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Calculation of thermal quenching parameters in BeO ceramics using solely TL measurements

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HIGHLIGHTS

- Thermal quenching effect was studied on BeO material for Hoya U-340 and BG-39 filters.
- Both W and C thermal quenching parameters were analyzed.
- Activation energies were determined for both quenched and unquenched peaks.

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ABSTRACT

The decrease of the luminescence efficiency with increasing temperature is known as the thermal quenching effect. Various materials exhibit thermal quenching, among which quartz and Al_2O_3:C stand as the mostly well known. BeO, which is used generally as OSL dosimeter, has been reported to undergo thermal quenching effect; so far in the literature only one thermal quenching parameter was determined using various OSL techniques. This study is the first attempt in the literature to calculate both *W* and *C* thermal quenching parameters. TL glow curves of BeO ceramic chips were studied as a function of heating rates ranging between 0.25 K/s and 8 K/s. The procedure of analysis includes various steps dealing with de-convolution of the quenched TL glow curves, correction for temperature lag, evaluation of the experimental thermal quenching parameter values, reconstruction as well as de-convolution of the reconstructed TL glow curves. The reconstruction step is really important, while its aim is twofold: (a) to provide the most stringent criterion for selecting the physically meaningful *W*,*C* values for BeO and (b) to check the kinetic parameters of the unquenched glow curve. The first peak of BeO is not affected by the thermal quenching, while both second and third peaks suffer from thermal quenching. *W* and *C* parameters were determined as 0.54–0.66 (±0.07) eV, $1.8 \cdot 10^5$–$2.4 \cdot 10^6$ ($\pm 4 \cdot 10^5$) for peak 2 and 0.74–0.86 eV and $2.8 \cdot 10^6$–$2.1 \cdot 10^7$ for peak 3 respectively for U-340 filter by using indirect fit method. The same process was applied for BG-39 filter and *W* and *C* parameters were compared to those found through U-340 filter. The values for the main dosimetric peak 2, are independent on the detection filter used, as well as in great agreement with the *W* values previously reported in the literature. The difference in the *W*, *C* parameters for those two peaks in BeO provide a strong argument to the fact that each one of these two peaks uses different recombination centers. According to the results of the de-convolution before and after the reconstruction, it is indicated that the influence of the thermal quenching is mainly reflected to the value of the activation energy for the case of peak 3, which is under-estimated without reconstruction.

1. Introduction

The decrease of luminescence efficiency with increasing temperature due to the increased probability of non-radiative transitions is known as the thermal quenching effect (Curie, 1963). The presence of thermal quenching effect can be easily monitored using the integrated area values of thermoluminescence (hereafter TL) glow peaks measured in various different heating rates. A possible decrease in integral values of the glow peaks stands as a probe for the thermal quenching. In the presence of the thermal quenching effect, not only TL glow curve area values, but also the TL

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kinetic parameters could be affected, especially the values of activation energy *E* and order of kinetics *b*. Thermal quenching effect is attributed to the mechanism described by the so called Mott-Seitz model, which takes account for the competition between the radiative and non radiative recombination (Curie, 1963; Subedi et al., 2010). According to this model, the thermal quenching internal efficiency versus temperature, *η(T)* is given by Curie (1963):

$$\eta\,(T) = \frac{1}{1 + Ce^{-\frac{W}{k\,T}}} \tag{1}$$

where *C* and *W* are called the "quenching parameters" and *T* is the temperature in units of K. *C* is a dimensionless constant which is the ratio of the non radiative transition probability to the radiative transition probability (Pagonis et al., 2010). The physical meaning of the *W* parameter, in units of eV, depends on the Mott-Seitz and Schön-Klasens models that explain the thermal quenching phenomena (Bøtter-Jensen et al., 2003). The Mott-Seitz model can be understood within the defect configurational coordinate diagram (Bøtter-Jensen et al., 2003). According to this model, *W* is the potential barrier required for the electron in the excited state to go to the ground state non radiatively, with emission of phonons only (Dexter et al., 1955). An alternative model to describe thermal quenching effect is the Schön-Klasens model, within which the luminescence emission occurs via delocalised transition but trapped holes are thermally unstable (Bøtter-Jensen et al., 2003). According to this model, *W* is the potential barrier required for thermal release of trapped holes which result in the decrease of concentration of luminescence centers as the temperature increases (Bøtter-Jensen et al., 2003). However, the efficiency decreases according to Eq. (1) as the temperature increases for both models.

Among the TL materials which exhibit thermal quenching, the most widely known are quartz and Al_2O_3:C. The thermal quenching studies on quartz were initiated by Wintle (1975), who measured the values of the *W* and *C* parameters for annealed natural quartz, being 0.64 eV and $2.8 \cdot 10^7$ respectively, by using radioluminescence. Petrov and Bailiff (1997) obtained a different set of *W* and *C* parameters for annealed synthetic quartz, as 0.78 eV and $3.1 \cdot 10^{10}$, also by using radioluminescence. Recently, Subedi et al. (2011) studied the thermal quenching effect on quartz samples of various origins and also investigated the effects on thermal quenching of different annealling temperatures. These latter authors have studied the thermal quenching effect on both natural as well as artificially irradiated TL signal, concluding that both signals yield similar thermal quenching experimental features. Subedi et al. (2010) had previously studied in detail the effect of the thermal quenching effect on TL glow curves of quartz. They have used reference TL peaks with selected values of *W* and *C* parameters, as these were proposed by Wintle (1975). These former authors showed that in the presence of thermal quenching, while the initial rise and peak shape methods underestimate the activation energies, the various heating rate method is less affected by its presence. Also, they tried to test the validity of the curve fitting method for thermally quenched TL peaks.

However, the most intense thermal quenching has been reported for aluminum oxide doped with carbon. Kitis et al. (1994) had studied the heating rate effects on the traping parameters of Al_2O_3:C and obtained the thermal quenching parameters, *W* and *C*, as 1.56 eV and $2.4 \cdot 10^{17}$ respectively by using TL measurements. However, these latter values are much overestimated, as Akselrod et al. (1998) obtained the thermal quenching parameters of $W = 1.08$ eV and $C = 3.6 \cdot 10^{12}$ for Al_2O_3:C by using luminescence lifetime measurements. From a physical point of view, these values are more realistic, as the *W* value is not higher than the activation energy of the main dosimetric trap in Al_2O_3:C. Later on, Dallas et al. (2008) reported a thermal quenching study on aluminum oxide grains exhibiting three different types of TL glow curve shape, namely wide, narrow and double-peak shapes. For all these three groups of samples, Dallas et al. (2008) have verified the *W* and *C* values reported by Akselrod et al. (1998). To a lesser extend, thermal quenching has also been studied for the cases of undoped CVD diamond (Chernov et al., 2012), doped magnesium oxide (Oliveira et al., 2013) and calcium carbonate (Kalita and Wary, 2015).

Beryllium oxide (BeO) has been used commercially in various fields. Due to its excellent heating conductivity and high electrical resistivity, it has been used for insulating heat sink in the electronic industry. It is a suitable material for crucibles and thermocouple tubes due to the high melting point (2570 °C) and high thermal shock resistance (Bulur and Goksu, 1998; Watanabe et al., 2010). BeO is an attractive material for radiation dosimetry due to mostly near tissue equivalence, yielding a Z_{eff} value of 7.13 (Scarpa, 1970; Sommer et al., 2007; Watanabe et al., 2010). The effective use of the BeO as a dosimetric TL material has been early exploited by many researchers (Moore, 1957; Tochilin et al., 1969; Scarpa, 1970; Crase and Gammage, 1975). Nevertheless, its toxicity in powder form and light sensitivity of TL signal makes difficult its usage as a TL dosimeter. BeO material shows three TL peaks at 75, 220, 340 °C with heating rate 5 K/s in the glow curve. The 220 °C peak is considered as the main dosimetric peak (Bulur and Goksu, 1998). These latter authors observed, for the first time, the optically stimulated luminescence (OSL hereafter) signal decreasing with readout temperature, indicating, among others, also the presence of thermal quenching. These authors have calculated the thermal quenching parameter *W* as 0.52 eV. Bulur and Yeltik (2010) obtained the same value for the same thermal quenching parameter, namely 0.55–0.59 eV using Linearly Modulated-OSL (LM-OSL) at elevated temperatures. Yukihara (2011) has reported the value of 0.56 eV for the thermal quenching parameter *W*, confirming thus previously published results obtained from Bulur and colleagues (Bulur and Goksu, 1998; Bulur and Yeltik, 2010). In a recent study, only the thermal quenching parameter *W* was calculated for BeO as 0.56–0.59 eV with Time Resolved OSL (TR-OSL) by Bulur and Sarac (2013). The studies on thermal quenching in BeO have been performed by applying various OSL methods, while the *C* parameter, which gives information about the probability of non radiative transitions was not determined until now.

The purpose of the this study is twofold: Firstly, to determine both thermal quenching parameters *W* and *C* for two different spectral emission wavelengths on BeO, by using just TL measurements. In addition, to determine the real shape of the unquenched glow curve of the BeO, by performing a reconstruction on the experimentally obtained TL glow curve shape. Both experimental and analysis approaches resemble much to those applied and reported by Dallas et al. (2008) and Subedi et al. (2011). However, in these latter two cases, previously published reference values for the thermal quenching parameters *W* and *C* were used from the literature. For the present case of BeO, only the *W* value is known; however, in the framework of the present study we treat it as unknown. Therefore, in the framework of the present approach, the calculation of *W* and *C* thermal quenching parameters will be performed without using any other reference value, namely from scratch.

2. Materials & experimental method

All BeO samples that were the subject of the present study were purchased from Thermalox 995, Brush Wellman Inc., U.S.A. These were in the form of rectangular discs, with dimensions of 4 mm and thickness of 1 mm. Five different discs were chosen for this study, based on reproducibility criteria. Annealing of the samples was performed in an oven at the temperature of 600 °C for 1 h, before measurement. Only thermoluminescence (TL) signals were