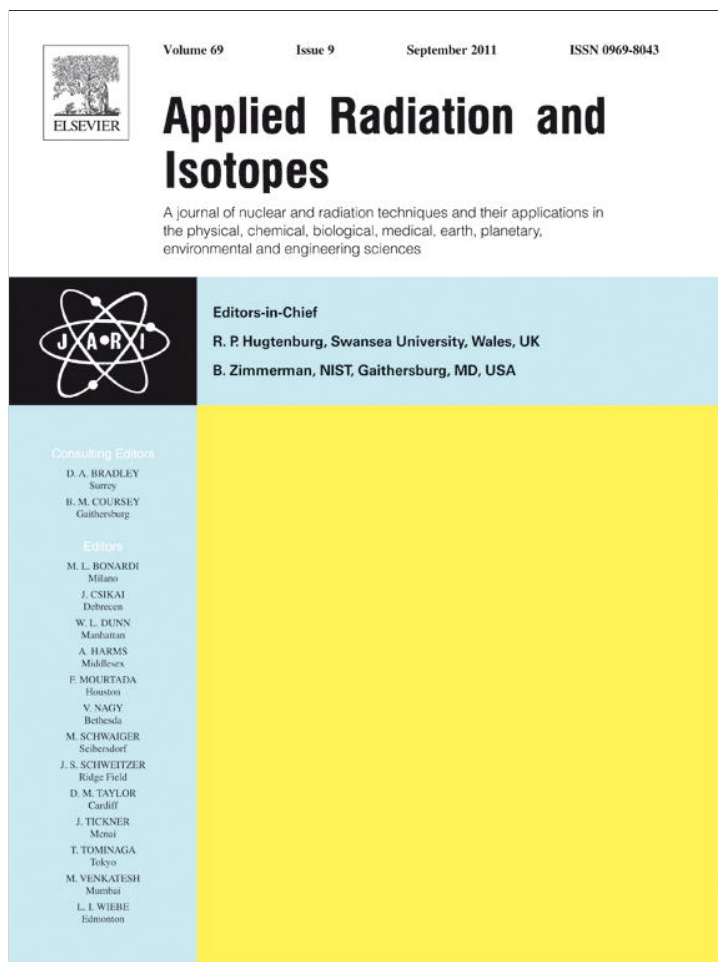


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## Dissolution and subsequent re-crystallization as zeroing mechanism, thermal properties and component resolved dose response of salt (NaCl) for retrospective dosimetry

George S. Polymeris<sup>a,\*</sup>, George Kitis<sup>b</sup>, Nafiye G. Kiyak<sup>a</sup>, Ioanna Sfamba<sup>b</sup>,  
Bhagawan Subedi<sup>b</sup>, Vasilis Pagonis<sup>c</sup>

<sup>a</sup> Faculty of Science and Arts, Physics Department, IŞIK University, 34980-Şile, Istanbul, Turkey

<sup>b</sup> Aristotle University of Thessaloniki, Nuclear Physics Laboratory, 54124-Thessaloniki, Greece

<sup>c</sup> Physics Department, McDaniel College, Westminster, MD 21157, USA

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## ABSTRACT

In the present study we report dosimetric properties of iodized salt aiming at using it as an accidental luminescent dosimeter. It was found that the very good sensitivity of its main dosimetric peak is strongly affected by thermal treatments. This is also the case for OSL emission. The sensitivity loss due to heating implies that caution should be exercised while applying single aliquot protocols for dose evaluation. The sequence of dissolution and subsequent re-crystallization was established to be an extremely effective zeroing mechanism for the TL signal. The linearity in the dose response was also monitored in the case of dissolved and subsequently re-crystallized salt. In the case of naturally occurring salt, zeroing of the TL signal due to dissolution as well as the linearity of dose response up to doses as large as 100 Gy were found to be very promising features for dating applications.

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### 1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) are basic application tools in radiation dosimetry. A special branch of the latter, i.e. retrospective dosimetry, uses heated dosimeters of known exposure times and of well-known age as well, such as bricks and/or tiles, in order to discriminate doses originating from sources other than the natural environment (Göksu et al., 2003). Its main application is accidental dosimetry, where one is interested in the determination of absorbed dose due to a radiation accident or other event, over and above the normal background radiation. Examples include the determination of absorbed doses during events such as nuclear weapons explosions, nuclear reactor accidents or other incidences of unintended radiation release (Haskell, 1993). Nevertheless, retrospective dosimetry deals also with geo-archaeological dating to a wide extend.

Retrospective dosimetry using unheated geological materials could also be feasible, provided that some basic requirements are fulfilled: the candidate material should exhibit a trapping level giving rise to natural TL (NTL) glow peaks above 150–180 °C,

i.e. in a temperature range that is sufficiently higher than the temperature of the unstable TL signals caused by artificial irradiation in the laboratory, so that these two signals will be clearly separable. In the case of OSL, these traps should be unstable over geological time as well as optically active in order to yield an OSL signal. Furthermore, the OSL signal of these traps has to be separable from the possible OSL signal of thermally stable traps or the thermally stable traps have to be light insensitive (Kiyak et al., 2010).

Some further desirable properties are: (i) stable sensitivity over many irradiation-TL readout cycles, so that single aliquot measurements can be applied, (ii) absence of thermal quenching effects, which can cause a substantial decrease in sensitivity as a function of the heating rate during TL measurements, (iii) its trapping levels must be empty (zeroing requirement), while (iv) an easy and fast resetting procedure should take place, (v) to have the lowest possible dose detection threshold and (vi) to have a linear dose response, at least in the dose region of interest (McKeever et al., 1995). These requirements should also be fulfilled in the framework of the application of common salt as a dosimeter and chronometer.

The attention on the TL properties of common, naturally occurring salt arose very early (McKeever, 1985). Photo-stimulated charge transfer was studied even earlier by Stoddard (1960), who demonstrated that a transfer to shallow traps (TL peaks below

\* Corresponding author. Tel.: +30 25410 78787; fax: +30 25410 63656.  
E-mail address: [polymers@auth.gr](mailto:polymers@auth.gr) (G.S. Polymeris).

80 °C) took place following low temperature (liquid nitrogen) irradiation. Further research on its luminescence properties has been carried out either within the context of radiation damage studies (Göksu et al., 1993; Ortega et al., 1993) or for potential use as a retrospective luminescence dosimeter (Bailey et al., 2000; Thomsen et al., 2002; Ankjaergaard et al., 2006; Murthy et al., 2006; Bernhardsson et al., 2009). According to the preliminary study of Bailey et al. (2000) on the OSL properties of NaCl, the OSL signal from NaCl seems to be promising for dating and dosimeter, in detecting doses as low as 1–100 mGy (Thomsen et al., 2002; Ankjaergaard et al., 2006; Bernhardsson et al., 2009). On the basis of their experimental results, Murthy et al. (2006) concluded that common salt can be considered as an accidental dosimeter for a period of 2 weeks. Dose-dependent behavior of the infrared stimulated luminescence (IRSL) decay from NaCl has been studied experimentally using single aliquots irradiated with a range of doses 2–200 Gy. It was observed that the maximum IRSL intensity was not changed by an increase in dose although the total intensity increased as expected (Tanir et al., 2007).

The physical basis of the luminescent signal has been discussed by many authors in the framework of the role of color centers, especially F-centers, keeping in mind the excellent correlation studies on TL and F-centers in alkali halides (McKeever, 1985). The emission mechanism associated with OSL from NaCl has been described by many workers (Bailey et al., 2000; Zhang et al., 2005) who have reported that the detrapping rate of the OSL signal from NaCl was non-linear and followed a complex pattern.

OSL properties of NaCl relative to dating and dosimetry have been investigated recently, mostly because NaCl is not only very easy to optically bleach, but also because it is highly soluble in water and this property suggests a possible geological application in which dissolution is the resetting event and the date obtained represents the date of desiccation (Bailey et al., 2000). To the best of the authors knowledge, a detailed study investigating the dissolution (and subsequent re-crystallization) of salt as a resetting mechanism has not been reported in the literature. Therefore, the study of the sequence of dissolution and subsequent re-crystallization as a zeroing mechanism for salt is one of the main goals of this study. Furthermore, the present study will aim at establishing the usability of salt as a natural retrospective dosimeter and chronometer, by further investigating some luminescent properties for several types of salts of Greek and Turkish origin. Therefore, in the framework of this objective, the study will stress mostly on the effect of thermal treatments on their both TL and OSL sensitivity to ionizing radiation. A similar work was earlier reported by Gartia (2009) in the framework of paleothermometry applications. Moreover, a component resolved TL and OSL dose response analysis, concerning mostly the signal of main dosimetric peak is also reported. Finally, the influence of the dissolution and subsequent re-crystallization to some of these aforementioned properties was also studied in the framework of the application of common salt as a dosimeter and chronometer.

## 2. Materials and methods

The salt samples used in the present work were five types of Greek, iodized salt with commercial names Kalas, Acte, Nike, Rea and Carfour, as well as one kind of Turkish iodized salt with the commercial name Billur tuz, laboratory coded T<sub>1</sub> and one Turkish natural salt. The latter comes from the Salt Lake (TuzGolu), which is the second largest lake in Turkey after Lake Van, with a salt ratio of 32%. Salt Lake has a 25,000 m<sup>2</sup> basin in the Konya region, which has the lowest surface-water levels in Turkey and has seen sharp drops in its underground water levels as well. “Tuz” means

“salt” in Turkish, and this lake actually produces one million tons of salt a year, or 64% of Turkey's total requirements. This lake is fed by the Melendiz river, several small streams and underground salt water springs. This location is also a home to the largest flamingo colony in Turkey.

All measurements were performed using the automated Risø TL/OSL reader (model TL/OSL-DA-15), equipped with an internal <sup>90</sup>Sr/<sup>90</sup>Y beta ray source of dose rate 0.1 Gy/s. Blue light emitting diodes (LEDs) (470 nm, 40 mW/cm<sup>2</sup>) were used for optical stimulation and the TL/OSL signal was detected through U-340 filters of 7.5 mm total thickness. A heating rate of 1 °C/s was used in all TL readouts in order to avoid temperature lag, up to a maximum temperature of 350 °C. All OSL measurements were performed in the Linearly Modulated mode, (LM-OSL), the ramping rate of which was selected as 0.04 mW/cm<sup>2</sup> s<sup>-1</sup>, increasing the stimulation light power from zero up to the maximum power (40 mW/cm<sup>2</sup>) over a stimulation duration of 1000 s. Unless otherwise stated, all OSL measurements were performed at room temperature, namely 25 °C. Unless otherwise stated, a test dose of 5 Gy of β<sup>-</sup> was used for all cases. The experimental protocols used will be described in the corresponding sections.

All TL glow curves and OSL decay curves obtained were analyzed into their individual components by following a computerized curve deconvolution procedure. In the case of TL glow curves the deconvolution was performed using the following general kinetics order (b) expression (Kitis et al., 1999):

$$I(T) = I_m b^{b/b-1} \exp \frac{E}{kT} \frac{T-T_m}{T_m} \times \left[ (b-1)(1-\Delta) \frac{T^2}{T_m^2} \exp \left( \frac{E}{kT} \frac{T-T_m}{T_m} \right) + Z_m \right]^{-b/b-1} \quad (1)$$

where  $\Delta = 2kT/E$ ,  $\Delta_m = 2kT_m/E$  and  $Z_m = 1 + (b-1)\Delta_m$ . Here  $E$  is the activation energy of the trap,  $I_m$  is the maximum intensity at a temperature  $T_m$ , and  $b$  is the kinetic order of the TL process.

All OSL curves were deconvoluted assuming general order kinetics for all possible components. The general order kinetics LM-OSL obtained by Bulur (1996) was used in the form given below, as it was further transformed so that instead of the parameters  $n_0$  and  $\sigma$ , it contains parameters obtained directly from the experimental curves (Polymeris et al., 2006; Kitis and Pagonis, 2008):

$$I(t) = \frac{I_m}{t_m} t \left( \frac{\beta-1}{2\beta} \frac{t^2}{2t_m^2} + \frac{\beta+1}{2\beta} \right)^{\beta/1-\beta} \quad (2)$$

where  $I_m$  and  $t_m$  are the values of OSL intensity and time at the maximum of the LM-OSL peak,  $\beta$  is the kinetic order and  $t$  the time of the optical stimulation. This LM-OSL equation was used for deconvolution of all experimental data. Curve fitting was performed by the MINUIT program (James and Roos, 1977), whereas the goodness of fit was tested by the Figure Of Merit (FOM) of Balian and Eddy (1977), given by:

$$FOM = \sum_i \frac{|Y_{Exper} - Y_{Fit}|}{A} \quad (3)$$

where  $Y_{Exper}$  is the experimental glow curve,  $Y_{Fit}$  is the fitted glow curve and  $A$  is the area of the fitted glow curve. The FOM values obtained were less than 1.5% in all LM-OSL curves, while for the corresponding TL curves of the order of 3–5%. The background for the LM-OSL measurements was simulated by an equation of the form:

$$bg = z_d \left( C + \frac{t}{P} \right) \quad (4)$$

where  $z_d$  is the zero dose OSL signal after blue stimulation and  $P$  the total stimulation time. The parameter  $z_d$  is evaluated experimentally, by measuring the background OSL signal at non-irradiated samples.  $C$  is a sample-dependent constant, very close to unity.

### 3. Experimental results and discussion

#### 3.1. TL glow curve shapes – deconvolution – sensitivity without previous heating

All samples studied, after artificial irradiation, yielded glow curves in which a distinctive glow peak at about 230 °C is observed. Typical glow curve shapes are shown in Fig. 1 for the case of Kalas salt (panel a) as well as the natural Turkish salt (panel b). The deconvolution analysis of all TL glow curves, performed by using the general order kinetics expression of Kitis et al. (1999), indicated 5 or 6 overlapping peaks. Among these, distinctive TL peaks are present in the temperature region between 200 and 300 °C. The main dosimetric TL peak was identified as peak no. 5 in most commercial salt samples (Fig. 1a) while in the case of natural salt TL

peaks no. 5 and 6 (Fig. 1b). The Greek salt sample under the commercial name of Kalas was proven to be the most sensitive sample, and is presented in Fig. 1b. The individual sensitivity of the main dosimetric glow peak of each salt sample after deconvolution analysis, normalized over the sensitivity of the most sensitive sample are given in the first column of Table 1. A test dose of 5 Gy is used for these sensitivity measurements. The lowest detection limit (LDL) is defined to be the signal, which is three times greater than the standard deviation of the background signal. The LDL of the most sensitive salt sample was evaluated to be of the order of 1–5 mGy. This result stands in good agreement with the respective LDL values reported by both Thomsen et al. (2002), as well as Bernhardsson et al. (2009). These authors reported LDL of the order of 1–100 mGy after applying the SAR protocol to their samples.

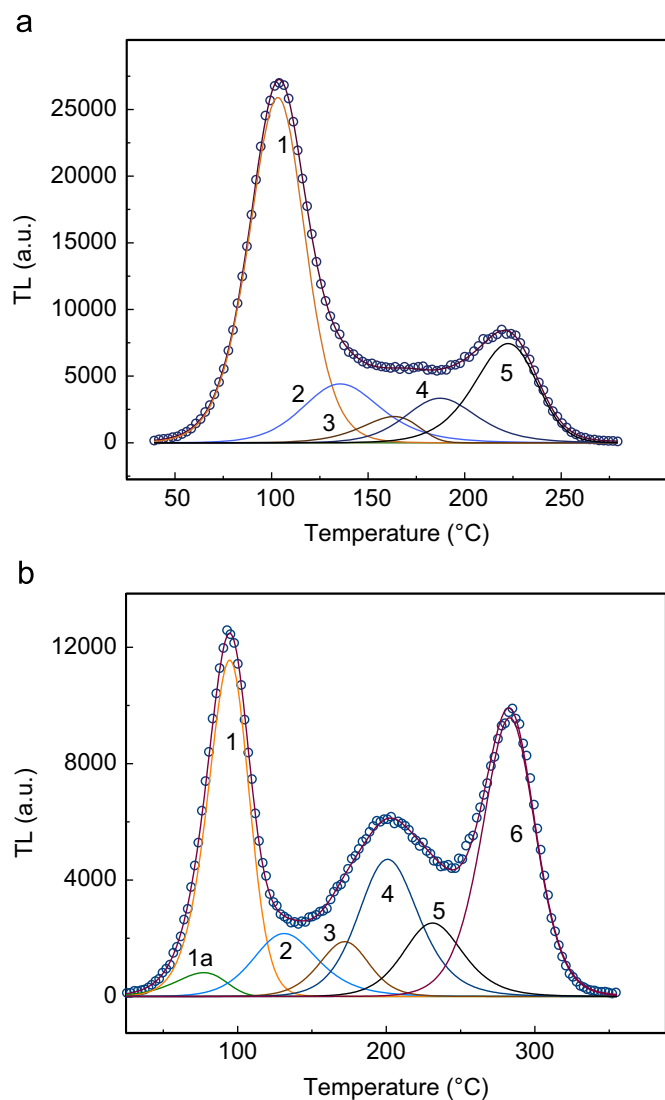
#### 3.2. Thermal properties

Unfortunately all salt samples were found to be extremely sensitive to thermal treatments. In the framework of the previous sections, every virgin (previously unheated) salt sample was irradiated by a test dose and then it was heated up to 350 °C to obtain the initial sensitivity. The same samples were irradiated by the same dose and were heated again up to the same temperature in order to obtain the new sensitivity induced after heating. It was observed that the new sensitivity of the heated sample was dramatically reduced relative to the initial sensitivity of the unheated sample, throughout the entire glow curve temperature region. This result is shown in Fig. 2 for the case of Kalas salt.

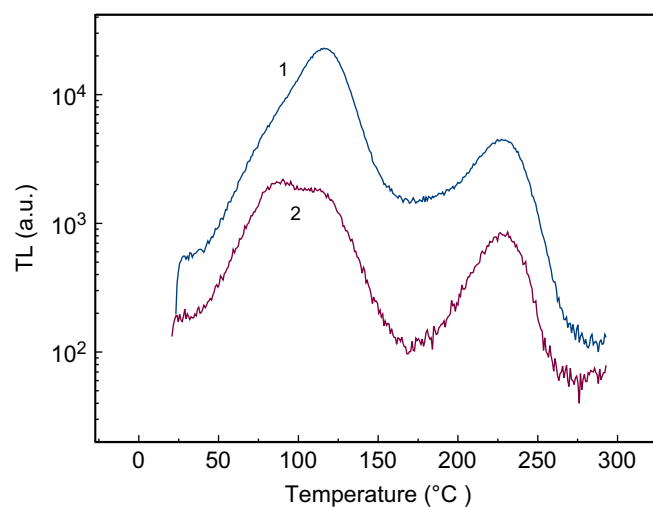
**Table 1**

Sensitivities of the iodized salts investigated relative to the sensitivity of the Kalas salt and the sensitivity loss between the first and second TL readout up to 350 °C at 1 K/s. The lowest detection limit of the Kalas salt is of the order of 1–5 mGy.

Salt	Relative sensitivity	Sensitivity loss after heating (%)
Kalas (GR)	1	86
Acte (GR)	0.7	47
Nike (GR)	0.41	40
Rea (GR)	0.3	49
Carfour (GR)	0.6	79
T1 (TR)	0.2	77
Natural (TR)	0.05	42



**Fig. 1.** TL glow curves of an iodized Greek salt (a) and natural Turkish salt (b) analyzed into its individual TL glow peaks. The FOM (%) values were less than 1.5%.



**Fig. 2.** TL glow curves without any previous heating (1) as well as after the heating applied by the readout to record previous curve (2) in the case of the most sensitive salt sample, i.e. Kalas salt sample.

The same effect was observed in all salt samples. The sensitivity loss of each salt sample before and after irradiation-TL readout cycles is given in the second column of Table 1 for the main dosimetric peak. Once the sensitivity drops substantially from the first to the second TL readout, then it follows a different behavior for more irradiation-readout cycles. These results are shown in Fig. 3, which shows that both the low temperature TL glow peak (curve a) as well as the dosimetric TL glow peak (curve b) are sensitized as a function of the irradiation-readout cycle.

Due to the interesting properties shown in Figs. 2 and 3, the thermal behavior of TL signal from the Kalas salt was further investigated using the following protocol (protocol A):

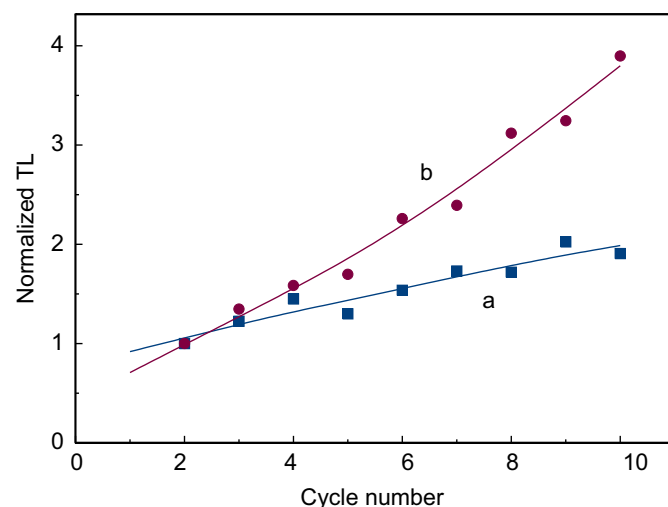
**Step 0:** record the sensitivity ( $S_0$ ) of an unheated sample for a test dose of 5 Gy. **Step 1:** heat an un-irradiated and unheated sample by applying a readout up to temperature  $T_i$ . **Step 2:** give the test dose on the sample of step 1. **Step 3:** TL readout (record sensitivity  $S$ ). **Step 4:** repeat steps 1–3 on a new un-irradiated and unheated sample for a new maximum readout temperature  $T_i$ , where  $T_i$  ranges between 150 and 300 °C.

The resulting ratios  $S/S_0$  are shown in Fig. 4A. The sensitivity of the heated samples is normalized over the sensitivity of the unheated sample. It is observed that the sensitivity of both peaks remains constant for readout temperatures up to 180 °C, and subsequently the sensitivities are sharply reduced in a very similar manner. The stability of the sensitivity of both peaks for readout temperatures up to 180 °C is a useful property, because it allows the height of the low temperature TL glow peak to be used as a mass normalization factor whenever necessary.

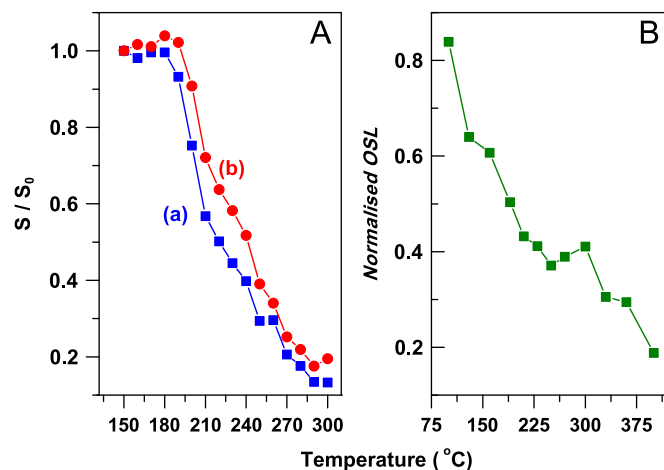
In the case of OSL, the thermal behavior was also studied according to the following experimental procedure (protocol B):

**Step 0:** application of a test dose of 5 Gy. **Step 1:** LM-OSL for 1000 s at RT in order to monitor the OSL signal without any previous heating. **Step 2:** TL readout up to temperature  $T_i$  where  $T_i$  ranges between 100 and 400 °C. **Step 3:** application of a test dose of 5 Gy. **Step 4:** LM-OSL for 1000 s at RT in order to monitor the OSL signal after the previous heating step. **Step 5:** Repeat steps 0–4 on a new un-irradiated and unheated sample for a new maximum readout temperature  $T_i$ .

The ratio of the signal monitored during Step 4 over the respective signal in Step 1 is plotted versus temperature in



**Fig. 3.** Sensitivity of the dosimetric TL glow peak normalized over the sensitivity of the second cycle (the first after heating) as a function of irradiation-readout cycle. Curve a after using a maximum readout temperature of 300 °C, while curve b by using a maximum readout temperature of 500 °C. Strong sensitization of the dosimetric peak is observed up to a factor of 2 in the former case (curve a) and by a factor of 4 in the latter (curve b).



**Fig. 4.** Influence of thermal properties on luminescence sensitivity of salt studied in the case of TL (panel A) as well as LM-OSL (panel B). In the former case, curve a stands for the low temperature shallow TL trap, while curve b corresponds to the dosimetric TL glow peak of salt as a function of the maximum readout temperature applied to unheated samples, normalized over the sensitivity of the unheated sample. In the case of panel B, the ratio of the OSL signal monitored during Step 4 over the respective during Step 1 is plotted versus temperature.

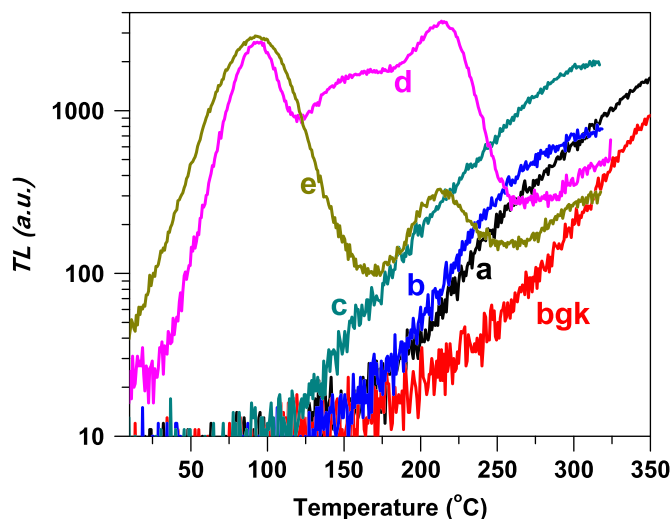
Fig. 4B. Due to the fact that OSL signals in salt are easy to bleach optically (Bailey et al., 2000), 1 ks is an adequately long stimulation interval for the signal to be totally removed. The data in Fig. 4B show that the LM-OSL sensitivity is decreasing even faster than the sensitivities in Fig. 4A. Even at the lowest readout temperature of 100 °C, the sensitivity ratio is of the order of 0.87, while it further decreases with increasing temperature. The data in Fig. 4B has important implications for correct application of the Single Aliquot Regenerative (SAR) OSL protocol (Murray and Wintle, 2000) to salt samples. Indeed, SAR OSL was applied previously to salt and results were already reported in the literature (Bailey et al., 2000; Thomsen et al., 2002; Bernhardsson et al., 2009; Ekendahl and Judas, 2011). In all aforementioned cases, the preheat applied during the measurements was around 180 and 220 °C, in order to remove signal resulting from less stable and shallow TL traps. However, according to Fig. 4, heating the salt at such temperatures would have resulted in de-sensitization of the OSL signal, inducing possible errors in the dose estimation. The SAR OSL protocol was invented in order to monitor and correct for sensitization in the case of OSL signal resulting from quartz. In the case of salt, with strong de-sensitization after the first heating, special caution should be exercised while applying SAR OSL protocols even with the lowest preheat temperatures possible.

### 3.3. Thermal quenching

The presence of the thermal quenching effect was studied by performing measurements as a function of the readout heating rate. It was found that the sensitivity of all salt samples was stable as a function of the heating rate and for heating rates between 1–10 °C/s. It is concluded that thermal quenching does not play a significant role in the luminescence of the samples studied here.

### 3.4. Dissolution and subsequent re-crystallization; a zeroing mechanism for the luminescence signals in NaCl

All commercial iodized salts usually remain in the marketplace for a period of up to three years. In this time period the environmental dose by itself is not adequate to fill the trapping levels. Curve (a) of Fig. 5 presents the NTL signal of the Kalas salt. It is obvious that only some residual thermoluminescence signal



**Fig. 5.** TL glow curve shapes in the case of the most sensitive salt sample, i.e. Kalas, measured after applying the corresponding sequence of procedures: (a) NTL without any other handling, (b) dissolution, subsequent irradiation and subsequent re-crystallization before TL readout, (c) irradiation, subsequent dissolution and subsequent re-crystallization before TL readout, (d) irradiation and subsequent measurement without any dissolution and (e) dissolution, subsequent re-crystallization and subsequent irradiation before TL readout. Curve bgk stands as background measurement.

is present especially in the high temperature region. The same result was yielded in all commercial salt samples. Therefore the zeroing requirement is fulfilled in case of commercial salt samples.

NaCl is highly soluble in water and natural halite deposits are found in areas of aridification (Bailey et al., 2000). This property suggests a possible geological application in which dissolution is the resetting event and the date obtained represents the time of the crystal formation (Bailey et al., 2000). However, this mechanism has not been previously thoroughly studied. In order to study this mechanism in detail, four portions of the most bright salt sample, i.e. Kalas, were treated differently as follows: the first portion was irradiated with a test dose and subsequently dissolved into ordinary water. The solution, which was produced, was then kept in the dark, so that the water was evaporated. Eventually, the TL signal of the re-crystallized salt was measured. The corresponding glow curve is labeled as curve b in Fig. 5. The second portion was treated similarly; however, irradiation took place immediately after dissolution. Curve c represents the corresponding glow curve. All dissolutions took place in the dark.

The third portion was irradiated and its TL signal was measured immediately afterwards, without dissolution, yielding glow curve d. Finally, the last portion was dissolved to distilled water, kept in the dark, so that the water was evaporated and the re-crystallized salt was then irradiated and measured. The latter case is represented by the glow curve e of Fig. 5. Glow curves b, c, d and e are presented along with glow curve of NTL (curve a) and one background curve (bgk) in Fig. 5 for the sake of comparison. Each of the glow curves plotted is the average of four individual measurements. As this figure reveals, the sequence of dissolution and subsequent re-crystallization stands as a very effective zeroing mechanism in the case of already irradiated salt. Furthermore, even in the case of irradiation during dissolution, the TL signal obtained is not distinguishable from the background signal.

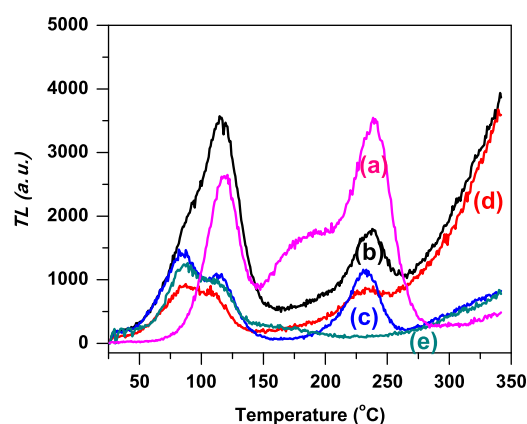
Moreover, dissolution (and subsequent re-crystallization) seems not to affect drastically the shape of the glow curve, as it becomes obvious by the comparison of glow curves d and e. The dosimetric glow peak at about 230 °C (glow peak no. 5 according

to Fig. 1a) is also present after dissolution, even though its sensitivity is much decreased afterwards. Consequently, the varying sensitivity of the main dosimetric peak observed in different salt samples could possibly be attributed to the different number of sequential dissolutions and subsequent re-crystallizations that each respective sample was subjected to. Finally, there is one obvious change in the shape of the glow curve, dealing with the presence of one extremely wide and prominent glow peak centered at around 120 °C. This experimental feature is clearly attributed to the sensitivity enhancement of the glow peak 2 (seen in Fig. 1a), which in conjunction with glow peak 1, result in the first wide glow peak.

### 3.5. Thermal properties after the sequence of dissolution and re-crystallization

In order to check the thermal properties of salt after dissolution, five portions of each sample were prepared and treated as follows: the first portion was irradiated by the test dose and measured immediately without any previous dissolution or heating. All other four portions were dissolved to water and subsequently were re-crystallized. The second portion was irradiated with a test dose and subsequently measured without any previous annealing. Finally, the remaining three portions were annealed before irradiation at 300, 350 and 400 °C, respectively. Fig. 6 presents the combined impact of the sequence consisting of dissolution, subsequent re-crystallization and heating on the glow curve shape and intensity of salt samples. Curve (a) corresponds to the glow curve of the Kalas salt received after neither heating nor dissolution, while curve (b) to the respective TL glow curve after dissolution, re-crystallization and subsequent irradiation of 5 Gy. Curves c, d and e correspond to samples, which after dissolution and re-crystallization, were subjected to pre-irradiation annealing at 300, 350 and 400 °C, respectively.

As inspection of Fig. 6 reveals, dissolution and subsequent re-crystallization decreases the sensitivity of the main dosimetric peak, while at the same time it increases the sensitivity of the group of glow peaks 1, 1a and 2, and especially peak 1a. This feature is also seen in Fig. 5. Furthermore, annealing after dissolution still affects strongly the sensitivity of the entire glow curve, causing strong de-sensitization. This is monitored for all the peaks of the glow curve. It seems that even after the procedure of dissolution



**Fig. 6.** TL glow curve shapes in the case of the most sensitive salt sample, i.e. Kalas, measured after applying a combination of dissolution and thermal treatments: curve a corresponds to the glow curve after neither heating nor dissolution, while curve b to the respective after dissolution, re-crystallization and subsequent irradiation of 5 Gy. Curves c, d and e correspond to samples, which after dissolution and re-crystallization, were subjected to pre-irradiation annealing at 300, 350 and 400 °C, respectively.

and subsequent re-crystallization, the salt samples seem to present the same variation in luminescence sensitivity in the case of the first and second TL readouts. As the annealing temperature increases, the sensitivity of the main dosimetric peak seems to decrease, while after a combination of dissolution and subsequent annealing at 400 °C, this specific peak is not present. However, de-sensitization of the main dosimetric peak is not so abrupt after re-crystallization. Regarding the group consisting of glow peaks 1, 1a and 2, sensitization seems to be stabilized for annealing temperatures greater than 300 °C, indicating the same results as without a previous dissolution. The same results were obtained for all samples under study.

The impact of heating to the TL sensitivity was studied for all salt samples also after dissolution of different salt concentrations and re-crystallization. Ten different solutions were prepared, consisting of salt as solute and water as solvent. Each one of these solutions contained different salt concentration, ranging between 3.6 mg per 10 ml and 72 mg per 10 ml in steps of 7.2 mg per 10 ml. These concentrations were selected so that none of these solutions was saturated. All dissolutions took place in the dark, with subsequent re-crystallization also in the dark. Two aliquots were selected from each solution, which were subjected to two sequential TL measurements at 350 °C. The de-sensitization was studied for both integrated signal of TL peak group consisting of 1, 1a and 2 glow peaks, as well as for the main dosimetric peak. Results are presented in Fig. 7. In the former case, the signal is decreased by almost one order of magnitude (curves a and b: corresponding to the first and second TL reading, respectively), indicating strong de-sensitization. This de-sensitization is found to be independent of the salt concentration in the solution. However, in the case of the main dosimetric peak (curves c and d: corresponding to the first and second TL reading, respectively), there is still some de-sensitization, but to a smaller degree than in the case without dissolution. According to Table 1, the sensitivity loss after first heating for the Kalas salt was estimated to be 87%. The combined effect of dissolution and TL measurement – heating results in a decrease in the sensitivity of the main dosimetric peak by almost 50%. In other words, dissolution and subsequent re-crystallization makes the salt samples less sensitive to thermal treatments in the case of the main dosimetric peak.

### 3.6. TL/OSL dose response

The TL dose response of all salt samples listed in Table 1 was studied for doses up to 100 Gy for the main dosimetric peak by applying deconvolution analysis. In the case of Kalas sample, the TL dose response of the main dosimetric glow peak was also studied after dissolution and re-crystallization. Glow curves after various doses are presented in Fig. 8 for both cases. Curves a–e correspond to irradiated samples without any previous dissolution, for doses ranging between 0.5 and 10 Gy. Since we are interested solely in the main dosimetric trap, the time elapsed between irradiation and measurement is 3 days. At the same figure, curves 1–6 correspond to samples that have been subjected to pre-irradiation dissolution and subsequent re-crystallization, for the same dose region. Results are presented in Fig. 9 in the case of the main dosimetric peaks of the Kalas salt (curve a, peak 5 of Fig. 1a) as well as for both main dosimetric peaks in the case of the natural Turkish salt (curves b and c,

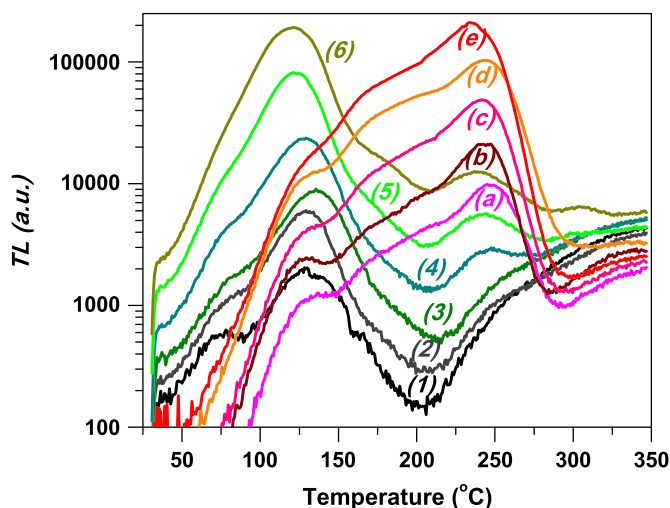


Fig. 8. TL glow curves at various different doses. Curves a–e correspond to irradiated samples without any previous dissolution, for doses (a) 0.5, (b) 1, (c) 2.5, (d) 5 and (e) 10 Gy, while curves 1–6 correspond to samples that have been subjected to pre-irradiation dissolution and subsequent re-crystallization, for the same doses. Curve 6 corresponds to 20 Gy.

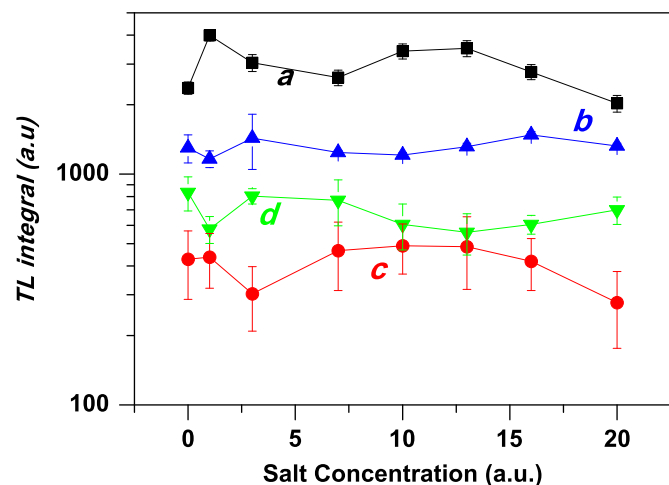


Fig. 7. TL de-sensitization after dissolution of different salt concentrations and re-crystallization, in terms of (a) the integrated signal of TL peak group consisting of 1, 1a and 2 glow peaks (curves a and b: first and second TL reading, respectively), as well as (b) the main dosimetric peak (curve c and d: first and second TL reading, respectively). 1 au corresponds to 7.2 mg per 10 ml.

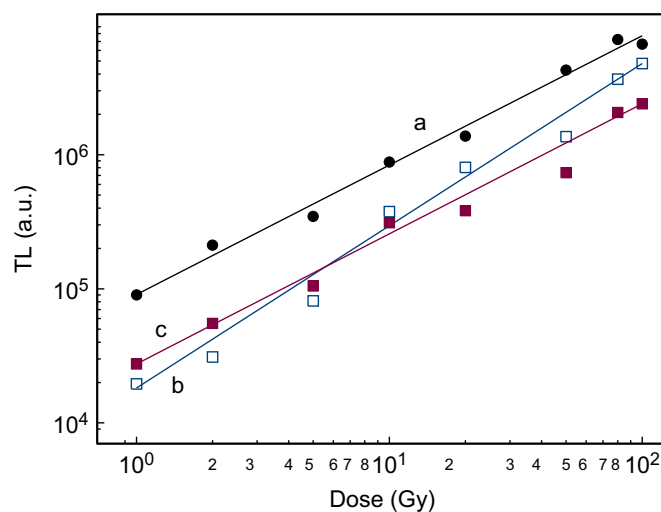


Fig. 9. TL dose response of the dosimetric TL glow peaks of salts. Curve a is for the dosimetric TL glow peak of Kalas salt. Curves b and c for the two dosimetric peaks of natural Turkish salt.

peaks 5 and 6 of Fig. 1b, respectively). The linear behavior of these dose response curves, extending up to almost 100 Gy, indicates that the natural salt is a suitable dating material. All other, commercial salt samples yielded TL dose response curve extremely linear for doses ranging from 0.25 up to 10 Gy. Thus commercial salt was proved to be a potentially useful accidental retrospective dosimeter. Despite the decrease in the sensitivity of the main dosimetric peak, the dose response after dissolution and re-crystallization is still linear in the same dose region (data not shown).

In the case of OSL, a component resolved dose response was obtained. Therefore, LM-OSL curve shapes were analyzed and resolved into their individual components. A typical example is shown in Fig. 10, for the Kalas salt sample. Based on LM-OSL curve fitting analysis, four non-first-order kinetics components were identified for all salt samples. The FOM values obtained for the goodness of fit, were better than 1% for all cases, indicating excellent fits. Similarly with TL, linearity in the same dose region is found also for the fast OSL component for the Kalas salt, as well as for the natural Turkish salt, as shown in Fig. 11. Therefore, the

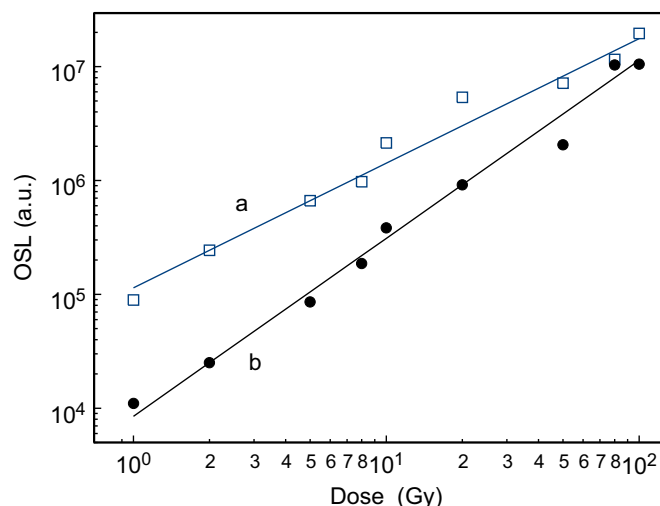


Fig. 11. OSL dose response of the faster OSL component of salt. Curve a for the Kalas salt and curve b for the natural Turkish sample.

dose response of these salts meets the linear dose response requirements for retrospective dosimetry.

#### 4. Conclusions

The conclusions regarding the potentiality of using salt samples as retrospective dosimeters can be summarized as follows:

- (1). Both common commercial as well as naturally occurring salt are excellent candidates for retrospective dosimetry, not only due to their widespread use and presence, but also due to desirable luminescence features like very fast optical bleaching and generally simple TL glow curve shapes.
- (2). The sensitivity loss due to heating during TL/OSL readout is probably problematic while applying single aliquot protocols for dose evaluation, especially in the case of TL. Retrospective dosimetry application using common salt may be more accurate if based on multiple aliquot protocols. However, this is not a major drawback due to the availability of salt samples in large quantities. Special caution should be exercised while applying SAR OSL protocols even with the lowest preheat temperatures possible.
- (3). The sequence of dissolution and subsequent re-crystallization was established in this paper as an extremely effective zeroing mechanism for the luminescence signals. Following the sequence, the TL glow curve does not change its shape. However, the sensitivity of the main dosimetric TL peak is reduced drastically by the dissolution and re-crystallization process. De-sensitization due to thermal treatments is also present following dissolution; however, it was found to be less drastic. Nevertheless, the issue of controlling aspects relating to the variation in environment at the time of crystallization needs further work.
- (4). All salt samples have very high TL/OSL sensitivity and a linear TL/OSL dose response, which makes them suitable for retrospective dosimetry. This linearity was also observed in the case of re-crystallized salt. In other words, the dissolution and subsequent crystallization does not change the dose response features of the salt sample.
- (5). In the case of naturally occurring salt, zeroing due to dissolution as well as the linearity of dose response up to 100 Gy could result in the effective use of NaCl as

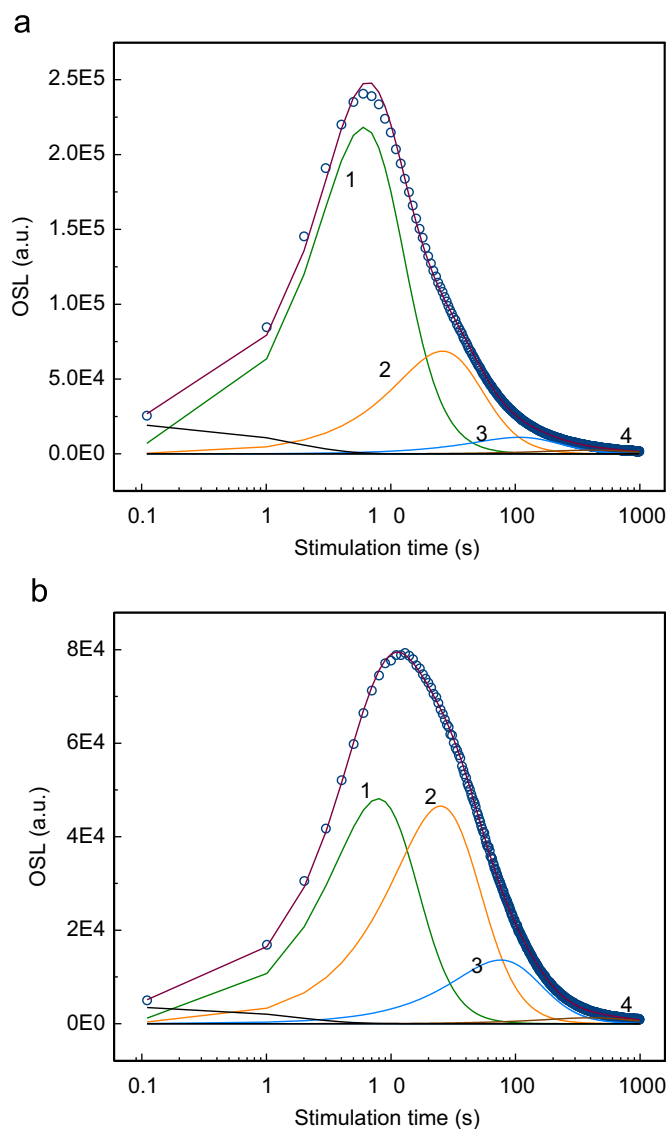


Fig. 10. LM-OSL analyzed into their four individual components of non-first-order kinetics (a) iodized Greek Salt and (b) natural Turkish salt. The FOM (%) values were of the order of 0.8%.

a chronometer in order to date of the last dissolution event or the date of last exposure to light for the material.

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