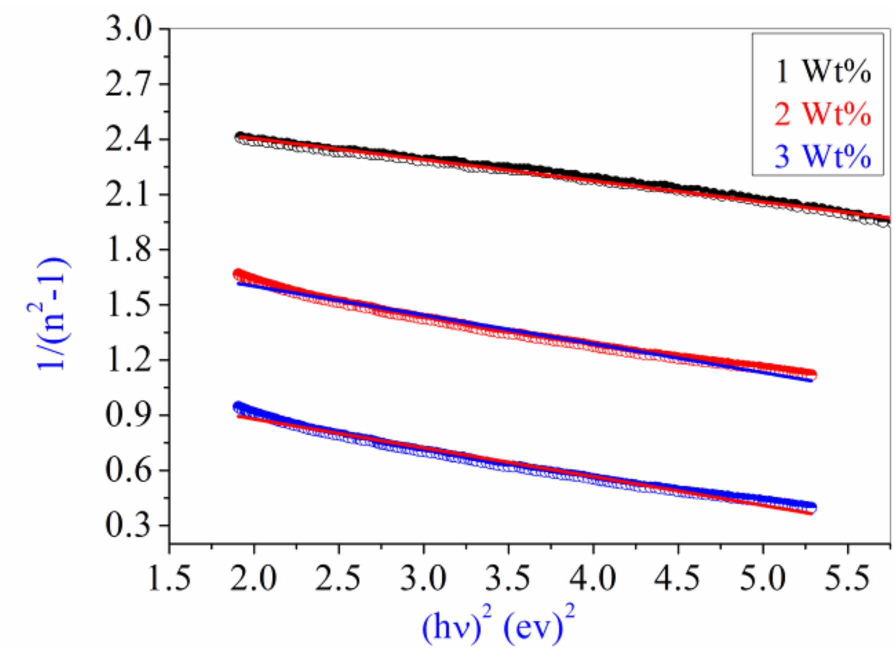

Fig. 5. $1/(n^2 - 1)$ against $(h\nu)^2$

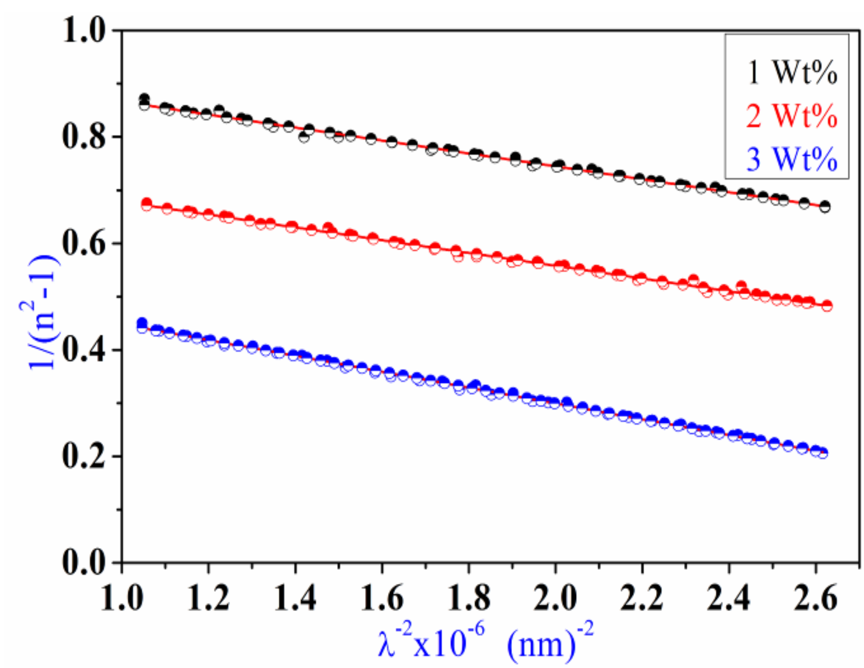

Fig. 6. $1/(n^2 - 1)$ against λ^{-2}.

The dispersion in refractive index can be also fitted by the following relation [37]:

$$n^2 - 1 = \frac{S_o \lambda_o^2}{1 - (\frac{\lambda_o}{\lambda})^2} \tag{6}$$

where λ is the wavelength of the incident lightis λ_o is the average oscillator strength and S_o, an average oscillator wavelength. Fig. 6 shows the curve of $1/(n^2 - 1)$ against $(1/\lambda^2)$. The value of S_o and λ_o were obtained from the slope $(1/S_o)$ and the infinite wavelength intercept $(1/S_o\lambda_o^2)$, the calculated values of S_o and λ_o are summarized in Table 1 indicating when increasing dye concentration, λ_o decreased while S_o increased and its values was found in order of 10^{13} m^{-2} for the films which is compatible with that results obtained by others [38] for a number of materials.

Table 1. Optical parameters of polymer film with different ratio of dye.

Dye wt %	E_o (eV)	E_d (eV)	$n_\circ$	ε_∞	$E_\circ/E_g$	$S_o \times 10^{13}$ (m^{-2})	$\lambda_\circ$ (nm)
1	2.41	3.15	1.52	2.31	0.919	6.07	162.24
2	2.52	3.52	1.55	2.40	1.003	4.33	233.54
3	2.61	3.78	1.56	2.45	1.069	3.23	159.58

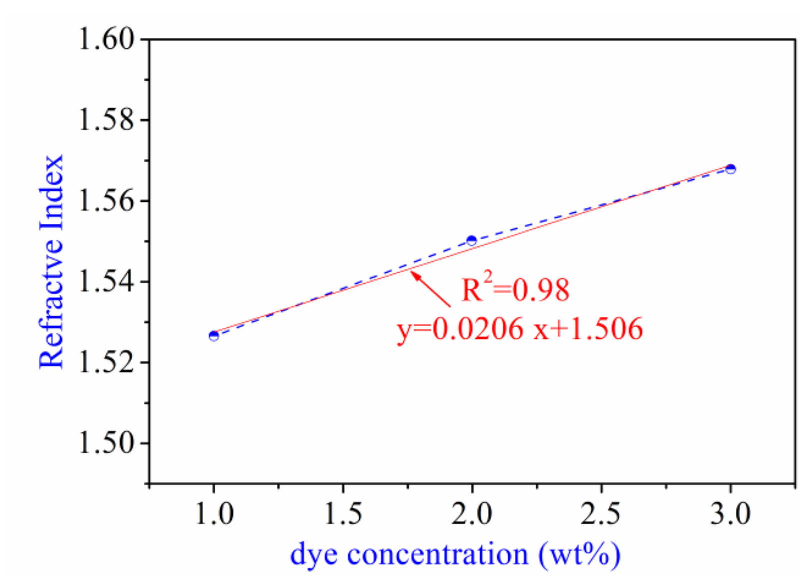

Fig. 7. Static $n_\circ$ as a function of volume fraction of 1,4-dihydroxyanthraquinone dye.

More recently, it has been reported that the linear connections between static refractive index, $n_\circ$ and the additive concentration imply that the additive particles are well disseminated throughout the host PMMA polymer [39, 40]. This was discovered by observing the linear relationships between and the additive concentration. Figure 7 illustrates how the static refractive index, $n_\circ$ of the polymer films changes with the volume fraction of dye powder used.

As can be observed, the static $n_\circ$ grew almost linearly as the dye concentration increased. Fig. 8 shows the regression value to be 0.98. The observed well-fitting regression of the data points shows that there was no appreciable aggregation and that the dye powder was dispersed uniformly throughout the PMMA host polymer. Other study groups of various well-dispersed polymer composite systems have shown the linear dependency of the refractive index of high wavelength with filler loading [41, 42].

4. CONCLUSION

Using a method known as solution casting, solid polymer composite films of PMMA are created. These films have varying weight percentages of 1,4-dihydroxyanthraquinone dye incorporated inside them. It has been discovered that the rise in 1,4-dihydroxyanthraquinone dye weight percent results in an increase in the optical quantities such as the absorption coefficient and the refractive index,. It has also been noticed that the energy of the optical band gap in PMMA diminishes with increasing 1,4-dihydroxyanthraquinone dye weight percentages.

Therefore, the incorporation of 1,4-dihydroxyanthraquinone dye may be of assistance in the process of engineering the band gap of PMMA, which may have applications in the field of electronics.

The analysis of the dispersion of the refractive index of the existing polymer composite films was carried out using the single oscillator model. There was shown to be a rise in the static refractive index.

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