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Effect of Gamma Irradiation on the Structural and Optical Properties of Bayfol CR1-4 Nuclear Track Detector

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Abstract
Bayfol is class of solid state nuclear track detectors (SSNTDs). Bayfol (a polycarbonate/ polyester blend) was irradiated with different doses of gamma ranging from 0 to 300 kGy. The effects of irradiation on the structural and optical properties of Bayfol were studied using X-ray diffraction (XRD) and UV spectroscopy. The XRD pattern was taken for the original (non-irradiated) sample and for samples irradiated with increasing doses of gamma irradiation. The "Intensity" and the "FWHM" were obtained; both values decrease up to a dose of 100 kGy and increase subsequently. This indicates a change in structure caused by gamma irradiation. The UV spectra of irradiated samples indicate that the absorption edge shifts towards the longer wavelengths relative to the original (non-irradiated) sample. This shows that there is a decrease in the band-gap and Urbach energy after irradiation. The refractive index decreases, with increased gamma dose, to a minimum at the 100 kGy dose and increases thereafter.
Keywords: Bayfol, gamma irradiation, X-ray diffraction, UV spectra

1. Introduction
Polycarbonate (PC) is an important engineering plastic because of its high optical transparency, mechanical strength, good electrical insulation and high resistance to heat. To improve resistance to chemicals, PC is blended with polyesters which have high resistance to chemicals [1].The resulting blends show the high impact resistance of PC with the resistance to corrosive chemicals of the polyesters. [2-5]. Bayfol is class of solid state nuclear track detectors (SSNTDs) composed of a polycarbonate/polyester blend. Research on the irradiation of polymers has included several areas: studies to improve the properties of polymers; studies of the impact on the mechanical, chemical, optical and electrical properties of the polymer; studies on the structural changes caused and how these changes explain the transformation of properties; and the study of changes caused by different types of irradiation [6-8]. Irradiation results in the degradation (chain scission) of the polymer chain, the formation of free radicals and the reuniting of these free radicals. The formation and transport of reactive species change the physical and chemical properties of the material [9-14]. The present work was undertaken to investigate the impact of gamma irradiation on the structural and optical properties (band gap, Urbach's energy and refractive index) of Bayfol CR1-4.

2. Experimental
2.1 Samples
Bayfol CR 1-4 is a polycarbonate (PC) with a chemical composition of $C_{16}H_{14}O_3$ blended with polyester (PC/PBT blend-film) with an average thickness of (250μm) and density (1.20 g/cm^3). It is manufactured by Bayer A.G., Leverkusen Germany.

2.2 Irradiation Facilities
The samples were exposed to gamma rays from a ^{60}Co source (manufactured by the Atomic Energy Canada Limited (AECL)) at the National Institute for Standards (NIS), Egypt. A dose rate of (2.4) Gy/min was used. The measurements were carried out 24 hours after the irradiation.

2.3 Experimental Apparatus
The X-ray diffraction measurements were carried out at room temperature using Shimadzu 6000 X-ray diffract meter equipped with Cu-k_α radiation of wavelength λ=1.5406 Å. The UV–Vis spectroscopy measurements were conducted by UV/Vis Spectrophotometer (Model Tomos UV-1800) in the wavelength range from 1100 nm to 190 nm, in transmission mode, using thin solid film of synthesized quantum composites.

3. Results and discussion
3.1. X-ray diffraction
XRD measurements (Fig. 1) were carried out on original (non-irradiated) and irradiated samples. The XRD patterns of the samples are characterized by blunted peaks extending in the 2θ range of (16^o -22^o). The maximum diffraction intensity is observed at (2θ = 19◦). The presence of blunted peaks indicates that Bayfol is a partly

crystalline polymer with a dominant amorphous phase. The integral intensity I (Fig. 2) is plotted versus the γ irradiation dose. The intensity decreases until a minimum value is reached at the dose of 100 kGy, followed by an increase, on increasing the dose. In the dose range of 20-100 kGy, there is a decrease in the crystalline phase due to cross-linking which destroys the crystalline structure. In the dose range of 100-300 kGy, there is an increase in the crystalline phase due to degradation induced by gamma irradiation. Chain scission ruptures the polymer chain, leaving free radical ends which acquire mobility. Radicals that are close together are able to recombine thus increasing the crystalline state [15-17]. Approximate indicative values of the full width at half maximal intensity (FWHF) were calculated and are also displayed in Fig 2. FWHF is inversely proportional to the crystal size (thickness of the lamella). The FWHF decreases on increasing the dose up to 100 kGy. This suggests an increase in the crystal size. The FWHF increases subsequent to 100 kGy, pointing to a decrease in the crystal size. The change in crystal size caused by irradiation is slight from 2.38-2.39 radian [17].

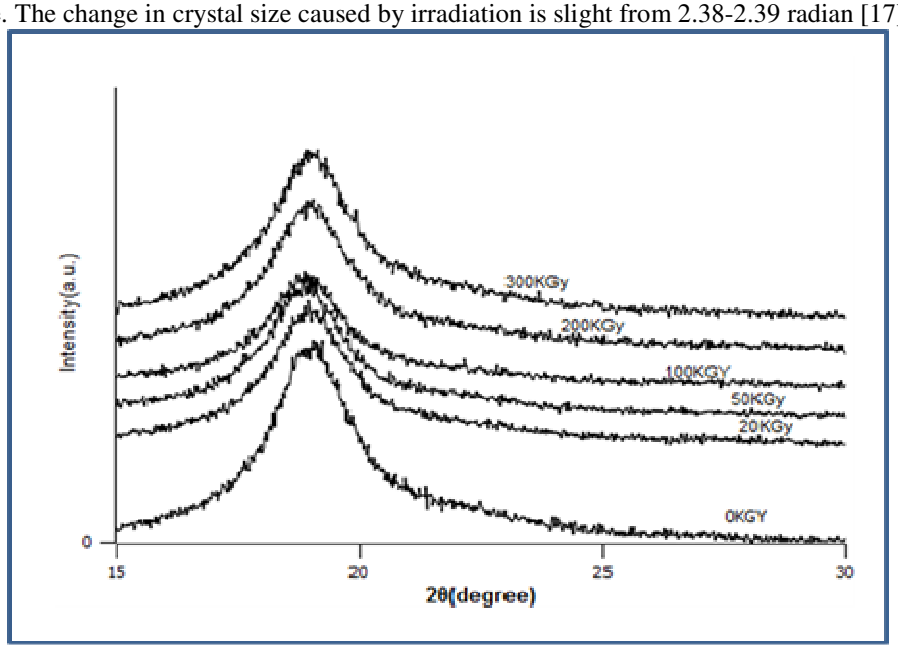

FIG. 1: XRD patterns of the Bayfol CR 1-4 before and after gamma irradiation.

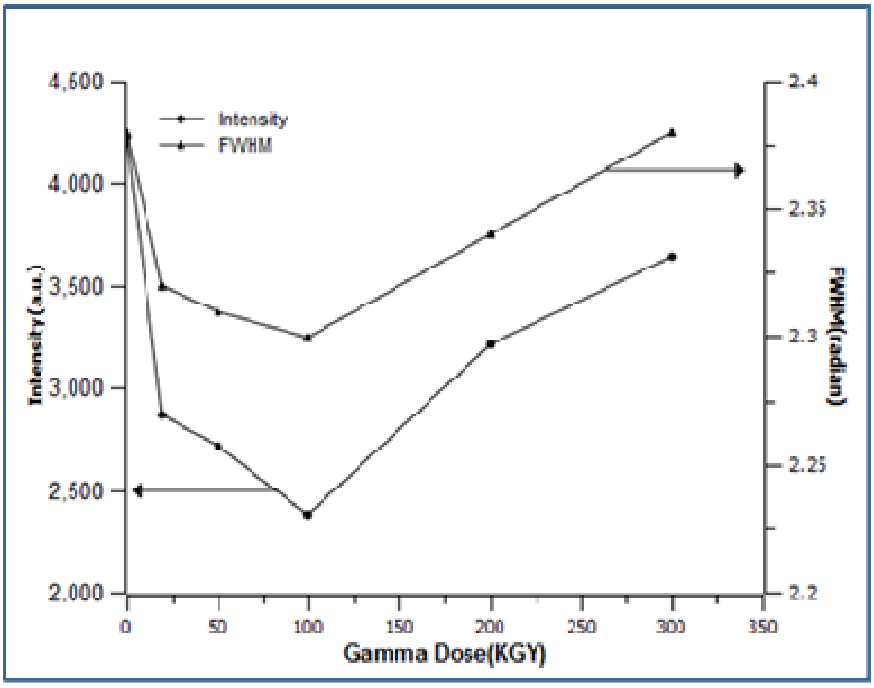

FIG. 2: Variation of the integral intensity (I) with gamma dose and the full width at half maximal intensity.

3.2. Absorption spectra

The UV spectrophotometric scans were measured in the wavelength range 200–900 nm for the original (non-

irradiated) sample and after γ-irradiation. Fig. 3 shows the absorption spectra for the samples. With increase in the optical absorption, due to gamma irradiation, the UV spectra of the polymers also show a shift upwards indicating a decrease in the band gap [18]. This shift in absorbance is probably caused by the formation of conjugated bonds (the possible formation of carbon clusters and/or defects). Increased irradiation increases defects and the formation of free-radicals which in turn increases the conductivity of the polymer [19-21].

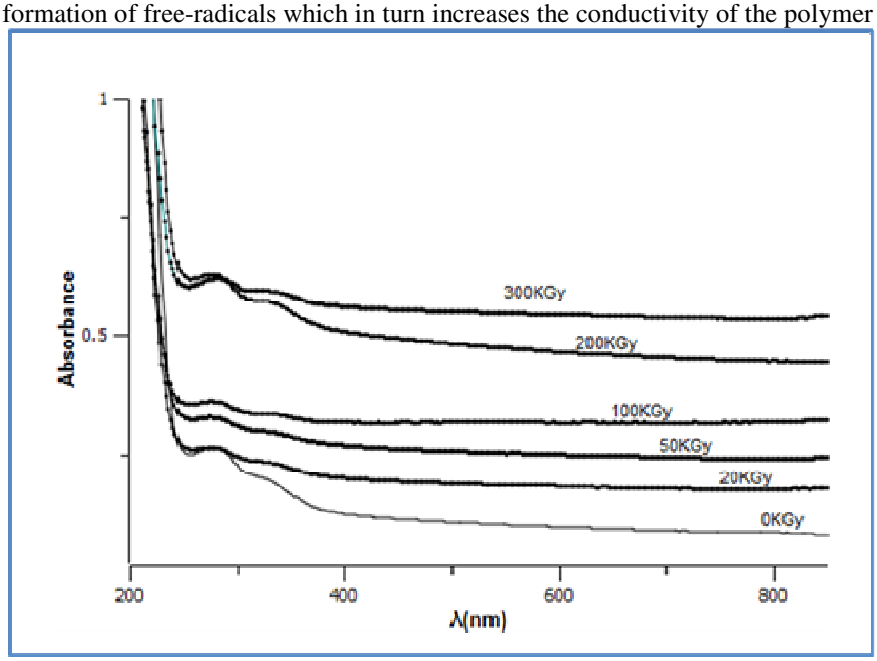

FIG. 3: Absorption spectra of the samples before and after gamma irradiation.

3.3. Optical energy gap

The absorption coefficient (α) is calculated from the optical absorption spectra using the relation [22]:, $\alpha(h\nu) = 2.303A / X$,where X is the film thickness in cm and "A" is defined by A=log(I_o/I) where I_o and I are the intensity of the incident and transmitted beams, respectively. The optical band gap was determined from the analysis of the spectral dependence of the absorption, near the absorption edge. The absorption coefficient for non-crystalline materials has the following frequency dependence [23]:

$$\alpha(h\nu) = A(h\nu - E_g)^r / h\nu \qquad (1)$$

Where (hν) is the energy of the incident photons, E_g is the value of the optical energy gap between the valence and the conduction bands, the exponent, r is an empirical index that characterizes the transition process. The exponent r takes values of 1/2 and 3/2 for the direct - allowed and forbidden transitions, respectively. It has the values 2 and 3 for the indirect –allowed and forbidden transitions, respectively [24]. The factor "A" is constant. The direct and indirect optical band gap were determined by plotting $(\alpha h\nu)^{1/2}$ and $(\alpha h\nu)^2$ as functions of photon energy (hν). Extrapolating the straight parts of these relations to the hν axis yields the corresponding direct and indirect band gaps as shown in Fig. 4. The direct band gap decreases from 4.87 eV to 4.38 eV and the indirect band gap from 5.54 eV to 5.11 eV for the non-irradiated sample and for the sample irradiated with 300 kGy dose respectively (Fig. 5). The decrease in optical band gap after irradiation may be attributed to the creation of carbon enriched clusters formed by the release of hydrogen as hydrogen molecules [25] and/or the creation of some intermediate energy levels due to structural rearrangements [26].The decrease in the energy gap implies an increase in the electrical conductivity of the irradiated polymers [27,28]. The number of carbon atoms per cluster (N) was calculated [29] and shows an increase with increased irradiation dose (Fig 5).

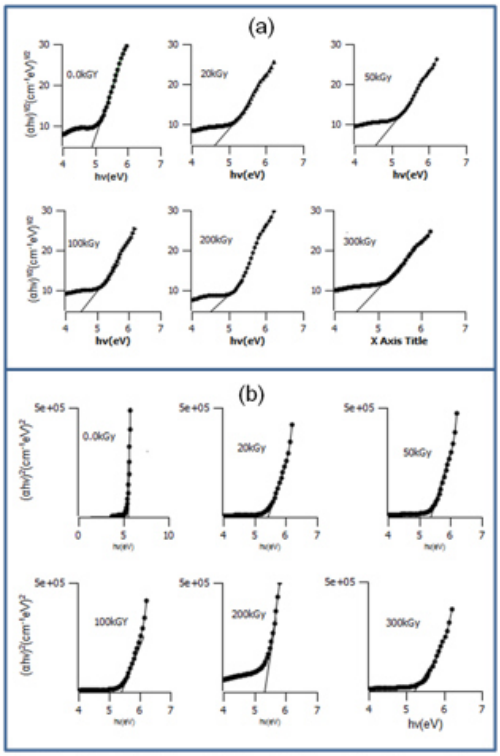

FIG. 4: $(\alpha h\nu)^{1/2}$(a) and $(\alpha h\nu)^2$ (b) as functions of photon energy(hν). Extrapolating the straight parts of the plots to the (hν) axis yields the corresponding direct and indirect band gaps.

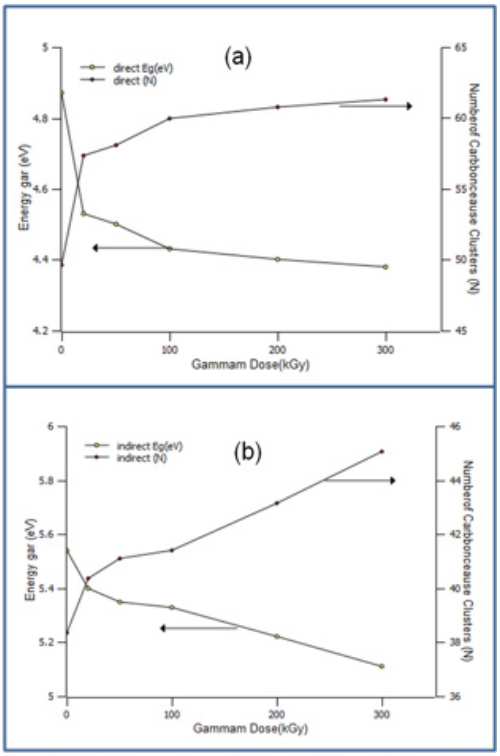

FIG. 5: The values of direct (a) and indirect (b) band gaps for Bayfol CR 1-4 before and after gamma irradiation.

3.4. Urbach Energy

The absorption coefficient α(ν) is described by the Urbach formula [30],

$$\alpha(v) = \alpha_o \exp\left(h\nu / E_u\right) \qquad (2)$$

Where α_o is a constant and E_u is the Urbach energy which is defined by the width of the tail of localized states in the forbidden band gap. The origin of E_u arises as thermal vibrations in the lattice [31]. The logarithm of the absorption coefficient α(ν) was plotted as a function of the photon energy (hν) for samples (Fig. 6a). The values of the Urbach energy (E_u) were calculated by taking the reciprocal of the slopes of the linear portion in the lower photon energy region of these curves. It was found (Fig. 6b) that E_u decreases with increasing gamma irradiation dose. The gradual decrease in the values of Urbach's energy may result from the creation of carbon enriched clusters [25], formation of defects and/or increase in the crystalline nature of the polymer with increase in the gamma irradiation dose [32,33].

3.5. Refractive Index

The refractive indices of solid films of Bayfol (thickness of 250 µm) -irradiated with gamma doses up to 300 kGy - were measured. The refractive index measurements were carried out using an Abbe refractometer (Type Reichert; mark II, Model-10480, New York). The wavelength of the light used is 5893 ^{o}A. The accuracy of measuring the values of refractive indices of the prism was ± 0.0001 at temperatures of 24-26 ^{o}C. Several values were measured for the same sample and the average value was used. The values obtained are given in Fig. 7 which illustrates the variation of the refractive index with the gamma dose. The refractive index decreases from 1.581 for the non-irradiated sample to a minimum value of 1.5713 around the 100 kGy dose due to chain scission, followed by an increase on increasing the dose up to 1.5843 at the 300 kGy irradiation dose. The gamma irradiation at the dose range of 100 - 300 kGy helps the formation of free radicals that are chemically active, allowing the formation of covalent bonds between different chains (cross-linking) [8,17].

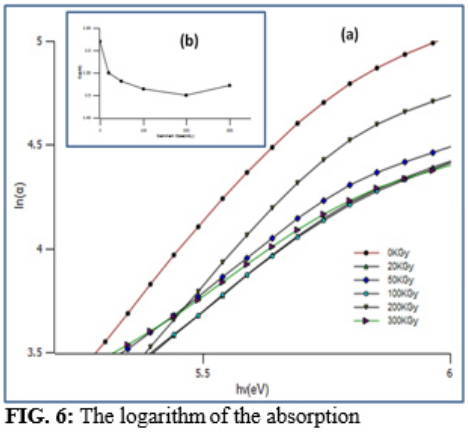

FIG. 6: The logarithm of the absorption coefficient α(ν) versus (hν) (a), and the Urbach energy (b) for samples before and after gamma irradiation

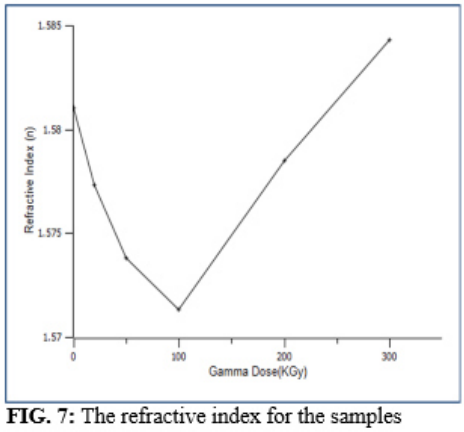

FIG. 7: The refractive index for the samples with the gamma dose.

4. Conclusion

In this work, the effect of increasing gamma irradiation dose on the structure and optical properties of Bayfol CR 1-4 has been investigated. The changes in the "Intensity" and the "FWHM" indicate a change in structure caused by the irradiation. In the dose range of 20-100 kGy, there is a decrease in the crystalline phase due to cross-linking which destroys the crystalline structure. In the dose range of 100-300 kGy, there is an increase in the crystalline phase due to degradation induced by gamma irradiation. The gamma irradiation, up to 300 kGy, decreases both the direct and indirect optical energy gap. The decrease may be attributed to the creation of carbon enriched clusters due to the release of hydrogen as hydrogen molecules and/or the creation of some intermediate energy levels due to structural rearrangements Also the Urbach energy E_u decreases with increasing the gamma irradiation dose. The refractive index decreases up to a minimum value due to chain scission, followed by an increase on increasing the dose, due to the formation of free radicals, allowing the formation of covalent bonds between different chains (cross-linking).

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