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Thermoluminescence Properties of Quartzite Rock after β-irradiation

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Abstract. In the present study, the thermoluminescence characteristics of natural quartzite collected from Karaisalı (Adana) in Turkey were investigated after β irradiation at room temperature with the purpose to use as radiation dosimetry. The glow curve of quartzite mineral shows two peaks at 110 ºC and 250 ºC and a shoulder at 375 ºC, explicitly. The resulting peaks, which were examined using the computer glow curve deconvulation (CGCD) method, were deconvuluted and kinetic parameters (activation energy E_a, frequency factor (*s*) and kinetic degree (*b*)) were determined. After CGCD methods, it was seen that the glow curve suporposed at least eight peaks. Additionally, kinetic parameters were determined using by Arrhenius plot obtained from initial rise (IR) method. In this study, it is shown that there is a correspondence between kinetic parameters calculated by IR method and peaks deconvoluated by CGCD technique in low temperature region (P1-P3). Additionally, reusability test was conducted to investigate its usability as a dosimetric material, and a change of 2% was observed in the results of 10 repeated times. Besides to the characterization of this sample X-ray diffraction (XRD) and scanning electron microscopy (SEM) - energy dispersive X-ray (EDX) results were investigated.

***Keywords*:** Quartzite, Thermoluminescence, β dose, Activation energy.

β Işınlaması sonrası Kuvarsit Kayacının Termolüminesans Özellikleri

Özet. Bu çalışmada, Türkiye'de Karaisalı (Adana)'dan toplanan doğal kuvarsit kayacının termolüminesans karakteristiği, bir radyasyon dozimetresi olarak kullanmak amacıyla oda sıcaklığında β ışınlaması yapıldıktan sonra incelenmiştir. Kuvarsit mineralinin ışıma eğrisi belirgin bir şekilde 110 ºC ve 250 ºC'de iki tepe noktası ve 375 ºC 'de bir omuzu göstermektedir. Bilgisayarla Işıma Eğrisi Ayrıştırma (CGCD) yöntemi kullanılarak tepeler ayrıştırılmış ve kinetik parametreler (aktivasyon enerjisi E_a, frekans faktörü (*s*) ve kinetik derece (*b*)) belirlenmiştir. CGCD yönteminden sonra, bu ışıma eğrisinin en az sekiz tepeden oluştuğu görülmüştür. Ayrıca IR yönteminden elde edilen Arrhenius grafiği yöntemiyle kinetik parametreler belirlendi. Bu çalışmada, düşük sıcaklıktaki tepe değerleri için CGCD tekniği ile ayrılan tepeler ile (P1-P3) IR yöntemi ile hesaplanan kinetik parametreler arasında bir ilişki olduğu gösterilmiştir. Ek olarak, dozimetrik materyal olarak kullanılabilirliğini araştırmak için tekrar kullanılabilirlik testi yapılmış ve 10 kez tekrarlanan sonuçlarda % 2'lik bir değişim gözlenmiştir. Bu çalışmaların yanında kuvarsit örneğinin karakterizasyonu için X-ışını kırımı (XRD) ve taramalı elektron mikroskobu (SEM-) enerji yayılımlı X-ışını analizi (EDX) sonuçları incelendi.

***Anahtar Kelimeler*:** Kuvarsit, Termolüminesans, β doz, Aktivasyon enerjisi.

1. INTRODUCTION

Thermoluminescence (TL) method is based on radiation energy absorbed by traps in insulators or semiconductors. This method has been used different applications, such as radiation dosimetry retrospective dosimetry, including personal monitoring, environmental monitoring [1-3]. Quartz is one of the natural minerals used in radiation studies frequently [4-7]. The

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luminescence emitted from quartz is complex and shows a variety of different components with diverse physical properties [8,9]. The diversity of the luminescence emitted from the quartz depends on the defects of the inner or impurity atoms.

As a result of examination of natural minerals by TL method, they proved to be useful as accidental or retrospective dosimetric material [10,11]. Natural minerals have varying TL properties depending on their genesis, geological history, impurity contents and chemical composition [12]. Quartz, one of the basic working materials of luminescence studies, has many different applications in the field, ranging from dating studies to food irradiation control [13, 14].

This paper focuses on the dosimetric properties of quartzite. For investigating dosimetric properties, it is aimed to investigate beta-ray dose responses, reusability test and calculation kinetic parameters using computerized glow curve deconvolution (CGCD) and initial rise (IR) methods by analyzing TL emission curves of quartzite rock. The structural characterization of the samples was supported by results of X-ray diffraction (XRD) method, scanning electron microscope (SEM) images and energy dispersive X-ray spectroscopy (EDS or EDX) techniques.

2. MATERIALS AND METHODS

2.1. Sample

The sample was taken from the Koçyazı formation between Karaisalı-Kozan area (North of Adana). The type locality of the formation is to the north of Feke (Adana) and have a thickness of up to 600 meters. The unit contains pink colored, medium bedded quartzarenite. The petrography of the quartzite sample shows that the sample composed of mainly quartz and minor mica (phengite) minerals with granular texture and a silicate matrix. The quartz grains have rounded edges indicating a transportation by fluids such as river or sea, where as the mica grains have needle-like shape.

2.2. X-ray diffraction (XRD) analysis

XRD analysis was used to identify the crystal structure. XRD patterns were recorded with a Cu-Kα radiation (λ = 1.5406 Å) on a Rigaku SmartLab brand X-ray diffractometer at a 40 kV potential and 30 mA current with a scan rate of 5 °min^{-1} at 10 °≤2θ≤90 °. XRD patterns of the studied sample are shown in Figure 1 and results obtained from the pattern are also indicated in Table1. In this sample was found to be quartzite and phengite minerals (Figure 1 and Table 1). Phengite is a series name for dioctahedral micas of composition $K(AlMg)_2(OH)_2(SiAl)_4O_{10}$, similar to muscovite but with addition of magnesium [15]. Quartzite sample space Group: P3121and phengite sample space Group: C12/c1. It's in the mineral 95.9% quartz-low and 4.1% phengite 2M1. Miller indices of the peaks were showed on the peaks in the Figure 1. (Q: Quartz-low and P:Phengite).

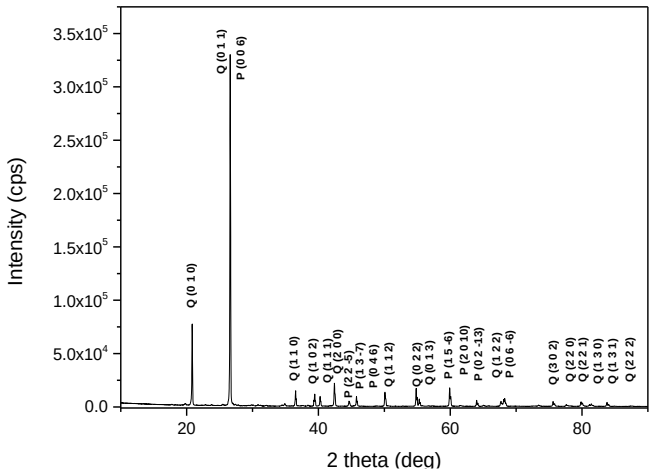


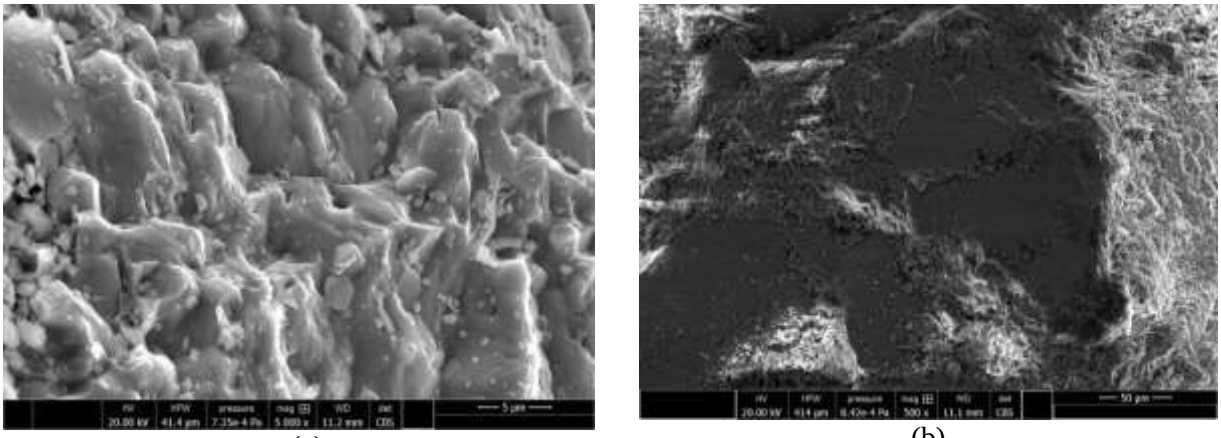
Figure 1. XRD graph for quartzite.

Table 1. The result of XRD patterns for natural quartzite.

	Formula	a	b	c	α	β	γ	V
Quartz low	SiO_2	4.919679	4.919679	5.412144	90	90	120	113.441
Phengite 2M1 (Fe-rich)	$H_{1.88}Al_{2.41}F_{0.12}Fe_{0.34}K_{0.94}Mg_{0.04}Na_{0.03}O_{11.88}Si_{3.26}$	5.225131	9.064596	20.031205	90	95	90	943.985

2.3. Scanning electron microscopy with energy dispersive X-ray spectrometry (SEM-EDX) analysis

In this study, SEM technique was used to obtain information about surface morphology and size distribution of quartzite. The obtained characterization of the sample by SEM together with EDX analysis (FEI Quanta 650 Field Emisssion SEM). EDX has been conducted in order to investigate the elemental composition of the material. In this process, sample was coated with gold using a fine coating ion sputtering device (Quorum-Q150R ES) to a thickness of 100 $^{o}Amin^{-1}$. Figure 2 (a) and (b) show the SEM images of the sample used in measurements for two different resolutions 5 µm and 50 µm, respectively. Figure 3 shows the emission spectra of this sample. The elemental composition obtained are tabulated in Table 2.



(a) (b)

Figure 2. SEM images of qurtzite sample from Karaisalı region (5 and 50 µm, respectively (a) and (b)).

Table 2. List of elements and their composition obtained in EDX analysis of quartzite.

Element	Weight %	Atomic %	Net Int.
Si	27.66	18.34	5507.35
O	48.52	56.46	2420.55
Al	3.07	2.12	562.84
Ca	1.67	0.77	200.22
K	1.42	0.68	196.51
C	11.32	17.54	159.98
Au	1.8	0.17	125.8
Mg	0.72	0.55	107.56
P	0.65	0.39	88.71
F	2.46	2.41	79.57
Na	0.7	0.57	57.76

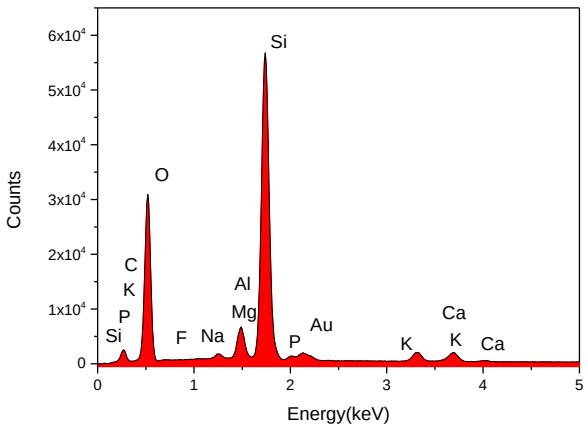


Figure 3. EDX spectra of quartzite sample.

2.4. Thermoluminescence (TL) Measurements

The TL measurements were carried out using an automatic Lexsyg Smart TL/OSL reader system which allows up to 40 samples having an internal EMI 9235QB Hamamatsu bialkaline photomultiplier tube (PMT). A filter (IRSL, TL wideband blue: BG39 + BG25 + KG3) was used in front of the PMT. Dose rate is 0.115 Gy/s. The heating system is able to heat the samples up to 700 ºC. In this study the sample was heated from room temperature (RT) to 450 ºC to obtain TL glow curves.

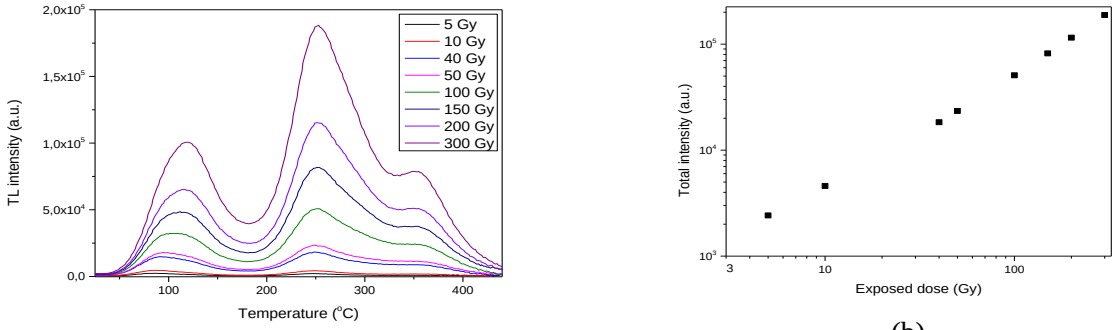
Samples were pestled in an agat mortar to avoid tribouminescence [16] then they were sieved the size range 90 and 140 µm and ~20 mg samples were pressed to obtain pellet by being held under a 2 ton pressure for 20 minutes. So, 8 mm diameter thin sample were used in TL experiments.

3. RESULTS

3.1. Dose response experiment

In this study, the glow curve readouts were performed at a linear heating rate (HR) of 2 °Cs^{-1} from room temperature (RT) to 450 °C. A gas flow of nitrogen was used to reduce the oxidation of the heating element during the readout. Also the background was subtracted from all TL data. Figure 4 (a) shows the variation of TL glow curves of sample as obtained after being exposed to beta particle irradiation in the dose range from 5 Gy up to 300 Gy at RT. For dosimetry applications the integrated area under the glow curve has to be proportional to the radiation dose.

The TL glow curves shows three peaks at approximately 110, 250 and 375 ºC (Figure 4 (a)). The peak with a maximum TL intensity is around 375 ºC. 110 and 250 ºC are seen directly but the other peak at 375 ºC has a shoulder form. The genarally known that TL glow peaks of quartz have occuring at 110, 160, 220, 325 and 375 ºC [17]. Due to the complexity of the luminescence emitted from the quartz, it is related to the diversity of quartz defects with regard to internal (e.g., Si and O vacancies) or impurity atoms (e.g., Al or Ti) [18]. It is also seen from Figure 4 (b) that the total area of glow peaks between RT and 450 ºC shows linear behavior (f(D)=1). Linearity results indicates good results (R^2 =0.99948).



(a) (b)

Figure 4. (a) Dose response curve results for quartzite sample 5- 300 Gy, heating rate 2 ºCs^{-1} (b) Dose linearity graph.

In order to characterize a TL material, kinetic parameters such as activation energy (E_a), kinetic order (b) and frequency factor (s) of its all glow peaks must be determined. The CGCD analysis has been widely used since 1980 to resolve a complex thermoluminescent glow curve into individual peak components. A lot of programme can be used to solve overlapping TL peaks for example GlowFit programme can be used only first degree glow peaks [19], TAMTAM programme [20] to solve overlapping peaks at first, second and general-order kinetics in dos system, TLanal software can decompose first, second and general order degree and mixed order

peaks in windows systems. In the given study, in order to determine the TL kinetic parameters of natural quartzite, the TLanal programme [21] was used as a CGCD method. This program has been used lot of studies [22-24]. After many attempts, it was found that the structure of the natural quartzite sample could be defined by at least eight glow curves (Figure 5). The three of these peaks (P1- P3) are used in the lower temperature region and the other three peaks (P4-P8) in higher temperature region. Kinetic paramaters E_a, *lns* and *b* using CGCD method were given in Table 3.

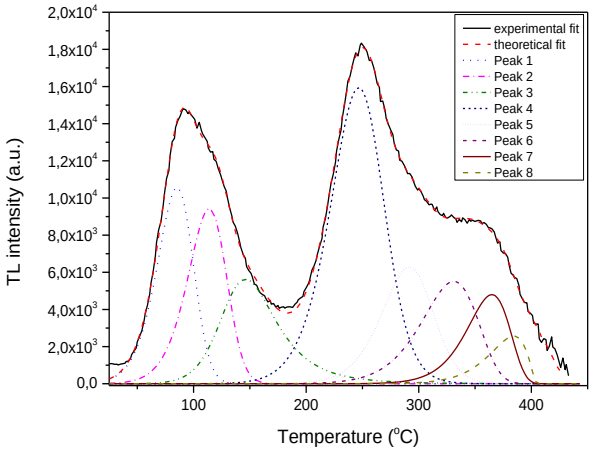


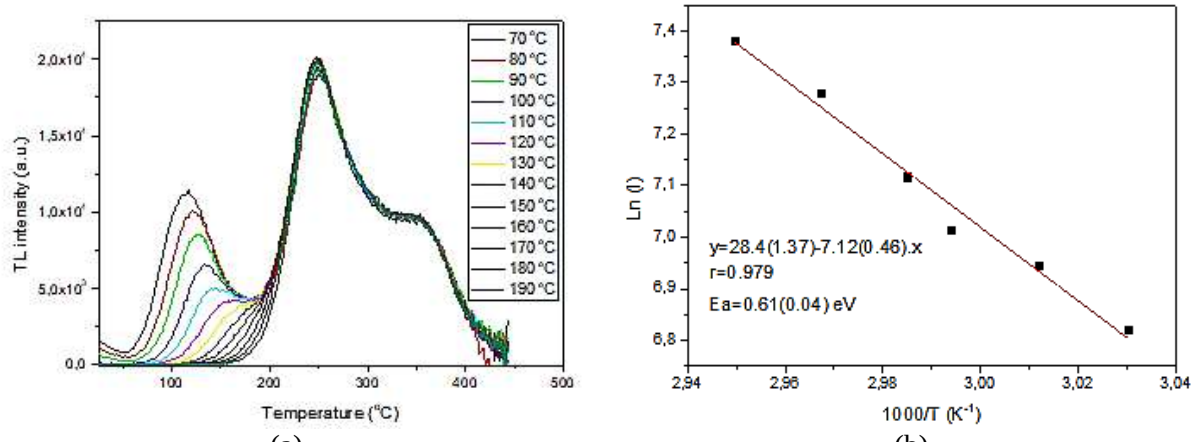
Figure 5. Deconvoluted TL glow curve for quartzite sample (Dose: 40 Gy, β=2 °Cs^{-1} FOM=1.8%).

Table 3. E_a, *lns* and *b* parameters of TL glow peaks of beta irradiated quartzite sample.

Peaks	*b*	E_a (eV)	*lns* (s^{-1})
P_1	1.20	0.73	21.78
P_2	1.17	0.78	21.57
P_3	2.62	0.97	24.84
P_4	1.40	1.13	23.03
P_5	1.36	1.38	26.09
P_6	1.00	1.23	21.10
P_7	1.01	1.76	29.82
P_8	1.02	1.70	27.77

3.2. Initial Rise (IR) Method

The kinetic parameters of overlapping dosimetric TL peaks were also tested using the IR method. This method is based on the number of trapped electrons (n_o) can be changes by only a small amount and thus it can be regarded as constant, so that the first- and general-order TL equations are simplified as $I(T)=constant\ x\ \exp(-E_a/kT)$, and the TL intensity is regardless of any kinetic parameter (*b*). In this equation, *I(T)* is *TL* intensity, k is the Boltzmann's constant and T is the temperature. After this results it's obtained Arrhenius plot (*ln*(*I*) versus *1/T*). This plot would yield a straight line with a slope of $-E_a/k$ can be calculated E_a, moreover *s* value can be calculated by means of the intercept (*ln(s/β)* on this graph. This method can be used in the initial region of the TL signal up to ~15% of peak maximum (I_m) [25].



(a) (b)

Figure 6. (a) TL glow curves after different thermal heatings from 70 to 190 °C with step of 10 °C (b) The graph of Arrhenius to after 70 °C preheat.

In this process, first step a selected exposure dose irradiation, second step relaxation and third step heating from RT to a chosen temperature T_{stop} and then it's obtained TL glow curve. Figure 6 (a) shows changes in the TL glow curves of 40 Gy (β=2 $^{o}Cs^{-1}$) irradiated samples after different thermal pretreatments from 70 °C to 190 °C in step of 10 °C. In this study, the T_m-T_{stop} study was carried out for low temperature TL glow curves (P1-P3). The logarithmic plot of TL intensity as a function of 1/T results were given in Figure 6 (b). Consequently, calculated E_a and s values are given in Table 4.

Table 4. Calculated activation energies (E_a in eV) and frequency factor (lns in s^{-1}) using the IR method.

Quartzite Tstop (^{o}C)	IR Range (^{o}C)	E_a (eV)	lns (s^{-1})
70	56-66	0.61±0.04	30.36
80	66-74	0.61±0.07	29.53
90	74-83	0.82±0.04	33.78
100	83-91	0.96±0.09	39.88
110	90-98	0.98±0.12	39.73
120	101-110	0.82±0.08	33.72
130	112-120	1.05±0.07	39.94
140	124-132	0.97±0.17	36.6

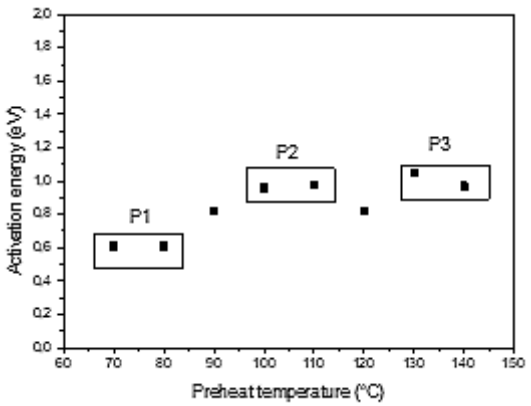


Figure 7. The application of IR method analysis to experimental data in Fig.6 (a).

Figure 7 shows the variation in the preheating temperatures versus the activation energies obtained with the IR method. In this graph each plateau shows a glow peak. So it can be seen that the complex glow curve is composed of three single glow peaks for low temperature region.

3.3. Reusability

Reusability is the best choice to determine the dosimetric property of the material by determining whether the luminescence signal observed in the material undergoes any change under the same conditions. Reusability of a good dosimetric material should have less repeated measurements than 5% under the same dose and reading condition [26]. In this study, the irradiation process of 40 Gy was repeated ten times for sample in the same conditions and the irradiated samples were used to observe the influence of repeated measurements on TL glow curves recorded with an increment of linear heating rate of 2 $^{o}Cs^{-1}$. The maximum variation range between the sequential measurements was less than 2% for sample (Figure 8). Therefore, these results show that this natural mineral is reusable in radiation dose assessment.

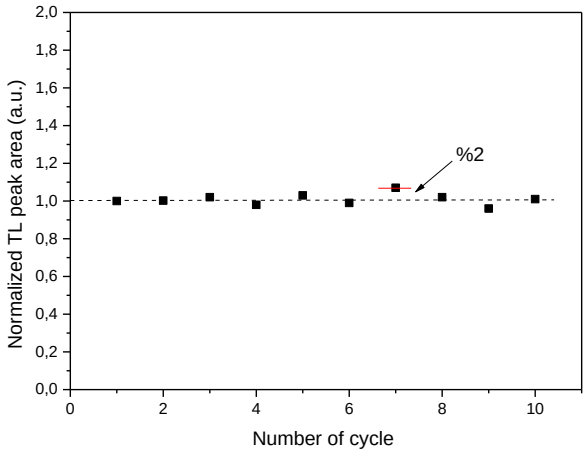


Figure 8. Reausubility result of quartzite sample.

4. CONCLUSIONS

For the characterization of the quartzite rock from the Karaisalı (Adana) region, quartzite and phengite minerals were found as a result of SEM-EDX and XRD measurements. Beta doses between 5-300 Gy were used to investigate its utility as a radiation dosimetry. When the change in the dose value versus the total signal intensity was examined, a linear change was observed for all doses given. TL glow peak which were irradiated 40 Gy β= 2 $^{o}Cs^{-1}$ is deconvoluated by

using CGCD methods and it was observed that the glow curve structure of this sample is eight glow peaks. Besides, E_a and *lns* were calculated for this method. These calculations were repeated for the decomposed peaks in the low temperature region by the IR method to compare these obtained results. It was observed that the results were consistent with each other. The reusability test was performed to observe signal changes in the repeated use of the dosimetric material. The maximum change was observed at 2%, which shows us that this sample can be used as a suitable dosimetric material.

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REFERENCES

[1]. McKeever S.W.S., Moscovitch M., Townsend P.D., Thermoluminescence Dosimetry Materials: properties and uses, (1995) Nucl. Technol. Publishing, Asford.

[2]. Göksu H. Y., Regulla, D. and Drexler, G. Present status of practical aspects of individual dosimetry. Part II: East European Countries, Radiation Protection–78 Part I, II (Luxemburg: Commission of the European Communities) Belgium (1994).

[3]. Planque G.D. and Gesell T.F., Thermoluminescence dosimetry—Environmental applications, The International Journal of Applied Radiation and Isotopes, 33-11 (1982) 1015-1034.

[4]. Aitken M. J., Thermoluminescence dating, (1985), Academic Press, London.

[5]. Scholefield R. B., Prescott J. R., Franklin A. D., Fox P. J., Observations on some thermoluminescence emission centres in geological quartz, Radiation Measurements, 23 2-3 (1994) 409-412.

[6]. Toktamiş H., Necmeddin A. Y., Topaksu M., Investigation of the stability of the radiation sensitivity of TL peaks of quartz extracted from tiles, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 262 (1) (2007) 69-74.

[7]. Yüksel M., Thermoluminescence and dosimetric characteristics study of quartz samples from Seyhan Dam Lake Terracces, Canadian Journal of Physics, In press, doi.org/10.1139/cjp-2017-0741.

[8]. Brito Farias T. M. D., Watanabe S., A comparative study of the thermoluminescence properties of several varieties of Brazilian natural quartz, Journal of Luminescence, 132 (10) (2012) 2684-2692.

[9]. Subedi B., Oniyab E., Polymeris G.S., Afouxenidis D., Tsirliganis N.C., Kitis G., Thermal quenching of thermoluminescence in quartz samples of various origin, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 269 (6) (2011) 572-581.

[10]. Cosma C., Timar A. , Benea V., Pop I., Jurcut T., Ciorba D., Using natural luminescent materials and highly sensitive sintered dosimeters MCP-N (LiF:Mg,Cu,P) in radiation dosimetry, Journal of Optoelectronics and Advanced Materials, 10 (3) (2008) 573 – 577.

[11]. Trindade N. M., Kahn H., Yoshimura E. M., Thermoluminescence of natural $BeAl_2O_4:Cr^{3+}$ Brazilian mineral: Preliminary studies, Journal of Luminescence, 195 (2018) 356-361.

[12]. Macedo Z.S., Valerio M.E.G., de Lima J.F., Thermoluminescence mechanism of Mn^{2+}, Mg^{2+} and Sr^{2+} doped calcite, Journal of Physics and Chemistry of Solids, 60 (1999) 1973-1981.

[13]. Fleming S.J., Study of Thermoluminescence of Crystalline Extracts from Pottery, Archaeometry 9 (1966) 170-173.

[14]. Kitis E., Zaragoza C.E., Furetta C., Thermoluminescence properties of Chile Guajillo (paprika) Mexicano. Applied Radiation and Isotopes, 63 (2) (2005) 247-254.

[15]. Availible at: https://en.wikipedia.org/wiki/Phengite
[16]. Garcia-Guinea J. and Correcher V., Luminescence spectra of alkali feldspars: influence of crushing on the ultraviolet emission band, Spectrosc. Lett., 33 (2000) 103-113.
[17]. Murray A.G. and Wintle A.S., Luminescence sensitivity changes in quartz, Radiat. Meas., 30 (1) (1999) 107-118.
[18]. Preusser F., Chithambo M.L., Götte T., Martini M., Ramseyer K., Sendezera E.J., Susino G.J., Wintle A.G., Quartz as a natural luminescence dosimeter, Earth-Science Reviews, 97 (1–4), (2009) 184-214.
[19]. Puchalska M. and Bilski P., GlowFit—a new tool for thermoluminescence glow-curve deconvolution, Radiation Measurements, 41 6 (2006) 659-664.
[20]. Bos A.J.J., Piters, J. M., Gomez Ros J.M. and Delgado A., 1993. (GLOCANIN, an Intercomparision of Glow Curve Analysis Computer Programs) IRI-CIEMAT Report, pp. 131-93-005 IRI Delft.
[21]. Chung, K.S., Choe, H.S., Lee, J.I., Kim, J.L. and Chang, S.Y., A computer program for the deconvolution of thermoluminescence glow curves. Radiation Protection Dosimetry, 115 (2005) 1-4.
[22]. Topaksu M., Yüksel M., Dogan T., Nur N., Akkaya R., Yegingil Z., Topak Y., Investigation of the characteristics of thermoluminescence glow curves of natural hydrothermal quartz from Hakkari area in Turkey, Physica B: Condensed Matter, 424 (1) (2013) 27-31.
[23]. Joseph Daniel, D., Kim, H. J., Kim S., Synthesis, X-ray, and thermoluminescence properties of $Li_3K_3Y_7(BO_3)_9$, Ceramics International, 44 (7) (2018) 8184-8189.
[24]. Yuksel M., Dogan T., Balci-Yegen S., Akça S., Portakal Z.G., Kucuk N., Topaksu M., Heating rate properties and kinetic parameters of thermoluminescence glow curves of La-doped zinc borate, Radiation Physics and Chemistry, 151 (2018) 149-155.
[25]. Furetta C. and Weng P.S., Operational Thermoluminescence Dosimetry, (1998), World Scientific, Singapore.
[26]. Furetta C. (2003). Handbook of Thermoluminescence, (2003), World Scientific, Singapore.