

Study the Effect of Gamma Ray on the Optical Properties of Polyvinyle Pyrrolidone (PVP) Doped With Safranin O Thin Film Prepared By Spin Coating Method

Hassan K. Ibrahim

Abstract— The effect of gamma rays on the optical properties of polyvinyle pyrrolidone (PVP) doped with safranin O thin film prepared by spin coating method on glass substrate at room temperature was investigated. The thin film was exposed to Cs-137 gamma-radiation source at dose rate 0.56 Gy/min at room temperature for 5 months .The analysis of the absorption data revealed existence of indirect transition in the optical band gap. The optical constants such as absorbance, transmittion, reflection, refractive index ,extinction coefficient ,real and imaginary parts of dielectric constants , optical and electrical conductivity ,dispersion energy E_d and oscillation energy E_0 ,the third order optical nonlinear susceptibility $\chi^{(3)}$,relaxation time τ and the surface, volume energy loss function and electronic interband transition strength have been investigated at film thickness 10 μm over spectral of (300-800)nm

Index Terms— Gamma rays, polymer, energy gap, optical constant, real and imaginary parts of dielectric constant, nonlinear susceptibility $\chi^{(3)}$, relaxation time ,loss function.

I. INTRODUCTION

Irradiation of solids with high energy radiation, like gamma-rays,electrons or neutrons expected to effect their physical properties [1].Studies on the changes in optical properties shows the effect of radiation are strongly dependence on the structure of the absorbing substances [2].

The study of the optical absorption spectra in the solids provides essential information about the band structure and the energy gap in the crystalline and non-crystalline materials[1] .Analysis of the absorption spectra in the lower energy part gives information about atomic vibrations while the higher energy part of the spectrum gives knowledge about the electronic states in atom [3] .Refractive index is an important optical parameter for the design of prisms ,windows and optical fibres [4]. The refractive indices of optical materials are considerable importance for applications in integrated optic devices such as switches [5]

Semi conducting polymeric mixtures take large area in different applications such as heating ,prevention of static electricity accumulation such as preventing dust attraction on electronic device cabinets and electrodes and used for

electromagnetic interference shielding of electronic devices and prevention of static electricity hazards in the handling of electronic chips and explosives [6],[7].

Many applications require the materials in thin form microscopic defect, which control surface smoothens and coating homogeneity, contribute to the scattering of light and associated degradation of the optical response [8].Similarly, structural disturbance at the atomic level can alter the optical properties through modifications to the complex refractive index of the material [9].

In this study the effect of gamma rays on the optical properties of polyvinyle pyrrolidone (PVP) doped with safranin O thin film prepared by spin coating method on glass substrate at room temperature was investigated . The optical constants such as absorbance, transmittion, reflection, refractive index ,extinction coefficient ,real and imaginary parts of dielectric constants , optical and electrical conductivity ,dispersion energy E_d and oscillation energy E_0 ,the third order optical nonlinear susceptibility $\chi^{(3)}$,relaxation time t and the surface, volume energy loss function and electronic interband transition strength have been investigated at film thickness 10 μm over spectral of (300-800)nm.

II. THE EXPERIMENTAL PART

Figure (1:a) shows the structure of polyvinyle pyrrolidone (PVP) which has formula C_6H_9NO [10] .

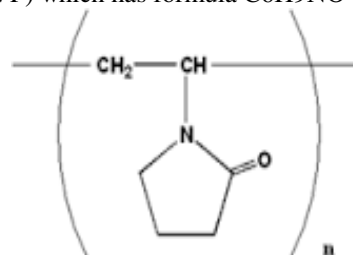


Figure (1:a):The expected chemical structure of polyvinyle pyrrolidone (PVP).

Figure (1:b) shows the expected structure of Safranin O that the molecular formula $C_{20}H_{19}N_4 Cl$ [11]:

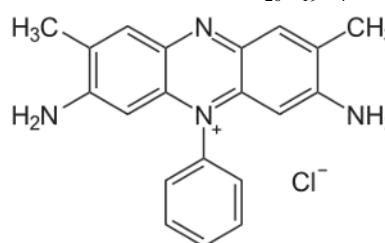


Figure (1:b):The expected chemical structure of safranin O

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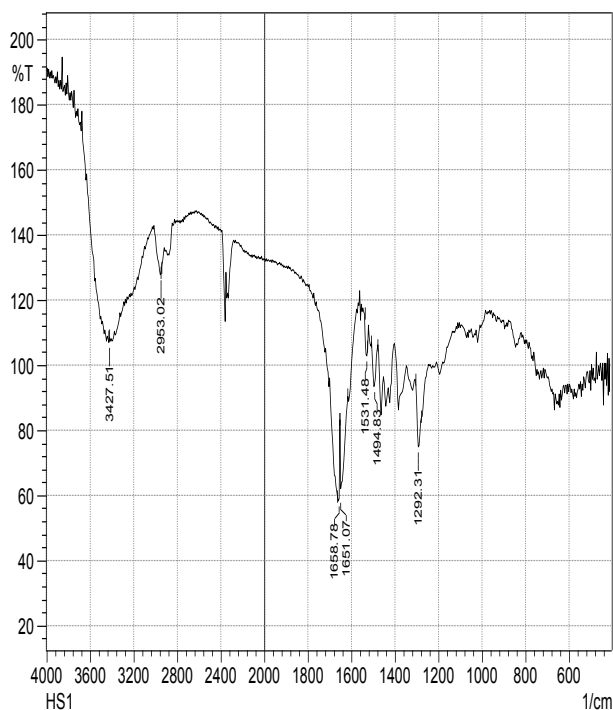
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1.5 M (0.315) gm of polymer (PVP) is first dissolved in Dimethyl Formamide (DMF) with stirred at room temperature for (2-4) h. 0.2 M (0.351) gm of safranin orange Azo dye dissolved in Dimethyl Formamide (DMF) with stirred at room temperature for 1h, then the dopant was added to the polymer with ratios 30%. The stirred solution was deposited on the substrate cited horizontally to get homogeneous thickness by spin coating method. The important thing before the deposition of films is the careful cleaning of the substrate which cleaned by immersing them in cleaning solution [10% $K_2Cr_2O_7 + H_2SO_4$] for 1h in ultrasonic path then washed with distilled water and organic solvent such as ethanol, acetone etc, then the substrate were dried before use. The film was irradiated with Cs-137 gamma source with an exposure rate 0.56 Gy/min for 5 months. A measurement of the spectral absorption (A) and transmittance (T) were recorded using UV-visible spectroscopy type CE 3055 in the wavelength (300-800)nm.

III. IR SPECTRA

The polyvinyl pyrrolidone (PVP) doped with safranin O dye thin film was identified by FT-IR spectroscopy in the range (4000-600) cm^{-1} as shown in the Fig(2).

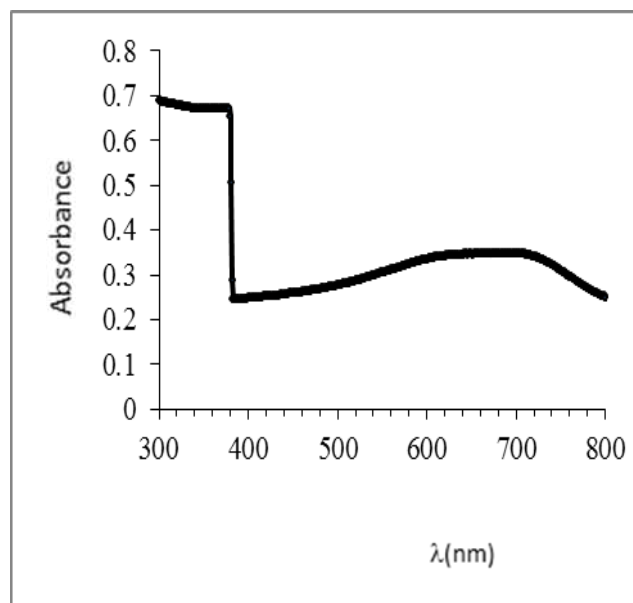


Fig(2): IR spectrum of polyvinyl pyrrolidone (PVP) doped with safranin O thin film

It is clear from the figure that O-H bending groups appeared at 1292 cm^{-1} . The band corresponding to $-N=N$ was recorded around 1494 cm^{-1} . The $C=C$ of the aromatic ring appeared around 1651 cm^{-1} . The $C=O$ gives band at 1658 cm^{-1} and C-H aliphatic gives band at 2953 cm^{-1} .

IV. RESULTS AND DISCUSSION

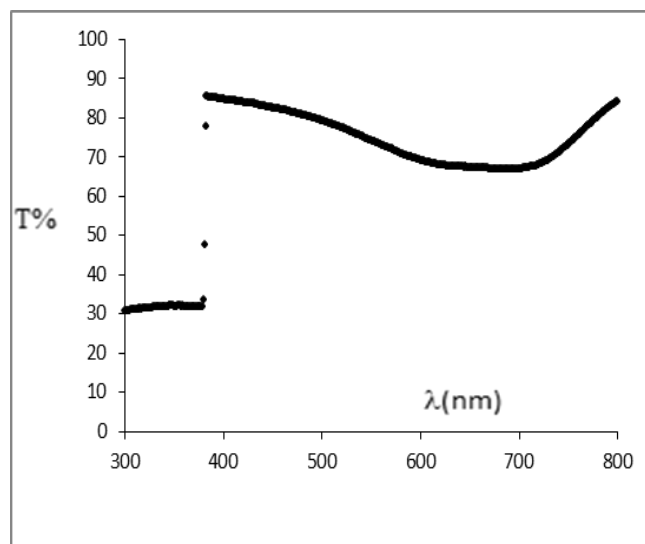
Fig(3) shows the relation between absorbance (A) of the film and wavelength. It is clear from the figure that the peak in the curve was appeared at wavelength ($\lambda=660\text{nm}$) which represents the transition $\pi \rightarrow \pi^*$.



Fig(3): Spectral absorption vis λ of polyvinyl pyrrolidone (PVP) doped with safranin O thin film.

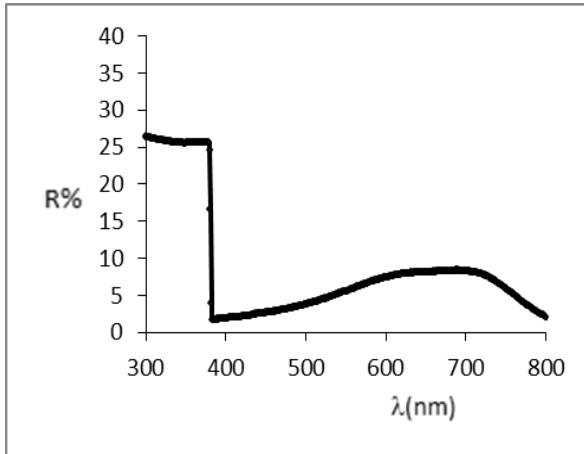
The transmittance spectrum in the UV-visible region showed that T% was decreased from 85% to 65% with increasing of λ from (400-700)nm. It is clear from the figure that is no light transmitted from the film in the wavelength between (590-725)nm, that mean all the light was absorbed by the polymer (PVP) doped with safranin O

At the longer wavelengths ($\lambda > 700\text{nm}$) the polymer became transparent and no light was scattered or absorbed (i.e $R+T=1$). The similar behaviour was observed for the polymer poly alpha naphthyle acrylate thin film [12].

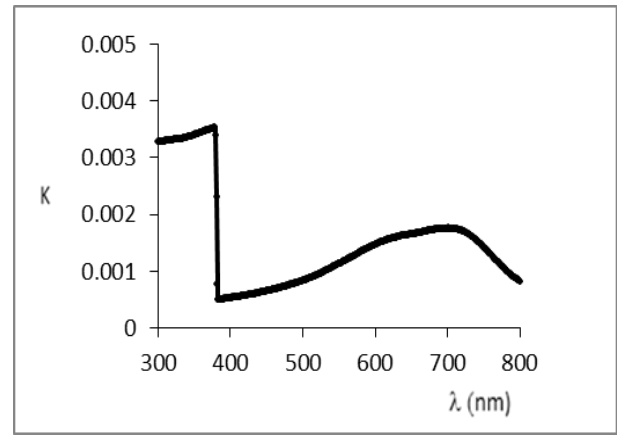


Fig(4): Spectral T% vis λ of polyvinyl pyrrolidone (PVP) doped with safranin O thin film

The reflectance of the film as a function of (λ) has been shown in the figure (5). It is clear from the figure that a dramatic increase of R% with increasing λ from (375-700) nm and the most reflectance occurs at wavelength around $\lambda=650\text{ nm}$ then R% was decreased as λ increased at the range (700-800)nm



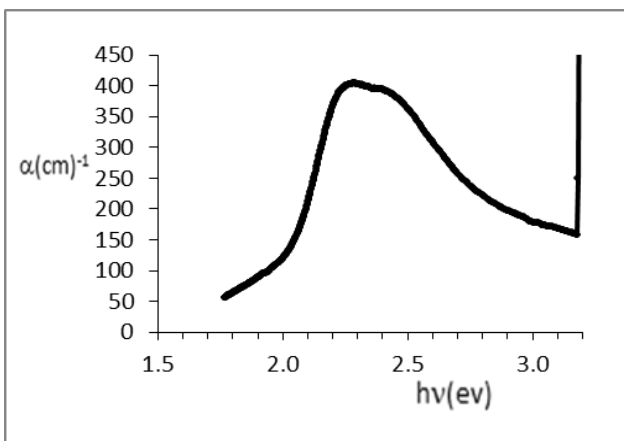
Fig(5):Spectral R% vis λ of polyvinyle pyrrolidone (PVP) doped with safranin O thin film.



Fig(7): The relationship between extinction coefficient and wavelength

The absorption coefficient (α) can be calculated from observed absorbance data using Beer Lamberts' formula ($\alpha=2.303A/d$) where A is the optical absorbance and d is the thickness of the film [13].

Fig(6) shows the relation between absorption coefficient (α) and photon energy one can observed from the figure that α was increased at low photon energy between (1.75-2.3) eV, moreover one can recognize a peak around 2.3 eV, followed by rapped decrease between(2.3-3.2) eV , then the curve was rocked up at high energies.

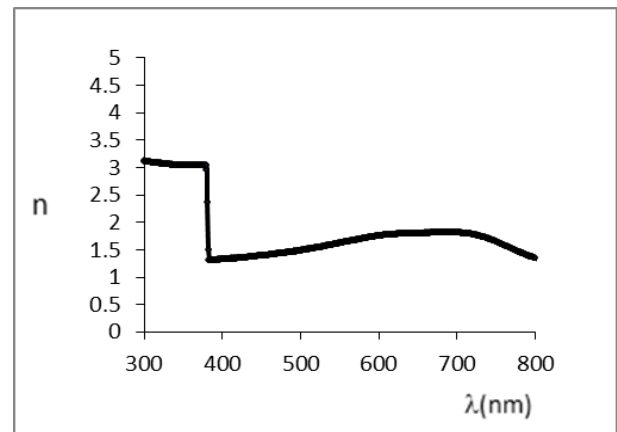


Fig(6): The relationship between absorption coefficient and wavelength

The extinction coefficient can be obtained from the relation ($k=\alpha\lambda/4\pi$), where λ is the wavelength of the light [14].The refractive index n can be described by the following relation [15]:

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

Figures 7 and 8 shows the dependence of refractive index ,n and the extinction coefficient k, on the wavelength , it is clear from the figures that the region (375-700)nm the values of n, and k, increases with increasing of wavelength while these values were decreased as wavelength increased in the region (710-800)nm, and the peak occurs around $\lambda=710$ nm.



Fig(8): The relationship between refractive index and wavelength

The absorption coefficient near fundamental absorption edge in both of crystalline and amorphous semiconductors is dependent on the photon energy . For direct transitions the absorption coefficient was taken on the following more general form as a function of photon energy [16].

$$\alpha h \nu = A (\alpha h \nu - E_g)^n \dots\dots\dots 2$$

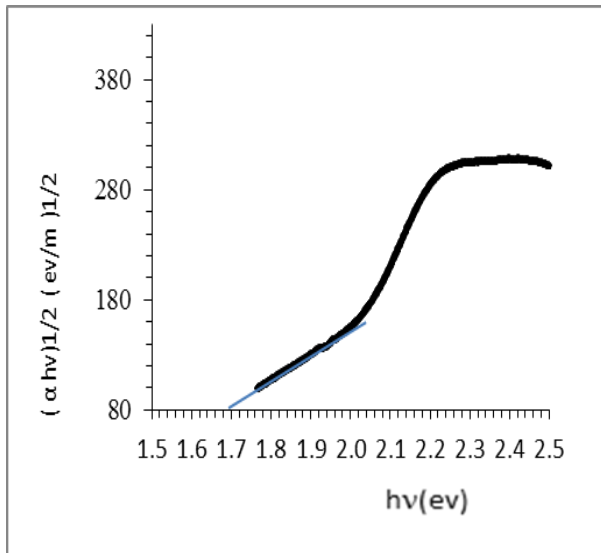
and for indirect transition

$$\alpha h \nu = B (\alpha h \nu - E_g)^n \dots\dots\dots 3$$

where ν is the frequency of the incident photon , n is the number which characterizes the optical processes .n has the value 1/2 for the direct allowed transition , 3/2 for a forbidden direct allowed transition and 2 for the indirect allowed transition , A and B are constants , and E_g is the optical energy gap. when the straight portion of the graph of $(\alpha h\nu)^n$ against $h\nu$ is extrapolated to $\alpha = 0$ the intercept gives the transition band gap [17] .

Fig (9) : shows the plot of $(\alpha h\nu)^{1/2}$ versus $h\nu$ (eV) for indirect transition. The indirect energy gap E_g was determined from tthis curve and given with value of $E_g= 1.90$ eV with phonon energy $E_p=1.68$ eV.

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Fig(9): The relation between $(\alpha hv)^{1/2}$ Versus $h\nu$

The real and imaginary parts of dielectric constants (ϵ_r and ϵ_i) was calculated by using equations [18],[19]:

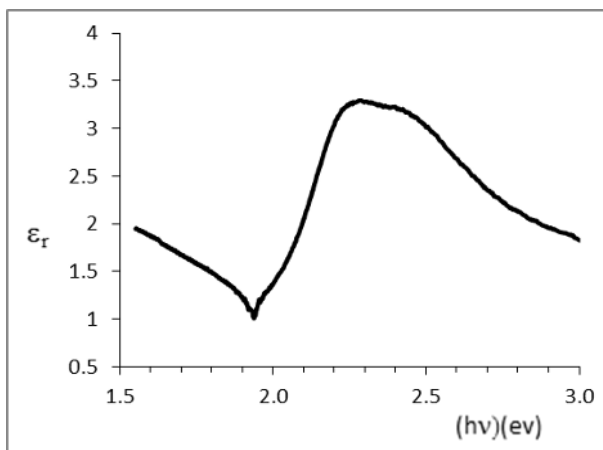
$$\epsilon_r = n^2 - k^2 \quad \dots\dots\dots 4$$

$$\epsilon_i = 2nk \quad \dots\dots\dots 5$$

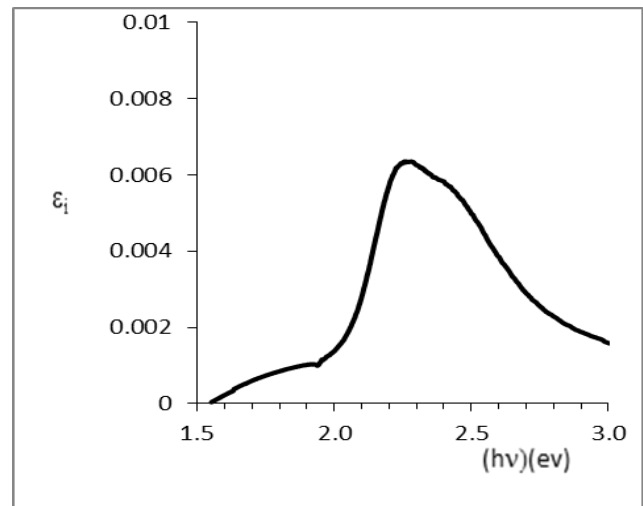
The dependences of ϵ_r and ϵ_i on the photon energy as shown in Figs. 10 and 11. Although The real and imaginary parts look like the same pattern but the values of the real part are higher than the imaginary part.

The relation between the dielectric constant and the photon energy indicates that some interactions between photons and electrons in the films are produced in this energy range. These interactions are observed on the shapes of the real and imaginary parts of the dielectric constant and they cause the formation of peaks in the dielectric spectra which depends on the material type. For example there was a high peak around $E=2.3$ eV for polyvinyl pyrrolidone (PVP) doped with safranin O thin film, but low peak observed for poly alpha naphthyle acrylate film. (as in 12).

The real and imaginary parts of dielectric constant were increased sharply in the high photon energy greater than 3.8 eV, similar behaviour was observed by others [20].



Fig(10): The relationship between real part of dielectric constant and photon energy

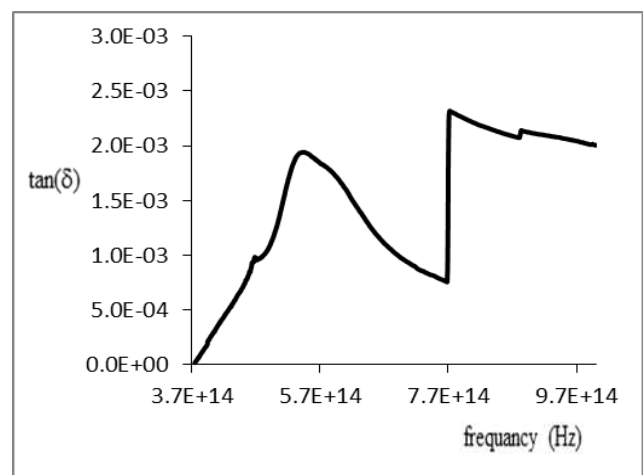


Fig(11): The relationship between imaginary part of dielectric constant and photon energy

The dissipation factor ($\tan \delta$) is a measure of loss-rate of power of a mechanical mode, such as an oscillation, in a dissipative system. For example, electric power is lost in all dielectric materials, usually in the form of heat. The dissipation factor $\tan \delta$ can be calculated according to the following equation (24) as in [21].

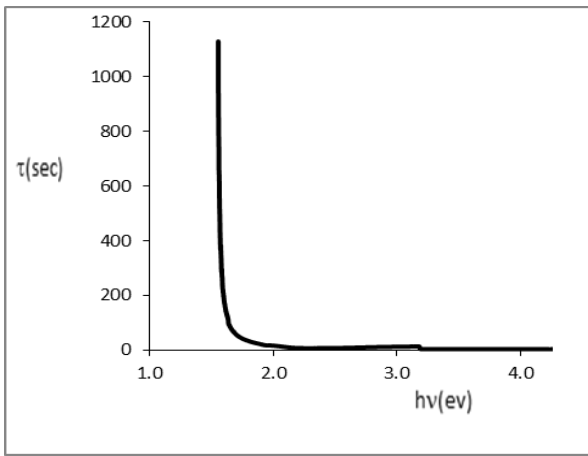
$$\tan \delta = \frac{\epsilon_i}{\epsilon_r}$$

The variation of dissipation factor of the investigated films with frequency is shown in Figure(12). It is found that the dissipation factor increases with increasing frequency (i.e. photon energy) in the absorption region, then there is fluctuation decrease in dissipation factor increasing of frequency

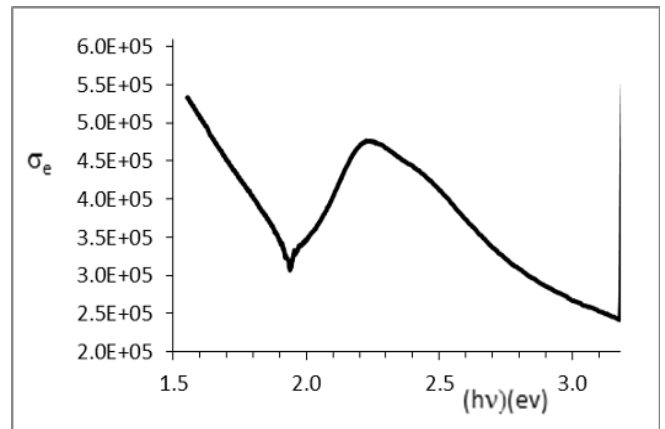


Fig(12): Dependence of dissipation factor $\tan \delta$ on the Frequency at room temperature

Fig(13) shows the dielectric relaxation time τ as a function of photon energy $h\nu$, it is clear from the figure there is a sharp decrease of τ with increasing of photon energy.



Fig(13): Dependence of dielectric relaxation time with photon energy



Fig(15): The relationship between optical conductivity σ_e and photon energy

V. OPTICAL AND ELECTRICAL CONDUCTIVITY

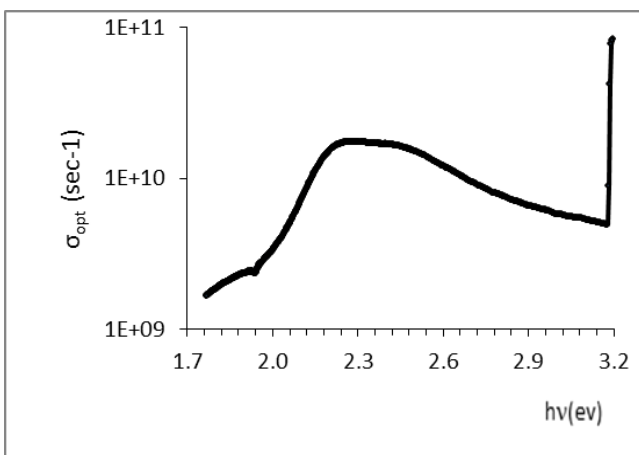
The optical and electrical conductivity σ_{opt} , σ_e were calculated depend on absorption coefficient α values as follow [22],[23]:

$$\sigma_{opt} = \frac{\alpha n c}{4\pi} \dots\dots\dots 7$$

$$\sigma_e = \frac{2\lambda\sigma_{opt}}{\alpha} \dots\dots\dots 8$$

where c is the velocity of light, α the absorption coefficient and n is a refractive index.

Figs.14 and 15 shows the variation of optical conductivity σ_{opt} and electrical conductivity, σ_e as a function of photon energy $h\nu$, one can conclude that the metal inclusion has greatly reduced the dielectric constant and increased the optical conductivity and the peak was recorded around $h\nu=2.3$ eV. The increased of optical conductivity at high photon energies is due to the high absorbance of sample thin films and also may be due to the electron excited by photon energy which also caused by hopping of the charge carriers between the localization state [24].

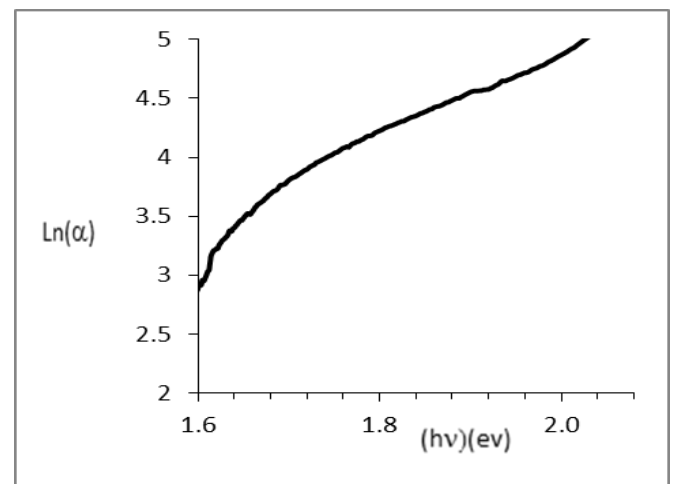


Fig(14): The relationship between optical conductivity σ_{opt} and photon energy.

Fig.(16) shows the variation of $\ln\alpha$ as a function of photon energy and the relation given by [25]:

$$\alpha = \alpha_0 \exp(h\nu/E_u) \dots\dots\dots 9$$

Where E_u is the Urbach energy, it can be evaluated by the reciprocal of the slope yields the magnitude of the (E_u), the value of E_u was found ($E_u = 280$ meV).



Fig(16): $\ln(\alpha)$ as A function photon energy ($h\nu$)

The relation between real part of dielectric constant ϵ_r and the square of wavelength λ^{-2} is given by[26]:

$$\epsilon_r = \epsilon_{\infty(1)} - B\lambda^2,$$

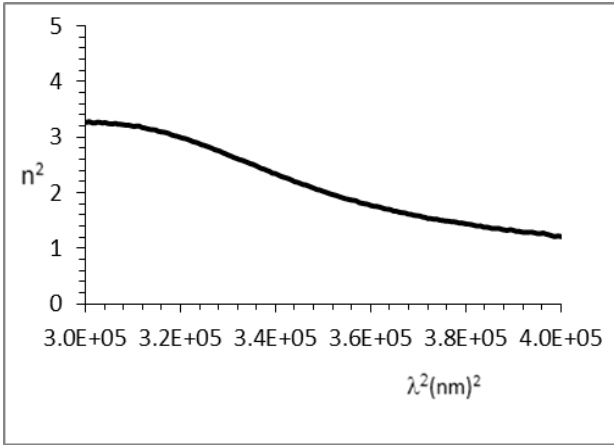
where

$$B = e^2 N / 4\pi^2 c^2 \epsilon_0 m \dots\dots\dots 10$$

where ϵ_{∞} the lattice dielectric constant or (the high frequency dielectric constant), N the free charge carrier concentration, ϵ_0 the permittivity of free space (8.854×10^{-12} F/m*), m^* the effective mass of the charge carrier and c the velocity of light. The real part of dielectric constants $\epsilon_r = n^2$ was calculated at different values of λ . Then, the obtained values of ϵ_r are plotted as a function of λ^2 as shown in fig(17) One can observe that the dependence of ϵ_r on λ^2 is linear at longer wavelengths. The extrapolating the linear part of this dependence to zero wavelength gives the value of $\epsilon_{\infty(1)}=2$ and from the slopes of these lines, values of N/m^* were calculated

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according to the Equation (10) of the constant B. The obtained value of N/m^* was $2.88 \times 10^{50} m^{-3} kg^{-1}$



Fig(17): plot of n^2 as a function of λ^2

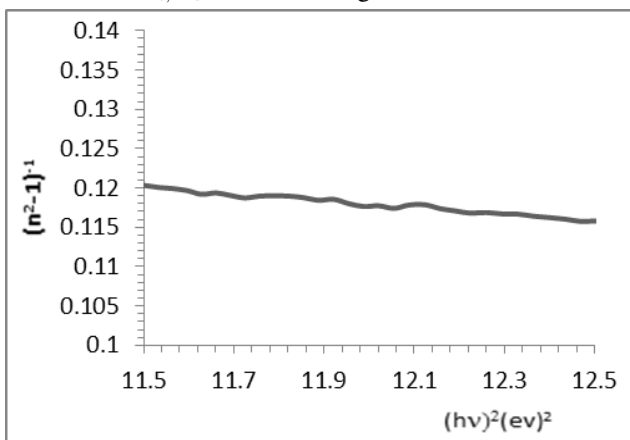
Based on the single-oscillator model, Wemple- DiDomenico [27],[28] is a semi-empirical dispersion relation to for determine the refractive index n at photon energy $h\nu$ can be written in the following relation:

$$n^2 = 1 + \frac{E_d E_0}{E_0^2 - (h\nu)^2} \dots\dots\dots 11$$

Where $(h\nu)$ is the photon energy, h is the blank constant, E_0 is the oscillator energy, and E_d is the oscillator strength or dispersion energy.. A plot of $(n^2-1)^{-1}$ vs. $(h\nu)^2$ was illustrated in Figure (18). The values of E_d and E_0 were obtained from the slope $(E_0 E_d)^{-1}$ and the intercept (E_0/E_d) with the vertical axis. Extrapolating the linear part towards long wavelengths, the point of interception with the ordinate at $(h\nu)^2=0$ yields the value of the dielectric constant at higher wavelength (ϵ_∞). The static refractive index (n_0) can be calculated using the relation $(n^2)_{(h\nu=0)} = E_d / E_0 + 1$.

The oscillator strength parameter, F , can be calculated using the following relation:
 $F = E_0 E_d \dots\dots\dots 12$

The values of E_0, E_d, F and ϵ_∞ are given in the table 1.



Fig(18): plot of $(n^2-1)^{-1}$ as a function of $(h\nu)^2$

The M_{-1} and M_{-3} moments of the optical spectra can be obtained from the relationship.

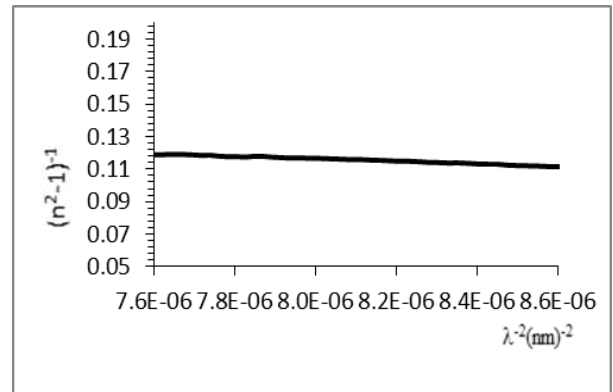
$$E_0^2 = \frac{M_{-1}}{M_{-3}}, E_d^2 = \frac{M_{-1}^3}{M_{-3}} \dots\dots\dots 13$$

The M_{-1} and M_{-3} moments were calculated using the above equations and are given in table 1.

The properties of the investigated PVP doped with safranin O thin film could be treated as a single oscillator at wave length λ_0 at high frequency. The high frequency dielectric constant can be calculated by applying the following simple classical dispersion relation[29]:

$$\frac{n_o^2 - 1}{n^2 - 1} = 1 - \left(\frac{\lambda_0}{\lambda}\right)^2 \dots\dots\dots 14$$

where n_0 is the refractive index at infinite wavelength λ_0 (λ_0 is the average interband oscillator wavelength), n the refractive index and λ the wavelength of incident photon. Plotting $(n^2-1)^{-1}$ against λ^{-2} which showed linear part, was below.



Fig(19): plot of $(n^2-1)^{-1}$ as a function of λ^{-2}

The intersection with $(n^2-1)^{-1}$ axis is $(n_0^2-1)^{-1}$ and hence n_0^2 at λ_0 equal to $\epsilon_\infty(2)$ (high frequency dielectric constant). The simple classical dispersion relation can be written as[30]:

$$n^2 - 1 = \frac{S_0 \lambda_o^2}{1 - \frac{\lambda^2}{\lambda_o^2}} \dots\dots\dots 15$$

where S_0 is the average oscillator strength which is equal to:

$$S_0 = \frac{n_0^2 - 1}{\lambda_0^2} \dots\dots\dots 16$$

The obtained values of S_0 and λ_0 are given in table(1).

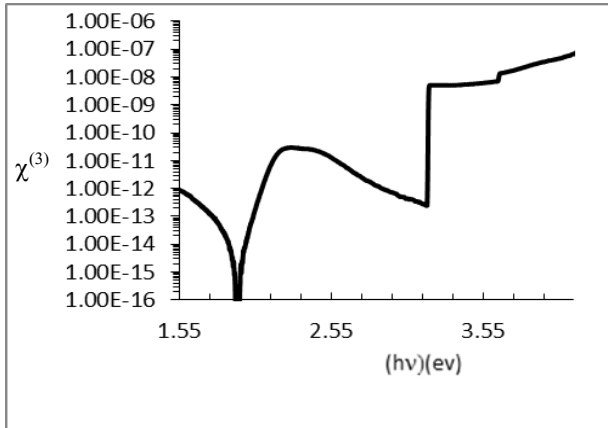
According to Wagner et.al.[31] the Miller rule is very convenient for visible and non linear infrared frequencies, which equalize the third order non polarizability parameter, $\chi^{(3)}$, the non linear optical susceptibility, and the linear optical susceptibility, $\chi^{(1)}$ through the equation:

$$\chi^{(3)} = A(\chi^{(1)})^4 = A[E_0 E_d / 4 \pi (E_0^2 - (h\nu)^2)^4] = A(n^2-1)^4 / (4\pi)^4 \dots\dots\dots 17$$

Where the constant $A = 1.7 \times 10^{-10}$.

The third order non linear optical susceptibility, $\chi^{(3)}$ of polyvinyl pyrrolidone (PVP) doped with safranin O was calculated from the equation (17) which is shown in the figure (20). One can observed from the figure that $\chi^{(3)}$ increases as the photon energy increase.

The estimated value of the third order non linear optical susceptibility, $\chi^{(3)}$ was calculated using equation(17) for $n_0=3.06$.The value of $\chi^{(3)}$ was 1.23×10^{-13} esu.



Fig(20): The third order optical non linear susceptibility as a function of photon energy.

Table (1): The optical parameters for polyvinyl pyrrolidone (PVP) doped with safranin O thin film.

| | | | |
|------------------------------|-----------------------|--------------------------------|------------------------|
| E_g (eV) | 1.90 | M_{-1} | 8.33 |
| E_{phonon} (eV) | 1.68 | M_{-3} (eV) ² | 0.28 |
| E_u (meV) | 280 | τ_0 (nm) | 252 |
| N/m^* (m ⁻³ Kg) | 2.88×10^{50} | S_0 (m) ⁻² | 1.32×10^{14} |
| E_0 (eV) | 5.4 | $\chi^{(3)}$ (esu) | 1.23×10^{-13} |
| E_d (eV) | 45 | $\epsilon_{\infty}(1) = n^2$ | 3.4 |
| F | 243 | $\epsilon_{\infty}(2) = n_0^2$ | 9.4 |
| n_0 | 3.06 | VELF | 9×10^{-4} |
| Re J_{cv} (eV) | 1.4×10^{-7} | SELF | 4×10^{-4} |
| Im J_{cv} (eV) | 8×10^{-5} | | |

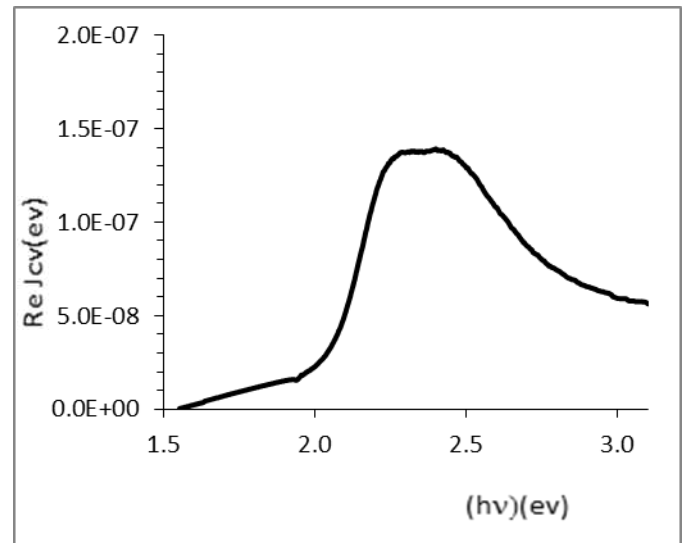
VI. INTERBAND TRANSITION STRENGTH

The complex interband transition strength can be calculated from the real and imaginary parts of the dielectric constant as the following relation [32]:

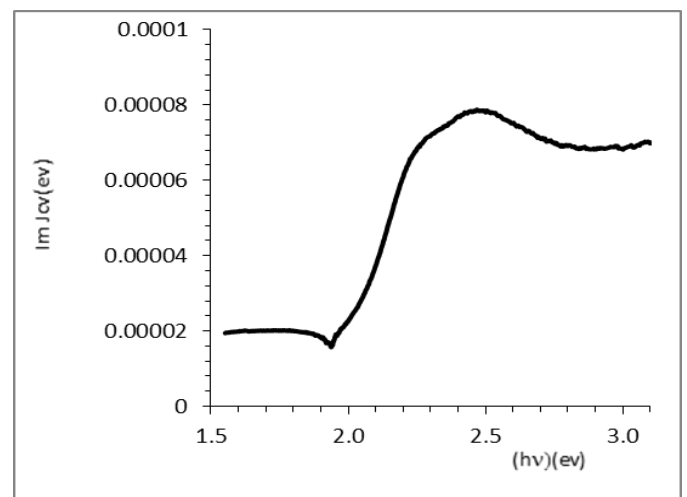
$$J_{cv} = J_{cv1} + J_{cv2} = \frac{m_0^2 4\pi^2}{e^2 h^2} \frac{(h\nu)^2}{2} (\epsilon_2 + i \epsilon_1)$$

where J_{cv1} , J_{cv2} are the real and imaginary part of interband transition strength, e is the charge of electron and m_0 is the mass of the electron ,the value of pre-factor ($m_0^2 4\pi^2/e^2 h^2$) is $8.289 \times 10^{-6} g cm^{-3} eV^{-2}$. It is clear from equation (17) that the real part of interband transition strength J_{cv1} is proportional to the imaginary part of the dielectric function . the imaginary part of interband transition strength J_{cv2} is proportional to the real part of the dielectric function. The figures 21 and 22 illustrate the relation between J_{cv1} , J_{cv2} and photon energy respectively, It is clear from the figures that is a dramatic increase in the interband transition strength J_{cv} with increasing of photon energy more than 4eV because the high absorption happens which increases the excitation of

the electrons , in addition J_{cv} has significant peak around the photon energy 2.3 eV. The values of J_{cv1} and J_{cv2} are given in the table 1.



Fig(21): The real part of interband transition strength as a function of photon energy.



Fig(22): The imaginary part of interband transition strength as a function of photon energy.

VII. SURFACE AND VOLUME ENERGY LOSS FUNCTION

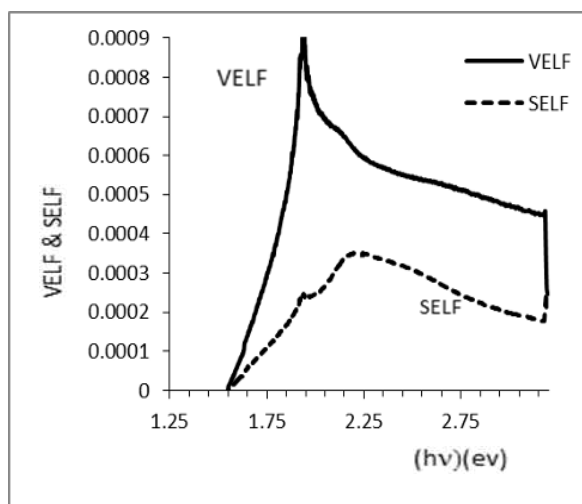
The surface and volume energy loss functions SELF,VELF respectively can be calculate depend on real and imaginary part of dielectric constant as following relations[33]-[34]:

$$\text{Surface} - \text{Im} \left(\frac{1}{1+\epsilon} \right) = \frac{\epsilon_2}{((\epsilon_1+1)^2 + \epsilon_2^2)}$$

$$\text{Volume} - \text{Im} \left(\frac{1}{\epsilon} \right) = \frac{\epsilon_2}{(\epsilon_1 + \epsilon_2)^2} \quad \dots\dots\dots 18$$

It is clear from the figure (23) that the peak of volume energy loss function is greater than the peak of surface energy loss function , it is also clear that the peak of SELF and VELF occurs at (1.95 eV) and (2.25eV) respectively. The values of SELF and VELF are given in the table 1.

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Fig(23): Surface and Volume energy loss functions as a function photon energy .

CONCLUSIONS

In the present investigation, the Optical properties of polyvinyl pyrrolidone (PVP) doped with safranin O thin film prepared via spin coating method on glass substrate at room temperature were calculated to obtain the optical constants such as absorbance, transmission, reflection, refractive index , long wavelength refractive index n_{∞} , extinction coefficient , real and imaginary parts of dielectric constants , optical and electrical conductivity , dispersion energy E_d and oscillation energy E_0 , the oscillator length strength s_0 , the optical moments M_1 and M_3 , the third order optical nonlinear susceptibility $\chi^{(3)}$, relaxation time τ and the surface, volume energy loss function and electronic interband transition strength.

The type of optical transition that responsible for optical absorption was indirect transition with energy gap $E_g=1.90$ eV and phonon energy $E_{\text{phonon}}=1.68$ eV. The dissipation factor $\tan \delta$ and optical conductivity σ_{opt} as well as electrical conductivity σ_e were increased with photon energy. , The third order optical nonlinear susceptibility $\chi^{(3)}$ has been calculated using Wagner model and the obtained value was 1.23×10^{-13} esu.

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