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Effect of x-ray radiation doses on the absorption spectra and optical energy gap of amino acid single crystal

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Abstract Crystals of Triglycine sulfate(TGS) irradiated with different x-ray doses were used to investigate x-radiation effect on optical absorption coefficient and optical energy gap. The absorption coefficient was calculated for unirradiated and irradiated TGS crystals. UV absorption edge positions of TGS crystals depend on x-dose, they shift to lower energy values with increasing the dose. This indicates the shifting of absorption band edge wavelength to lower energies with the increase in x-dose. The optical absorption can be interpreted in terms of band-to-band transition in the visible-ultraviolet region. Values of the allowed indirect optical energy gap **(**E_g**)** of TGS were calculated as a function of x-dose. Value of E_g decreases from 4.94 eV to 4.7 eV with increasing x-doses from 0 up to 1.4 kGy. This investigation reveals that TGS crystal can be a suitable material with desired optical properties relevant for dosimetric applications.

Keywords TGS crystal, Absorption coefficient, x-irradiation, Optical energy gap

Introduction

Triglycine sulfates (TGS), a compound of the simplest protein amino acid [1-3]. Possibilities of practical application of TGS single crystals as radiation-sensitive elements of solid state dosimetric systems for high energy ionizing radiation are analyzed. It is established that the most suitable for this purpose are sensors based on the electrical and optical properties of the investigated materials. The optical absorption edge, electrical polarization and dark dielectric constant were used as radiation-sensitive in most experiments [4].

There is an increasing interest in TGS crystals since it may have useful properties for various optoelectronic applications in the visible and infrared spectral ranges. It exhibits strong absorbing ability in the most part of infrared region It is a material of choice for high speed broad band infrared pyroelectric detectors and vidicon. TGS crystal has good pyroelectric properties.. At room temperature, its pyroelectric coefficient value is the highest. [5-7].

Non-ionizing radiation is radiation without enough energy to remove tightly bound electrons from their orbits around atoms. Examples are microwaves, sound waves and visible light. Ionizing radiation is radiation with enough energy so that during an interaction with an atom, it can remove tightly bound electrons from their orbits, causing the atom to become charged or ionized. Examples are gamma rays and neutrons [5]. x-Rays are electromagnetic waves or photons not emitted from the nucleus, but normally emitted by energy changes in electrons. These energy changes are either in electron orbital shells that surround an atom or in the process of slowing down such as in an x-ray machine.

Studies on the changes in optical properties of ferroelectric thin films, irradiated with ionizing radiations, yielded valuable information regarding the electronic processes in these materials. High-energy radiation, such as x-rays, change the physical properties of the materials through which they pass. The changes are strongly dependent on the structure of the absorbing substances. Ionization occurs and charged species, both ionic and free radical are formed. The study of radiation-induced defects is not only important in observing the changes in the physical properties degradation or efficiency improvement in its applicability in a radiation environment, but it is also critical in getting basic information on vacancies, interstitials and their interaction with impurities. In the present study, results on the optical absorption spectra and optical energy gap of TGS crystals was carried out on the search for the suitable material with desired properties relevant for dosimetric applications [8].

Experimental Technique

TGS crystals were grown at about 320 K by gradually lowering the temperature of solutions according to the reaction:

$$3NH_2CH_2COOH+H_2SO_4 \longrightarrow (NH_2CH_2COOH)_3(H_2SO_4) \quad \text{.... (1)}$$

Transparent, flawless crystals of optical quality and with well-developed faces were obtained. b-plate samples were cut from the grown crystals. The samples had a b-surface area of about 20-30 mm^2 and a thickness of 0.8-1 mm. The samples used for optical measurements were clear, transparent and free from any noticeable defects.

A group of samples had been irradiated by x-rays at room temperature using linear accelerator (Siemens Mevatron) at dose rate about 22 Gy /min, with 6 MeV energy. Accumulated dose was done by successive exposures from 0.004 up to 1.4 kGy.

The optical transmission spectra (T) was measured in the photon energy range from 3 to 5.6 eV using Shimadzu UV-VIS dual beam scanning spectrophotometer. The incident unpolarized light was nearly perpendicular to b-plane. The surrounding medium was air. Two samples of different thicknesses were used, the thicker one in the sample beam and the thinner in the reference beam. This technique was used to eliminate the effect of reflection losses automatically and the result is the transmission ratio of the two samples. The absorption coefficient (α) was calculated by means of the relation:

$$\ln T_{1\text{-}2} = \alpha \Delta d \qquad \text{...(2)}$$

where $T_{1\text{-}2}$ is the transmission ratio and Δd is the thickness difference.

Results and Discussions

Figure (1) shows the room temperature measurements of the transmittance spectra (T) for TGS crystals in the wavelength range from 220 to 360 nm at different x-doses (from 0.004 to 1.4 KGy). It is clear that an abrupt increase in the transmittance exists at 245 nm (near the absorption edge).

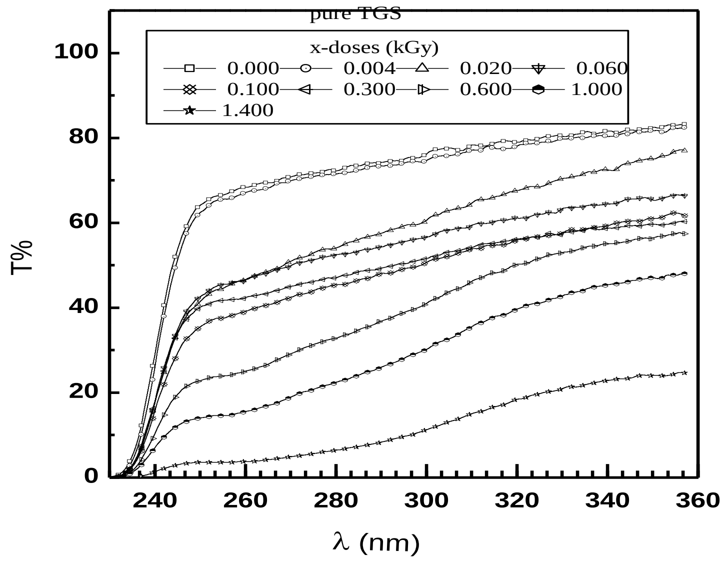

Figure 1: Spectral distribution of the transmittance (T) versus the incident wavelength (λ) for un-irradiated and irradiated with different x-dosespure TGS crystals.

The optical transmittance spectra of TGS crystal irradiated with different x- doses (from 0.004 to 1.4 KGy) as a function of the wavelength in the range from 220 to 360 nm measured at room temperatureare plotted in Fig. 1.

It was shown that TGS crystals highly transparent over the 240-360 nm region. The average optical transmittance values were higher than 70% in this range. It is clear that an abrupt increase in the transmittance exists at 245 nm (near the absorption edge) for unirradiated and Irradiated TGS crystals had a sharp absorption edge onset between 220 and 240 nm. It is observed that, the optical absorption edge shifts toward higher wavelengths with increasing the x-ray radiation dose.

Optical absorption measurements is a standard technique for investigating band structure and it is therefore of interest to study absorption in thin films. The absorption spectra in the lower region (IR), are useful in studying the molecular vibrations. The higher energy region (UV) can be useful to manifest the electronic states of the atoms and other important phenomena affected by irradiation (Madi et al, 1999).

The absorption coefficient α of a crystalline solid obeys the following relationship [9]:

$$E\omega \propto (E - E_g)^n \qquad ...(3)$$

Where E_g is the optical gap, E is the photon energy and n being an index which characterizes the optical absorption process. For direct allowed transition $n=1/2$, for direct forbidden transition $n=3/2$, for indirect allowed transition $n=2$ and, finally, for indirect forbidden transition $n=3$.

The general characteristic of these spectra is that they are composed of an almost flat baseline (absorption negligible) and a steep cut off (big absorption). Fig. (2). UV absorption edge positions of TGS crystals depend on x-dose, they shift to lower energy values with increasing the dose. This indicates the shifting of absorption band edge wavelength to lower energies with the increase in x-dose. The optical absorption can be interpreted in terms of band-to-band transition in the visible-ultraviolet region.

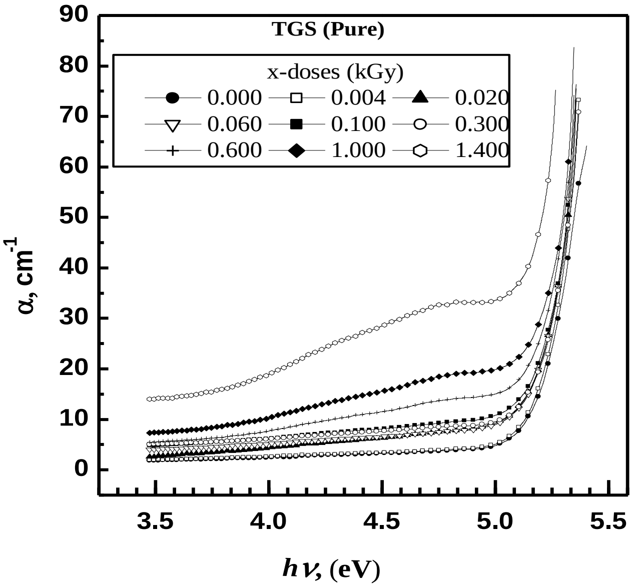

Figure 2: Spectral distribution of the absorption coefficient (α) against the incident photon energy (h ν) for un-irradiated and irradiated with different x-doses pure TGS crystals

Analysis of absorption data was carried out to determine the predominant optical transition. According to the power law of equation (2), the value of the exponent n was found to be equal 2. This means that the optical transition for TGS crystals is indirect allowed one. The indirect allowed transition is the most probable type of transition near the fundamental edge of pure TGS crystal.

The resulting absorption (α) of unirradiated TGS crystal and irradiated crystals with different x-doses in the energy range from 3.5 to 5.6 eV, is shown in Figure (2). The general level of absorption increases under the influence of irradiation by x-rays. This is probably due to the scattering by x-irradiation induced defects in the crystal. An induced band centered at about 4.75 eV could be observed. The maximal absorption coefficient of the band increases with increasing the dose of x-irradiation.

It can be seen that the absorption increases slowly with increasing photon energy in the range E = 3.5 to 5 eV. At photon energies E> 5 eV all the spectra showed a steeper increase of the absorption coefficient which shifted to lower photon energies with increasing x-doses. It can assumed that this increase in (E=hν) is due to the onset of interband transitions at the fundamental edge. E_g for pure TGS crystals irradiated with different x-doses are plotted in Figure (2).

The values of $(\alpha E)^{1/2}$ were calculated and plotted against E=hν. Fig. 3 shows a typical Tauc's [10] plot. The straight portion of the curve was then extrapolated and its intersection on the abscissa was determined as shown in the inset of Fig. 3. This value corresponds to the energy gap (E_g).

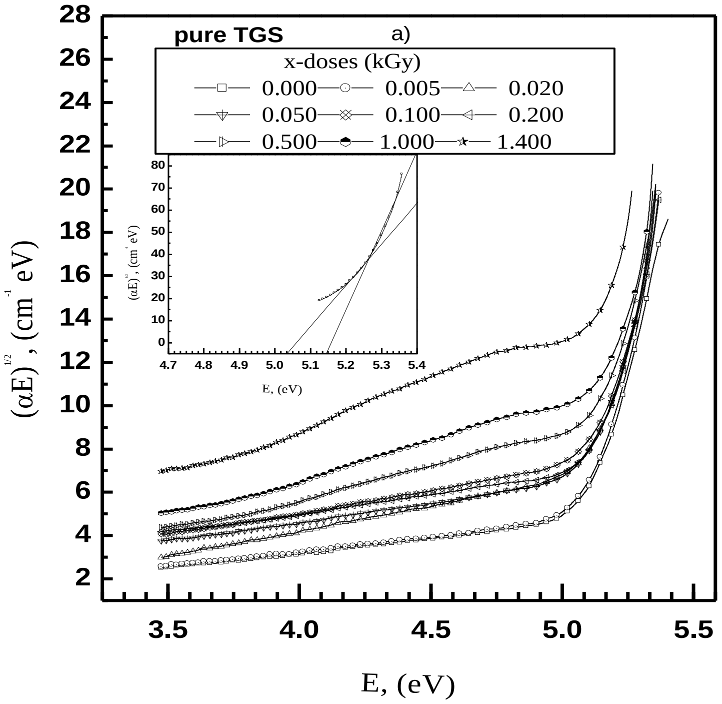

Figure 3: Spectral distribution of $(\alpha E)^{1/2}$ at different photon energy (E) forun-irradiated and irradiated with different x-doses pure TGS crystals. Inset shows the fitting of the linear portion employed in calculating the optical energy gap

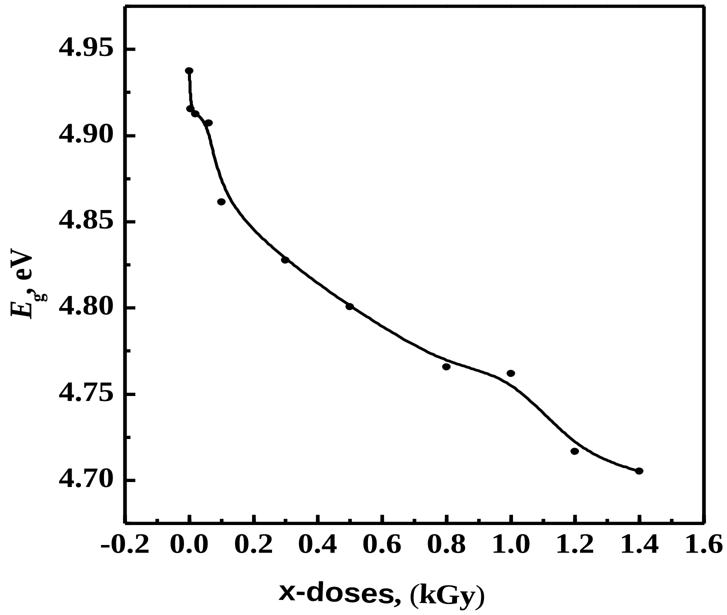

Figure 4: Variation of the indirect optical energy gap (E_g) of TGS crystals irradiated with different x-doses

For x-irradiated samples, E_g was found to be shifted towards lower energy values as show in Fig (4). This decrease in E_g may be attributed to point defects created in the crystals during x-irradiation by direct interaction of Compton electrons with lattice atoms and by multiple collisions. The slight change in the band gap resulting from increasing x-irradiation may be due to the irregular changes in the defect concentration present in the structure in addition to the possibility of recombination processes. The large quantities of energy deposited during irradiation in any target material leads to a complex series of chemical reactions, and these can have a very significant effect on the chemical and physical properties of the irradiated compound. During irradiation process, formation of ions, activated atoms and molecules and free radicals may result.

The maximal of the band observed at 4.75 eV in the absorption coefficient figure increases with increasing the x-radiation dose. This absorption band could be attributed to the free radicals CH_2COO^- formed at lattice sites of glycine-I since these radicals are unstable in low irradiation doses [11].

Gaffar and Abu El-Fadl [12] studied some optical parameters of TGS crystals doped with orthonitroaniline (ONA) and the effect of γ-rays on these parameters. They showed that such study might be promising since the individual effect of ONA and irradiation on TGS crystals is quite valuable. They also concluded that, the cut-off wavelength of unirradiated TGS crystals doped with ONA lies at 225 nm. In case of g-irradiated samples the cut-off is moved slightly towards the higher wavelength region (at about ≈ 235 nm). Also the general level of absorption in TGS crystals increases under the influence of irradiation by γ-rays. This is probably due to the scattering by g-irradiation-induced defects in the crystal.For γ-irradiated TGS crystals, E_g opt. was found to be shifted towards lower energy region as the g-dose increases. They attributed this decreases in E_g to point defects created in TGS during γ-irradiation by direct interaction of compton electrons with lattice atoms and by multiple collisions.

The effects of x-beam irradiation with different doses on microhardness and its related physical constants on $[K_y(NH_4)_{1-y}]_2ZnCl_4$ mixed crystals has been studied by Abu El-Fadl et al. [13]. They reported that the tests were performed for x-doses from 0.2 kGy up to 1.6 kGy for loads from 20 to 160 g. Their experimental results showed that, the hardness decreased as the x-doses increased. Variation of the microhardness follows the normal ISE trend for low x-doses and un-irradiated crystals then follows the reverse ISE trend for high x-doses.

Conclusion

***** Optical absorption near the fundamental edge is measured for TGS crystals

* The indirect allowed transition is the most probable type of transition near the fundamental edge of TGS crystals.

* Monotonic decrease of the edge (E_g) was observed after x-irradiation.

*Crystal irradiated with high doses shows a board absorption band centered at about 4.75 eV.

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