Study of the Thermal Neutrons& Γ-Irradiatation on the Optical Constants of Pm-355

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Abstract

The optical reflection, transmission,and UV/VIS- absorption spectra had been recorded in the wavelength (200-500nm) for PM-355 before and after irradiation with thermal neutrons by using (Am-Be) neutron source of flux ($5000n/cm^2$ .Sec) for a week, then γ-ray irradiation by using ^{60}Co-dose within range of (20-120Mrad) at normal conditions. The absorption spectra of irradiated samples showed radiation induced absorption changes by photodegradation.There was an increment in absorption proportional with irradiation dose.It is attributed to the interfaces traps, which are formed by thermal neutrons & γ-irradiation. It was found that the absorption and reflectance spectra were in opposite behavior of transmittance.The optical constants (α,K_{ex},n,ε,E_g,ΔE, and E_u) were calculated for all samples.The effect of irradiation on the optical constants of the PM-355 before and after irradiation had been discussed and investigated.

Keywords: optical constants, optical properties, PM-355, photodegradation.

Introduction

Solid polymers have been considerably studied in view of their wide potential application for novel systems and devices [1-4]. It is well known that molecular structure and the physical properties of polymers could be modified by ionizing radiations [5,6]. Ionization of atoms and scission of molecules occur leading to the formation of charged species both ionic and free radicals. Optical absorption studies are important to provide details of the electronic band structures, localized states and type of optical transitions [7-10].

Beer-Lamberts' law is fundamental for quantities absorption spectroscopic in the UV/VIS and constitutes a special case of the general law of absorption of radiation equation (1) in homogenous matter[11]:

$I=I_0e^{-\alpha x}$ .... (1)

Equation (1) implies that the decrease in light intensity only depends upon the intensity at depth (x) and on a constant (α) characteristic of the substance (absorption coefficient) [12].

The absorbancy is the logarithm of the ratio between the intensities of the light beam before entering and after leaving the mater, log10 [I_0/I]. Abbreviated E (for Excitation) or A.

Let the light incident on a sample be of intensity [I_0], and the emergent light [I]. Then the fraction transmitted [$T=I/I_0$] and the percentage transmittance [12]:

$T\%=100\ I/I_0$ ....(2)

The absorption coefficient α of material involved is:

$\alpha = 2.303A/x$..(3)

Where:

x = The sample thickness in cm

A = Defined by:

$A = \log(I_0 / I)$ ....(4)

The extinction coefficient (K) was calculated using the following equation [13]:

$K=\alpha\lambda/4\pi$..(5)

Analysis of optical absorption spectra could reveal the energy gap E_g between the Conduction Band (CB) and the Valence Band (VB) due to direct and indirect transitions of both crystalline and amorphous materials. The absorption edge coefficient α is a function of photon energy and obeys Mott and Davis's model (Mott and Davis, 1970):

$\alpha h\nu = B(h\nu - Eg)^r$ ....(6)

Where:

hν = The energy of the incidence photon

h = The Planck constant

E_g = The optical energy band gap

B = Constant known as the disorder parameter, which is nearly independent of the photon energy Parameter r is the power coefficient with the value that is determined by the type of possible electronic transitions, i.e., r = 1/2, 3/2, 2or 1/3 for direct allowed, direct forbidden, indirect allowed and indirect forbidden respectively [14,15].

The direct optical band gap can be evaluated from the linear plots of $(\alpha h\nu)^2$ versus $h\nu$. The extrapolations of the lines of $(\alpha h\nu)^{1/2}$ versus ($h\nu$), for which $(\alpha h\nu)^2 = 0$, give the direct optical band gap.

The reflectivity was determined from the values of transmittance (T) and absorbance (A) using the relation (R+A+T=1).

The reflectance can be expressed in terms of optical contents, (n) and K

$R= ((n-1)^2+ K^2)/ (n+1)^2+ K^2$ ....(7) or

$n=(((1+R)/(1-R))^2-(K^2+1))^{½}+ (1+R)/(1-R)$

When K <<n

$n=(1+R^{½})/(1-R^{½})$ ....(8)

The complex dielectric constant is real and imaginary part of the refractive index and given by the following equation [13]:

$\varepsilon= \varepsilon_1+ \varepsilon_2=(n+iK)$ ....(9)

where ε_1 and ε_2 are the real and the imaginary parts of ε, in which

$\varepsilon_1=n^2-K^2$ ....(10)

$\varepsilon_2=2n K$ ....(11)

The optical activation energy, ΔE, is the energy width of the tail of localized states in the band gap was valuated using the Urback-edges method given by the formula [15]:

$\alpha(\omega) = \alpha_0 \exp(\hbar\omega / \Delta E)$ ....(12)

Where:

α_0 = A constant

$\omega = 2\pi\nu$

The activation energy ΔE of irradiated samples were determined from the slope of the straight lines of $\ln(\alpha)$ versus photon energy $h\nu$[17].

Materials and Methods

We used commercially available PM-355 sheet($1*2cm^2$),and of thickness (0.05cm). It was irradiated to the thermal neutrons of flux ($5000n/cm^2$ .Sec) by using neutron source(Amercium-Berlium) for one week, and ^{60}Co γ-ray irradiation within range of (20-120Mrad) at normal condition (temperature (25°C), and relative humidity(55%)). These filmes were evaluated spectrophotometrically by using UV/160 Shimadzu spectrophotometer, which operates in the wavelength range of 200 nm to 1100 nm before and after irradiation with neutrons source and ^{60}Co γ-ray irradiation within range of (20-120Mrad).

Results and Discussion

The study of optical absorption spectra is one of the most productive methods in developing and understanding the structure and energy gap of the polymers.Figs.(1-3) show the absorption, reflectance, and transmittance spectra of (PM-355)before and after irradiation with thermal neutrons of flux ($5000n/cm^2$ .Sec) and γ-ray dose within range of (20-120Mrad).

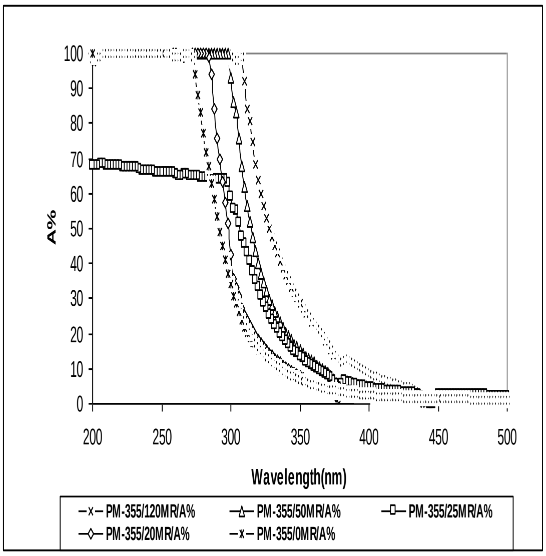

Fig.(1) Absorption spectra versus the wavelength for (PM-355) before and after irradiation with thermal neutrons of flux($5000n/cm^2$ .Sec)and γ-ray dose within range of(20-120Mrad).

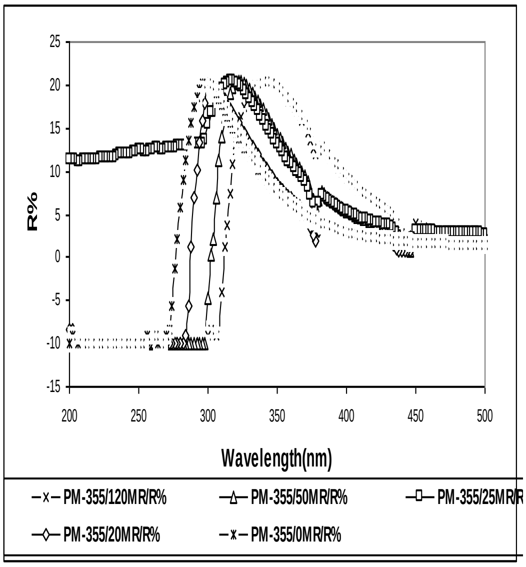

Fig.(2) Reflectance spectra versus the wavelength for (PM-355)before and after irradiation with thermal neutrons of flux($5000n/cm^2$ .Sec)and γ-ray dose within range of(20-120Mrad).

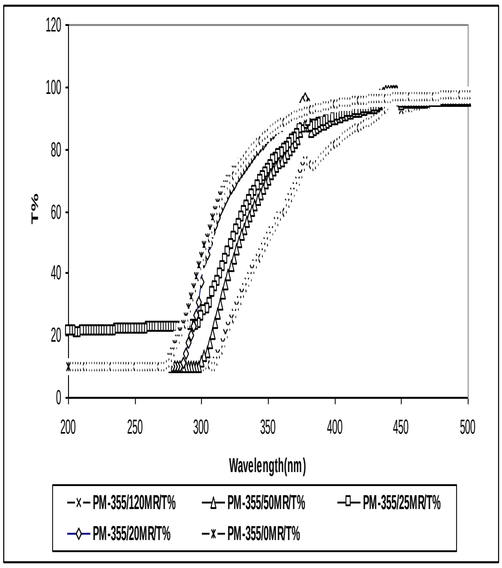

Fig.(3) The transmittance spectra of the (PM-355)before and after irradiation with thermal neutrons of flux(5000n/cm^2 .Sec) and γ-ray dose within range of(20-120Mrad).

It can be observed that the absorption peaks are of systemstic increasing with absorbed doses, and shift towerd the longe wavelengths.It was attributed to the ionizing radiation induces chemical reaction in polymers which result in changes in both molecular structure and macroscopic properties.Therefore,when PM-355samples were irradiated, degradation products are formed electron and hole take place causing changing in optical absorbance Fig.(1), which in a agreement with Khan,and Ahmad results [18]. It was seen that the absoption spectra of PM-355 before and after irradiation with thermal neutrons of flux (5000/cm^2) and γ-ray dose within range of (20-120Mrad) were in the UV-region, that's mean that all samples are transparent. Also the reflectance, and transmittance spectra were studied, it was obvious that the absorption and reflectance spectra were in opposite to that of transmitance Figs.(1-3).

The absorption coefficient of the samples involved was calculated using equation (3). Fig.(4) shows the absorption coefficient of the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad) Fig.(4) .

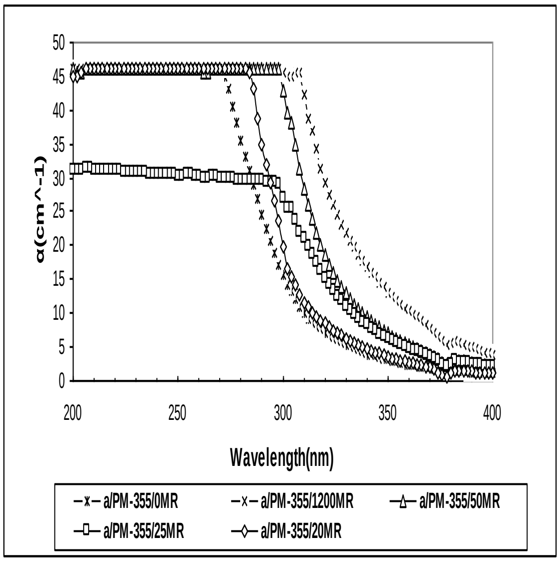

Fig.(4) The absorption coefficient versus the wavelength for the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

It was clear from the figure that absorption coefficient was increasing with irradiation dose. It was attributed to the photodegradation induced absorption changes, which was caused by increasing the localized state by radiation.The extrapolations of the lines of $(\alpha h\nu)^2$ versus $(h\nu)$, for which $(\alpha h\nu)^2 = 0$, give the direct optical band gap Fig.(5). It was found that the energy gap decreasing with the irradiation dose, which was attributed to increasing in localized state with irradiation dose Fig.(6).

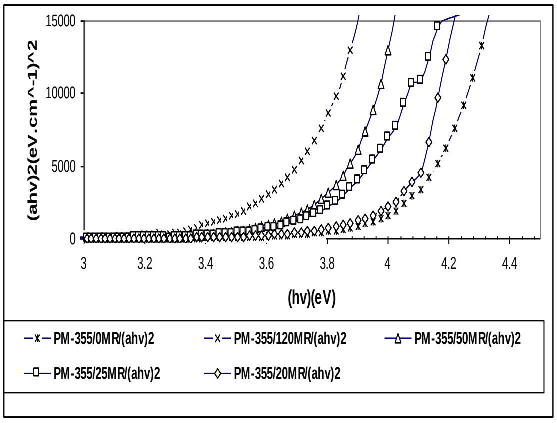

Fig.(5) Direct allowed transition $(\alpha h\nu)^2$ versus $(h\nu)$, for PM-355 before and after irradiation with thermal neutrons of flux(5000n/cm^2 .Sec)and γ-ray dose within range of (20-120Mrad).

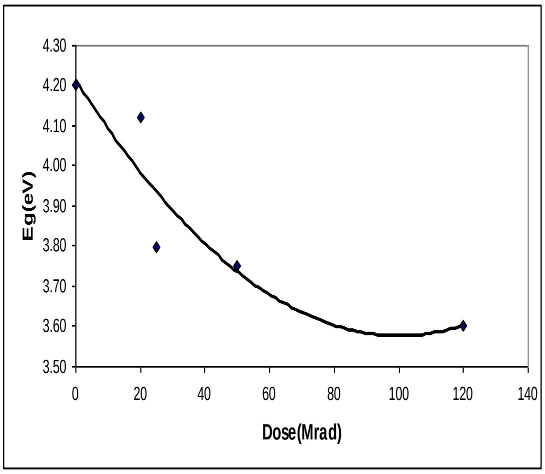

Fig.(6) The energy gap variation versus the irradiation dose for PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

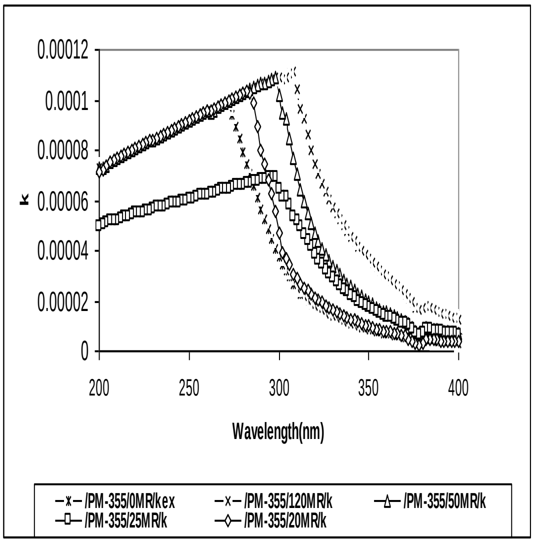

Fig.(7) The extinction coefficient versus wavelength for the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

The behavior of extinction coefficient (K) is nearly similar to the corresponding absorption coefficient as shown in Fig.(7).

The increasing in extinction coefficient with irradiation dose was due to increasing in absorption coefficient for the previous reasons,its peaks displaced towered long wavelength,that was attributed to decreasing in energy gap with irradiation dose, the absoption coefficient for PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad) were calcaculated at(λ_c =1240/Eg$_{)}$, and absorption coefficeint decreased with wavelength, so it was decreased with irradiation dose (λ_c longer). Figs.(8-9).

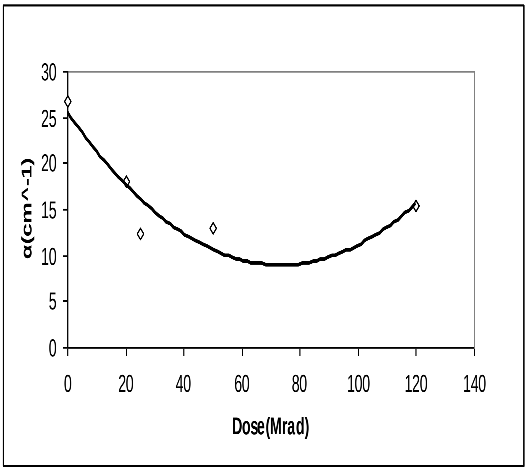

Fig.(8) The variation of α versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

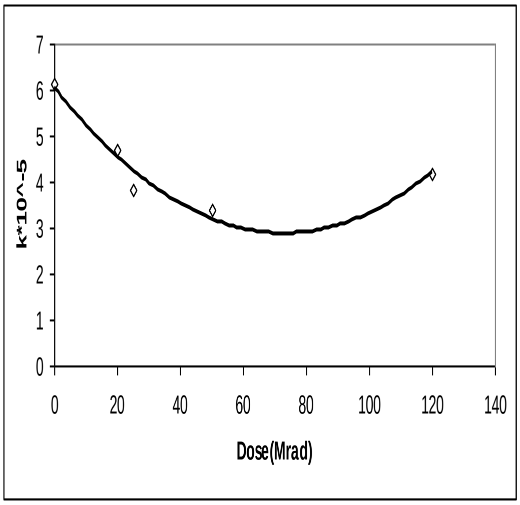

Fig.(9) The variation of K versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

Studying the refractive index will complete the fundamental study of the optical properties ,and optical behavior of the material. Fig.(10) shows the variation of the refractive index with wavelength for PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad). This figure shifts towerd the long wavelengths with increasing the irradiation dose,which is nearly similar to the corresponding refraction spectra (refractive index depends on reflectivity(eq.(8)). This behavior of the wavelength shif is due to decreasing in energy gap with irradiation dose,which causes increasing in localized states and increasing in refractive index Figs.(11-12).

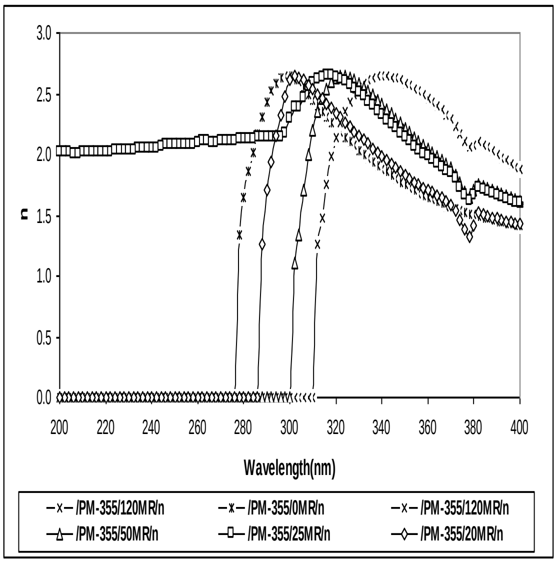

Fig.(10) The refractive index versus wavelength for the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2$.Sec) and γ-ray dose within range of (20-120Mrad).

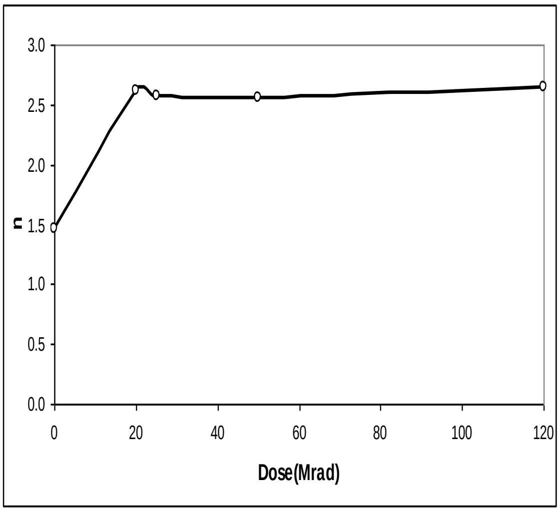

Fig.(11) The variation of refractive index versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2$.Sec) and γ-ray dose within range of (20-120Mrad).

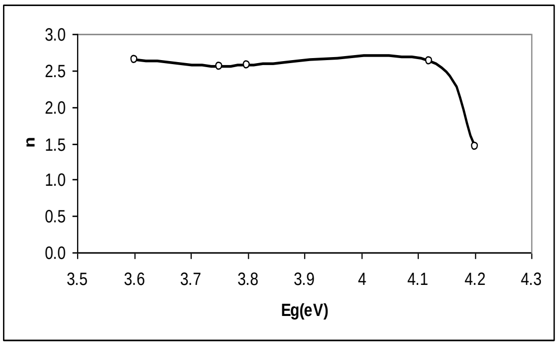

Fig.(12) The variation of refractive index versus energy gap the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2$.Sec)and γ-ray dose within range of (20-120Mrad).

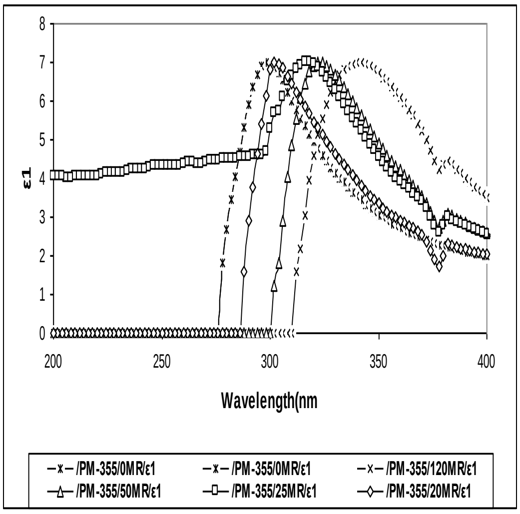

Fig.(13) The variation of real part of the dielectric constant versus the wavelength for the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2$.Sec) and γ-ray dose within range of (20-120Mrad).

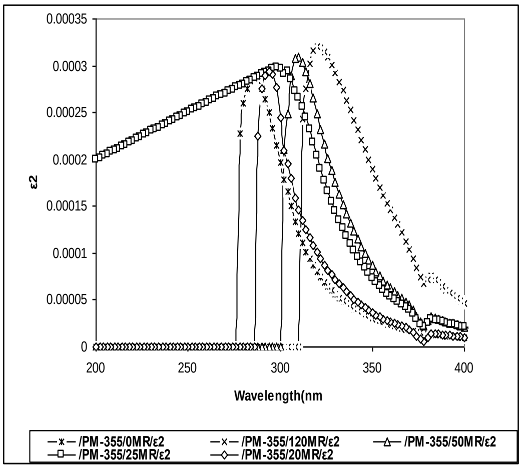

Fig.(14) The variation of imaginary part of the dielectric constant versus wavelength for the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2$.Sec) and γ-ray dose within range of (20-120Mrad).

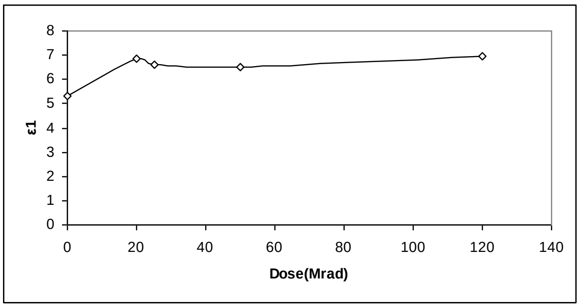

Fig.(15) The variation of real part of the dielectric constant versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

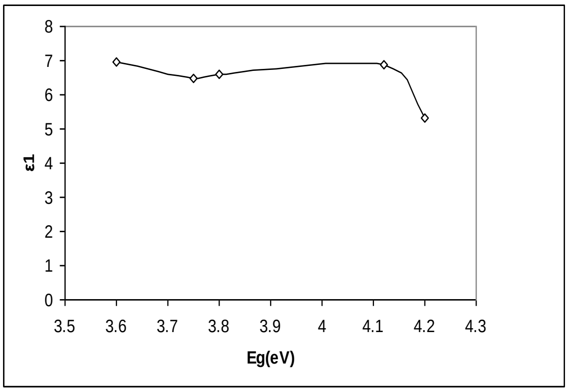

Fig.(16)The variation of real part of the dielectric constant versus energy gap for the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

Figs.(13-14) show the variation of the real and imaginary parts of the dielectric constant respectively for the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad). The behavior of ε_1 is simmilar to that of refractive index because of the smaller value of K^2 compared to n^2 Figs.(15-16) ,while ε_2. depends mainly on K's values, which is related with the variation of the absorption coefficient Figs.(17-18).

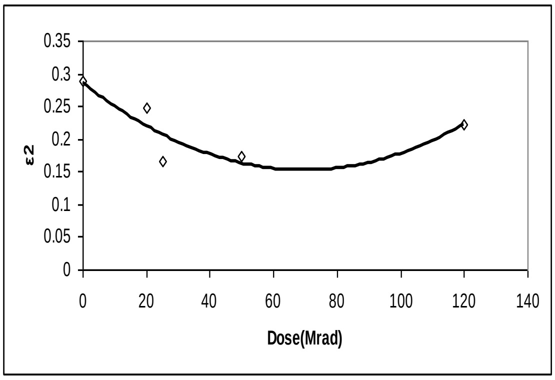

Fig.(17) The variation of imaginary part of the dielectric constant versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux (5000/cm^2) and γ-ray dose within range of (20-120Mrad).

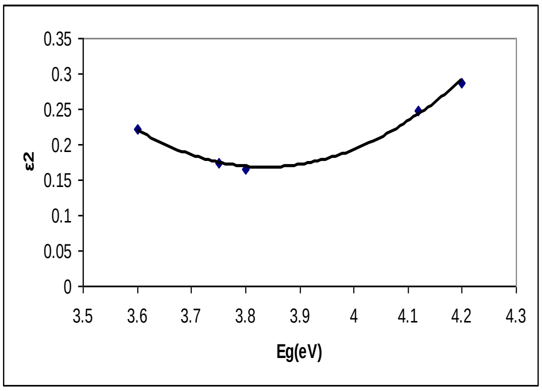

Fig.(18) The variation of imaginary part of the dielectric constant versus energy gap for the PM-355 before and after irradiation with thermal neutrons of flux (5000n/cm^2 .Sec) and γ-ray dose within range of (20-120Mrad).

The activation energy ΔE of irradiated samples were determined from the slope of the straight lines of ln (α) versus photon energy hν of Fig.(19).

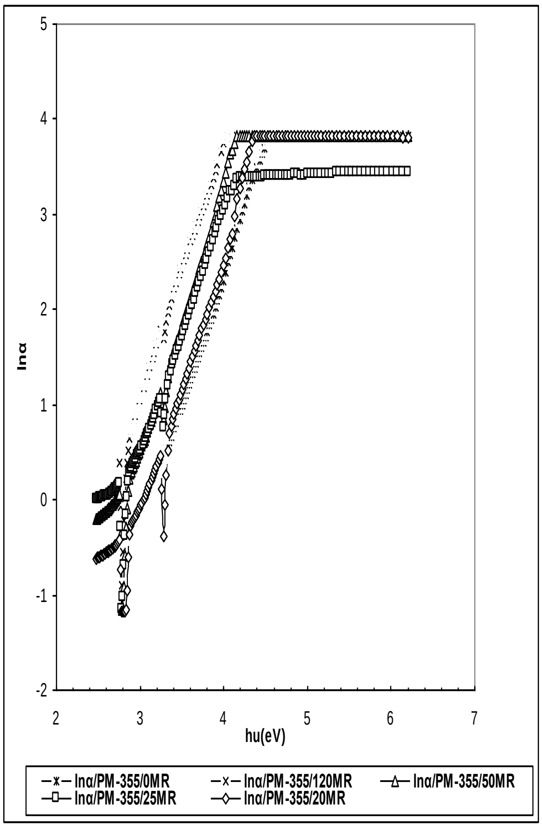

Fig.(19) The variation of of ln (α) versus photon energy $h\nu$ the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2.Sec$) and γ-ray dose within range of (20-120Mrad).

The results of ΔE values for different doses are shown in Fig.(20). It can be seen that ΔE decreases with the increase of the radiation dose.

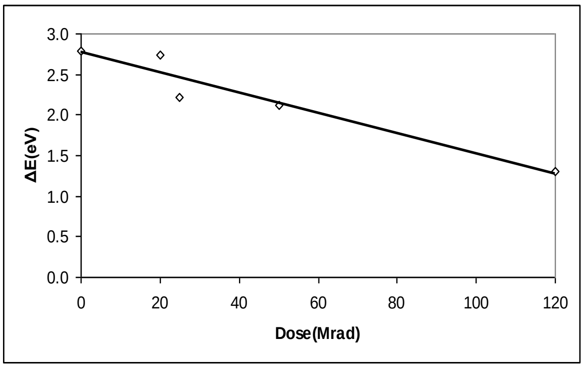

Fig.(20) The variation of ΔE versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2.Sec$) and γ-ray dose within range of (20-120Mrad).

The density of localized states in the band can be evaluated from the Urback energy (E_u) at ($\alpha<10^4cm^{-1}$), which refered to there is absorption tails at energies smaller than optical energy gap.The reciprocal of the slop gives the value (E_u) Fig.(21). It was found that the Urback energy (E_u) increasing with the irradiation dose, which is atributed to the interfaces states were induced by irradiation [19-20]. The calculated optical constant of the PM-355 before and after irradiation with thermal neutron source and γ-ray within range of (20-120Mrad),which were listed in Table (1).

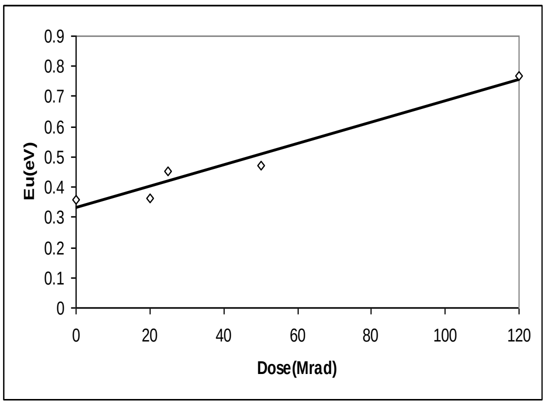

Fig.(21) The variation of (E_u) versus irradiation dose the PM-355 before and after irradiation with thermal neutrons of flux ($5000n/cm^2.Sec$) and γ-ray dose within range of (20-120Mrad).

Table(1)
The optical constants of the PM-355 before and after irradiation with thermal neutron source of flux ($5000/cm^{-1}$) and gamma ray with the range of (20-120Mrad).

Dose (Mrad)	E_g (eV)	λ_c (nm)	α (cm^{-1})	$K_{X10^{-5}}$
0	4.2	288.1	26.8	6.14
20	4.12	301	18	4.7
25	3.8	326.32	12.44	3.83
50	3.75	330.67	12.9	3.39
120	3.6	344.44	15.31	4.19

Dose (Mrad)	n	ε_1	ε_2 $_{X10^{-3}}$	ΔE (eV)	E_u (eV)
0	1.46	5.33	0.288	2.79	0.36
20	2.62	6.88	0.247	2.74	0.37
25	2.57	6.6	0.166	2.22	0.45
50	2.56	6.5	0.175	2.12	0.47
120	2.64	6.95	0.221	1.3	0.77

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الخلاصة

سجلت أطياف الامتصاص والنفاذية وحسبت أطياف الانعكاسية للنماذج قبل وبعد التشعيع بالنيوترونات الحرارية.

ولمدة اسبوع وبأستخدام مصدر تشعيعها بأشعة كاما باستخدام (كوبلت- 60) وبجرعة تقع ضمن المدى (20-120 ميكاراد) ضمن الظروف الاعتيادية. أظهرت أطياف الامتصاص تغيرا محتثا بالتشعيع.ان الزيادة بالامتصاصية تتناسب طرديا مع الجرعة الاشعاعية . وقد اعزي ذلك الى المصائد البينية الناجمة عن التشعيع بالنيوترونات الحرارية واشعة كاما. أن أطياف الامتصاصية والانعاكسية ذات سلوك معاكس لسلوك أطياف النفاذية. حسبت وفسر تأثير التشعيع على الثوابت البصرية لقبل وبعد التشعيع ($\alpha, K_{ex}, n, \varepsilon, E_g, \Delta E$, and E_u .PM-355)، التفكك الضوئي (PM-355 الكلمات المفتاح:الثوابت البصرية، الخواص البصرية.