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Na_2O doped borate silica glass as a dosimetric material for gamma ray

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Glass series of (45-x) ZnO-45B_2O_3-10SiO_2-xNa_2O, 0.05≤ x ≤ 0.7 were prepared using the melt quenching method. Powdered forms of the chemicals were obtained with each batch weighing 20 g. Borate silica glasses doped with Sodium are examined in term of TL (thermoluminescence) properties to find their possibility to use as radiation dosimeter's glass. The sample with 0.1% Na_2O concentration has a higher TL response among the concentrations for a delivered dose 50 Gy of Co^{60} gamma radiation. The annealing procedure and heating rate are determined. TL response within the dose range of 0.5–4Gy and 10-100 Gy, sensitivity, reproducibility, minimum detectable dose, Effective Atomic Number and fading are investigated. The outcomes indicate that this glass has a possible to be used as a radiation dose evaluation.

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1. Introduction

The usage of glass materials as ionizing radiation detectors proposed by many researchers in different fields due to its many valuable properties such as easy in handling, chemical inertness and rigidity etc. Also, glass can be in contact or very close to the exposed persons and therefore, can be used as emergency dosimeters[1]. Ionizing radiation has many effects on glass structure, one of which is its ability to generate electron/hole pairs which are trapped within the glass. When the glass heated, electron/hole pairs migrate and recombine to release light. Since the emission of light is usually proportional to the radiation dose absorbed, the light released can provide a means by which the exposed dose can be measured. This phenomenon is the basis of thermoluminescence (TL) dosimetry [1, 2]. Borate compounds have attracted much attention due to its high sensitivity, low cost and ease of preparation. Previously studies show that some of borates perfect materials for medical and environmental dosimeters. [3-5]. The creation of compounds containing borate is affected by the inclusion of some metal cation additives and modifying the optical properties of the borate glasses [6-8]. In this report we present the improve TL characteristics of zinc borate silica doping with Na_2O as material for thermoluminescence dosimeter (TLD).

2. Materials and methods

Zinc borate glass system (ZBS) was prepared, the details are described in our earlier paper [2, 9, 10]. Glass series of (45-x) ZnO-45B_2O_3-10SiO_2-xNa_2O, 0.05≤ x ≤ 0.7 were prepared using the melt quenching method [1, 11]. Powdered forms of the chemicals were obtained with each batch weighing 20 g. The batch powder was subjected to mechanical milling for 1 hour and then transferred to an alumina crucible and melted in an electric furnace at temperature 1300 ºC for 1 hour and the details are described in our earlier report[2]. Cobalt-60 gamma radiation was used to irradiate the samples. This source which is available at the Universiti Kebangsaan Malaysia (UKM) has half-life of 5.3 years. The gamma source has characteristic dose rate of 9.234 kGy h^{-1}. The reader model 4500 (Harshaw USA) available at the Material Laboratory, Department of Physics, Universiti Teknologi Malaysia was used to obtain the TL measurements. The data was read out after 24 h of irradiation to prevent the occurrence of shallow electron traps and avoid glow peaks at very low temperatures. Each experimental data point represents an average value of 3 to 5 when reading out the sample's measurements. The TL glass properties, comprising the annealing procedure, optimum concentration and sitting time temperature profile (TTP) heating rate were investigated for the best batches of the proposed glasses [2]. All raw materials for prepare glass compositions are presented in Table 1.

Table 1. The raw materials for prepare glass compositions

Glass	Batches Composition (mol%)				
	ZnO	B_2O_3	SiO_2	Na_2O	
S1	44.95	45	10	0.05	
S2	44.9	45	10	0.1	
S3	44.7	45	10	0.3	
S4	44.5	45	10	0.5	
S5	44.3	45	10	0.7	

3. Result and discussion

3.1. XRD Analysis

Fig. 1 shows the x-ray diffraction pattern of optimum doped samples, it is clear that glass revealed bands with no discrete or continuous sharp peaks. This confirms that the doped glass sample is in non-crystalline phase.

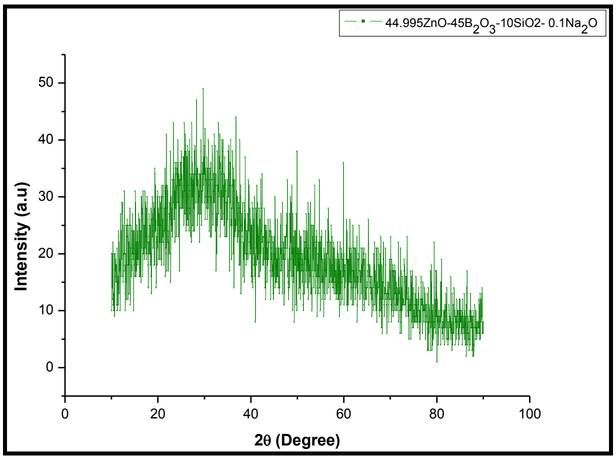

Fig. 1. XRD spectra of doped compound (ZBS:Na_2O) (color online)

3.2. FE-SEM Analysis

(FESEM) analysis usually contains the generation of an X-ray spectrum from the entire scan area of a scanning electron microscope [12, 13]. Fig. 2 shows that the characteristic micrograph of optimum doped ZBS with Na_2O glass samples, dispersion of grains can be detected from the micrograph structure for present glass samples. This shows the existence of permeable structure in the glass.

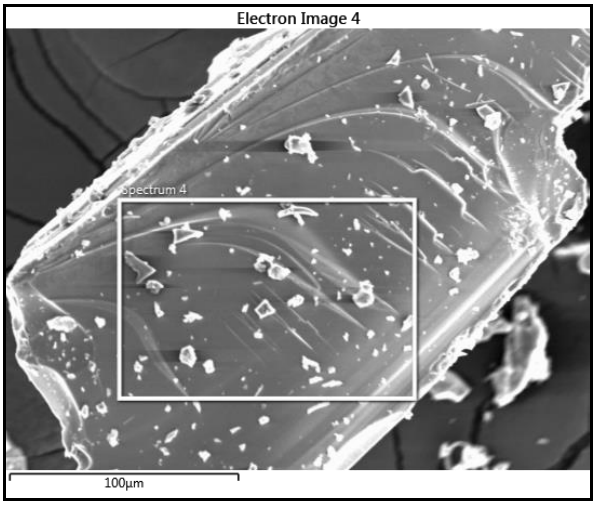

Fig. 2. FESEM micrograph of (ZBS:Na) glass sample

3.3. Optimization of Sample Composition

Borate silica glass samples ZBS doped with Na_2O were prepared with varying mol percentage of Na_2O as listed in Table 1.

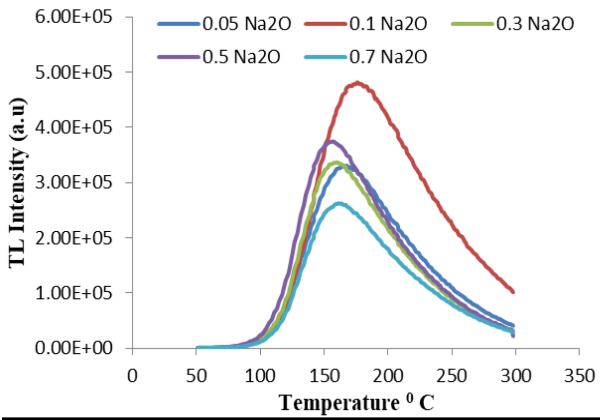

Fig. 3. TL glow curve of doped Na_2O with ZBS glass sample (color online)

The samples exposed to 50 Gy dose of Co^{60} gamma radiation. Figs. 3and 4 show the glow curve intensity and TL response for all samples with different concentrations of Na_2O respectively. A TL peak temperature around (160-175) ^{o} C is observed for the glow curves with a seeming shift in the positions of the glow curves to higher temperature as shown in Fig. 3. The maximum TL response with lowest standard deviation is set up to be the sample with 0.1 mol% Na_2O as shown in Fig. 4, making it the most thermoluminescence efficient one.

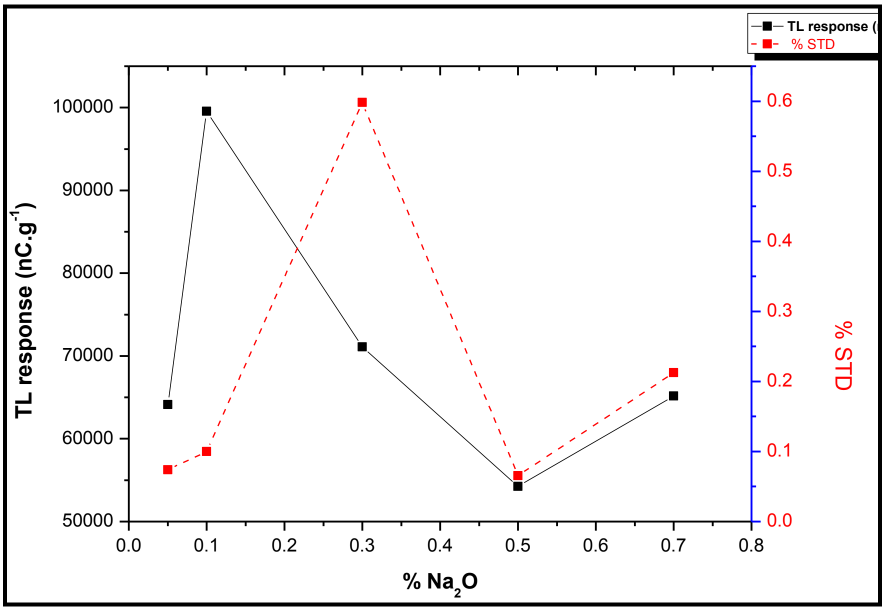

Fig. 4. TL response of doped Na_2O with ZBS glass (color online)

3.4. Annealing Procedure

Annealing procedure of doped glass samples ZBS:Na was carried out in order to obtain the highest sensitivity and to remove previous irradiation [4, 14]. This process was done using several temperatures range of 100 °C - 400 °C at a fixed duration of 30 min for all temperature. Afterward, the samples were exposed to a test dose 50 Gy of Co^{60} gamma ray and TL response was noted. Figure 5 shows the plot of TL response and standard deviation of doped ZBS:Na glass samples against the annealing temperature. From the plot it can be seen that the maximum TL response with lowest standard deviation obtained at temperature of 100 °C. To define the best annealing time, the samples were annealed at a temperature of 100 °C and range of annealing time from 15 min to 90 min. the samples then exposed to 50 Gy of Cobalt-60 gamma radiations. Then, the TL response analysis was carried out. The result is revealed in Fig. 6, it is obvious that the optimum annealing duration is 15 min with a lowest standard deviation of 0.15%.

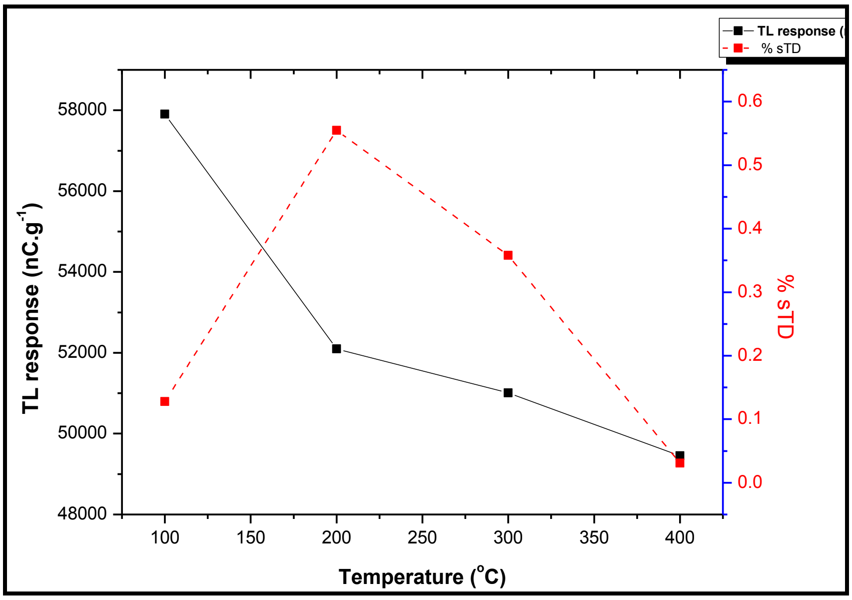

Fig. 5. Variation TL response of (ZBS:Na) with annealing temperature (color online)

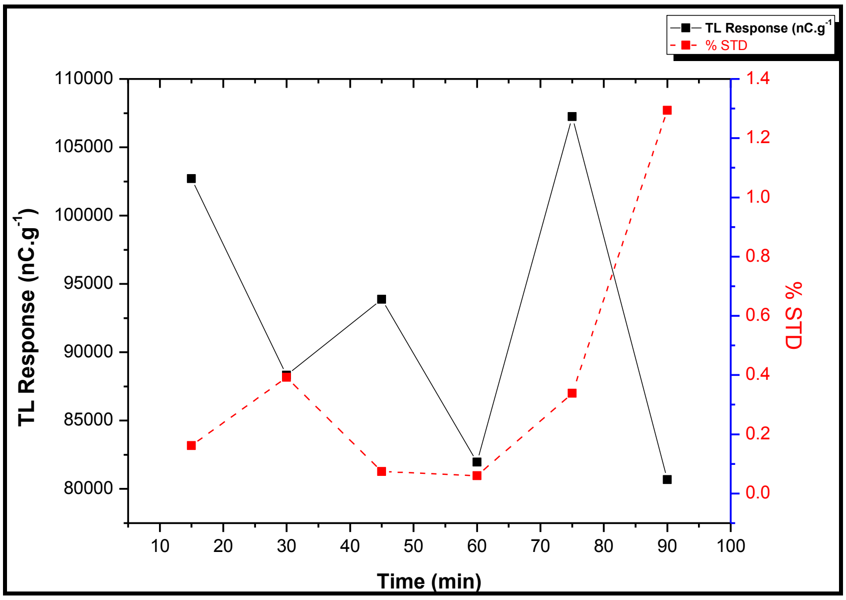

Fig. 6. Variation TL response of (ZBS:Na) with annealing time (color online)

3.5. Heating Rate

The phenomena for thermal quenching of TL intensity because of higher heating rates arise. So, there is requiring for the optimum heating rate at which maximum recombination will happen. This can be refered to the relation between the time required for releasing electrons and the amount of electrons released by the thermal stimulation. The effect of heating rate on TL response of the new phosphor was studied. The effect of various rates of heating rate from (1to 10 °C/s) on the structure of the proposed dosimeter at a fixed dose of 50 Gy , the TL response and standard deviation as a function of heating rate is shown in Fig. 7. The heating rate 3 °C/s is considered as the optimum heating rate in this case since it has maximum TL response with lowest standard deviation.

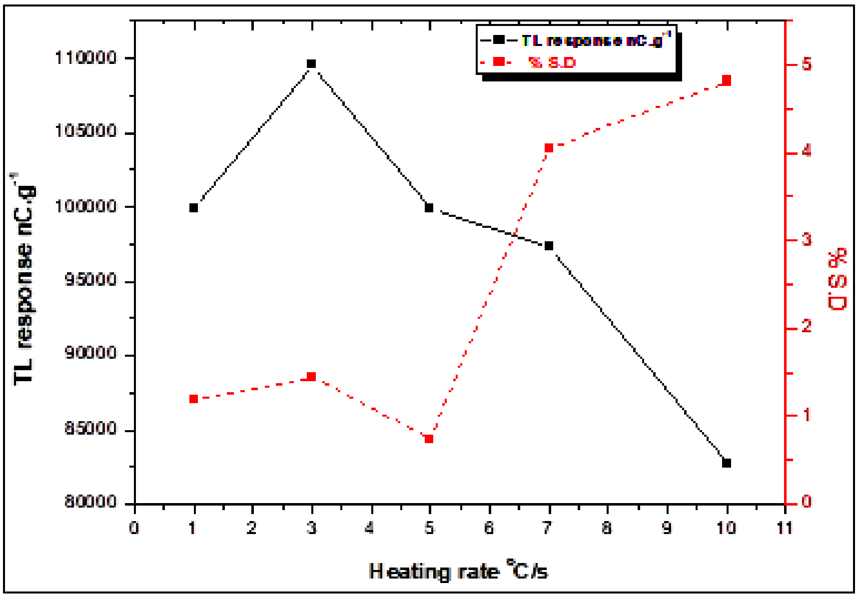

Fig. 7. TL response of (ZBS:Na) with various heating rates (color online)

3.6. Dose Response

The important feature of thermoluminescent materials in dosimetric applications is linear dose response, with the thermoluminscence of a material determined based on a linearity range [15]. The TL responses of ZBS doped with Na_2O within the range 0.5Gy - 4.0Gy and 10Gy – 100Gy doses of Co^{60} gamma radiations represented in figure 8 and 9. The TL response rises linearly with an increase of dose for glass phosphor. This shows good linear dependence of the TL response to the gamma radiation doses. The linearity is a significant advantage for dosimetry application then it confirms accurate valuation of the dose [4, 16, 17].

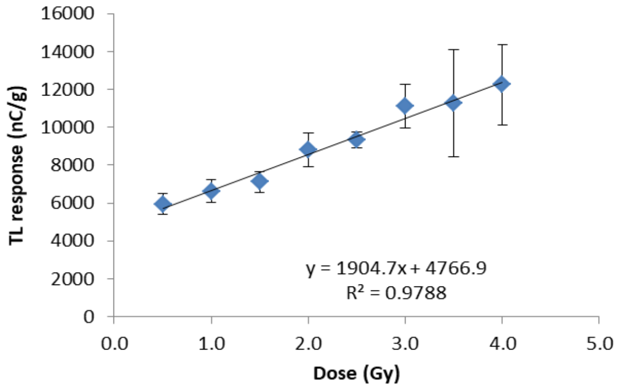

Fig. 8. (ZBS:Na) glass samples exposed to dose range (0.5-4) Gy of Co^{60} gamma ray (color online)

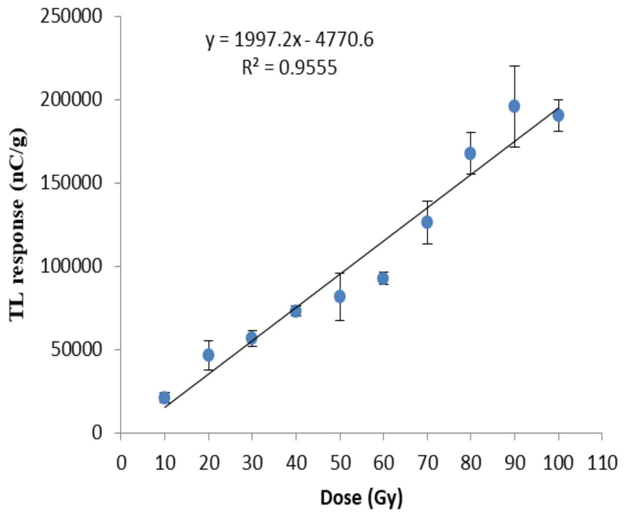

Fig. 9. (ZBS:Na) glass samples exposed to dose range (10-100) Gy of Co^{60} gamma ray (color online)

3.7. Sensitivity

TL sensitivity is defined as the TL intensity per unit mass of the dosimeter and per unit radiation dose [18, 19]. TL sensitivity of any compound under different doses of radiation is identified as the ratio of TL response to the mass of dosimeter and exposed dose. The slope of the graph that plotted between TL response and various doses can serve us to determine the sensitivity of the compound. The sensitivity of the glass sample under study to Cobalt-60 gamma radiation of both ranges (0.5-4) Gy and (10-100) Gy were found to be 1904.7 nC Gy^{-1} g^{-1} and 1997.2 nC Gy^{-1} g^{-1} respectively.

3.8. Reproducibility

The composition (ZBS:Na) glass samples were subjected to nine sequence of irradiation in order to evaluate the reusability of the accessible dose measurements, Co^{60} gamma radiation at a dose of 4 Gy was used for this purpose. The reproducibility characteristics of the current composition under study are displayed in Figure 10, TL signal intensity of the individual dose is approximately the same after numerous measurements. The sample is like not to suffer any physiochemical change due to frequent irradiation and annealing processes. The result confirmed that these glass phosphors are reusable in radiation dose assessment.

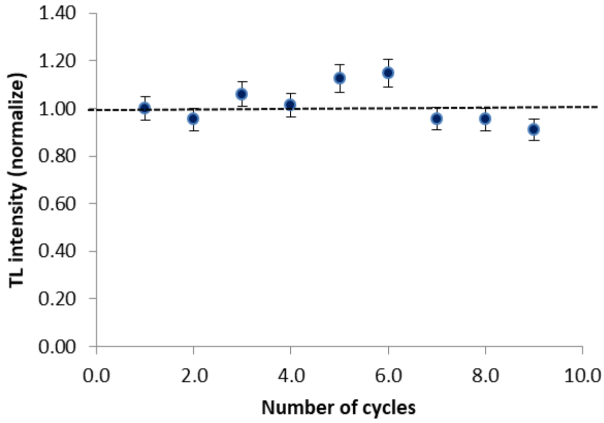

Fig. 10. Reproducibility of ZBS:Na glass samples irradiated with Co^{60} gamma radiation (color online)

3.9. Minimum Detectable Dose

Minimum detectable dose (MDD) is the minimum dose of ionizing radiation which is detected by the dosimeter. The minimum detectable dose (MDD) of the present material was calculated using equation (1) [20].

$$D_o = (B^* + 2\sigma_B)F \tag{1}$$

where D_o is the minimum detectable dose, B^* is the average background TL of the un-irradiated dosimeter. σ_B is the standard deviation of the background signal, and F is the calibration factor expressed in the Gy nC^{-1} and using equation (2).

$$F = \frac{dose(Gy)}{A(nC)} \tag{2}$$

Five samples were annealed and TL readings were recorded without any irradiation. The average background signal (B^*) was found to be 0.773 nC and the standard deviation of average background (σ_B) was found to be 0.055. The samples were irradiated by 4 Gy of Cobalt-60 gamma radiations. By using equation 2, the calibration factor F was found to be 3.8 x 10^{-4} Gy nC^{-1}. Substituting the values of average background, standard deviation of average background and calibration factor in equation 1, the minimum detectable dose (MDD) of the study sample was found to be 0.338 mGy.

3.10. Effective Atomic Number

The effective atomic number (Z_{eff}) of ZBS:Na glass sample was calculated using Mayneord's equation 3 [21].

$$Z_{eff} = \sqrt[b]{a_1(Z_1)^b + a_2(Z_2)^b + a_3(Z_3)^b + \cdots a_n(Z_n)^b} \quad (3)$$

$$a_i = \frac{n_i(Z_i)}{\sum_i n_i(Z_i)} \quad , \; n_i = \left[\frac{N_A . Z_i}{A_i}\right] . W_i \quad (4)$$

where a_i is the fraction of electron for each element to the total electrons of the composition, n_i is the number of electrons in one mole for every element. N_A is the Avogadro's number of $6.023*10^{23}$ mol^{-1}, Z_i, A_i and W_i are the atomic number, mass number and fractional weight of each element. The exponent values b is in a range from 2.94 and 3.5 depending on the energy applied. The value of the effective atomic number for the present glass sample was found to be 21.74.

3.11. Fading

Thermal fading is a decrease of the TL intensity signal due to the effect of ambient temperature after a period. The response of thermoluminescent detectors (TLDs) may show some variations during their storage, both before and after irradiation . At each temperature there is a probability that charge carriers escape from the trapping centres within a TLD, this process is called fading of the TL signal. During the time between annealing and exposure, the defect structures, acting as trapping and recombination centres, may undergo some transformations, leading to changes of sensitivity. The thermal fading of the present compositions under study is investigated at 4 Gy dose of Co ^{60} gamma radiation. The samples annealed and irradiated, then they kept in the dark place at room temperature in order to reduce possible release of trapped electrons by high temperature and light. Fig. 11 shows the plot of the residual signal against storage time up to 6 weeks. It can been seen from the graph that the irradiated glass samples (ZBS:Na) show residual signal reduction by 14.33% after one week, 20.31% after two weeks, 28.73% after three weeks, 30.6% after four weeks, 35.9% after five weeks and 40.4% after six weeks of irradiation at dose 4 Gy.

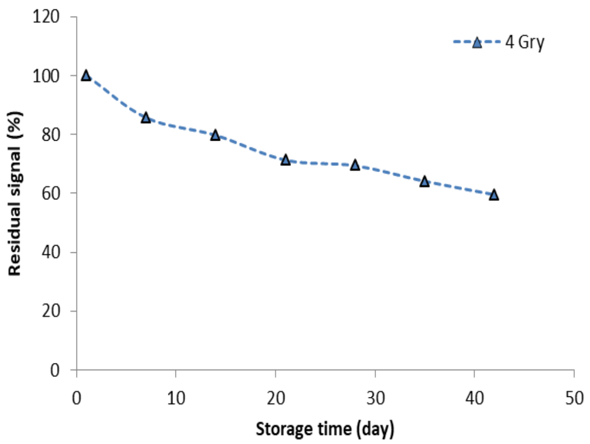

Fig. 11. Fading characteristics of (ZBS:Na) glass exposed to Co^{60} gamma radiation

4. Conclusion

Basic TL dosimeter properties of Borate silica glass doped with various concentrations Na_2O, were prepared using the melt quenching method. The characteristic micrograph of optimum doped ZBS with Na_2O glass samples, dispersion of grains can be detected from the micrograph structure for present glass. Analysis of the TL glass dosimetric properties of the doped revealed that TL peak temperature around (160-175) ^{o} C is observed for the glow curves with a seeming shift in the positions of the glow curves to higher temperature. The optimum sample amoung all concentrations is 44.9ZnO-45B_2O_3-10Si_2O-0.1Na_2O. The best heating rate was found to be 3 °C/s and simple annealing procedure was carried out. Dose response linearity, sensitivity, reproducibility, mimimum detectable dose, Effective atomic number Z_{eff} and thermal fading were achieved. The outcomes propose that present glass phosphor appropriate to use as a TL dosimeter.

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