Correlation between thermoluminescence and optically stimulated luminescence of α-Al_2O_3:C,Mg

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ABSTRACT

The optically stimulated luminescence (OSL) and thermoluminescence (TL) signals of an Al_2O_3:C,Mg single crystal were investigated as a function of the heating temperature and illumination time before readout, respectively. The TL and OSL results were analyzed in complementary ways, including the area under the curve, whole range and partial integration, and peak intensity. A correlation between the continuous decay of the main TL peak intensity (at 185 ºC at a heating rate of 1 ºC/s) against blue light (470 nm) illumination times and the decay of the OSL signal for higher temperatures was found. Both these results showed that the emptying of the OSL active trap was closely related to the emptying of the trap related to the main TL peak, indicating a correlation of both phenomena.

Keywords: Al_2O_3:C,Mg; OSL; thermoluminescence

1. INTRODUCTION

Thermoluminescence (TL) corresponds to the light emission upon heating of insulating or semiconducting materials previously exposed to ionizing radiation, besides incandescence (blackbody radiation) [1]. These materials find application in radiation dosimetry, particularly in medical physics (radiotherapy, radiation diagnosis and nuclear medicine) [2]. Optically stimulated luminescence (OSL) is similar to TL but in this case luminescence is stimulated by the absorption of optical energy instead of thermal energy [3]. In recent years, OSL has established itself in radiation dosimetry based on the development of dosimeters Al_2O_3:C and BeO [4], together with the search for new OSL dosimetric materials [5-7]. In OSL, optical stimulation releases charge carriers from the traps [3] and the material emits a light signal related to the absorbed irradiation dose [4]. In order for TL and OSL to occur, there must be at least one type of electronic trap that captures charge carriers in a localized energy level within the band gap, and a recombination center from where light is emitted.

Al_2O_3:C,Mg is well-known as a fluorescent nuclear track detector (FNTD) that was originally introduced for optical data storage by Akselrod *et al.* [8]. This material has been successfully used in dosimetry of neutrons, protons and heavy charged particles [9, 10] with superior sensitivity and functionality when compared to other FNTD materials [11]. This material is sensitive to charged particles within a broad linear energy transfer (LET) range requiring little or no post-exposure chemical processing, and being reusable [11]. An additional advantage of Al_2O_3:C,Mg is that laser-induced fluorescence allows for non-destructive fast readout using confocal scanning microscopy [9, 12]. This way, images can be processed automatically with tracks appearing in the form of bright spots on a dark background that can be counted with an image processing software [13, 14].

Al_2O_3:C,Mg single crystals contain high concentrations of F and F^+ centers [8, 10]. In contrast to the predecessor Al_2O_3:C, whose F luminescence centers have a long lifetime of ~35 ms, Al_2O_3:C,Mg luminescence occurs due the presence of high concentrations of F^+ centers with a considerably shorter lifetime <7 ns. Such a short lifetime enables its use as a radiation dosimeter in applications requiring fast luminescence response like dose mapping and real-time optical fiber dosimetry [15, 16]. Also, Al_2O_3:C,Mg crystals contain high concentrations of F_2^{2+}(2Mg) [14] that form F_2^+(2Mg) due to an ionizing radiation induced radiochromic transformation [12, 17]. Since the transformation rate is proportional to the absorbed dose, this material ends up storing the cumulative radiation dose that can be read at 750 nm [18].

Al_2O_3:C,Mg has been investigated for TL and OSL applications [11, 19, 20]. The TL glow curve of Al_2O_3:C,Mg shows a main peak around 170 ºC (1 ºC/s), that is a result of emissions at 325, 415, 520 and 750 nm likely corresponding to F^+, F, F_2^{2+}(2Mg) and of F_2^+(2Mg) centers, respectively [16, 21, 22]. Recently, the luminescence of these defects and their role as recombination centers in the TL process was investigated as a function of temperature, from room temperature (RT) up to 400 ºC [21].

The goal of this work is to investigate the correlation between the OSL and TL signals of Al_2O_3:C,Mg through the following measurements: (i) the TL signal emitted after partial readouts of the OSL signal, and (ii) the OSL signal remaining after partial TL measurements.

2. MATERIALS AND METHODS

The sample investigated was an Al_2O_3:C,Mg single crystal grown by the Czochralski technique by Landauer, Inc., Crystal Growth Division, Stillwater, OK, USA.

The single crystal was cut into a 8 x 1.6 x 0.5 mm^3 rectangular parallelepiped with one polished side. The sample mass was 48 mg.

OSL and TL measurements were carried out using a commercial automated TL/OSL reader made by Risø National Laboratory (model DA-20). TL glow curves were obtained using a heating rate of 1 or 5 ºC/s, from RT to 300 ºC. OSL emission was stimulated using blue light emitting diodes (470 nm, FWHM = 20 nm) delivering 80 mW/cm^2 at the sample position in CW mode. Each OSL measurement was carried out during 60 s with 90% of the maximum LED power. The TL and OSL signals were detected with a bialkali photomultiplier tube behind an UV-transmitting, visible-absorbing glass filter (Hoya U-340, 7.5 mm thick) that blocked the stimulation light while transmitting part of the OSL/TL signal, and a 5 mm dia. mask. Irradiation was performed at room temperature using the built-in $^{90}Sr/^{90}Y$ beta source of the TL/ OSL reader (dose rate of 10 mGy/s) with a total dose of 100 mGy (10 s total exposure). Two protocols were used to investigate the TL-OSL correlation, as follows:

Protocol A: partial OSL

1. Heating up to 300 ºC at 5 ºC/s (to empty the traps), followed by cooling to RT
2. Irradiation for 10 s (100 mGy) at RT
3. CW-OSL measurement during a time interval t_{stop}
4. TL up to 300 ºC at 1 ºC/s

Steps #1 to 4 were repeated with the illumination time (t_{stop}) in step #3 increasing from 1 to 60s, in 1 s steps. First of all, a measure without any influence of illumination time (partial OSL) was performed.

Protocol B: partial TL

1. Heating up to 300 ºC at 5 ºC/s (to empty the traps), followed by cooling to RT
2. Irradiation for 10 s (100 mGy) at RT
3. TL measurement at 1 ºC/s until stop temperature T_{stop}
4. CW-OSL measurement at RT for 60 s

Steps #1 to 4 were repeated with T_{stop} varying from 125 to 225 ºC, in 5 ºC steps. First of all, a measure without any influence of temperature (partial TL) was performed.

The TL and OSL results were analyzed in complementary ways, including the area under the curve (TL and OSL, whole range and partial integration), and maximum intensity (TL main peak at 185 ºC; OSL initial signal). Since all measurements were carried out with the same sample, results were not normalized by the sample mass.

3. RESULTS AND DISCUSSION

Figure 1 shows the TL glow curves of Al_2O_3:C,Mg single crystal obtained with different illumination times, according to protocol A. The glow curves were dominated by emission near 185 ºC (TL main peak), in addition to other low intensity peaks at lower temperatures. Previous works showed that the shallow traps related to these peaks are due to the presence of Mg in the lattice [16], and that the main peak followed a first order kinetics TL mechanism with a trap activation energy of 1.36 eV and frequency factor in the range of 10^{14} s^{-1} [15].

In this work, we noted that the position of the main peak and the TL intensity in the 60-125 ºC range were not affected by the illumination time, which possibly indicates that the traps related to these peaks are not optically active. On the other hand, a significant decrease in the intensity of the main peak even for short illumination times was observed and assigned to optical bleaching due to the partial OSL readouts. The TL behavior as a function of the illumination time was further investigated through the analysis of the area of the TL glow curve from RT to 60 ºC and of the main TL peak (125-225 ºC), together with the peak intensity value of the TL main peak at 185 ºC (Fig. 2). The intensity of the main peak decreased continuously for the whole illumination time interval (from 1 to 60 s), while the intensity of the RT-60 ºC TL signal increased initially and only after about 3 s of illumination it started to decrease. Further, the time decay of these two regions of the TL glow curves was different, with the main peak decreasing with a faster rate. The signal of the main TL peak essentially vanished after about 30 s while that of the RT-60 ºC TL region was still measurable up to 60 s. In order to better illustrate the different behavior of both regions of the TL glow curve, the partial areas were normalized to the respective area of the glow curve obtained with no illumination, as shown in Fig. 2c. As an example, it was noted that after an illumination time of 10 s the area of the TL main peak was reduced to 17 % of the initial value while 72 % of the area of the RT-60 ºC TL signal remained. The increase of the intensity and slower decay rate observed in the RT-60 ºC TL signal was tentatively assigned to a photo-transference of charge carriers from the deep traps to the shallower traps during illumination (at RT) [23]. As reported by Kalita et al [24], this material presents TL peaks at temperatures higher than the maximum temperature used in this work (300°C), and the charge carriers trapped in the centers related to these peaks may be released by the stimulation light and

be retrapped in the shallower traps, contributing to the augment of the low temperature part of the glow curve. At longer illumination times, the carriers trapped in the shallower traps related to the TL peak depicted in the inset of Figure 1 are also released.

The thermal cleaning of the OSL signal following protocol B is presented in Fig. 3 for selected OSL curves (for visual clarity), where a decrease of the OSL intensity can be seen for higher T_{stop} temperature values. These curves were analyzed in terms of their maximum intensity value (*i.e.*, initial OSL signal value) and the total area under the curve, as shown in Fig. 4 as a function of T_{stop}. The behavior of both quantities was similar, with both exhibiting three distinct temperature regimes: the first, from RT to 150 ºC range where the OSL signal remained essentially constant, followed by an intermediate temperature regime (from 150 to 200 ºC) where there was a steep decrease in the OSL signal, and a third regime (>200 ºC) where the OSL signal slowly decayed to zero. The constancy of the integrated and the maximum OSL signal with heating treatments up to about 150 ºC (Fig. 4) showed that the kinetics of the shallower traps (*i.e.*, TL signal from RT up to ~125 ºC) did not play a major role in the generation of the OSL signal. Suporting that traps related to these TL peaks are probably not optically active at this stimulation wavelength. On the other hand, the as T_{stop} temperature reaches the range corresponding to the TL main peak (125 to 150 ºC), a large decay of the OSL signal (integrated and maximum values) is observed, corroborating the hypothesis that the traps related to the main TL peak are also emptied by the blue stimulation light. Moreover, the absence of TL signal above ~225 ºC corresponded to the absence of OSL signal. These results suggested the OSL signal to be exclusively related to the trap related to the main TL peak.

Finally, Fig. 5 presents the integrated OSL and TL signals as a function of the illumination time. In terms of the TL signal, both the main peak intensity (integration

interval: 125-225 ºC), and whole glow curve intensity are shown. As expected, the OSL signal increased with illumination time, with a small trend to a higher values, while the TL signal continuously decreased to zero. The behavior of TL and OSL signals was complementary, suggesting that heat or light stimulation were releasing charge carriers from the same traps. The photo transference from deep traps probably is the phenomenon responsible for the observed small trend of OSL signal to higher values for longer illumination times.

4. CONCLUSIONS

In this work, the correlation between the TL and OSL signals of an Al_2O_3:C,Mg single crystal was investigated. The decay of the main TL peak at 185 ºC (at a heating rate of 1 ºC/s) with blue light (470 nm) illumination and the decay of the OSL signal with thermal treatments show a correlation between both phenomena. These results showed that the emptying of the OSL active trap was closely related to the emptying of the trap related to the main TL peak. Moreover, while heating the irratiated sample up to 150 °C did not affect the OSL signal significantly, heating at higher temperatures rapidly exhausted OSL emission.

5. ACKNOWLEDGMENTS

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6. REFERENCES

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FIGURE CAPTIONS

Figure 1 - TL glow curves of Al_2O_3:C,Mg single crystal obtained under different illumination times using protocol A. The TL signal without previous illumination (TL0s) time is included for comparison. The insertion shows the curve with highlight for the sample without illumination and 1 second of illumination added. See text for details.

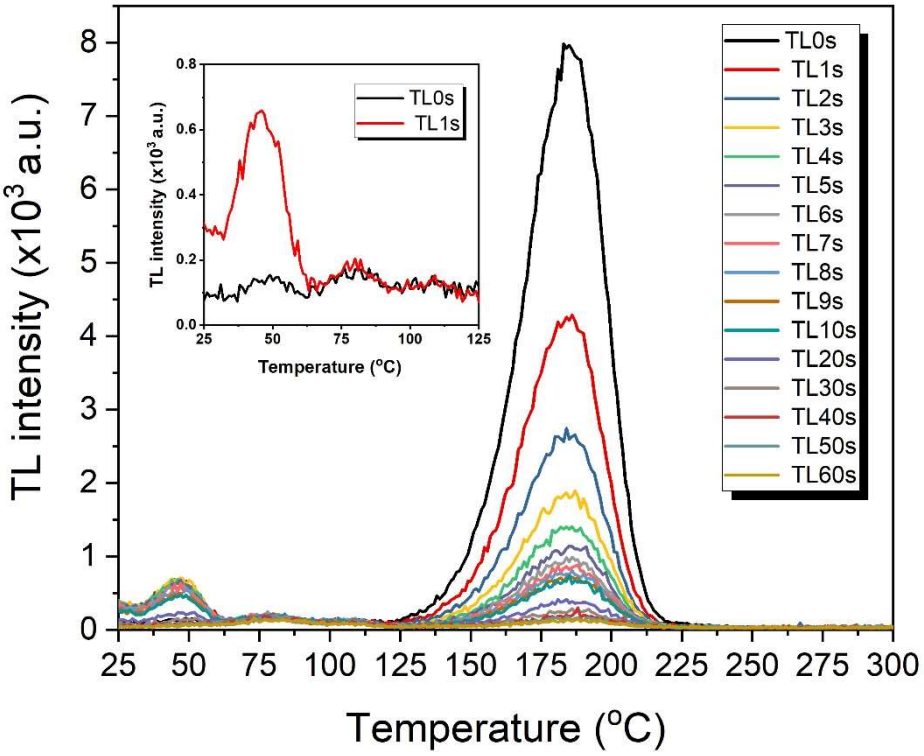


Figure 2 - a) Peak intensity value of the TL main peak (at 185 ºC); b) area of the TL glow curve from RT to 60 ºC (open circles), and of the main TL peak (125-225 ºC; solid triangles); and c) TL area of the main TL peak (125-225 ºC; solid triangles) and of the glow curve from RT to 60 ºC (open circles) normalized to the respective area of the glow curve obtained with no illumination as a function of illumitaion time.

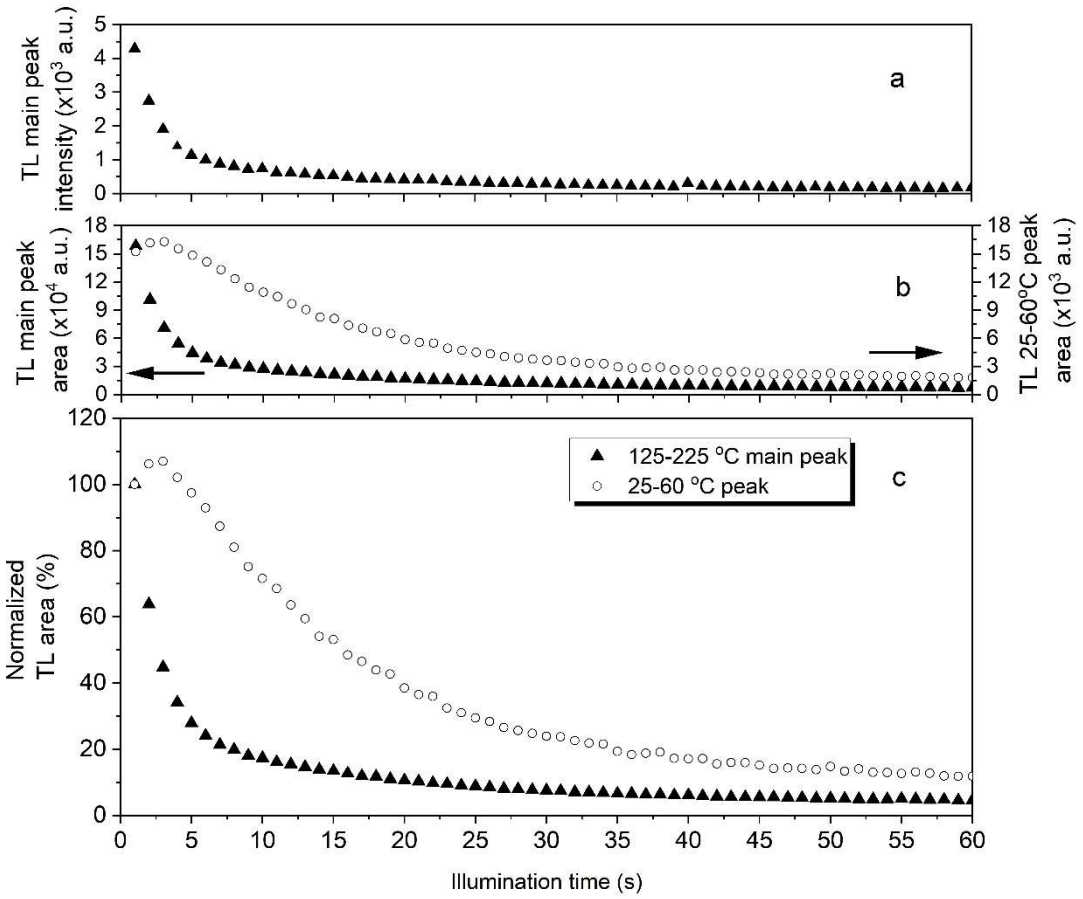
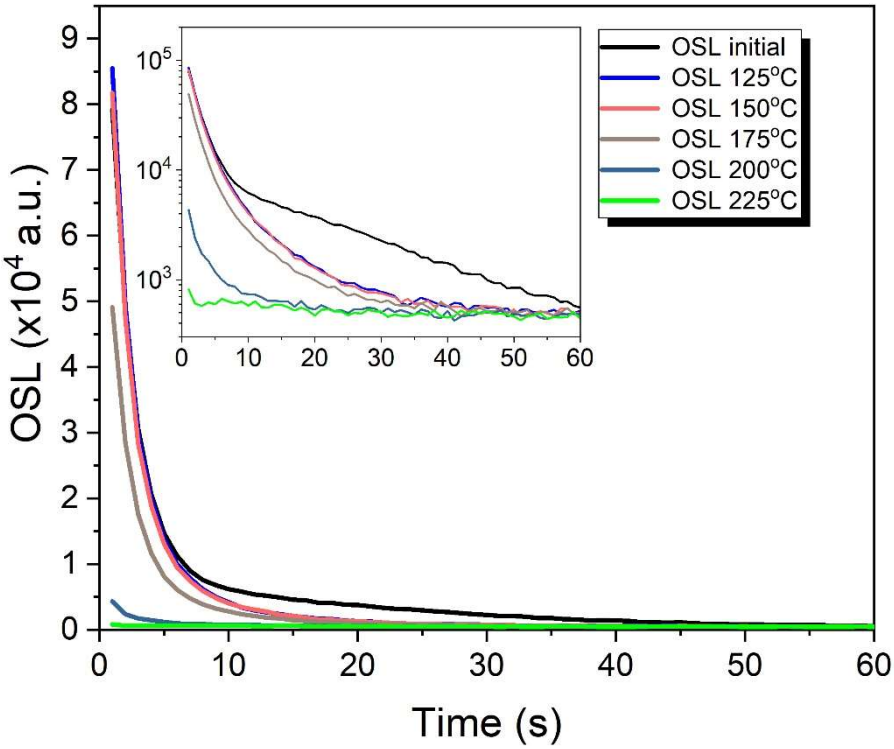
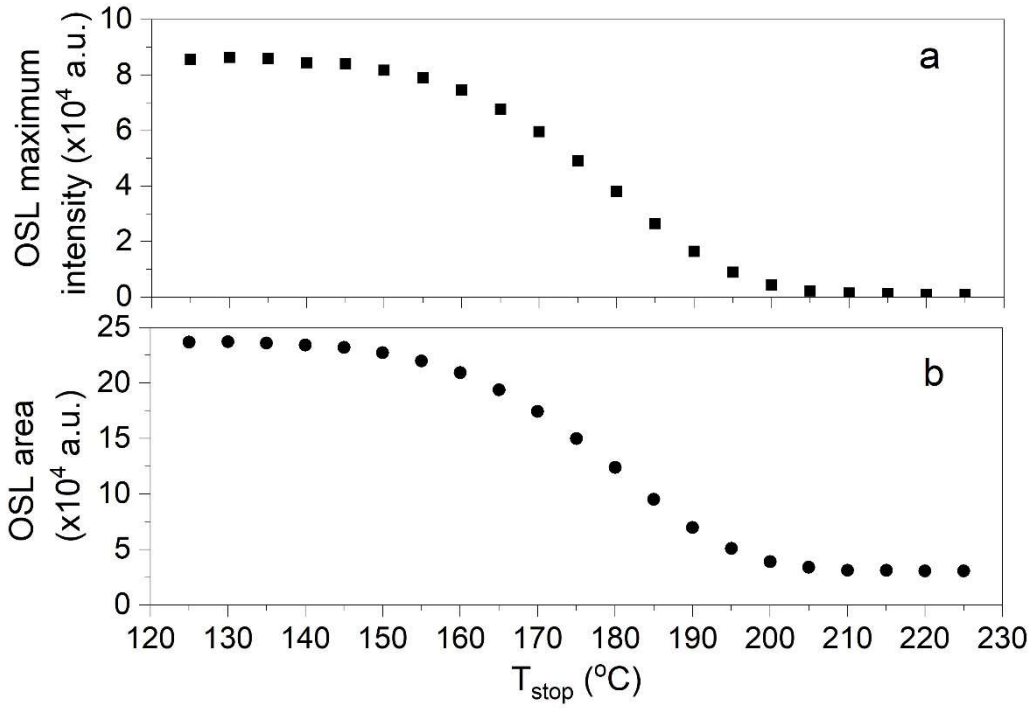


Figure 3 - OSL signal of Al_2O_3:C,Mg single crystal obtained at room after heating till selected temperatures (T_{stop}) using protocol B. The nomenclature OSL XXX ºC corresponds to the limit temperature at which the sample was heated before the measurement at room temperature.The inset presents the same data in semi-log scale to facilitate visual analysis. The OSL signal without previous thermal treatment (OSL initial) is included for comparison. See text for details.



298 **Figure 4 –** a) The OSL maximum intensity (*i.e.*, initial OSL signal value), and b) total

299 area under the OSL curve as a function of T_{stop}.



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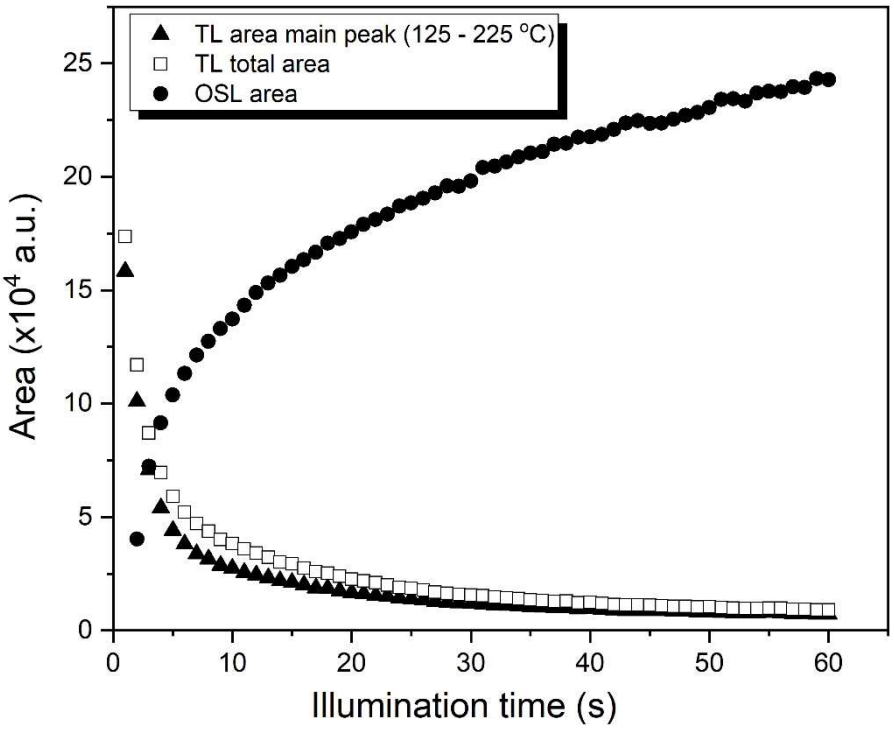
311 **Figure 5 –** Integrated partial OSL signal (solid circles) and integrated TL signal after
312 OSL illumination (whole glow curve (open squares), and main peak (solid triangles)) as
313 a function of the illumination time.



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