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## **Radiation Physics and Chemistry**



journal homepage: www.elsevier.com/locate/radphyschem

## Comparative study of the thermoluminescence properties of natural metamorphic quartz belonging to Turkey and Spain



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### HIGHLIGHTS

• The thermoluminescence (TL) peak of metamorphic guartzes was investigated.

• Comparable differences were seen between low and high dose levels.

AD and CGCD methods were used

#### ARTICLE INFO

Article history Received 1 September 2013 Accepted 22 October 2013 Available online 30 October 2013

Keywords. Metamorphic quartz Thermoluminescence Doselinearity Pre-heat

### ABSTRACT

The aim of this study is to investigate the sensitization of the thermoluminescence (TL) peak of metamorphic quartzes from Adiyaman in Turkey (TMQ) and from Madrid in Spain (SMQ). Quartz samples of two different origins were  $\beta$ -irradiated between ~6.689 Gy and 4816 Gy at room temperature. X-ray diffraction analysis has indicated that both TMQ and SMQ have the same crystal structure. Chemical analyses of both TMQ and SMQ were performed using the XRF technique. The preheat processes were carried out at 125 °C for 10 s in the TL measurement. TMQ and SMQ samples have different TL properties in two ways. First TMO has four first order TL glow peaks while SMO has five first order TL peaks and secondly, the observed dose sensitivity of TMQ samples is higher than the SMQ samples.

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### 1. Introduction

Ouartz, as one of the most abundant minerals, is an important rock-forming mineral. It is estimated that about 12% of the mass of the Earth's crust is made of it. Quartz is chemically an almost inert and passive substance at the surface and a very active agent under conditions deep within the Earth's crust. The changes of pressure (P)-temperature (T) conditions, natural irradiation or alteration cause variations in the structure of quartz during the hydrothermal and metamorphic processes (Gotze, 2009). At higher temperatures and pressures, it participates in many complex chemical reactions during rock and mineral formation. Metamorphosis is the only major process in which quartz is either produced or consumed and it disappears from the environment during the formation of new minerals (<http://www.quartzpage.de/gen\_rock. html>, 2013). New formation of natural crystal known as the

0969-806X/\$ - see front matter © 2013 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.radphyschem.2013.10.014

metamorphic quartz generally in aquatic media occurs at high pressures and temperatures as respectively > 20 MPa and > 200 °C up to 6000 bars and temperatures over 500–600 °C (Gotte et al., 2011).

Quartz, one of the natural dosimeters used in luminescence studies, has great importance by means of mineral formation for quantifying the radiation history of materials in a variety of applications such as testing the authenticity of art objects, nuclear accident dosimetry (Bailiff et al., 2000), food irradiation control (Yazici et al., 2008) and dating of archeological materials and sediments (Adamiec, 2005; Porat et al., 2007; Preusser et al., 2009). Using quartz as a dosimeter, it is important to characterize it in terms of general properties such as dose response, preheat related to it's mineral structure (Pagonis et al., 2002; Toktamis et al., 2007; Topaksu et al., 2013).

The dosimetric characteristics of TL materials mainly depend on the kinetic parameters quantitatively describing the trapping-emitting centers responsible for the TL emission. The studies of natural/ synthetic quartz show that quartz displays a number of TL peaks when it was heated from room temperature up to 500 °C after irradiation (Spooner and Questiaux, 2000; Yazici and Topaksu, 2003).

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The shock-metamorphosed quartz, caused by the impact of an asteroid or meteor quartz, showed TL properties with maximal at 365 nm, 470 nm and 610–680 nm band. Electron and hole centers which originate from vacancies including those from the substitution of  $Al^{3+}$  and/or  $Fe^{3+}$ , for  $Si^{4+}$  by the electron paramagnetic resonance (EPR) have been found in them (Serebrennikov et al., 1982). Mineralogical formation, crystallinity index and the content of the impurities seem to be the main parameters of influence in the shape intensity of the cathodoluminescence (CL) and TL glow curve emission of hydrothermal and metamorphic quartzes (Topaksu et al., 2012).

In this study, we compared TL behaviors of metamorphic quartz samples which were collected in Turkey and Spain. For this purpose, X-ray diffraction (XRD) and X-ray fluorescence (XRF) analyses were performed in order to give a rough description of the crystalline structure and elemental-chemical composition of the TMQ and SMQ samples. Experimental TL glow peaks from two different metamorphic quartzes were also decomposed using the GlowFit programme (Puchalska and Bilski, 2006) with the computer glow curve deconvolution (CGCD) method. Then the  $\beta$ -dose linearity of these TL glow peaks were tested from  $\sim$ 20 Gy to 4816 Gy. In addition to these, the temperature of TL glow peaks, the order of kinetics b, the trap depth/activation energy (E) and frequency factor (*s*) were determined.

#### 2. Material and methods

All measurements were made on an automated Risø TL/OSL-DA-20 reader having an EMI 9235 QA photomultiplier tube (PMT) attached to filter pack consisting of Hoya U-340 (290-370 nm) filter. To prevent the scattered stimulation light from reaching the PMT, the reader is equipped with a 7.5 mm Hoya U-340 detection





Table 1

e XRD results for TMQ and SMQ samples.											
Alloy		Crystal structure	Unit cell p	Volume	<i>Z</i> *						
Name	Close formula		а	b	с	α	β	γ			
TMQ Quartz	SiO <sub>2</sub>	Hexagonal	4.9146	4.9146	5.4065	90	90	120	113.089	3	
SMO Ouartz	SiO <sub>2</sub>	Hexagonal	4.9141	4.9141	5.4060	90	90	120	113.056	3	

filter which has a peak transmission around 340 nm. β-irradiation was performed using an 1.48 GBq (40 m Ci) <sup>90</sup>Sr/<sup>90</sup>Y beta source with a maximum energy of 2.27 MeV. It was calculated that the absorbed dose rate of quartz at the sample position on the carrousel is 6.689 Gy/min.

The crystalline structures of the samples were analyzed by Rigaku Miniflex II model X-ray diffractometer at 30 kV (scanning rate:  $2^{\circ}$ /min, 15 mA with  $\lambda = 1.5406$  Å Cu-K $\alpha$  radiation) using X-ray diffraction (XRD) technique.

To give a rough description of the elemental-chemical composition of the TMQ sample, Wavelength Dispersive X-ray Fluorescence (WDXRF) analysis was performed using an X-ray Fluorescence spectrometer Rigaku ZSX Primus II.

### 2.1. Sample preparation

The three samples used for this study were gently crushed to prevent crystal damaging effect (Toyoda et al., 2000). After that, samples were washed with distilled water, then dried in the incubator and sieved to obtain the suitable size of fine grain  $(90 \ \mu m < \text{grain size} < 140 \ \mu m)$ . Finally, the grains were deposited with aluminum discs of 10 mm diameter and 0.5 mm thickness as aliquots for measurements. The samples were weighed  $\sim$  5.0 mg powder samples and these samples were used for measurements. The incandescent background was subtracted from the TL data.

#### 3. Result and discussions

X-ray diffraction pattern is shown in Fig. 1 and the obtained results from the pattern are also demonstrated in Table 1. All diffraction peaks match well with the characteristic peaks of SiO<sub>2</sub>. X-ray diffraction measurements show that TMO and SMO have the same hexagonal crystal structure. The type and concentration of different impurities are also observed and given in Table 2 as a result of XRF analysis of the TMQ and SMQ samples. Si, Cu, Fe, Mg and S appeared as the major trace elements of the sample.



Chemical analysis of two natural metamorphic quartzes from TMQ and SMQ performed by XRF.

Elements	SMQ (%)	TMQ (%)
SiO <sub>2</sub>	99.400	94.621
$Al_2O_3$	0.320	2.817
K <sub>2</sub> O	0.010	1.174
Fe <sub>2</sub> O <sub>3</sub>	0.060	0.776
TiO <sub>2</sub>	0.140	0.295
MgO	-	0.210
CaO	0.060	0.042
Na <sub>2</sub> O	_	0.036
$P_2O_5$	0.010	0.020
MnO	-	0.009
Trace	ppm	ppm
Zr	76	80

TMQ sample Space Group: P3221 (154) and SMQ sample Space Group: P3121 (152).

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