Determination of Thermoluminescence Kinetic Parameters of Bauxite by Computer Glow Curve Deconvolution Method (CGCD)

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Abstract: The purpose of the present work is to obtain fundamental information on the trapping levels of bauxite by the thermo luminescence TL technique. Studies of the dependence of the glow intensity on excitation dose, isolation of single glow peaks and the determination of the trap parameters are reported. Glow curve obtained for bauxite showed a characteristic peak at about $T \approx 412K$, and the peak height increased with increasing irradiation dose. Glow curve analysis indicated that the glow curve in the temperature region between room temperature and 573K could be well described as a superposition of seven peaks. The seven peaks were described by first-order kinetics. The activation energies "E' and frequency factor "S" for individual glow peaks of bauxite at different irradiation doses were calculated. It was found that the experimental glow curve of bauxite had main dissymmetric peak at about maximum intensity $I_{'m}$= 23897 (arb. unit) and redundancy maximum temperature T_m=415K). Furthermore, the deconvolution of bauxite TL glow curve had one main dosimetric peak (peak 2) at about 409.5 K. This means bauxite can be used as radiation dosimetry for low-level gamma radiation.

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1. Introduction

The thermoluminescence material is thus the material that during exposure to ionizing radiation absorbed some energy which is stored. The stored energy is released in the form of visible light when the material is heated (Bos., 2001).The energy absorbed from ionizing radiation allows electrons to become free and then to move through the crystal lattice, some of them can be trapped at imperfections of the lattice and remain trapped for a short or long period of time according to the energy associated with the defects. Subsequent heating of the crystal can release some of these trapped electrons associated with emission of light. Controlled measurement of the emitted light (glow curve) is normally used for determination of the radiation dose absorbed by the crystal. Furthermore, in order to understand its behavior it is necessary to determine the so-called kinetics parameters (the activation energy of the traps, *E*, the frequency factor, *s*, and the kinetic order of the process, *b*). Most of the methods for determining values of these parameters are based on the analysis of the glow curve. More recently, a computer-based method has been introduced by the computerized glow-curve deconvolution (CGCD) (Horowitz and Yossian., 1995; Berger et al., 2006 and Yazici, & Ozturk., 2003).

There are a few studies considering the thermoluminescence properties of the Bauxite material. (Villarreal-Barajas et al., 2002) Studied the themoluminescence glow curves of aluminum oxide thin films to beta particle radiation. They found that only one peak at 435K; Kortov (2007) studied the thermoluminescence kinetics in dosimetric aluminum oxide crystals. They found that the main dosimetric peak appeared at T_{max} = 450 K and they had established differences in the order of the kinetics within different temperature ranges of the dosimetric peak. Zhang et al., (2006) studied thermoluminescence characteristics of α- Al_2O_3:C ceramics they found three clear TL glow peaks at 405, 493 and 610 K . The present work investigates the physical properties of bauxite as radiation dosimeter, the dependence of the glow intensity on excitation dose, isolation of single glow peaks, and the trap parameters.

2. Experimental Procedure

Samples preparation and irradiation

Samples were packaged in polyethelyne capsule and packaged in a small envelope before irradiation. All samples were 0.5 mg for each reading. The proposed samples were irradiated using Cesium-137 (^{137}Cs) γ –rays ($E\gamma = 0.662$ *MeV*, $t_{1/2}$=30 years) in the radiation protection department, nuclear research center, Egyptian Atomic Energy Authority with absorbed dose rate in air changes with the distance of the samples from the surface of the source. The gammas beam 1.5 Ci (at the date of production 6/1982). Cesium-137 irradiation facility manufactured by Atomic Energy of Canada. The dose rate of the ^{137}Cs γ-source was 1.5 mGy/ hr during the experiment. The samples were irradiated at room temperature by gamma radiation emitted from Cesium-137 with doses 0.2, 0.25, 0.50, 0.75, 1.0 and 1.25 Gy respectively. TL glow curve different doses was measured with a heating rate 10^{o} C/s, using TL Analyzer 4000 Harshaw.

X-ray diffraction (XRD)

The structure of the bauxite samples were checked before use at room temperature by means of X-ray powder diffraction technique (XRD) using Shimadzu Diffractometer XRD 6000, Japan, with Cu-$K\alpha_1$ radiation (λ = 1.54056 Å). The data were collected by step-scan modes in a θ -2θ range between 10° and 80° with step size of 0.02° and step time of 0.6 seconds. Pure Silicon Si 99.9999% was used as an internal standard. Part of the Bauxite has been calcined at 200 °C for one hour in order to investigate the effect of heating of the crystal upon gamma irradiation doses.

Glow Curve Analyzer

In this work we used a new computer program, *GlowFit*, for deconvoluting first-order kinetics thermoluminescence (TL), eq.(1) describes the formula of the model used in “GlowFit” to simulate the glow curve (Bos et al., 1993).

$$I(T) = I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \exp\left(\frac{E}{kT_m}\left(\alpha\left(\frac{E}{kT_m}\right) - \frac{T}{T_m}\exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right)\alpha\left(\frac{E}{kT}\right)\right)\right)$$

……… (1)

Where:

α-, is a quotient of 4th order polynomials (Abramowitz and Stegun,, 1972) ,

I'_m ,T_m are the maximum intensity and temperature respectively

k is Boltzmann constant.

Glow Fit can also determine the quality of fitting between experimental and deconvolution curves, which called figure of merit (FOM).

The main advantage of *GlowFit* is the ability to resolve complex TL glow curves consisting of strongly overlapping peaks such as those observed in heavily-doped LiF:Mg,Ti (MTT) detectors. *GlowFit* is a Microsoft Windows-operated user-friendly program (Puchalska and Bilski., 2006).

Deconvolution and Determination of Trapping Parameters

Information about the behavior of traps in a luminescent material is usually derived by fitting the glow curves in the thermo luminescence spectrum of the material to a first order formula. From the fit one seeks to obtain values for the activation energy E, and the frequency factors s, of glow peak. A comparison between the experimental and theoretical glow curves to correct the derived parameter values. These comparisons to obtain some useful insights of experimentally observed thermoluminescence spectra. Also, computerized glow curve deconvolution (CGCD) program with (*GlowFit*) was used for segregating the Thermoluminescence Dosimetry (TLD) glow curve into its component glow peaks and for determining of its trapping parameters. Overlapped peaks due to the probability of rewrapping. CGCD software is based on a simple model of one electron trap and one hole center, whereas, *GlowFit* depends on deconvoluting first-order kinetics TL glow-curves (Bos., 2006; Chit ambo., 2004).

3. Results & Discussion

The experimental glow curve of bauxite as a function of temperature at different irradiation doses and heating rate 10^{o} Cs^{-1} is presented in Fig.1. The irradiated samples were read out with a harshaw 4000 TLD reader. It can be noticed that the experimental glow curve has a main dosimetric peak at about maximum intensity I'_m= 23897 (arb. units) and maximum temperature T_m=412K . These value was close to those determine by Villarreal-Barajas et al., (2002) for aluminum oxide thin films to beta particle radiation ,the main dosimetric peak at 435K and It was 450 K for aluminum oxide crystal Kortov et al., (2006) and it was at 405 K for α-Al_2O_3:C ceramics according to Zhang et al., (2006).

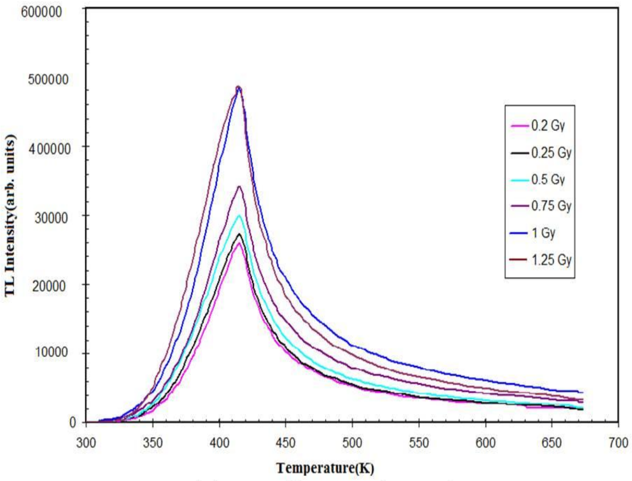

Fig. 1 Experimental glow curve of bauxite as a function of temperature at different irradiation doses and heating rate was 10 °Cs^{-1}.

The intensity was measured at different irradiation doses and heating rate10oCs^{-1} It was analyzed using the computer *GlowFit* program to resolve the individual peaks assuming first order kinetics by CGCD program. Experimental glow curve and its deconvoluted peaks as a function of temperature at fixed heating rate at 10 $^{o}Cs^{-1}$ are illustrated in Fig.2. The glow curves of the sample irradiated by 0.2 Gy were best described as a superposition of seven glow peaks. The solid line gives the experimental glow curve and dashed line show the best fit. The quality of fitting between experimental and deconvolution curves, (FOM) = 3.02%, this value is considered a good fit according to Misra and Eddy (1979); Yazici and Topaksu.(2003).

The recorded experimental glow curve has been deconvoluted into seven peaks by using the aiding program *GlowFit* software. Peak 2 is the main dosimetric peak at about maximum intensity I'_m= 19822 (arb. Units) and maximum temperature T_m=409.5 K. The trap parameters of the dosimetric peak are obtained at the kinetic order b=1, activation energy “E" at 0.928 eve, the frequency factor s at 2.5x1011, and the area under the peak 216764. The evaluated trapping parameters (maximum intensity I_{max}, maximum temperature T_{max}, activation energy *E*, frequency factor *s*, and the area under the peak Area using the program (CGCD) for the seven peaks at irradiation dose 0.2 Gy are shown in the Table (1). The glow curve was best described as a superposition of seven glow peaks with kinetic order b =1, activation energies of (0.739, 0.928, 1.096, 1.172 , 0.717, 0.493 and 0.521eV, respectively) and the frequency factors ,(6.16 $\times 10^{9}$,2.5$\times 10^{11}$, 5.05$\times 10^{12}$, 6.46$\times 10^{12}$, 15875513,16543.16and3635.88.

The TL response as a function of the irradiation dose is shown in Fig.3. From the figure, one can see that, the absorbed dose increased with increasing irradiation dose for 0.2, 0.25 , 0.50 , 0.75 and 1.0 Gy and then decreased. In a thermoluminescence that has been irradiated with an ionizing radiation, there were electrons trapped in the various trapping levels. The release of these trapped electrons on heating and the subsequent light emission when they combined with holes gave a measure of the absorbed dose (Miyoshi et al., 2003) . The trapped electron increases as irradiate dose increase so the TL response increase too up to saturation state at 1 Gray. The trap electrons release of the electrons may take place even in the absence of heating at any increasing of radiation dose more than 1 Gray. This means that bauxite can be used as a radiation dosimeter for gamma radiation low- level.

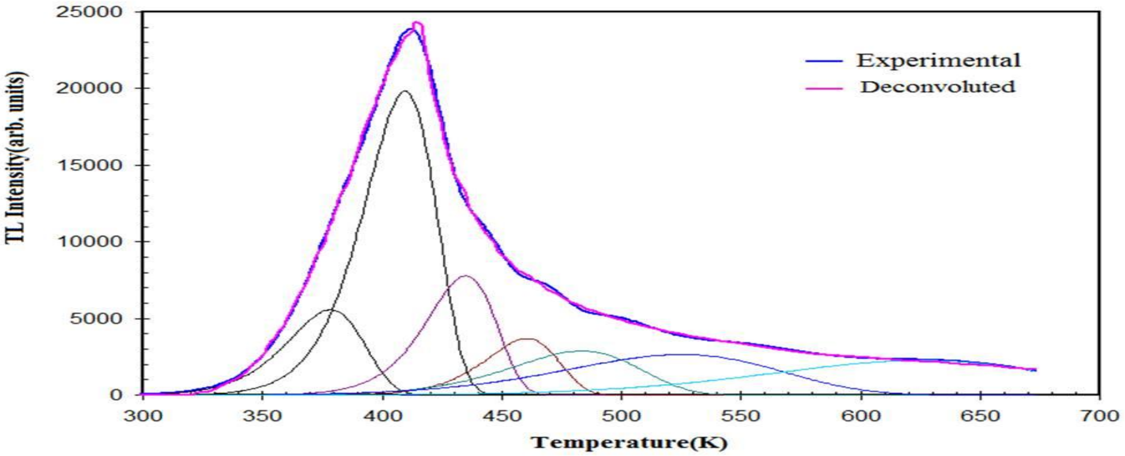

Fig.2 : Bauxite glow curves obtained with a heating rate 10 oCs-1 and dose rate 0.2 Gy.

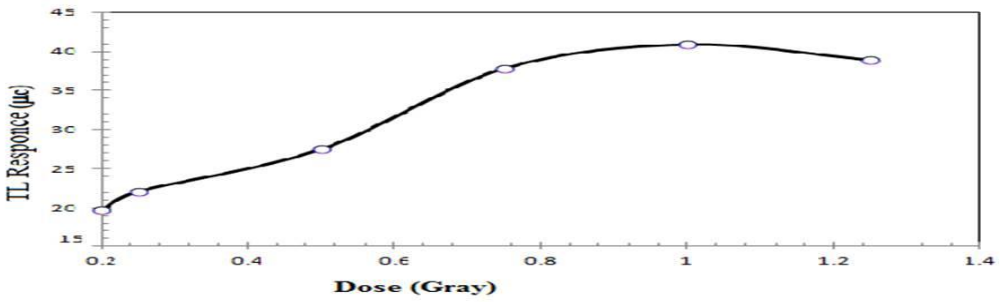

Fig. 3 TL response of bauxite as a function of the absorbed dose

Table 1: The evaluated trapping parameters of Bauxite using the Program (CGCD) with kinetic order b=1.

Peak No.	Max. Intensity **($I\cdot_m$)** (arb. units)	Max. Temp. **T_{max}** (K)	Activation energy **(E) (eve)**	Frequency factor **(s) (s^{-1})**	**Area** under the peak
1	5550	378.4	0.739	6.16×10^{09}	64410
2	19833	409.5	0.928	2.5×10^{11}	216764
3	7780	435	1.096	5.05×10^{12}	81707
4	3683	460.5	1.172	6.46×10^{12}	40563
5	2871	483.6	0.717	15875513	54867
6	2641	525.2	0.493	16543.16	82739
7	2261	625.8	0.521	3635.88	79544

The X-ray diffraction patterns and the Bauxite calcined at 200 °C are shown in Fig. 4. Apparently, the major bauxite hydrated phases, such as gibbsite, boehmite, kaolinite and goethite are presented. The four phases were indexed using the CRYSFIRE and CHEKCELL software. Moreover, Fig.4 also shows that calcinations of bauxite at 200 °C did not change the bauxite structure or decompose its initial phases.

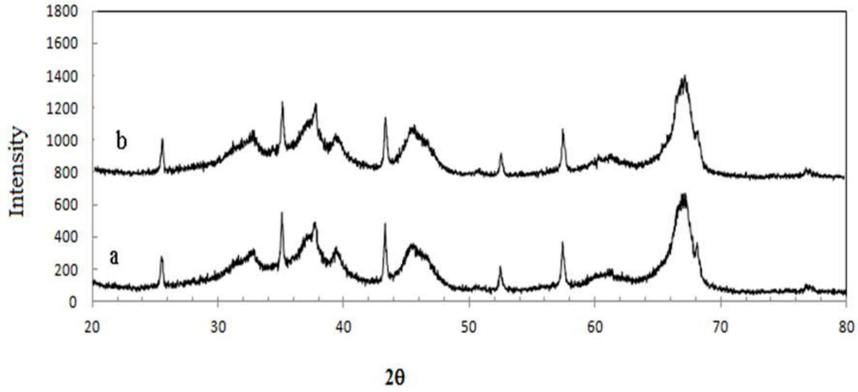

Fig.4: X-ray diffraction patterns of bauxite samples recorded at a) room temperature and b) after calcination at 200 °C for one hour

Conclusion

The kinetic parameters of bauxite thermoluminscence were determined by Computer Glow Curve deconvolution Method (CGCD). The TL responses of bauxite increases with increasing irradiation dose till 1Gy then decreased. The glow curves of the sample irradiated by 0.2 Gy were best described as a superposition of seven glow peaks. The quality of fitting between experimental and deconvolution curves, (FOM) = 3.02% with kinetic order b=1, activation energies of (0.739, 0.928, 1.096, 1.172, 0.717, 0.493 and 0.521 eV , respectively) and the frequency factor, *s* (6.16×10^{9}, 2.5×10^{11}, 5.05×10^{12}, 6.46×10^{12}, 15875513, 16543.16 and 3635.88). The experimental glow curve has main dosimetric peak at maximum intensity I_m= 23897 (arb. Units) and maximum temperature T_m=412K). The deconvoluted Bauxite TL glow curve has one main dosimetric peak (peak 2) at about 409.5 K. This means that bauxite can be used as radiation dosimeter.

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