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Dose response features of quenched and reconstructed, TL and deconvolved OSL signals in BeO

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ABSTRACT

Despite the fact that Beryllium oxide (BeO) has been suggested by several research groups as an alternative passive radiation detector and dosimetric material, various concerns about supralinear dose response features were raised for both thermoluminescence (TL) and optically stimulated luminescence (OSL) signals of this material. This paper presents a detailed dose response study on BeO, for two different experimental conditions, namely for (i) the TL signal following irradiation at room temperature (RT), 125 and 200 °C and (ii) the CW – OSL signal measured at three different measurement temperatures (RT, 100 and 220 °C), following two different preheating temperatures (T_{pre}= 100 and 220 °C). The selection of these temperatures is based on the TL glow curve structure for BeO. The analysis was performed using (a) the actual experimental luminescence signals which suffer thermal quenching, and (b) by using the reconstructed luminescence signals, after correcting for thermal quenching. Deconvolution was performed only for the OSL signals. Intense supralinearity features for both TL peaks and OSL components are reported. Reconstruction affects greatly the dose response features of all TL signals, enhancing supralinearity. The dose response linearity coefficient \( \mu \) of the dominant OSL component C₁ is found to be independent of the preheating temperature. This provides a strong argument for the fact that the charge associated with this component originates mainly from the trap responsible for the TL peak 3 in BeO with \( T_m \) beyond 300 °C. Based on our
Experimental results, the supralinearity features of BeO are attributed to competition for free electrons by electron traps. The results of the present study suggest that competition takes place during both the irradiation and heating stages of a dose response experiment in BeO. Finally, preheating at 200 °C and subsequent OSL measurement at 100 °C is highly recommended as the optimum configuration for BeO, not only for roughly linear dose response but also in order to minimize transfer effects.

Key Words: BeO, TL, OSL, dose response, reconstruction, supralinearity, irradiation temperature

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1. Introduction

Beryllium oxide (BeO) is currently widely used as a passive radiation detector and as a personal/environmental dosimeter, due to the following reasons [1-8]:

1) BeO has a near-tissue equivalent value of $Z_{\text{eff}} = 7.14$, i.e., very close to the value of $Z_{\text{eff}} = 7.35–7.65$ for biological tissues, and the value of $Z_{\text{eff}} = 7.51$ for water. This means that this material has a minimal (< 20 %) over- and under-response to low energy photons, which is optimal for a plethora of applications in different fields of medicine, such as radiotherapy or radiation diagnostics [9].

2) Both thermoluminescence (TL) and optically stimulated luminescence (OSL) signals from BeO can be effectively used in dosimetric applications [2, 10-13].

3) Despite being an insulator, it has a high thermal conductivity, which exceeds even that of certain metals. This assures a rapid and uniform heat transfer during the readout stage of TL measurements. The TL sensitivity of BeO is approximately equivalent to that of TLD-100 [14], making thus BeO optimal for environmental monitoring. Also, the absence of overlapping low temperature peaks leads to simplified annealing procedures.

4) It is a tough ceramic material, formed in a high temperature environment into discs or rods, is insensitive to most chemical agents, insoluble in water and has a high mechanically resistance [15].

5) The availability of BeO ceramics has increased, and it has a low cost.

6) In its solid undisturbed form, BeO is practically harmless. However, the health hazard due to high toxicity increases when it is being smashed, crushed, or transformed into the reactive dispersed form [16].

7) Finally, it was demonstrated that the dosimeters perform well and the radiation dose can be estimated even if the samples are accidently exposed to light. This is done by means of thermally transferred luminescence signals in BeO [17,18].

Concerning its luminescence and dosimetric properties, Ceramic BeO (Thermalox® 995, Brush Beryllium Co.) is superior to other commercially used BeO ceramics, because of its high-quality control and relative freedom from spurious signals, and also a stable long-term response [11, 17-19]. Gammage and Garrison [20] identified Al$^{2+}$ substituting for Be as the TL trapping entity for the material by Thermalox, while its impurity composition (in units of ppm) as supplied by the manufacturer is Al(54), Ca(61), Cr(10), Cu(3), Fe(100), Mg(945), Si(2150), Ti(4) [6]. Besides Thermalox, BeO is also commercially available from other companies; Innovacera Advanced Materials Co., China produces BeO in rod shape, which is the most appropriate form for Electron Paramagnetic Resonance (EPR) measurements [21]. Recently, Aşlar et al. [22] reported on an unusual BeO sample, which yields different TL glow curve structure and OSL decay properties from the Thermalox material. These authors have
attributed these differences to the increased content of Cr, Mg and Sn and to sintering and/or calcination process during manufacturing of the dosimeters.

One interesting luminescence feature of BeO deals with a very strong supralinearity of the dose response curves, with Thermalox being one of the materials yielding quite intense supralinearity. Tochilin et al. [1,2] have early reported a supralinearity index $f(D)_{\text{max}} = 15$ at $10^2$ Gy following gamma irradiation using $^{60}\text{Co}$ source, and a value of $f(D)_{\text{max}} = 4$ for X-rays with energy 9 keV. Crase and Gammage [19] have reported that suprlinear behavior in Thermalox BeO can start at dose as low as 0.1 Gy. On the other hand, Yasuno and Yamashita [23] have reported $f(D) = 1$, indicating a linear TL dose response for BeO:Na and BeO:Li, from 1 mR to 1000 R. However, these phosphors exhibit enhanced fading. For example, a peak at 165 °C in BeO:Li faded rapidly to 50% of its initial height in several hours storage in the dark, and the peak temperature simultaneously shifted to 180°C, indicating composite trapping structures. BeO:Na exhibits a less extreme but similar fading. Strong dose response supralinearity was also reported for the main dosimetric TL peak 2 in BeO [24], a specific TR – OSL component [25] and in both TL and OSL signals at high doses following only beta irradiation [26]. A quadratic, supralinear dose response behavior was also reported by Martinez Baltezar and Azorin Nieto [27] for the deconvolved TL peak 2 in BeO, in the dose range 1.3-23.4 Gy. Lately a new type of anion-deficient Al$_2$O$_3$–BeO ceramic compound has been reported, which is used for high-dose dosimetry and has a supralinear TL and OSL dose response, even beyond 1 kGy [28, 29].

Although the commonly used Thermalox brand shows strong supralinearity in TL dose response, such supralinearity features have not been reported for the case of the OSL dose response. Bulur and Göksu [13] were the first to point out that the dose response of the CW-OSL signal in BeO was linear up to 10 Gy, in contrast to the supralinearity exhibited by the corresponding TL response signal [11]. Similar linear OSL dose response was also reported by Caraça Santos et al. [30], who investigated a fiber-coupled Beryllium oxide ceramic. However, these researchers found a supralinearity factor of 29.4% at doses as high as 15 Gy. On the other hand, Sommer and Henniger [31] and Sommer et al. [32] have reported that the OSL dose response of BeO chips is mostly linear, covering more than six orders of magnitude and with saturation beginning to appear at 5 Gy. In the lower dose range of 25μGy and 2.5 mGy, Bulur and Yeltik [33] have shown that the CW-OSL dose response of BeO ceramics is linear, making the material good enough to be used for personal radiation monitoring. Using a $^{90}\text{Sr}/^{90}\text{Y}$ beta source, Malthez et al. [34] found that the OSL dose response of BeO followed a linear behavior. The same study reported a linear TL dose response in the range of 1-10 mGy, using 28 keV X-rays and 1.25 MeV Co-60 photons. Also, Groppo and Caldas [26] showed that there is a difference between the TL and OSL linearity of BeO when used with alpha, beta and X radiation. Specifically, the dose response curves have a greater linear
range in the case of the OSL than the TL signals. The OSL signals indicate supralinear components which appear only at the high doses of beta radiation, while there is a supralinear TL response in the high doses, and for all three types of radiations. This latter feature was the most important reason for the use of OSL signals from BeO for dosimetric applications in breast radiotherapy [35] and mammography [36].

As far as the environmental monitoring application is concerned, several studies have shown that BeO exhibits a low, albeit substantial, fading of the luminescence signals. Specifically, there is a maximum of 5% fading in the signal in the first 24 hours of storage in the dark at room temperature, followed by stability of the signal over a lot of months. This specific shortcoming of BeO was thoroughly discussed in the framework of the latest Solid State Dosimetry conference that took place in Hiroshima, Japan in 2019. More specifically, for the OSL signal of commercial BeO that is being measured at room temperature without any preheating, a substantial fading has been reported; this fading could be as high as 5-8%. Towards overcoming or circumventing this drawback, OSL measurements at elevated temperatures following an appropriate preheating have been suggested. Nevertheless, various concerns regarding the linearity features of such OSL signals were raised. The various dose response features that were so far reported in the literature for BeO are presented in Table 1, and concern various stimulated luminescence signals. A careful study of the contents of this Table indicates that:

(a) In all TL and OSL studies of BeO, irradiation takes place at room temperature (RT).
(b) The majority of the OSL measurements are performed at RT, following a specific preheating protocol.
(c) In addition to TL and OSL signals, Table 1 shows dose responses for time resolved OSL (TR – OSL), thermally-transferred OSL (TT – OSL) and a special case of OSL signals measured at RT following a TL up to 400 °C [13]. This special signal was also termed as thermally transferred OSL (TT – OSL, [17, 18]), and also represents a case of thermally assisted OSL (TA – OSL, [37]) with the OSL measurement taking place at RT or ambient temperatures.
(d) There is only a unique case in Table 1, where linearly modulated OSL (LM – OSL) is used for measuring both OSL and TT – OSL signals. Finally, all dose response curves in Table 1 were constructed without either a deconvolution analysis, or a correction for thermal quenching. A correction for thermal quenching has been introduced by Yukihara [38] for the TL signal of BeO in his study on TT – OSL.

Despite the intense research that has been devoted to the luminescence properties of BeO, surprisingly the dose response properties of OSL signals measured for various combinations of preheating and measurement temperatures are not reported in the literature yet. Therefore, in order to fill the existing literature gaps in the bibliography dealing with the dose response in BeO, the primary aim of the present study is to perform a component-resolved dose response study in two cases. First, we study the TL signal in BeO following irradiation at various elevated temperatures, including RT. Second, we report on the
CW – OSL signal measured using a combination of preheating and measurement temperatures. Finally, secondary goals of the present work include (a) studying the influence of thermal quenching on the dose response, by carrying out the corresponding thermal quenching correction of these dose responses and (b) suggesting the optimum temperature configuration regarding preheating and measurement for the OSL signal in BeO.

2. Materials and Methods

The BeO ceramics used in this study were in the form of rectangular discs, with dimensions of ~ 4 mm in length and thickness of 1 mm. All BeO ceramics used in this study were purchased from Thermalox 995, Brush Wellman Inc., U.S.A. Annealing of all samples was performed in an oven at the temperature of 900 °C for 30 min., in order to empty both the dosimetric and deeper traps, [18, 21, 38]. The samples were cooled immediately after the heating process, by putting the crucible in contact with a heat sink.

All OSL measurements were performed at the Institute of Nuclear Sciences, Ankara University, Turkey, using a RISØ TL/OSL reader (model TL/OSL-DA-20), with Reader ID 267 [39]. This system is equipped with a high-power blue LED light source (470±20 nm, 40 mW cm⁻²), a 0.129±0.03 Gy/s ⁹⁰Sr/⁹⁰Y β-ray source emitting beta particles with a maximum energy of 2.27 MeV and an EMI 9635QB PM Tube [40]. All OSL measurements were performed at the Continuous Wave stimulation mode (CW-OSL, [41]) over 100 s, at various combinations of preheating (T\text{pre}) and measurement temperatures (T\text{meas}), using a 7.5 mm thick Hoya U-340 filter (270–380 nm, FHWM 80 nm). This stimulation duration was proven adequate to deplete the OSL signal reaching a flat (background) level.

TL measurements were performed at two different laboratories, namely: (a) at the Institute of Nuclear Sciences, Ankara University, Turkey, using a RISØ TL/OSL reader and with the same filter as in the OSL measurements and the appropriate irradiator and (b) at the Nuclear Physics Laboratory of the Physics Department, Aristotle University of Thessaloniki, Greece, using a Harshaw (Bicron) TLD reader model 3500. All TL measurements as well as the preheating for the OSL measurements were performed in a nitrogen atmosphere. TL was measured with a constant heating rate of 2 °C/s in order to avoid significant temperature lag [42] between the temperatures of the heater and that of the sample. TL measurements were performed from RT up to a maximum temperature of 400 °C. The dose response of the following signals was studied, using 12 different doses, ranging between 2 mGy and 4 Gy:

Case (a): TL signal following irradiation at RT
Case (b): TL signal following irradiation at 125 °C,
Case (c): TL signal following irradiation at 200 °C,
Case (d): CW-OSL at RT following a preheat at 100 °C,
Case (e): CW-OSL at 100 °C following a preheat at 100 °C,
Case (f): CW-OSL at RT following a preheat at 220 °C,
Case (g): CW-OSL at 100 °C following a preheat at 220 °C,
Case (h): CW-OSL at 220 °C following a preheat at 220 °C,

In order to construct the dose response curves, each point corresponds to the average of two individual measurements. In the first case (a) above, the TL signal was measured using both experimental set-ups for comparison purposes.

The selection of either irradiation or preheating temperatures was based on the TL glow curve of a typical Thermalox BeO material. As this TL glow curve is a simple curve without overlapping glow peaks, the temperatures were selected in order to isolate the signals from deeper traps. Thus, the temperatures of 100-125 °C and 200-220 °C were selected, in order to deplete completely the TL peaks 1 and 2 respectively. These temperatures may vary slightly at different laboratories, because the TL glow curves were measured using different equipment, and therefore a divergence of 20 – 25 °C was anticipated.

For all OSL measurements, irradiation was performed at RT. High temperature irradiation was performed using the glow oven of an old TL analyzer type 711 of the Littlemore Co. [43]. The glow oven has the ability to both heat the sample, as well as to keep the temperature at a steady elevated level. After achieving the required temperature, irradiations were carried out with a 90Sr/90Y beta source delivering a dose rate of 0.38 Gy/min. The experimental details for each signal are presented in Table 2. In all cases, the duration of each preheat was 10 s.

3. Method of analysis

3.1 Deconvolution procedure

Based on the previous experience of our group on the features of the TL glow curve of BeO, deconvolution analysis of the TL glow curves was not a mandatory task for the present study, as TL glow curves indicate the presence of two non-overlapping single TL glow peaks with delocalization temperature below 200 °C and around 300 °C [13, 17, 18, 21, 44, 45]. Following the nomenclature of these previous articles, the TL glow peak below 200 °C will be termed TL peak 2, while the second TL peak at higher temperature will be termed TL peak 3 [44, 45]. Typical TL glow curves of BeO are presented in Fig. 1A. The TL peak 1 (around 70 °C) is unstable and decays quickly at room temperatures; thus, it was not studied, as it is not appropriate for dosimetry [24].

On the contrary, a deconvolution analysis was applied in all OSL decay curves, independent of the measurement temperature. The CW-OSL signal is complex, with contributions from two different components termed \( C_1 \) and \( C_2 \) hereafter [21, 33], with \( C_1 \) being the CW-OSL component yielding the
lower lifetime, and therefore a corresponding higher photo-ionization cross section. All CW-OSL decay curves were analyzed using the analytical framework of the one trap one recombination center (OTOR) model. The general analytical expression given by Kitis and Vlachos [46], corresponding to delocalized transitions via the conduction band, is the following:

\[ I(T) = \frac{N \cdot R}{(1-R)^2} \left( \frac{p(t,T)}{W(z)} + \frac{W(z)}{W(z)} \right) \]  

where the \( N \) is the total concentration of trapping states and \( W(z) \) is the Lambert W function [47], \( R = A_n/A_m \) with \( A_n \) and \( A_m \) being the retrapping and recombination coefficients correspondingly (in cm\(^3\)s\(^{-1}\)). The \( p(t,T) \) corresponds to the stimulation probability. The function of the \( p(t,T) \) for the CW-OSL signal, gets \( p(t,T) = \lambda, \) \( \tau = 1/\lambda \) is the component lifetime and \( \lambda \) (1/s) stands for the respective decay constant. The argument \( z \) of the Lambert function \( W(z) \) is given by:

\[ z = \exp\left( \frac{R}{1-R} - \ln\left( \frac{1}{1-R} + \frac{A t}{1-R} \right) \right) \]  

where \( A \) is the optical stimulation probability (in s\(^{-1}\)) given by

\[ \frac{1}{A} = \frac{1}{\tau} = \frac{1}{\lambda} = \tau_{CI} = (\sigma \varphi)^{-1} \]  

with \( \sigma \) representing the OSL cross section and \( \varphi \) denoting the fluence of the stimulation photons. The free fitting parameters of Eqs.1&2, besides the initial luminescence intensity, include the component lifetime parameter \( \tau \) and the dimensionless parameter \( R \) which defines the order of kinetics. This parameter \( R \) is similar to the kinetic order coefficient \( b \) in the general order kinetics (GOK) model [41, 48]. When \( R \) has a positive value close to 0, it corresponds to first order of kinetics, and when \( R \) has values close to 1, this indicates second order of kinetics. The photo-ionization cross section of each OSL component was calculated according to the equation 3. All curve fittings were performed using the software package Microsoft Excel, with the Solver utility [49], while the goodness of fit was tested using the Figure Of Merit (FOM) of Balian and Eddy [50] given by:

\[ \text{FOM} = \sum_i \left[ \frac{Y_{\text{exp}} - Y_{\text{fit}}}{A} \right] \]  

where \( Y_{\text{exp}} \) is the experimental data, \( Y_{\text{fit}} \) is the fitted data and \( A \) is the area of the fitted curve.

### 3.2 Reconstruction as correction for thermal quenching

Thermal quenching is the term that describes the decrease of the luminescence efficiency with increasing stimulation temperature [51]. BeO stands among the luminescence phosphors that exhibit intense thermal quenching. For a review of related articles on this specific material, the readers could refer to Aşlar et al. [44] and references therein. Luminescence efficiency \( \eta(T) \) is described by the following equation [51]:
\[ \eta(T) = \frac{1}{1 + C \cdot e^{-\frac{W}{kT}}} \]  

where \( C \) and \( W \) are called the “quenching parameters”, \( k \) is the Boltzmann constant and \( T \) is the temperature in units of K. \( C \) is a dimensionless constant representing the ratio of the non-radiative transition probability to the radiative transition probability [52, 53]. The physical meaning of the \( W \) parameter, in units of eV, is a measure of the energy between an energy level in the excited state to an energy level that is degenerate to both the excited state and the ground state from which a radiationless transition to the equilibrium state of the ground state can occur [54].

The raw (or quenched) TL or OSL intensity is given by:

\[ I_q(T) = I_{unq}(T) \cdot \eta(T) \]  

where \( I_q(T) \) and \( I_{unq}(T) \) are either the TL glow curves or OSL decay curves, which are quenched and unquenched, respectively. The unquenched TL or OSL luminescence intensity is unknown, but it can be identified from the TL signal using the following formula [55]:

\[ TL_{unq}(T) = \frac{TL_q(T)}{\eta(T)} \]  

Each experimental data point of each TL glow curve, corresponds to a specific temperature, and thus to a different value of the thermal quenching efficiency according to equation (5). Therefore, according to the reconstruction procedure, each experimental data point of the quenched TL glow curve is divided by the corresponding thermal quenching efficiency. In other words, for the reconstruction of TL, each entire glow curve was used, instead of the integrated intensity of the peaks. On the contrary, for the case of OSL measurements, the entire integral should be corrected, by dividing with the thermal quenching efficiency of the temperature of measurement, namely:

\[ OSL_{unq}(T) = \frac{OSL_q(T)}{\eta(T_{meas})} \]  

### 3.3 Linearity features of the dose response

In all cases, the integrated intensity of the aforementioned TL peaks / OSL components was also used to construct the corresponding dose response curves. These curves were fitted using the following equation [56]:

\[ Lum (TL \ or \ OSL) = m \cdot D^\mu \]  

where \( m \) is a proportionality coefficient and \( \mu \) is a constant which indicates the linearity features of the dose response curve [57]. Hereafter, \( \mu \) parameter will be termed as the linearity coefficient, with \( \mu = 1 \).
indicating the (ideal) case of linearity, $\mu > 1$ indicating supra-linearity properties and $\mu < 1$ sub-linearity [56, 58].

4. Results and Discussion

4.1. TL glow curves, OSL decay curves and decay kinetics

Fig. 1A presents TL glow curves measured for various cases, namely following irradiation at room temperature (curves $a_1$ and $a_2$), at 125 °C (curve $b$) and at 200 °C (curve $c$). In all cases a dose of 500 mGy was used. While both curves ($a_1$) and ($a_2$) correspond to irradiation at room temperature, curve ($a_1$) was measured using a Risø system, while curve ($a_2$) was measured using the Harshaw 3500 system.

TL peak 2 is apparent when irradiation takes place at RT and 125 °C. However, when the irradiation takes place at 220 °C, only TL peak 3 is present in the glow curve. It is worth mentioning that TL peaks 2 and 3 do not yield the same $T_m$ values for the two experimental set-ups used. Both TL peaks measured using the Risø system yield higher $T_m$ values as well as wider TL peaks than the Harshaw TL system, despite the fact that the TL glow curves were measured using the same heating rate of 2 °C/s. These differences could be attributed to various issues including thermal between the sample and the heater plate or sample and heater plates [42, 59]. In the case of Harshaw readers, the sample is heated directly on top of the heating planchette [60]. On the contrary, in the Risø geometry the chip dosimeters are placed on stainless steel cups, and these cups in turn are placed on top of the heating element; thus, the sample is heated indirectly. Better thermal contact is achieved for the Harshaw readers, resulting in lower values for both $T_m$ and FWHM, namely closer to the “real” structure of the TL glow curve/peaks.

Another interesting feature is revealed by this same figure, indicating that the TL glow curves that were measured using the Risø system are slightly more intense than the TL glow curves measured using the Harshaw 3500 system. This observation is valid for doses higher than 0.1 Gy, despite the fact that the detection optics in the Risø system consists of the Hoya U-340 filter, and the detection optics in Harshaw does not include any filter.

Figs. 1B and 1C present the same TL glow curves as in Fig. 1A, namely following irradiation at room temperature (curves $a_1$ and $a_2$), at 125 °C (curve $b$) and at 200 °C, albeit reconstructed, namely following a correction for thermal quenching according to equation 7. Fig. 1B focuses solely on TL peak 2, while Fig. 1C presents data for TL peak 3. Each figure presents also the corresponding thermal quenching efficiency versus temperature (left axis in both figures).

The reconstruction was performed using different $W$ and $C$ values for each TL peak, by using the results reported by Aşlar et al. [44]. These authors reported $W$ and $C$ values within specific range for each TL peak. Therefore, in the present study the median of each range of values was used. Correction for
thermal quenching via reconstruction results in shifting both the $T_m$ and FWHM parameters to higher values, for each TL peak. It also results in an increase of both $I_m$ and integrated TL peak intensity [44, 55]. In addition, these changes are more profound for the case of TL peak 3, as the $T_m$ of this peak is very close to the temperature where the gradient of the efficiency changes versus temperature.

According to Table 2, the error on the average $T_m$ value in the original (quenched) data is less than 1-2%, especially for the case of the Harshaw reader [60]. Nevertheless, in the reconstructed TL glow curves the shifting of the $T_m$ in the TL glow curves is within a wide range, and this results in a large corresponding standard deviation. This latter feature can be easily observed from both Figs. 1B, 1C and Table 2.

Fig. 2 presents the OSL decay curves according to cases (d) to (h) of Table 2, and for the same dose of 0.5 Gy. The main Fig. 2 aims in presenting the different OSL intensities of the OSL signal, while the inset presents the same OSL decay curves normalized over the initial intensity, in order to show the changing shape of the decay curve. The experiment involves measurements at three different temperatures ($T_{\text{meas}}$= RT, 100 and 220 °C), following two different preheating temperatures ($T_{\text{pre}}$= 100 and 220 °C). The OSL decay curve corresponding to case (h) (namely both preheating and measurement at 220 °C) corresponds to the lowest, faint intensity. Among the rest of the cases, for each measurement temperature (RT and 100 °C), the OSL is slightly more intense following preheating at the lower temperature of 100 °C, compared to those preheated at 220 °C. This latter feature provides a first experimental indication that the OSL signal mostly originates from TL peak 3.

The normalized OSL curves in the inset of Fig. 2 indicate different slopes at the measurement temperature, with a mild decreasing dependence of the slope (and the corresponding OSL decay constant) on the OSL measurement temperature. This change in the slope of OSL decay curves is not as steep as in the case of isothermal TL [61], but is clearly present [62]. In all cases, independent of either the preheating or the measurement temperatures, the OSL decay curves reach a flat signal level below 100 counts.

All OSL decay curves were deconvolved, and two individual components were required for achieving excellent fits. In all cases, the OSL consists of two components, termed $C_1$ and $C_2$. The former component is the dominant one in all OSL signals of the present study, contributing $82\pm5\%$ to the total OSL signal. Both OSL components are described by first order of kinetics, as the corresponding average values of the retrapping parameter $R$ do not extend 0.1. As OSL curves are measured at constant elevated temperatures, reconstruction of the OSL signal is achieved by dividing with the value $\eta(T_{\text{meas}})$ using equation 8. As this value does not depend on the stimulation temperature, reconstruction affects solely the OSL intensity, and does not affect the features of the decay curves.

Fig. 3 presents the lifetime for each component for the OSL measurements, as a function of the dose. These results stand in excellent agreement with the standard OSL theory of first order kinetics, indicating
that these lifetimes depend strongly on the measurement temperature, but do not depend on the preheat temperature or on the dose. In addition, the results agree with both the theory of delocalized transition models and with the results in the recent paper by Polymeris et al. [61], verifying once again that in BeO all electrons recombine via the conduction band and not via tunneling recombination processes.

For each case (d) to (h) of Table 2 and for each component, a distribution of lifetime values results in an average value, along with the corresponding 1σ standard deviation. These values, being used for the calculation of the photo-ionization cross section and the corresponding error, are shown in Table 2 as a unique, average value. For this calculation, the conversion factor of Polymeris et al. [63] was used.

These aforementioned kinetic results from the deconvolution analysis agree well with the corresponding results presented by Bulur and Yeltik [33] and Aşlar et al. [21]. Fig. 4 presents the dependence of the cross section \( \sigma \) on the temperature of the OSL measurement. By measuring the OSL signal at 220 °C, the cross section \( \sigma \) increases by one order of magnitude. However, the intensity is dramatically decreased by the same factor, and even more.

### 4.2. TL dose response curves and features

Fig. 5 presents the TL dose responses for the original (quenched) TL peaks P2 and P3, and for all cases from (a) to (c) shown in Table 2. In case (c), the irradiation takes place at 200 °C (curve c), and only TL peak 3 is measured.

Table 2 presents the linearity coefficients \( \mu \) for each quenched TL peak. It is quite interesting to note that the dose response of TL peak 2 is supralinear in all cases. When irradiation takes place at RT, the dose response indicates the same \( \mu \)-value which is larger than unity, and is also independent of the equipment used. When irradiation takes place at 125 °C, the dose response becomes less steep but is still slightly supralinear, indicating a value of \( \mu =1.12 \). On the other hand, the behavior of TL peak 3 is linear only when irradiation takes place at 220 °C, while in all other cases linearity is not present.

The supralinear behaviour of both TL peaks (mostly the TL peak 2 but occasionally TL peak 3 as well), is due to competition effects between the charge traps in the material; competition for free electrons by electron traps. According to Bulur and Göksu [13] and Bulur [24], such type of competition effects occurs during heating, but during irradiation are also possible [64]. In both cases, the competing traps are believed to be the deeper traps. Detailed analysis of the TL glow curve up to 650 °C, in conjunction with the presence of (a) intense transfer effects [21], (b) TT – OSL [17, 18] and (c) TA – OSL [37] have shown that more than one trap exist within the temperature region of the TL glow curve above 450 °C in BeO.
In the present study, the supralinearity of the TL P2 could be attributed to competition for free electrons by electron traps during heating. Kitis et al. [65] and Chen and Hag-Yahya [66] offered an explanation based on the competition between the traps of LiF:Mg,Ti (TLD-100) materials. With increasing irradiation temperatures, the shallow traps are gradually more delocalized, and deeper traps become more filled than the others. As these deeper traps act as competitors, both the deeper traps and competitors are gradually filled, thus reducing the effect in accordance with the charge balance in the material. Consequently, the charge carriers occupy the main traps with less competition, resulting in an enhanced TL signal intensity and superlinear behavior.

The supralinear dose response behavior of both TL peaks is highly enhanced after the reconstruction analysis based on thermal quenching corrections. Each TL glow curve was corrected according to equation 7, and later on the dose response curves were constructed by integrating the signal for each individual TL peak. Fig. 6 presents the dose response curves of TL peak 2 and TL peak 3 following the reconstruction analysis. For the TL peak 2, the linearity coefficient \( \mu \) has a value of around 3 for irradiation at room temperature, and a value of around 2.2 for irradiation at 125 °C.

The most important dose response feature is seen in the TL dose response of the reconstructed TL peak 3. As the right-hand side plot of Fig 6 shows, all dose responses yield (i) an initial stability region where the signal seems not to be sensitive to the dose, (ii) a subsequent region of supralinear dependence and (iii) a final saturation area. These features seem to be prevalent and independent of either \( T_{\text{irr}} \) or the equipment used.

Table 2 presents the estimated \( \mu \) value for region (ii) of each reconstructed TL P3 dose response. It is worth emphasizing that the data points of all four dose response curves seem to form a single dose response curve, independent of both measurement set-up and irradiation temperature. While the integrated intensities of the reconstructed TL glow peak 3 are more or less similar for the four cases (a) to (c) for each dose, the intensity for the case (a1) is somehow more intense. As curve (a1) was measured using a Risø system, the \( T_m \) of TL P3 for this case is shifted to higher temperatures. Thus, the impact of the reconstruction is more intense for these TL peaks, resulting in enhanced integrated TL intensities.

### 4.3. OSL dose response curves and features

The component-resolved OSL dose response curves for both OSL components are presented for all cases (d) to (h) in Fig. 7 (right-hand side plot and left-hand side plot for components C1 and C2 respectively). This figure indicates some really interesting results. In fact for the cases (d) to (g), namely for OSL measurement temperatures of RT and 100 °C, the dose response of the dominant component C1 indicates linearity coefficients \( \mu \) around 0.92 (Table 2). This latter value is, within errors, independent of
both preheating as well as measurement temperatures. This experimental result provides a strong argument towards the significance of TL peak 3 as the main reservoir that feeds this OSL component.

Fig. 7 indicates that when OSL measurements take place at 100 °C, the corresponding integrated intensity is lower than the respective intensity for OSL measurements at RT. Nevertheless, this decrease does not exceed 11 – 15 %, with this percentage corresponding to the contribution of the main TL peak (P2) to OSL component C₁. This result is quite interesting, as it does not contradict the conclusion of Aşlar et al. [21] that the OSL component C₁ in BeO is using charges from both traps of TL peaks 2 and 3, with a significant amount for TL Peak P2. In addition, this result stands in excellent agreement with previous work which concluded that the main source for the OSL signal is not the trap corresponding to TL peak 2 [9, 13, 17, 38].

A notable exception is seen when the OSL measurement takes place at 220 °C. For this case, the parameter μ of the dose response increases substantially for both components, but especially for component C₁. This increase can be explained as due to the contribution of the TA – OSL signal to the overall OSL signal, as it was argued by Aşlar et al., [21].

Transfer of charge is also possible from the so called very deep traps (VDTs), namely traps with delocalization temperatures higher than 500 °C. While measuring TA – OSL signals, the sample is stimulated optically at an elevated temperature. Thus the contribution of the TA – OSL to the main OSL signal depends strongly on the OSL measurement temperatures, for both OSL components. However the contribution to the OSL component C₁ becomes detrimental when the measurement temperature exceeds 180 °C [21]. When stimulation takes place at these temperatures, both the dosimetric and very deep traps are simultaneously emptying. Fig. 8a presents the linearity coefficient μ for the component C₁ versus the measurement temperature.

The results of the present study indicate clearly that the μ parameter for component C₂ depends on both the preheating and measurement temperatures. This is strongly supported by Fig. 8b, where a contour plot is presented for this value, versus the measurement and preheating temperatures. It is important to note once again that the contribution of component C₂ to the overall OSL signal is of the order of 20% or even less. Nevertheless, this component is still important, not only from a dosimetric point of view, but also for understanding the physics and the origin of the charge that constitutes this component. The contour plot of the linearity index of C₂ indicates that for this specific component, supralinearity is ubiquitous. According to Refs [21, 37], the contribution of the TA – OSL to the OSL signal for this component becomes significant even from very low stimulation temperatures of 100 °C. However, the intense supralinearity that it is seen at the edges of the contour plot, implies that both competition during irradiation and during heating could take place for the charge constituting to C₂. The indirect conclusion from these features indicates that the charge of component C₂ originates from more than one traps, with
contributions that depend on the selection of preheating and stimulation temperatures. Similar conclusions were also reported for the cases of perovskite [67], CaF$_2$:N [68] and recently quartz [69] and magnesium oxide [70].

5. Conclusions

BeO dosimeters are important materials for the future of luminescence dosimetry. The discrepancy regarding whether luminescence signals in BeO show supralinear dose response is quite a prominent feature in the related literature. The present study was motivated by the lack of dose response curves in BeO for (a) TL signals of this material following irradiation at temperatures above RT, (b) OSL signals of this material for various combination of preheating and measurement temperatures and (c) the same signals following correction for the thermal quenching effect in the dose response curves of BeO.

Therefore, dose response experiments and analysis were performed for the cases of (i) the TL signal following irradiation at RT, 125 °C and 200 °C (ii) the CW – OSL signal measured using a combination of preheating and measurement temperatures within the same temperature region. All signals were reconstructed and the impact of such a correction approach was studied for each TL peak and OSL component following deconvolution analysis for the latter case. Besides the specific combination that includes both preheat and OSL measurement at 220 °C, all the other cases provide OSL signals with rich statistics.

Strong supralinearity was seen in many dose response curves for various TL peaks and OSL components. Reconstruction affects greatly the dose response features of all TL signals, enhancing supralinearity. Besides the dosimetric properties of the corresponding signals, specific conclusions could be drawn for the luminescence mechanisms in BeO, namely:

1. The independence of the linearity coefficient $\mu$ of the dominant OSL component $C_1$ on the preheating temperature provides a strong argument for the fact that the charge associated to this component originates mainly from the trap responsible for the TL peak 3.

2. The dependence of the linearity coefficient $\mu$ of the second OSL component $C_2$ on both preheating and measurement temperature, as highlighted by the contour plot of Fig. 8, in conjunction with the large supralinearity effects in the luminescence dose response curves, provide experimental indication of the presence of competition for free electrons by electron traps during both irradiation and heating of the samples.

3. Finally, based on the results of the present study, the optimum way to measure the OSL signal in BeO includes a preheating at 200 °C and measurement at temperatures lower than 110 °C. For this configuration both OSL components yield a roughly linear dose response. Measurement at 100 °C is highly recommended in order to minimize transfer effects.
References


FIGURE CAPTIONS

**Figure 1**: Plot (A) presents examples of entire raw TL glow curves corresponding to a dose of 0.5 Gy for the cases (a) to (c) according to Table 2 and section 2. Plot (B) presents both quenched and reconstructed
TL glow peaks P2 from plot (A); the same for plot (C) and P3. Index Rec denotes the reconstructed TL glow curves. In all cases, TL glow curve $a_1$ was measured using a Risø TL/OSL reader while TL glow curve $a_2$ was measured using a Harshaw 3500 reader.

Figure 2: Examples of OSL decay curves corresponding to a dose of 0.5 Gy for cases (d) to (h) according to Table 2 and section 2. Inset presents the same decay curves normalized over the initial intensity in order to show the different shape of the OSL measurements according to the measurement temperature.

Figure 3: OSL deconvolution results regarding the lifetime $\tau$ for each OSL component and each combination of preheating and measurement temperatures for all cases (d) to (h). Filled data points correspond to the component $C_1$ while open data points correspond to component $C_2$.

Figure 4: Photo-ionization cross section versus the temperature at which the OSL measurements take place; squares correspond to the case (d), circles correspond to the case (e), triangles correspond to the case (f), stars correspond to the case (g) and diamonds to case (h). Filled data points correspond to the component $C_1$ while open data points correspond to component $C_2$.

Figure 5: TL dose response curves for quenched TL peak 2 (left-hand side) as well as quenched TL peak 3 (right-hand side) for the cases (a) to (c). Curve $a_1$ was constructed using the Risø TL/OSL reader while curve $a_2$ using the Harshaw 3500 reader. All plots are presented in log-log scale; for the linearity coefficients $\mu$, the readers could refer to Table 2.

Figure 6: TL dose response curves for reconstructed TL peak 2 (left-hand side) as well as reconstructed TL peak 3 (right-hand side) for the cases (a) to (c). Curve $a_1$ was the only one that was constructed using the Risø TL/OSL reader. In all cases, reconstructed dose responses of TL peak 3 yield (i) an initial stability region where the signal seems not to be sensitive to the dose, (ii) a subsequent region of supralinear dependence and (iii) a final saturation area. All plots are presented in log-log scale; for the $\mu$ coefficients the readers could refer to Table 2.

Figure 7: OSL dose response curves for components $C_1$ (left-hand side) and $C_2$ (right-hand side) for the cases (d) to (h) according to Table 2 and section 2. All plots are presented in log-log scale; the linearity coefficient $\mu$ for each case is also presented in the figure.
Figure 8: Left hand side plot presents the linearity coefficient $k$ of component $C_1$ versus the measurement temperature, as this parameter does not depend on the $T_{pre}$. Right hand side plot presents the contour plot of the linearity coefficient $\mu$ of component $C_2$ versus both preheating and measurement temperatures.

Highlights

1. TL dose response in BeO following irradiation at various temperatures (RT, 125 and 200 °C).
2. OSL dose response in BeO using a combination of different measurement and preheating temperatures.
3. Dose responses were studied for the reconstructed luminescence signals as well.
4. Supralinearity features are monitored for both signals.
5. Competition takes place due both irradiation and heating stages.

Table 1: BeO dose response features of various luminescence signals already existing in the literature.

<table>
<thead>
<tr>
<th>Signal</th>
<th>Authors [Reference]</th>
<th>Deconvolution</th>
<th>Tirr</th>
<th>$T_{pre}$</th>
<th>Measurement $T$</th>
<th>Dose range</th>
<th>Linearity &amp; Saturation</th>
</tr>
</thead>
<tbody>
<tr>
<td>TL and OSL</td>
<td>Tochilin et al., [2]</td>
<td>Yes</td>
<td>RT</td>
<td>-</td>
<td>RT to 400°C (TL)</td>
<td>10 mR - 10^6 R</td>
<td>Lin., Supr. (10 mR-50 R) and Saturation at 10^6 R</td>
</tr>
<tr>
<td>TL</td>
<td>Crase and Gammage, [19]</td>
<td>No (indiv. Peak’s integral)</td>
<td>RT</td>
<td>167 °C</td>
<td>RT to 400°C</td>
<td>1mR-100R</td>
<td>Lin. (up to 1000R)</td>
</tr>
<tr>
<td>TL</td>
<td>Yasuno and Yamashita, [23]</td>
<td>No (indiv. Peak’s integral)</td>
<td>RT</td>
<td>-</td>
<td>RT to 400°C</td>
<td>50R-5000R</td>
<td>Lin. (up to 2000R)</td>
</tr>
<tr>
<td>CW-OSL</td>
<td>Bulur and Göksu, [13]</td>
<td>No (Integrated OSL signal)</td>
<td>RT</td>
<td>125 °C</td>
<td>50 °C</td>
<td>5.6mGy-12.8Gy</td>
<td>Lin. (5.6mGy-12.8Gy)</td>
</tr>
<tr>
<td>OSL after TL</td>
<td>Bulur and Göksu, [13]</td>
<td>Yes</td>
<td>RT</td>
<td>400 °C</td>
<td>50 °C</td>
<td>25 mGy-400 mGy Lin.</td>
<td></td>
</tr>
<tr>
<td>CW - OSL</td>
<td>Sommer and Henniger, [32]</td>
<td>No (maximum intensity)</td>
<td>RT</td>
<td>50 °C</td>
<td>RT</td>
<td>20 μGy-100 Gy Lin.</td>
<td></td>
</tr>
<tr>
<td>TL</td>
<td>Bulur, [24]</td>
<td>No (indiv. Peak’s integral)</td>
<td>RT</td>
<td>180 °C</td>
<td>RT to 450 °C</td>
<td>0.25 – 10 Gy Lin.</td>
<td></td>
</tr>
<tr>
<td>CW - OSL</td>
<td>Sommer et al., [31]</td>
<td>No (maximum intensity, integral or integral over a defined part)</td>
<td>RT</td>
<td>50 °C</td>
<td>RT</td>
<td>10 μGy-100 Gy Lin.</td>
<td></td>
</tr>
<tr>
<td>CW-OSL</td>
<td>Bulur and Yeltik, [33]</td>
<td>No (initial OSL signal)</td>
<td>RT</td>
<td>160 °C</td>
<td>RT</td>
<td>25 mGy - 2.5 mGy</td>
<td></td>
</tr>
<tr>
<td>TL &amp; CW-OSL</td>
<td>Malthez et al., [34]</td>
<td>No (maximum intensity for OSL, integral of peak for TL)</td>
<td>RT</td>
<td>No for TL 250 °C for OSL</td>
<td>RT to 250°C (TL) RT (OSL)</td>
<td>2.7-11 mGy</td>
<td></td>
</tr>
<tr>
<td>TR-OSL</td>
<td>Bulur and Saraç [25]</td>
<td>Yes (fast and slow TR – OSL components)</td>
<td>RT</td>
<td>100 °C</td>
<td>RT</td>
<td>0.5 – 1000 Gy Supr.</td>
<td></td>
</tr>
<tr>
<td>TL and OSL</td>
<td>Groppo and Caldas, [26]</td>
<td>No</td>
<td>RT</td>
<td>-</td>
<td>RT to 450°C (TL) RT (OSL)</td>
<td>a (0.2-200Gy) b (0.1-200Gy) X (0.01-1Gy) Supr.</td>
<td></td>
</tr>
<tr>
<td>CW - OSL &amp; TT-OSL</td>
<td>Yukihara, [17]</td>
<td>No (integrated signal over 120 s stimulation, minus background)</td>
<td>RT</td>
<td>250 °C only for TT - OSL</td>
<td>RT</td>
<td>0.8 – 100 mGy</td>
<td></td>
</tr>
<tr>
<td>M - OSL &amp; TT - OSL</td>
<td>Yukihara, [18]</td>
<td>No (integrated signal minus background)</td>
<td>RT</td>
<td>260 °C only for TT - OSL</td>
<td>RT</td>
<td>50 mGy – 25 Gy</td>
<td></td>
</tr>
<tr>
<td>TL</td>
<td>Martinez Baltezar, and Azorin Nieto, [27]</td>
<td>Yes</td>
<td>RT</td>
<td>-</td>
<td>RT to 400°C</td>
<td>1.3 – 23.4 Gy TL P2</td>
<td></td>
</tr>
</tbody>
</table>

Abbreviations and nomenclature: **Lin.** = Linearity, **Supr.** = Supralinerarity, **Sub.** = Sublinearity, **CW-OSL** = Continuous Wave OSL, **TR – OSL** = Time Resolved OSL, **TT – OSL** = Thermally Transferred OSL, **LM – OSL** = Linearly Modulated OSL, **RT** = Room Temperature
Table 2: Technical, experimental and analysis details of all luminescence signals (both quenched as well as reconstructed) that were the subject of the present study. The parameters that are presented in Italics correspond to the TL peak 3 and CW – OSL component $C_2$. Values inside the parenthesis indicate 1 standard deviation error value. The values of the parameter $k$ were calculated via fitting using the equation 9 within the dose range that is presented in the corresponding column.