# A Review of Accumulated Radiation Exposure Impact in Selected Medical Materials & Thermoluminiscence Dosimeter (TLD) Crystal

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### **ABSTRACT**

The aim of the following review was to highlight and indicate the accumulated exposure dose could induce certain degradation on the accuracy of TLD crystal as research and personnel radiation dosimeter. The method of the study depends mainly on the previous literature and experimental results done by different authors in the field. The resultant analysis deduced that: the accumulated exposure dose on organic compounds in different forms of matter (dry, liquid, gas) induced controllable (desirable/undesirable) chemical and physical properties as well as in the TLD chips which is influenced by certain opacity, reduced crystallinity and general structural changes which are in turn influence the accuracy and sensitivity of measuring radiation dose.

Keywords: Radiation effects, TLD and Degradation.

### 1. Introduction

Ionizing radiation has been utilized in different fields (medical and industries) based on the induced effect that occur in different state of matter i.e. gaseous, liquid or dry solid state; no doubt or debate, it could induce certain effect in many applicable medical materials and even the materials used for radiation measurement and detection. In this realm: Hossam, [1], carried out study about the Effects of gamma irradiation on the crystallization, thermal and mechanical properties of Poly(L-Lactic Acid)/Ethylene-co-Vinyl Acetate (PLLA/EVA) blends after receiving 100 kGy. They revealed the induced effects by Differential Scanning Calorimetry (DSC) and Thermgravimetric analysis (TGA) crystallization behaviors and thermal stability before and after exposed to different doses of gamma irradiation. The effect of degradation in form of weight loss for EVA and PLLA/EVA blend has been shown in clearly in Figure (1 & 2) which revealed that: the hydrolytic degradation rate of PLLA/EVA blend can be widely controlled by exposing the PLLA/EVA to gamma irradiation and also by EVA content.

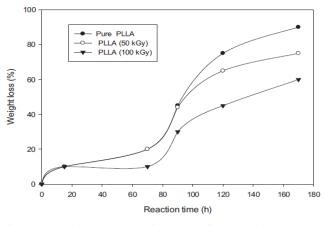


Figure 1: Weight loss (%) of PLLA before and after exposed to gamma irradiation (Hossam, [1])

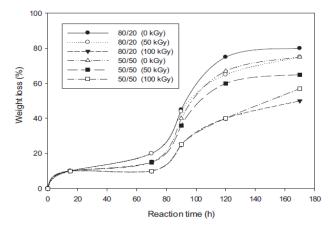


Figure 2: Weight loss (%) of polymer blends PLLA/EVA before and after exposed to gamma irradiation (Hossam, [1])

On the study done by Kalidasan et al, [2] where gamma irradiation (5, 10, and 20 kGy) of sodium borate single crystals Na<sub>2</sub>[B<sub>4</sub>O<sub>5</sub>(OH)<sub>4</sub>]-(H<sub>2</sub>O)<sub>8</sub>(monoclinic system) showed an induced crystal defect which in turn influence thermoluminescence glow curves and as well the vibrational modes of the sodium borate single crystals, dielectric permittivity, conductance and dielectric loss versus frequency graphs of these crystals have been analyzed to know the effect of gamma ray irradiation on these parameters:

The x-ray rocking curves confirm the formation of defects after gamma ray irradiation in the sodium borate single crystals, where the crystallinity being reduced following irradiation towards the amorphous state (Figure 3). And the thermo-luminescence glow curves due to gamma ray induced defects have been observed with tortuously and increases following the irradiation dose (Figure 4). While the molecular structure also has been noticed in an increasing mode at 5 kGy, then decreased for 10 kGy and totally vanish at 20 kGy; however the increment of Raman intensity could be ascribed

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to charge localization and enhancement in the saturation of bonds (Figure 5). Even the properties such as conductance, dielectric, and di-electric permittivity versus applied frequency graphs showed the sign of reduction and defects at 10 kGy of gamma irradiation (Figure 6).

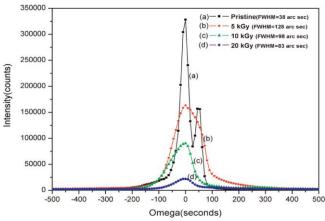


Figure 3: X-ray rocking curves of pristine and 5, 10,20kGy doses of gamma ray irradiated sodium borate single crystals

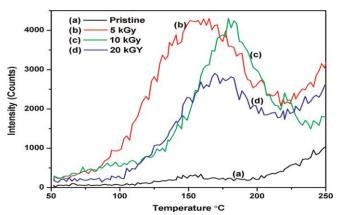


Figure 4: Thermo-luminescence spectraofpristineand5, 10, 20 kGy doses of gamma ray irradiated sodium borate single crystals

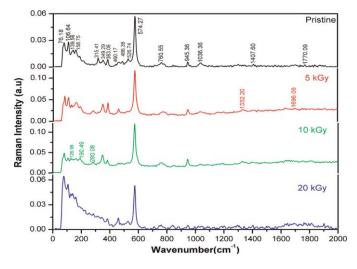
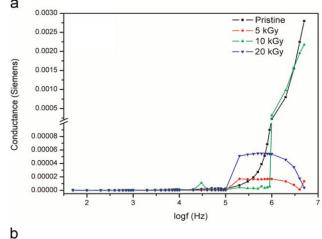
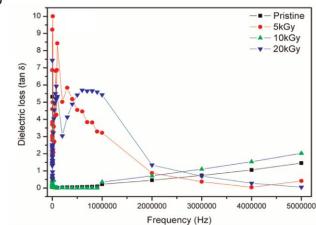


Figure 5: FT-Raman spectra of pristine and 5, 10, 20 kGy doses of gamma ray irradiated sodium borate single crystals





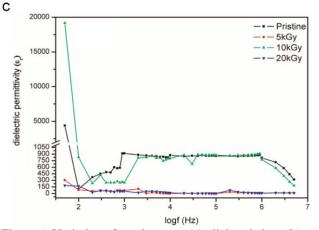


Figure 6: Variation of conductance (a), dielectric loss (b), and dielectric versus permittivity (c) versus frequency of pristineand5, 10, 20 kGy doses of gamma ray Irradiated sodium borate single crystals

Dey et al, [3] showed the effects of gamma irradiation (50, 100, 150, 250 and 500 krad) in the Blend film of polyvinyl alcohol (PVA), in which they found that: the tensile strength of pure PVA film increases following the increment of radiation dose and the highest tensile strength (32MPa) for blends was observed for 5% gelatin containing PVA film at 50 krad which was 19% higher than that of non-irradiated blend (Figure 7) and the elongation at break for both pure and blended PVA were increased following the irradiation increment up to the highest elongation break (165%) for 10% gelatin containing PVA film at 100 krad as shown in Figure

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(8), then the elongation at break decreased with the increase of radiation doses due to radiation induced degradation.

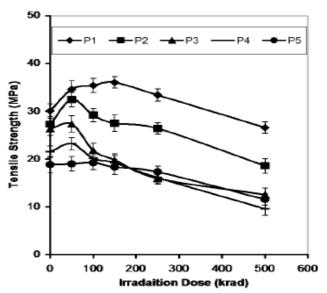


Figure 7: Tensile strength (TS) of PVA based gelatin film against different gamma irradiation dose (Krad)

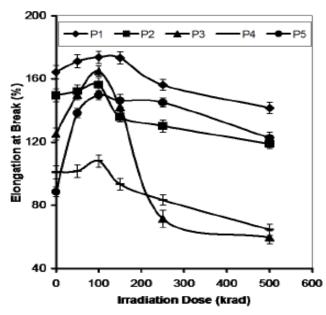


Figure 8: Elongation at break (Eb) of PVA based gelatin film against different gamma irradiation dose (Krad)

In the same realm, Sen et al, [4] showed the degradation of commercial isobutylene-isoprene rubbers after irradiated by gamma radiation (10 kGy) in view of viscosity loss significantly up to 100 kGy of irradiation dose with no further change beyond this dose. However with the presence of air; degradation has been observed even at low dose relative to  $N_2$  presence as shown in Figure (9).

Other radiation effects have been studied by Sandra et al, [5] (2013) as controlled degradation of isoprene/isobutene in rubbers for recycling purposes; in which they evaluated gamma-irradiation effects for re-use or recycling purposes in elastomeric bromobutyl compositions irradiated at 5, 15, 25, 50, 100, 150 and 200 kGy.

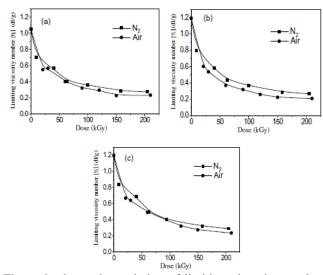


Figure 9: shows the variation of limiting viscosity numbers with irradiation dose, for low dose rate irradiation (0.18 kGy/h) in air and N2; (a) Ex165, (b) Ex 268, (c) BK1685N (Sen et al, 2003)

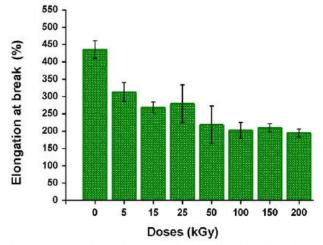


Figure 10: Elongation at break for irradiated and non-irradiated Bromobutyl rubber

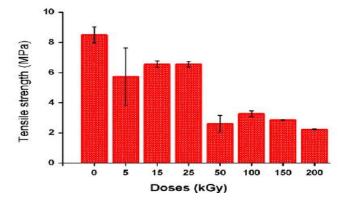


Figure 11: Tensile strength for irradiated and non-irradiated Bromobutyl rubber

They revealed that: The elongation at break has been reduced obviously following radiation dose from 5-200 kGy due to chain scission as in Figure (10), while the tensile strength showed a gradual reduction even at low doses (5 kGy) which

is ascribed to prevalence of bond scission leading to consequent mass molar reduction (Figure 11); however for doses from 5 to 25 kGy; scission and cross linking simultaneous could be observed as co-events and for doses above 50 kGy; the prevalence of chain scission and further polymeric chain degradation were observable.

The general factors that influence the structures of organic compounds (polymers) have been summarized as environmental agents by Fried, [6] (2003) with relative examples as shown in Table (1)

Table 1: shows the effects of environmental agents on organic compounds (Polymers) [6]

Agent	Susceptible polymer	Example
Biodegradation	Short-chin polymers,	Polyurethanes,
	Nitrogen-containing	polyether-
	polymers, polyesters	polyurethane
Ionizing radiation	Aliphatic polymers	PMMA,
	having quaternary	polyisobutylene
	carbon	
Moisture	Heterochain polymers	Polyesters,
		polyamides
		polyurethanes
Organic liquid and	Amorphous polymers	Polystyrene,
vapors		PMMA
Ozone	Unsaturated	Polyisoprene,
	elastomers	polybutadiene
Sunlight	Photosensitive	Polyacetals,
	polymers	polycarbonate

In the field of radiation measurement (dosimetry), the thermoluminiscence detector (TLD) has been mostly applicable with only eight common types as (Beryllium Oxide (BeO) [7], Lithium Borate (Li<sub>2</sub>B4O<sub>7</sub>), Lithium Fluoride (LiF) [8], and Magnesium Borate (MgB<sub>4</sub>O<sub>7</sub>) with low atomic number (Z) which consider as tissue equivalent materials and the non-tissue equivalent materials imply Calcium Sulphate (CaSO<sub>4</sub>) [9], Calcium Fluoride (CaF<sub>2</sub>), Aluminum Oxide (Al<sub>2</sub>O<sub>3</sub>) and magnesium orthosilicate (Mg<sub>2</sub>SiO<sub>4</sub>) which are high atomic number (Z) with high sensitivity and used for environmental monitoring.

These TLDs also could be influenced by irradiation and showed considerable changes that in turn will affect their applications for long period for radiation measurement. Hence Mohammed et al, [10] showed in the scanning electron microscope image of LiF:Mg,Ti (TLD crystal) (a) annealed before irradiation (b) annealed after irradiation by 15 Gy (Figure 12) where the morphology of LiF:Mg:Ti crystal appeared as more clear with no darkening in its surface compared with that appeared in (b).

In such view and since the high temperature would altered the thermoluminescence glow curve after irradiation, and as well the energy band gap (Eg) for the TLD crystal influenced by irradiation i.e. the pure TLD crystal before irradiation, it's Eg would be in the range of solid state, while after irradiation it's Eg decreases to be in the range of semiconducting material due to immigration of electrons from valence band towards conduction band and trapped in the forbidden band; hence the

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assumption of radiation induce opacity and crystallization changes in TLD crystal are inevitable which are in turn will affect the accuracy of TLD crystal radiation measurement after long period of application or specific radiation dose.

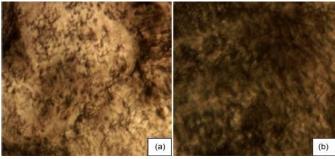


Figure 12: Shows the scanning electron microscope image of LiF:Mg:Ti (a) annealed before irradiation, (b) annealed after irradiation by 15 Gy.

The ionizing irradiation also has an impact on the crystallinity of some selected materials, in such realm, the polyvinyl alcohol (PVA) crystallinity has been reduced due to  $\gamma$ -irradiation (100 kGy) towards amorphous phase as shown in x-ray diffraction (XRD) spectrum (Figure 13) done by Eman, [11], in which the pattern of XRD intensity peaking at  $2\theta=19.66^{\circ}$  decreases gradually from control to irradiated samples and shifts to the lower angle side with another intensity peaking at  $2\theta=22.76^{\circ}$  which in turn increases gradually with the increase of dose. This result may reflect the fact that the polymer matrix suffer from some kind of structural rearrangement due to irradiation treatments.

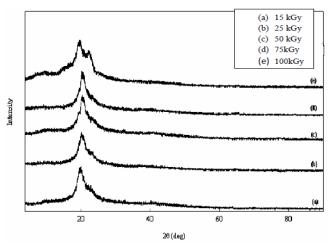


Figure 13: Show amorphous phase in XRD spectrum of PVA irradiated at different doses

The effects of  $\gamma$ -radiation on PVA crystallinity have been also studied by Mohammed et al, [12]; in which the dominant peak of XRD pattern of pure PVA film irradiated at different doses up to 50 kGy showed a prominent peak at  $2\theta = 19.5^{\circ}$  (Figure 14), corresponding to the typical crystallinity of PVA as discussed in the literatures [13, 14]. The smooth XRD pattern indicates that the pure PVA samples have no scattering influencing the spectra due to its pureness. When it was irradiated with  $\gamma$ -rays, there is continuous reduction of the crystalline structure towards amorphous phase at higher dose

of 50 kGy. The crystallinity of PVA was destroyed with increasing dose suggesting that the PVA backbone alignment could be hindered by bond scission of the C-H and C-OH side chains.

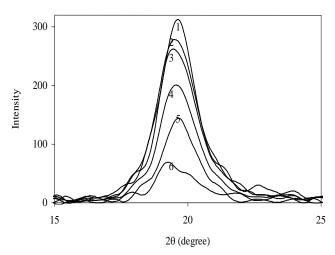


Figure 14: shows the XRD spectrum of pure polyvinyl alcohol PVA irradiated at different doses

#### 2. CONCLUSION

From the general highlighted literature; the irradiation effect could be generalized to imply all organic compounds (polymers) as well as those utilized in radiation dosimetry such air, TLD crystal etc. which in turn will influence the persistence, consistence, accuracy and sensitivity of radiation measurement after specific period; therefore the quality assurance of recalibration has to be considered accordingly.

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