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Investigation of Optical and Electrical Properties of DAM-ADC Nuclear Track Detector Induced by Gamma Irradiation

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Abstract: Effect of gamma irradiation on the optical and electrical properties of Diallyl maleate- allyl diglycol carbonate (DAM-ADC) polymer was investigated. Samples of DAM-ADC were irradiated with gamma doses in the range of 100-500 kGy. The optical characterization of samples have been studied through the measurements of UV-visible absorption spectra. The refractive index and the absorption index were computed using the obtained data of transmittance and reflectance. The complex dielectric constant, dissipation factor, volume energy loss, surface energy loss functions, and optical conductivity were calculated and interpreted. The optical absorption data showed that dispersion energy, high frequency dielectric constant, and optical conductivity increase with increasing gamma while the optical energy band gap decreases. The results reflects that DAM-ADC polymer is a good candidate in optoelectronic devices based on its conductivity and dispersion parameters. In addition, it can be used as gamma radiation dosimetry in some specific range of doses.

Keywords: DAM-ADC, UV-Visible, Volume Energy Loss, Dissipation Factor, Optical Conductivity

1. Introduction

During the last few decades, the importance of polymer materials has increased because of their low cost and weight, easy processability, high quality surfaces, easy fabrication of thick and thin samples. Polymers based on solid state nuclear track detectors are being widely used in many applications: alpha and neutron dosimetry, cosmic ray studies and astrophysics, porosity and microfilters, uranium prospection and radon emanation measurements, radiobiology and nuclear medicine measurements made on meteorites, alpha particles energy resolution, low linear energy transfer radiation dosimetry, radiological protection and monitoring, and optoelectronic devices [1–9].

Diallyl maleate (DAM), allyl diglycol carbonate (ADC), and mixtures of them (15% DAM–85% ADC) were cast in to polymer plates under three kinds of polymerizing conditions. The copolymer plates of DAM and ADC showed intermediate characteristics which are suitable for control of the sensitivity as nuclear track detector [10–13]. DAM-ADC polymer is characterized as transparent material has high compact strength. This leads it an advantageous material for several applications such as microelectronics and biosensor production technologies [9].

Numerous efforts have recently been made to investigate the influence of gamma radiation on polymers, in order to find out the suitability of using polymers as gamma radiation dosimeters [14-17]. Interaction of radiation with polymers leads to chain aggregation, chain scission, formation of double bonds and molecular emission. As a consequence of this, the physical and chemical properties such as: optical, electrical, mechanical, chemical and track properties of the polymer are modified [18-22]. Mainly the effectiveness of these changes depends upon the structure of the polymer and the conditions of irradiation with gamma source like energy and fluence. The study of these changes may enhance their applications in different fields, like the evolution of high radiation doses [23].

The main objective of this study is to investigate the effect of gamma radiation on optical and electrical properties of DAM-ADC polymer. The absorption spectra are measured in the wavelength range 190–1100 nm. The optical constants, refractive index *n* and the absorption coefficient *k* over the spectral range are determined from the transmittance $T(\lambda)$ and reflectance $R(\lambda)$ data. The complex dielectric constant $\hat{\varepsilon}$, the volume energy loss (VELF) and surface energy loss (SELF) functions are computed. The dissipation factor $\tan\delta$ and optical conductivity σ_{opt}. characteristics are calculated. In addition, the optical energy gaps E^{opt}. of the pristine and gamma irradiated samples are also estimated.

2. Experimental Work

2.1. Materials

The co-polymer of Diallyl maleate and allyl diglycol carbonate (DAM-ADC) detector which used in the present study has been manufactured by Yamamoto Kogaku Co., Ltd., Japan. It has a chemical composition $C_{22}H_{30}O_{11}$ with thickness about 1000 μm and density 1.2 g/cm^3. The main constituents of the detector plate are 15: 85% from DAM and ADC, and a small amount of di-isopropyl peroxy dicarbonate (IPP) was used as a polymerizing initiator. Samples of DAM-ADC detector with area of 1.0 cm x 1.0 cm were carefully cut with laser beam.

2.2. Sample Irradiation

DAM-ADC samples were irradiated at room temperature using 1.25 MeV ^{60}Co gamma source of dose rate 4 kGy/h. Samples were irradiated with doses in the range from 100 kGy up to 500 kGy. The irradiation source is available in Egyptian Atomic Energy Authority (EAEA), Cairo, Egypt.

3. Results and Discussion

3.1. UV-Vis Spectroscopy

UV–Vis measurements of pristine and irradiated DAM-ADC samples were performed with Jasco V-576 (Japan) model double-beam spectrophotometer where its wavelength range is from 190 to 1100 nm, keeping air as a reference.

The nature changes in the UV–Vis absorption spectra of pristine and gamma irradiated DAM-ADC samples are shown in Figure 1. It is clear that a shift of absorption edge towards longer wavelength with increasing gamma absorbed dose can be readily observed. The absorption peak with increasing gamma dose is seen to change into a broad one. This behavior is generally interpreted as produced by the formation of extended systems of conjugate bonds, i.e. possible formation of carbon clusters. The absorption bands in the investigated range of wavelength are associated to the π-π^* electronic transitions. This type of transitions occurs in the unsaturated centers of the molecules, i.e. in compounds containing double or triple bonds and also in aromatics. The excitation of π electron requires smaller energy and hence, transition of this type occurs at longer wavelengths [24-26].

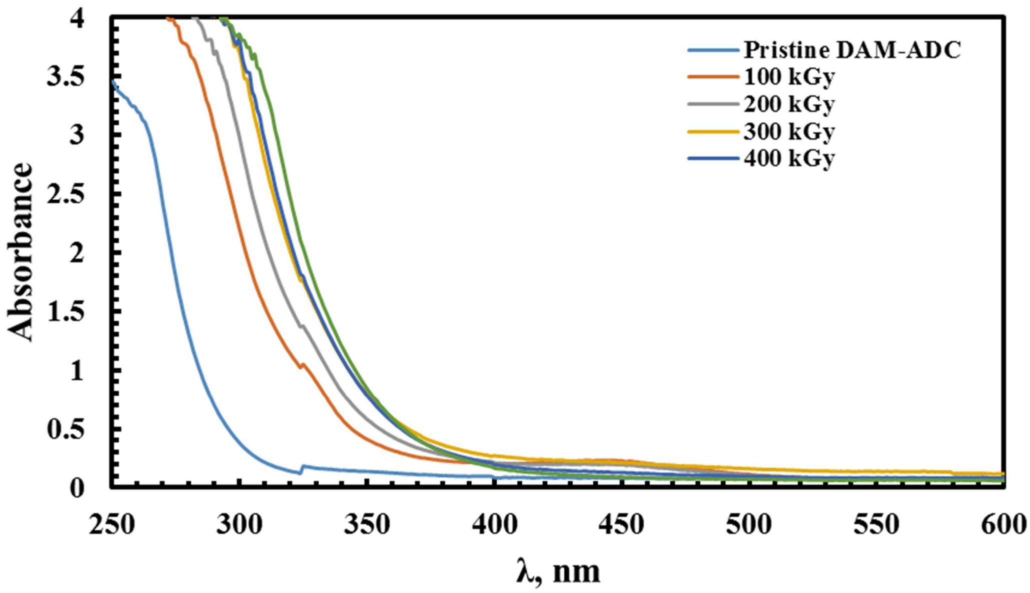

Figure 1. *UV–visible absorption spectra of the pristine and gamma-irradiated DAM-ADC polymer.*

Figure 2 shows the spectral distribution of transmittance $T(\lambda)$ and reflectance $R(\lambda)$ for pristine and gamma irradiated DAM-ADC samples. The results clearly show that the gamma irradiation shifts the transmission edge towards higher wavelengths indicating a decrease in the optical energy gap value. It is also observed that the intensity of transmittance peaks within the absorption region increases by irradiation. This is attributed to the presence of intraband transitions at localized states in the energy gap.

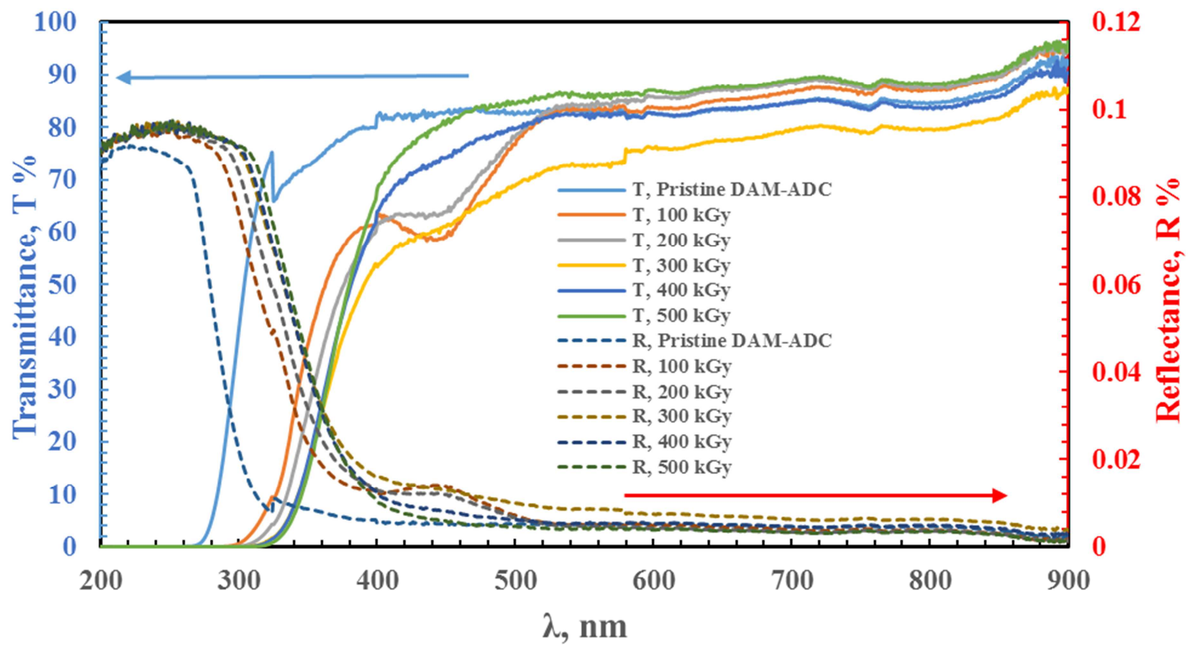

Figure 2. *Spectral distribution of transmittance T(λ) and reflectance R(λ) for the pristine and gamma-irradiated DAM-ADC polymer.*

3.2. Refractive Index *n* and Absorption Index *k*

The refractive index n and absorption index k are computed by using the obtained data from transmittance and reflectance data. The normal reflectance for any absorbing material can be given by the following equation [27]:

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

The refractive index computes directly from Eq. (1) [28]:

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

The values of the absorption index k can be calculated from the absorption coefficient α using the relation $k = \alpha\lambda / 4\pi$ [29], where $\alpha = 2.303(A/t)$, A is the measured absorbance and t the thickness of the sample. Dependence of the absorption index k and refractive index n on the wavelength for the pristine and gamma-irradiated DAM-ADC polymer are plotted in Figure 3 and Figure 4, respectively. The obtained values of both n and k show that these parameters are dependent on irradiated gamma dose.

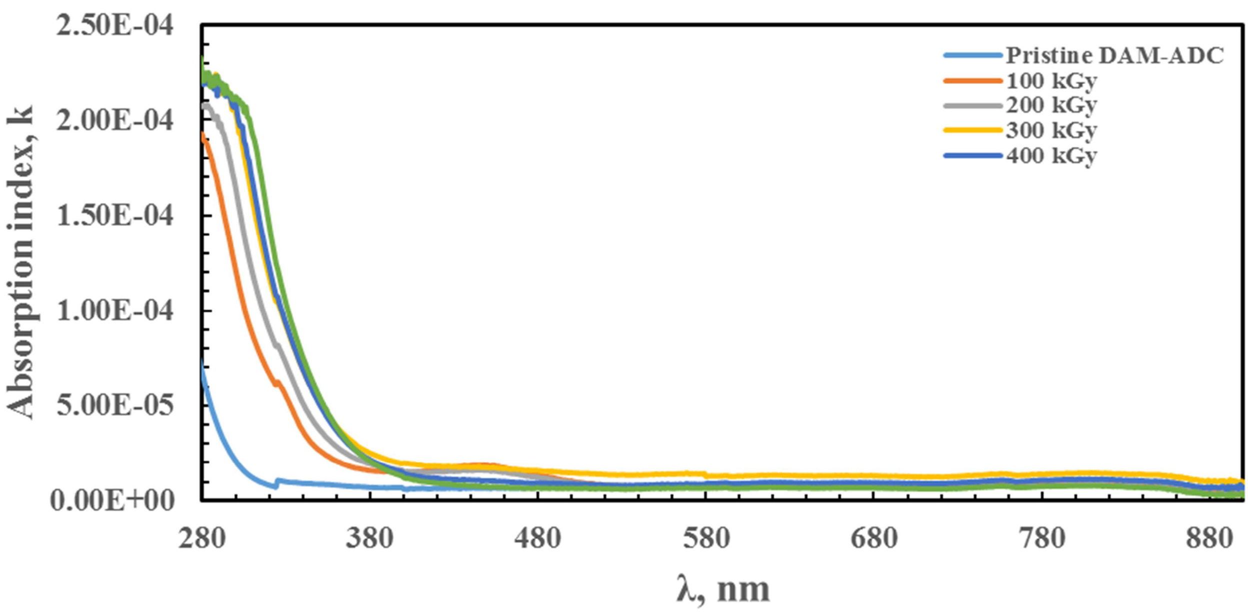

Figure 3. *Dependence of the absorption index k on the wavelength for the pristine and gamma-irradiated DAM-ADC polymer.*

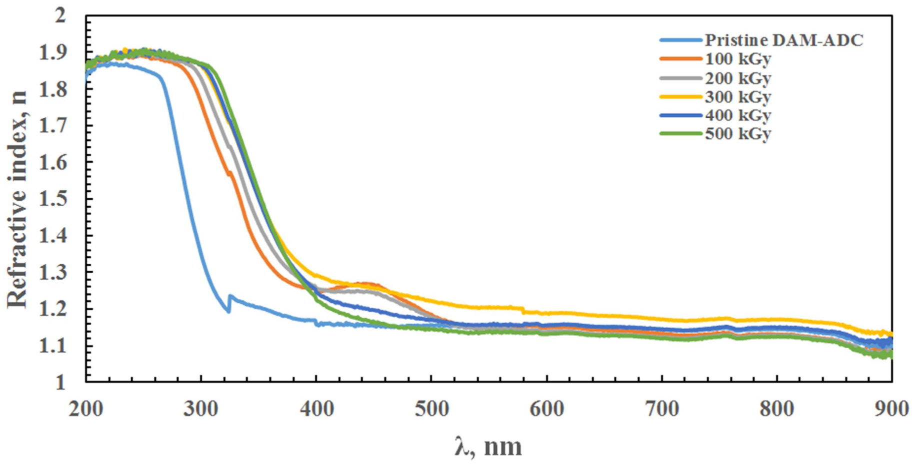

Figure 4. *Dependence of the refractive index n on the wavelength for the pristine and gamma-irradiated DAM-ADC polymer.*

3.3. Dielectric Constant and Dissipation Factor

The dielectric constant $\hat{\varepsilon}$ is an important quantity for the design of highly efficient optoelectronic devices [9]. This is because the frequency dispersion of the dielectric constant $\hat{\varepsilon}$ characterizes completely the propagation, reflection and loss of light in multilayer structures. Thus, this constant provides us with information about the electronic structure of the material. The optical properties of any solid material are characterized by the complex refractive index $\hat{n} = n(\lambda) + ik(\lambda)$ and the dielectric function $\hat{\varepsilon} = \varepsilon_1(\lambda) + i\varepsilon_2(\lambda)$. The real part ε_1 (represents the normal dielectric constant) and imaginary part ε_2 (represents the absorption associated of radiation by free carrier) of dielectric constant were determined by the following relations in the absorption region $(k \neq 0)$ [30, 31]:

$$\varepsilon_1 = n^2 - k^2 = \varepsilon_\infty - \left(\frac{e^2 N}{4\pi c^2 \varepsilon_0 m^*}\right)\lambda^2, \quad (3)$$

and

$$\varepsilon_2 = 2nk = \left(\frac{\varepsilon_\infty \omega_p^2}{8\pi^2 c^3 \tau}\right)\lambda^3, \quad (4)$$

where ε_∞ is the lattice dielectric constant, λ is the wavelength, e is the charge of the electron, N is the free charge-carrier concentration, ε_0 is the permittivity of free space, m^* is the effective mass of the electron, and c the velocity of light. ω_p is the plasma frequency (a resonant frequency for free oscillations of the electrons about their equilibrium positions), and it can be given by ($\omega_p = e^2 N / \varepsilon_0 \varepsilon_\infty m^*$) [30]. The dependences of ε_1 and ε_2 on the photon energy $h\upsilon$ for pristine and gamma irradiated samples are depicted in Figure 5 and Figure 6, respectively. It is seen that both ε_1 and ε_2 follow the same pattern, increase with increasing photon energy, and the values of real part are higher than the imaginary part. This may be due to the change in the crystallite size and the internal strain of the irradiated DAM-ADC samples. This change causes the formation of peaks in the dielectric spectra which depends on the material type and irradiated gamma dose.

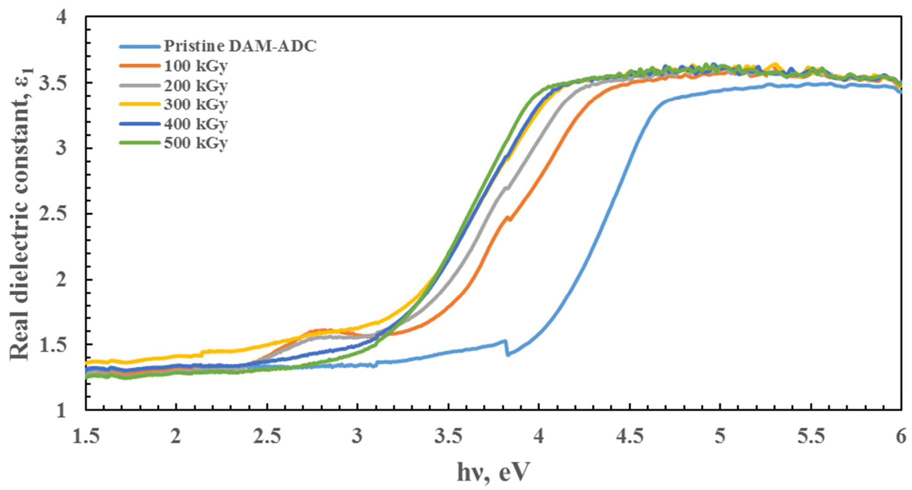

Figure 5. *Plots of ε_1 vs. photons energy (hv) for the pristine and gamma-irradiated DAM-ADC polymer.*

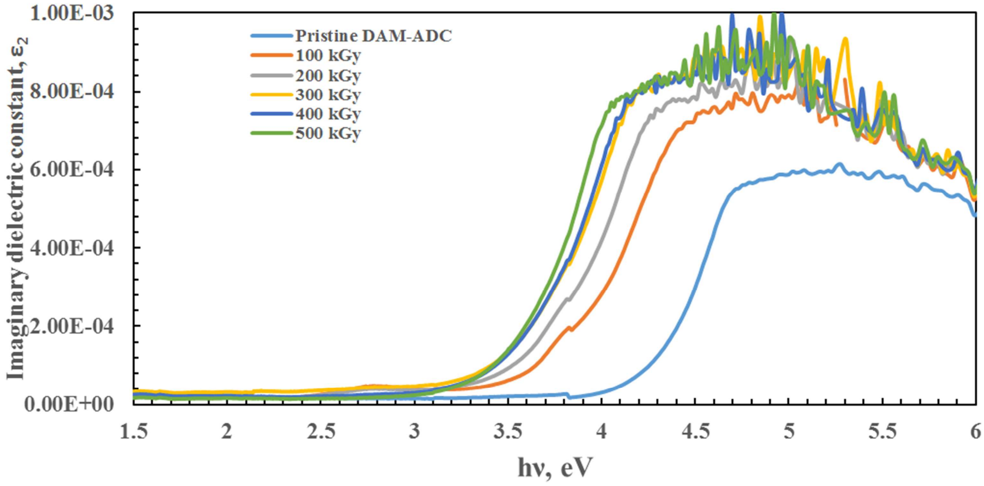

Figure 6. *Plots of ε_2 vs photons energy (hv) for the pristine and gamma-irradiated DAM-ADC polymer.*

The dissipation factor *tan δ* is the ratio of the absorption associated of radiation by free carrier to the normal dielectric constant or the rate of power loss of a mechanical mode, such as an oscillation in a dissipative system. This factor computed according to the following equation [30]:

$$\tan\delta = \frac{\varepsilon_2}{\varepsilon_1} \tag{5}$$

The variation of the dissipation factor *tan δ* for pristine and gamma irradiated DAM-ADC polymer samples on the photon energy $h\upsilon$ has the same behavior of both real and imaginary parts of dielectric constant. This supports the effect which occurs in the irradiated samples.

3.4. Energy Loss Functions

The volume energy loss (*VELF*) and the surface energy loss (*SELF*) functions are important physical quantities which are used to describe the rate of energy loss for electron passing through a material. These quantities were studied for the unirradiated and gamma irradiated DAM-ADC samples as a function of the photon energy $h\upsilon$ are illustrated in Figure 7 and Figure 8, respectively. VELF and SELF quantities are related to the real and imaginary parts of dielectric constant by the following relation [32]:

$$VELF = -\mathrm{Im}\left(\frac{1}{\hat{\varepsilon}}\right) = \frac{\varepsilon_2}{\varepsilon_1^2 + \varepsilon_2^2} \tag{6}$$

and

$$SELF = -\mathrm{Im}\left(\frac{1}{\hat{\varepsilon}+1}\right) = \left(\frac{\varepsilon_2}{(\varepsilon_1+1)^2 + \varepsilon_2^2}\right) \tag{7}$$

It is clear that the energy loss by free charge carriers when traversing through the bulk material has the same behavior as when they traverse the surface and there is no significant difference between them at lower and higher photon energies. In addition, it observed that the values of *VELF* increase more than *SELF* at the particular peaks which characterized the unirradiated and γ- irradiated DAM-ADC polymer samples.

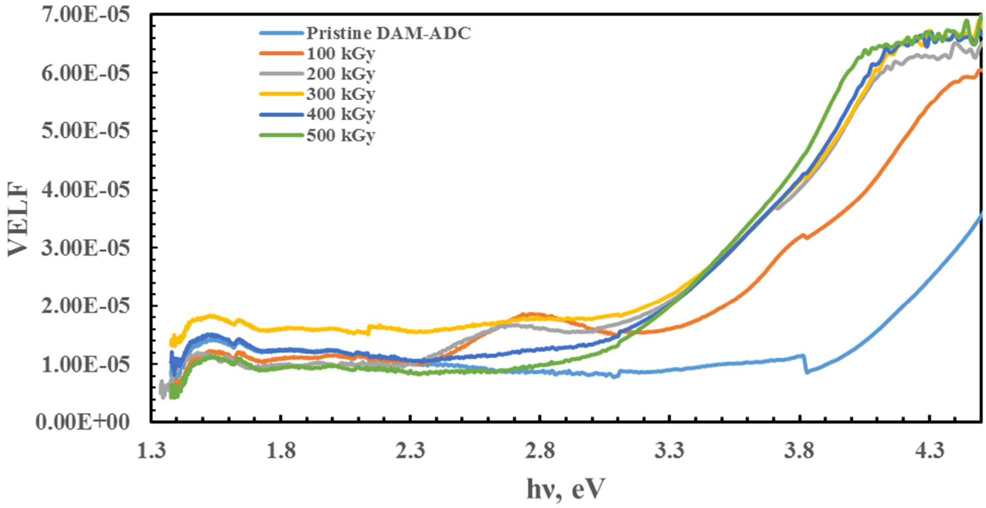

Figure 7. *Dependence of VELF function on the photon energy (hv) for the pristine and gamma-irradiated DAM-ADC polymer.*

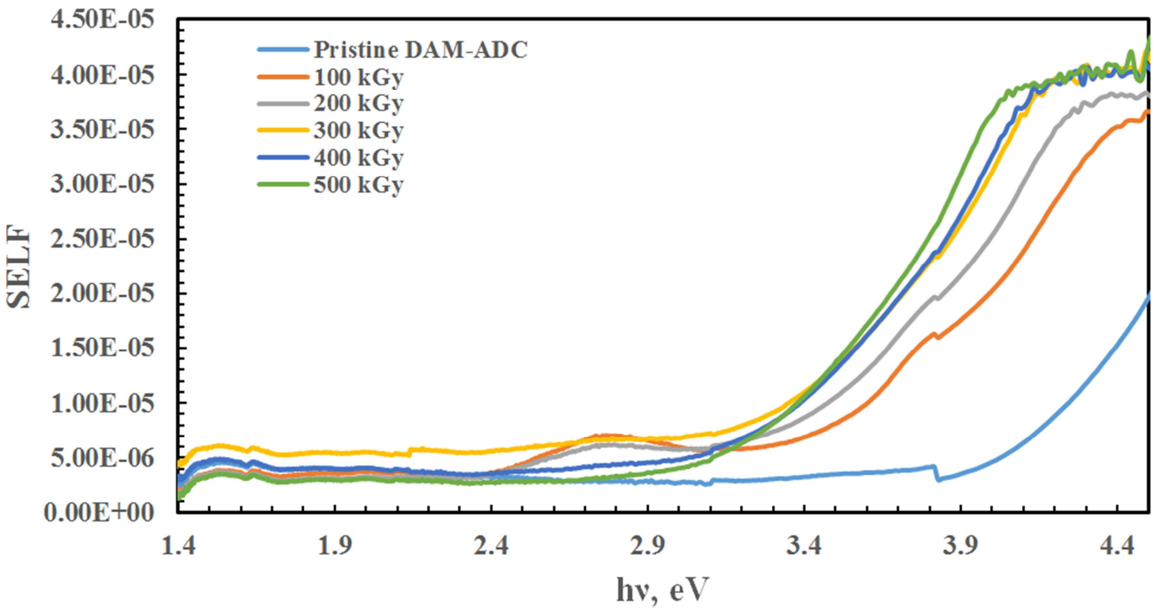

Figure 8. *Dependence of SELF function on the photon energy (hν) for the pristine and gamma-irradiated DAM-ADC polymer.*

3.5. Optical Conductivity

The optical conductivity σ_{opt} describes the optical properties of materials and used to detect the allowed interband optical transitions of materials. The complex optical conductivity ($\hat{\sigma} = \sigma_1(\lambda) + i\sigma_2(\lambda)$) is related to the complex dielectric constant ($\hat{\varepsilon} = \varepsilon_1(\lambda) + i\varepsilon_2(\lambda)$) by the following relations [33]:

$$\left.\begin{aligned} \sigma_1 &= \omega\varepsilon_0\varepsilon_2 \\ \sigma_2 &= \omega\varepsilon_0\varepsilon_1 \end{aligned}\right\} \tag{8}$$

where σ_1 and σ_2 are the real and the imaginary parts of the optical conductivity. Figure 9 and Figure 10 depict σ_1 and σ_2 parts as a function of the photon energy $h\upsilon$, respectively for the unirradiated and gamma irradiated DAM-ADC polymer samples. It is clear that the real part of optical conductivity σ_1 and the imaginary part of dielectric constant ε_2 follow the same pattern. In addition, the imaginary part of optical conductivity σ_2 and the real part of dielectric constant ε_1 have the same pattern for the studied samples. The results show that there are distinct peaks for σ_1 and σ_2 characterized the unirradiated and γ- irradiated DAM-ADC samples, the origin of these peaks may be attributed to the optical interband transitions.

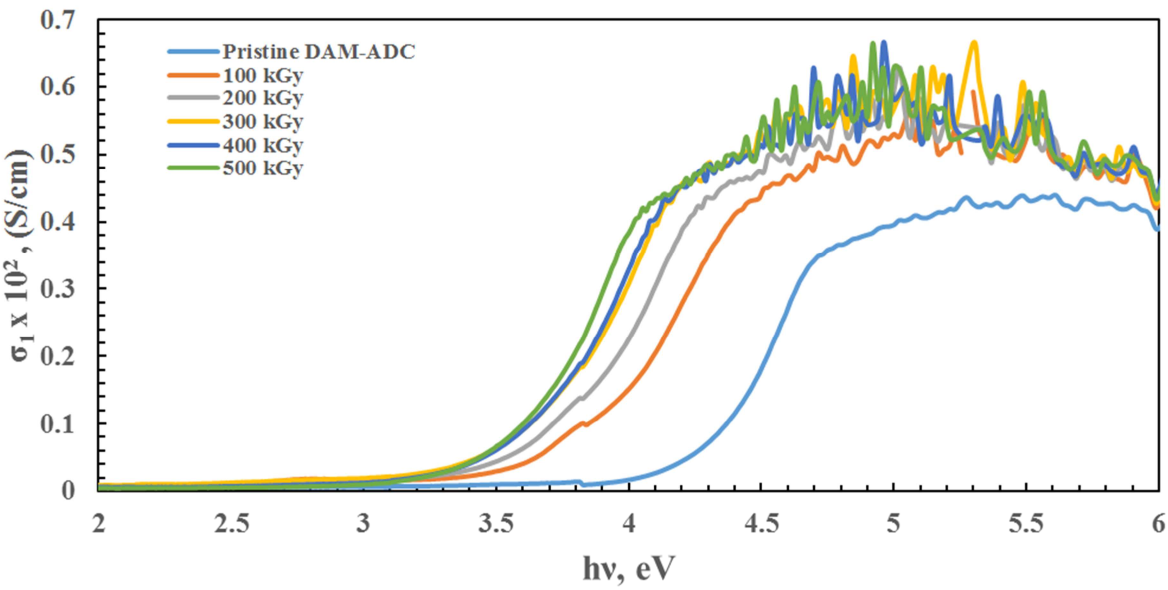

Figure 9. *Dependence of σ_1 on the photon energy (hν) for the pristine and gamma-irradiated DAM-ADC polymer.*

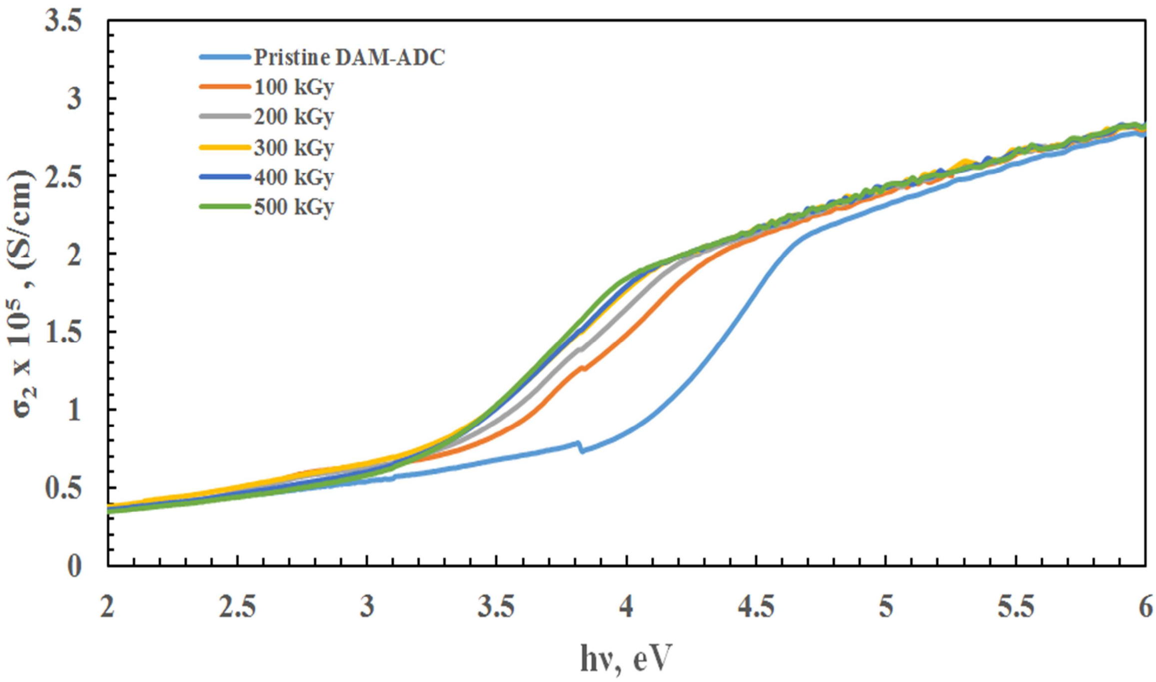

Figure 10. *Dependence of σ_2 on the photon energy (hν) for the pristine and gamma-irradiated DAM-ADC polymer.*

3.6. Optical Energy Band Gap $E^{opt.}$

The spectral distribution of the optical absorption coefficient α(ν) particularly near the fundamental absorption edge is useful in order to investigate the optical induced transitions and for the provision of information about the structure and optical energy band gap in both crystalline and non-crystalline materials. Therefore, the optical band gap and nature of optical transitions can be obtained from the dependence of the absorption coefficient α(ν) on the photon energy $h\upsilon$. The absorption coefficient is associated with inter band transitions, thus the absorption data follows a power-law behavior which is given by [34, 35]:

$$\alpha(\nu)h\upsilon = C\left(h\upsilon - E^{opt.}\right)^n \tag{9}$$

where C is a constant that independent on the photon energy, but depends on the transition probability, and *n* is a number which characterizes the transition process. It is well known that γ- radiation can cause ionization or excitation of the optical electrons and possibly displacement of atoms from their sites in the lattice of solid. Thus when DAM-ADC polymer are irradiated with γ- radiation, crosslinking and chain scission are produced.

In this work, the indirect allowed optical energy band gap $E^{opt.}$ for pristine and irradiated DAM-ADC samples was determined using Eq. (9), putting n=2, and plotting $(\alpha h\upsilon)^{1/2}$ versus the photon energy ($h\upsilon$). With extrapolation of the linear part of the curve to lower energy, it is possible to determine the optical energy band gap. Figure 11 shows the dependence of $(\alpha h\upsilon)^{1/2}$ on the photon energy ($h\upsilon$) for DAM-ADC pristine and γ- irradiated samples. It clear that the values of the indirect allowed optical energy band gaps are 4.30 eV for the γ- unirradiated sample and 3.83, 3.74, 3.66, 3.62, and 3.60 eV for γ- irradiated samples with doses 100, 200, 300, 400, and 500 kGy, respectively. Results indicate that the optical energy band gap $E^{opt.}$ decreases with increasing the irradiation gamma dose. This may be due to the fact that, when polymer exposed to irradiation doses, it starts to crosslink, a formation of new covalent bonds can be obtained. Then, different chains are formed, i.e cross linking starts which are in turn hinder the motion of molecules and reduce its activities and in turn decrease the optical energy band gap. In addition, a formation of free radicals are obtained, which causes a decrease in the optical energy band gap. As the gamma dose increases from 100 to 500 kGy, the crystalline structure is assumed to be perturbed and leading to an increase in the degree of disorder. At higher dose (500 kGy), the chains scission become predominant relative to the crosslinking [36]. It is known that $E^{opt.}$ decreases with increasing the degree of disorder of amorphous phase. Furthermore, the band tailing shifts to higher energies and extends into the forbidden band as shown in Figure 11.

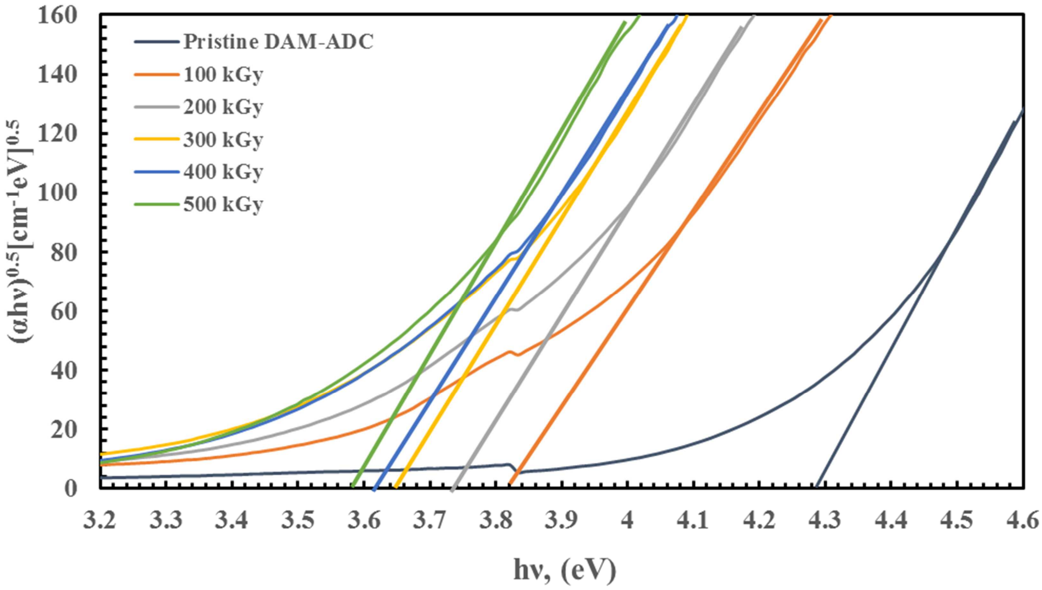

Figure 11. *Variation of* $(\alpha h\nu)^{0.5}$ *with (hν) for pristine and DAM-ADC samples irradiated with different doses of gamma rays.*

Figure 12 illustrates the variation of the optical energy gap as a function of γ- dose for DAM-ADC samples. There is a linear relationship between the optical energy gap values and γ- doses in the range of 100-300 kGy ($R^2 = 0.9988$). One can conclude that the DAM–ADC track detector is useful in the field of γ- detection and can be used as a dosimeter of gamma in the range of 100-300 kGy.

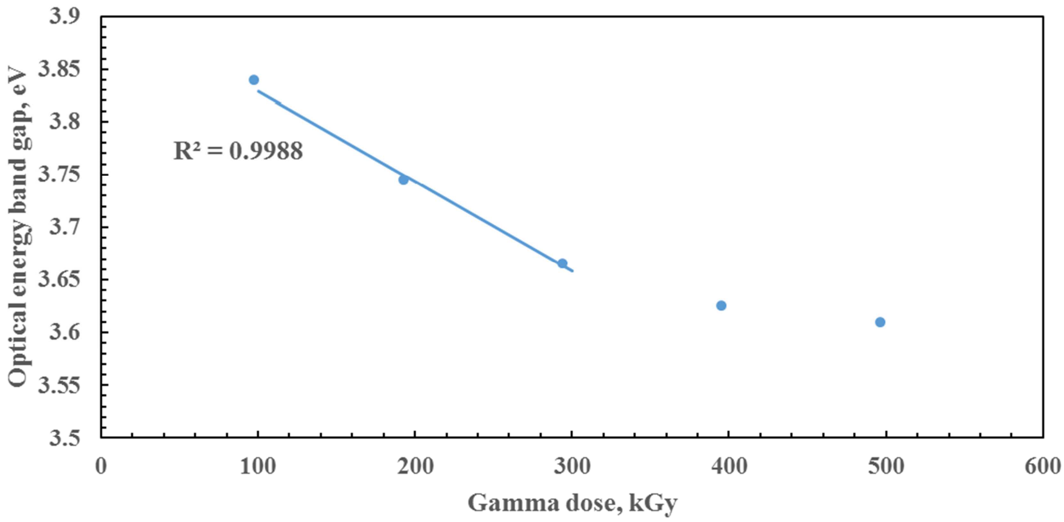

Figure 12. *Variation of the optical energy gap as a function of gamma dose for DAM-ADC samples.*

4. Conclusion

The effect of gamma irradiation on the optical and electrical properties of DM-ADC nuclear track detector was studied. Samples of DAM-ADC detector were irradiated at room temperature with gamma doses in the range of 100-500 kGy using 1.25 MeV ^{60}Co source of dose rate 4 kGy/h. The optical properties of pristine and gamma irradiated samples have been studied through the measurements of UV-visible absorption spectra at normal incidence of light in the wavelength range 190-1100 nm. The spectra show observable shift of absorption edge towards longer wavelength with increasing gamma absorbed dose. The refractive index n and the absorption index k were computed using the obtained data of transmittance $T(\lambda)$ and reflectance $R(\lambda)$. VELF and SELF study indicate that energy loss by free charge carriers when traversing through the bulk material has the same behavior as when they traverse the surface in particular for relativity lower energies. It means that DAM-ADC polymer detector has isotropic response i.e, surface like bulk response. The results of the optical conductivity indicate that DAM-ADC polymer is a good candidate in optoelectronic devices based on its band gap and dispersion parameters. The optical energy gap decreases as γ-irradiation increases. A linear relationship between the optical energy gap values and gamma doses in the range of 100-300 kGy is observed. Therefore, the DAM–ADC track detector can be used as a gamma dosimeter in this range.

Hightlight

a. DAM-ADC polymer samples were irradiated with different gamma doses.
b. UV-visible spectra of the irradiated samples were performed.
c. Dielectric constant, energy loss function, and optical conductivity of the irradiated samples were calculated
d. The optical energy band of unirradiated and irradiated DAM-ADC samples was determined.

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