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Investigation on optical properties in the surface of KCl_xBr_{1-x} mixed crystals irradiated to gamma radiation (A new approach)

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Background: Similar to the thermoluminescence (TL) spectra, the optical parameters variations can also be used in TL crystals for dosimetry purposes. These optical parameters can include absorption, transmission and reflection coefficients. *Materials and Methods:* Single crystals of KBr and KCl which have been used in this research were grown from laboratory grade extra pure precursor powders. Crystal growth was performed by using a Czochralski crystal growth apparatus. The grown crystals were irradiated using a ^{60}Co source to doses of 0.5, 1.0 and 1.5 kGy. Reflection spectra were measured by a Shimatzu UVPC 3101 model spectrometer in 5 and 45 incident degree angles. *Results:* Optical properties of KCl_xBr_{1-x} mixed crystals surfaces irradiated with gamma radiation were determined. Refraction indices and dielectric coefficients of the crystals in the optical region of 250-750 nm were studied. It was observed that both irradiated and non-irradiated mixed crystals had similar surface behaviour in the order of $n \approx 1.4$ and $\varepsilon \approx 2.2$ in the whole range of 250-750 nm, while colour intensities of irradiated samples were different for various irradiation doses. The reason could have been due to the permanent heat transfer effects with the outer surface layers of the crystal and the rapid relaxation of the excited states in defected surface of the crystal. *Conclusion:* Reflection spectra along with the crystal surface optical parameters could not be efficient for the dosimetry purposes and additional information such as absorption or transmission data would also be necessary **Iran. J. Radiat. Res., 2009; 7 (3): 165-169**

Keywords: *Gamma radiation, optical characteristics, reflection spectroscopy, PACS, fading.*

INTRODUCTION

There has been high attention in researches on thermoluminescence (TL) characteristics, and optical absorption of alkaline crystals as a function of absorption doses of different radiation sources (1-2). In addition to the rapid response to the radiated rays TL crystals as a solid environment can also have wide range sensitivity to radiation in the range of mGy to kGy. Nowadays most of the alkaline single crystals, as the TL crystals, with high chemical purity can be easily prepared. TLD is not able to measure instant doses and the signals erased during read out. Due to these weaknesses, the selection of optical behaviour of this family of crystals, as a dosimetry index, can produce more accurate measurements at low irradiation doses without the mentioned disadvantages (3-4). With suitable electronic circuit design, instant and accumulative dosimetry will be possible. Therefore, the optical behaviour of crystals can be used as an effective tool for this purpose. In this article we have shown that it is possible to introduce an accurate method to determine optical characteristics of a family of KCl_xBr_{1-x} crystals' surfaces due to the gamma ray irradiation effects.

MATERIALS AND METHODS

In order to find specific optical behaviours of the crystals irradiated with gamma radiation, different mixed crystals of KBr and KCl crystals were considered with different relative weights in the mixed samples. Single crystals of KCl_xBr_{1-x}, with x= 0.1, 0.3, 0.5, 0.7 and 1, were grown by Czochralski method and by using high purity KCl and KBr salt powders purchased from Merck.

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Five precursor mixed powders with the above mentioned compositions of KCl and KBr were melted separately in a ceramic crucible at temperatures up to 860°C. A small KCl crystal was chosen as a seed crystal and mounted to the cooling bar of the Czochralski crystal growing apparatus. In order to avoid any damage to the seed crystal and also initiate the crystal growth, the melting temperature was allowed to decrease to 850 °C. To enlarge the crystal diameter, the melting temperature decreased from 0.2 °C/min down to 825 °C. The crystal growth continued with the rate of 5 mm/hr until the desired dimensions were obtained.

After the completion of crystal growth, the oven temperature was allowed to be cooled to the rate of 5°C/min down to 500 °C. This procedure was necessary to avoid any possible crack in the grown crystals. An annealing procedure was also performed and the crystals were kept at 500 °C for 12 hours, [3-4]. A typical XRD diffraction pattern of the grown single crystals of KCl is shown in figure 1.

According to this new approach, reflection spectra of KCl_xBr_{1-x} crystals, as a function of gamma irradiation doses for different x values, were recorded at two incident angles of 5° and 45° in the spectral range of 250-750 nm. The real and imaginary parts of the refractive and dielectric complex functions of the samples were calculated by the recorded data

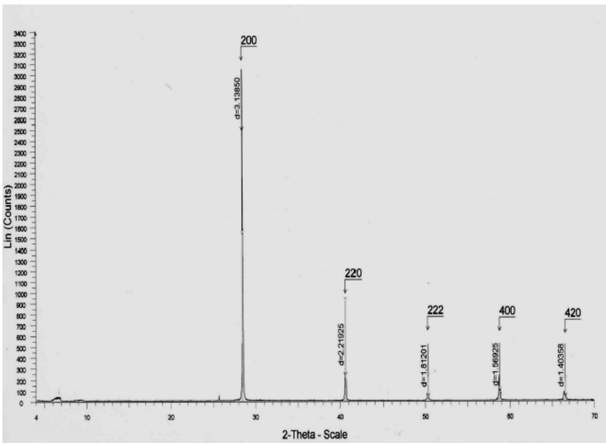

Figure 1. A typical X-ray pattern of pure KCl single crystal.

according to the mathematical relations. The calculated parameters of the refractive and dielectric indices for KCl_xBr_{1-x} family of crystals, before and after gamma irradiation at three doses of 0.5, 1 and 1.5 kGy were determined.

Due to the long exposure periods up to about six hours for delivering different doses of gamma irradiations up to 1.5 kGy, performing the reflection spectroscopy of the samples before fading process was necessary. The presence of gamma irradiation source and the reflection spectrometer at its nearby was very essential to obtain more accurate results. Accordingly, each time we performed the gamma irradiation process and recorded the reflection spectra of the samples on one group of crystals which were irradiated on one levels of 0.5, 1 and 1.5 kGy gamma radiations only.

Five rods of single crystals with different x compositions were grown. Each of the rods of grown raw crystals was cut in to four similar pieces. A set of crystals of each group were selected as the reference, non irradiated samples, and the other three sets were used for gamma irradiation purposes at three dose levels of 0.5, 1 and 1.5 kGy.

In order to have more precise reflection spectra of the samples, one side of the crystals were completely matted and colored in black and then gamma irradiation and reflection spectroscopy procedures were performed.

Gamma irradiation process was carried out in one of the Medical centers in Isfahan by a Theratron Phonix (ACEL, Canada) ^{60}Co gamma radiation source. The period of irradiation samples, at a distance of 45.5 cm, were t_1 (0.5 kGy) = 125min, t_2 (1 kGy)= 250min, t_3 (1.5 kGy)= 375min, respectively. The first group of the irradiated single crystal samples were sent immediately to the nearby centred to record their reflection spectra.

Reflection spectroscopy was performed by a Shimadzu spectrometer (Model UVPC3101) on incident reflection angles of

5° and 45° in the spectral region of 250 – 750 nm for the crystals. Refraction and dielectric indices of all crystal surfaces were calculated using reflection spectroscopy data at 5° and 45° angles by applying the following formula:

$$R = \frac{1}{2}(R_S + R_P) \quad (1)$$

$$N(\omega) = n(\omega) + i\,k(\omega) \quad (2)$$

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\,\varepsilon_2(\omega) \quad (3)$$

Where: R_s is the S polarized light reflection R_p is the P polarized light reflection and φ is the incident angle.

If $R_{5^\circ} = R_{0^\circ} = R_{5,0} = R_{P,0}$

$R_{P,45^\circ} = R^2_{3,45^\circ}$

$$R_{45^\circ} = \frac{1}{2}(R_{s,45^\circ} + R_{P,45^\circ}) = \frac{1}{2}R_{s,45^\circ}(1 + R_{s,45^\circ}) \quad (1)$$

$$R_{S,45^\circ} = \frac{1}{2}[(1 + 8\,R_{45^\circ})^{\frac{1}{2}} - 1] \quad (2)$$

$$a^2 - b^2 = n^2 - k^2 - \mathrm{Sin}2\varphi$$
$$ab = nk \quad (3)$$

$$R_S = \frac{(a-\cos\varphi)^2 - b^2}{(a+\cos\varphi)^2 + b^2}$$

$$\varepsilon_1 = n^2 - k^2$$

$$\varepsilon_2 = 2nk \quad (4)$$

$$R_{S,0} = \frac{(n-1)^2 - k^2}{(n+1)^2 + k^2} \quad (5)$$

$$R_{S,45} = \frac{(a-\cos 45)^2 - b^2}{(a+\cos 45)^2 + b^2} \quad (6)$$

By using all the above equations it is possible to determine an overall formula in the following forms:

$$N(\omega) = f(R_0, R_{45}) \quad (7)$$

$$\varepsilon(\omega) = g(R_0, R_{45}) \quad (8)$$

Considering these two equations, there would be no need to use polarizors in the whole range of the spectrum, also N (ω) and ε(ω) parameters can be calculated in a very short time and in a very accurate manner by using two groups of independent reflection data at two incident angles of 5° and 45°.

RESULTS AND DISCUSSION

According to the above mentioned formulas (1-6) it will be possible to determine the optical behaviour by reflection spectroscopy. In this method, it is supposed that, the optical parameters of the surfaces and the bulk material are similar.

During irradiation procedure, the whole crystal was irradiated uniformly by ^{60}Co. It was accordingly expected that the crystal surfaces and its bulk to have similar optical behaviour effects. By this assumption the reflection spectra of different samples of none gamma-ray irradiated and samples of KCl_xBr_{1-x} family of single crystals with different gamma level doses were recorded for different x compositions. Refractive and dielectric indices of the single crystals were calculated and compared. It worth's noting that the imaginary part of the refractive index, i.e. the extinction coefficient, and also the imaginary part of the dielectric coefficient, 2nk, were also been calculated. As shown in figures 2-9, these parameters were nearly zero in the above mentioned spectral region. The y axes in the figures 2-9 were extended to cover the values of the related physical parameters.

The calculation of the refractive and dielectric indices of the samples revealed that negligible changes in the optical region of 250-750 nm could occurred. Therefore, these changes were not proportional to the amount of gamma irradiation doses. The calculated parameters of N(ω) and ε(ω) over the spectral region, and for gamma-ray irradiated and non-irradiated single crystals showed a uniform behaviour. A comparison of our results with the given values in other reports confirmed our calculations [7, 8].

Considering the results of the reflection spectra, as well as the calculated optical and dielectric parameters of the crystals, before and after gamma-ray irradiation processes, the followings can be concluded:

- After the irradiation, in spite of colour centres formation with different intensity in

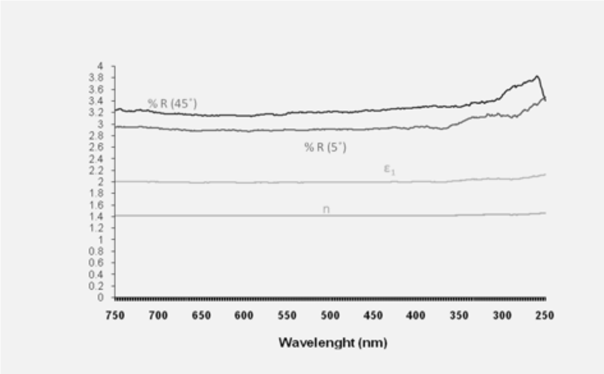

Figure 2. Reflection, refractive and dielectric coefficients spectra of pure KCl, as blank sample, against wavelength in the optical region of 250 -750 nm.

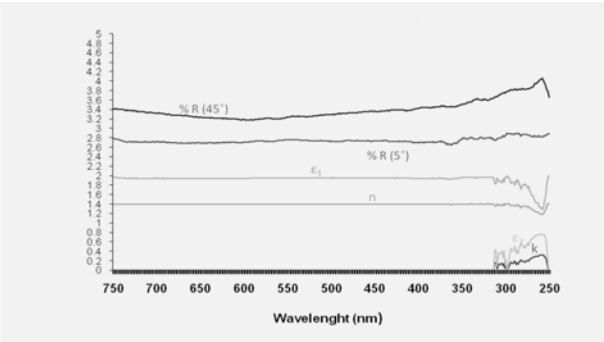

Figure 3. Reflection, refractive and dielectric coefficients spectra of pure KCl, against wavelength in the optical region of 250 -750 nm irradiated with 0.5 kGy dose of gamma radiation.

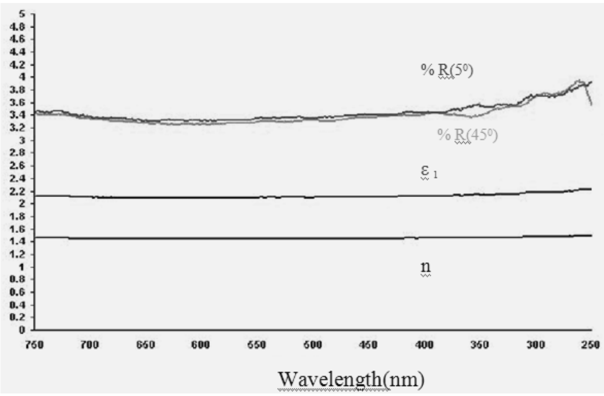

Figure 4. Reflection, refractive and dielectric coefficients spectra of pure KCl, against wavelength in the optical region of 250 -750 nm irradiated with 1 kGy dose of gamma radiation.

the samples with different x values, the optical characteristics of the crystal surface in the visible region over 400-700 nm region remained constant.

- No regular changes were observed over 250-750 nm region during irradiation absorption in crystals.

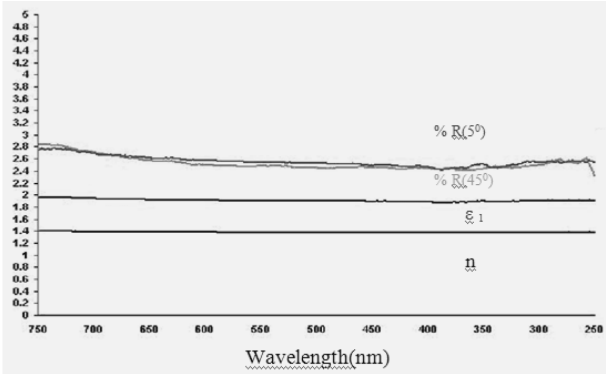

Figure 5. Reflection, refractive and dielectric coefficients spectra of pure KCl, against wavelength in the optical region of 250 -750 nm irradiated with 1.5 kGy dose of gamma radiation.

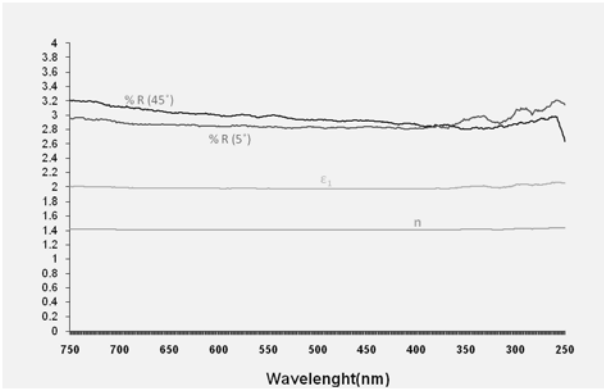

Figure 6. Reflection, refractive and dielectric coefficients spectra of mixed crystal of $KCl_{0.5}Br_{0.5}$ against wavelength in the optical region of 250 -750 nm.

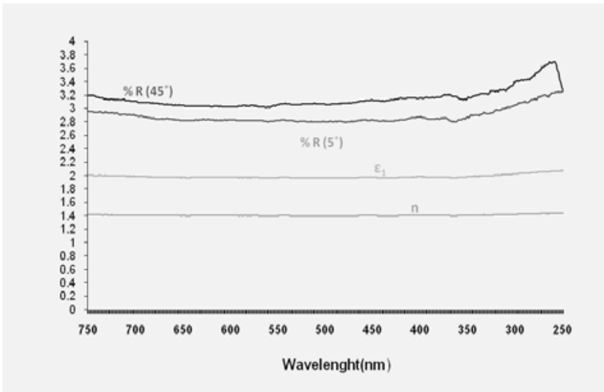

Figure 7. Reflection, refractive and dielectric coefficients spectra of mixed crystal of $KCl_{0.5}Br_{0.5}$ against wavelength in the optical region of 250 -750 nm, irradiated with 0.5 kGy dose of gamma radiation.

- According to the crystal defects on the surface layers of the irradiated samples and heat effects which caused fading process after irradiation of the external surface layers of the crystal change into their initial conditions in a very short period of time.

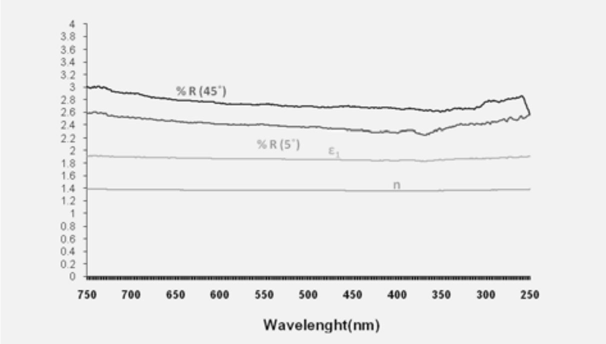

Figure 8. Reflection, refractive and dielectric coefficients spectra of mixed crystal of $KCl_{0.5}Br_{0.5}$ against wavelength in the optical region of 250 -750 nm, irradiated with 1.0 kGy dose of gamma radiation.

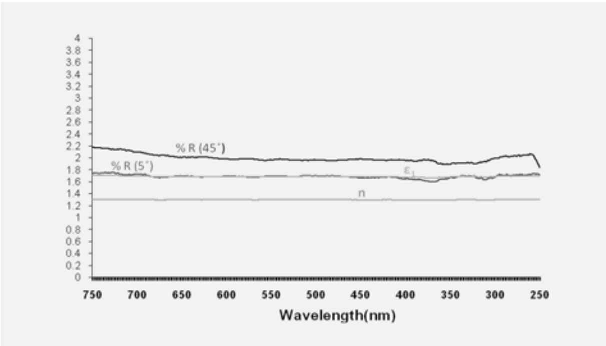

Figure 9. Reflection, refractive and dielectric coefficients spectra of mixed crystal of $KCl_{0.5}Br_{0.5}$ against wavelength in the optical region of 250 -750 nm, irradiated with 1.5 kGy dose of gamma radiation.

However, it prevented the exchange of heat in the internal layers of the crystal at the same time which also avoided the fading process in the interior of the crystals.

- Despite the different variety of the darkness in the bluish purple colour of the single crystals due to the different gamma-ray irradiation doses, no remarkable changes were observed in the reflected spectra of the samples. So, it could be estimated that optical behaviour characteristics of the external and internal surface layers would be different after the irradiation processes.

In fact, it can be claimed that the calculated $\varepsilon(\omega)$ and $N(\omega)$ parameters were related to the surface layers of the crystals and could not be extended to the whole body of the crystals.

The other point which should be mentioned here is that the reflection spectra of these crystals at visible spectral region of 400-700 nm after irradiation processes remained almost constant.

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