Effect of Particle Size on TL Dosimetric Properties of Household Salt

Ülkü Rabia Yüce a,*, Birol Engin b

a Turkish Atomic Energy Authority, Sarayköy Nuclear Research and Training Center, 06983 Saray, Kazan, Ankara, Turkey, ulku.yuce@taek.gov.tr

b Dokuz Eylül University, Faculty of Science, Department of Physics, 35160 Buca, İzmir, Turkey, birol.engin@deu.edu.tr

*Corresponding author. Tel.: +90 3128101705; Fax: +90 3128154307

E-mail address: ulku.yuce@taek.gov.tr
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Abstract

This work investigates the effect of particle size in the thermoluminescence (TL) response of household salt that classified into five size fractions between 63 and 250 µm. Irradiation induced two TL peaks at 110°C and 240°C for each particle size fraction. Besides these peaks, two additional glow peaks were also observed at 80°C and 300°C for small particle size fractions < 90µm. The complexity of the glow curve pattern increased with decreasing particle size (<125 µm). The intensity of the dosimetric TL peak at 240°C increases with increasing particle size. The maximum beta radiation sensitivity of salt samples was obtained for the particle sizes of 125-180 µm and 180-250 µm. For different particle sizes, the stability of dosimetric TL peak at room temperature was studied over a storage period of 30 days. The kinetic parameters of dosimetric TL peak were determined at different particle sizes using the initial rise (IR), peak shape (PS) and variable heating rate (VHR) methods and the results are discussed in detail. Moreover, the variations of the glow peaks depending on the heating rate were also studied.

Keywords: Salt, Thermoluminescence, Retrospective dosimetry, Particle size, Beta irradiation
1. Introduction

Thermoluminescence (TL) in alkali halides has been studied extensively during the past. F-centers are the major contributor for the glow curve of alkali halides (McKeever, 1985; Ortiz et al., 2005), it does not mean that such defects can act alone. As a matter of fact some other defects might be also responsible for some part of the glow curve. Those can be mainly interstitials and vacancies among others, but trapped electrons or holes in pre-existing impurities or other point imperfections might be important (Ortiz et al., 2005). When irradiated alkali halides are heated, they give rise to TL because of the thermal annealing of color centres with suitable radiative recombination at the emission center (McKeever, 1984). In this category, sodium chloride (NaCl) is one of the popular materials since it has promising dosimetric properties. The attention on the TL properties of common, naturally occurring salt arose very early (McKeever, 1985). Recently, a lot of research has been focused on the TL and OSL (Optically Stimulated Luminescence) dosimetric properties of the common household salt (NaCl) crystals (Thomsen et.al.,2002; Gartia et.al.,2004 ; Murty et.al., 2006; Tanr et.al., 2007; Bernhardsson et.al., 2009 ; Fujita et.al., 2011; Ekendahl and Judas, 2011 ; Christiansson et.al., 2011; Polymeris et.al., 2011; Spooner et.al., 2011, 2012; Hunter et.al., 2012; Masterhazy et.al., 2012; Timar-Gabor and Trandafir, 2013; Christiansson et.al., 2014; Druzhyna et.al., 2016). These literature works concluded that table salt or house-hold salt has properties suitable for accidental dosimeter. For example, Spooner et al. (2011) collected 19 salt samples from 5 different continents for TL measurements. They suggested that doses of ~1 mGy can be measured using some salts. Luminescence properties for several types of salts of Turkish and Greek origin were investigated by Polymeris et al. (2011). The dose detection limit of the most sensitive salt sample was evaluated to be of the order of 1-5 mGy. This result stands in good agreement with Thomsen et al., (2002) and Bernhardsson et al. (2009) who reported dose detection limit of the order of 1-100 mGy using the single aliquot regeneration
(SAR) protocol. In a very recent work on Israeli salts (Druzhyna et al., 2016), the standard deviation increased below ~1 mGy and the dosimetric glow curve showed large statistical fluctuations which rendered the TL measurements unreliable at 0.5 mGy.

Sample preparation of salt for TL dosimetry usually involves mechanical operations, like grinding for powdering the salt. These operations may introduce defects and changes in the morphology of the salt, with the consequent induction of TL signals which may affect the radiation-induced dosimetric signal evaluation. The possibility of inducing free radicals by mechanical operations in bone and tooth enamel has been reported a long time ago (Marino and Becker, 1968; Polyakov et al., 1995). On the other hand, several researchers have suggested that grinding and milling treatments of some natural minerals cause significant changes in the TL spectra of mineral. The introduction of intrinsic defects by mechanical grinding and milling in natural minerals, such as natural calcite, produces new TL peaks or selectively enhances some glow peaks (Khanlary and Towsend, 1991). Göksu et al. (1988) studied the changes in TL and cathodoluminescence (CL) glow curves and spectra due to calcite defects and heat treatment. They found that introducing surface defects on calcite crystals or heating the single crystal samples at temperatures above 350ºC caused an increase in the TL signal after irradiation. The defects induced by the surface deformations and heat treatments had a broad spectrum in the 300-500 nm region, centered on 400 nm. Ranjbar et al. (1999) showed that the TL intensity of the coarse grain quartz powder produced by mortar and pestle increases with decreasing particle size down to a size of 38 μm, after which it decreases with decreasing particle size of the material. In the case of inorganic polymineral dust samples collected from beta irradiated oregano, the TL intensity increases with the increase in particle size (Beneitez et al., 1994). The luminescence spectra of aliquots of natural transparent alkali feldspars, which were crushed at different energies, show the irreversible collapse of the UV 290 nm peak (Garcia-Guinea and Correcher, 2000). The
grinding and milling of these samples also alters the temperature position and intensities of the UV-blue spectra emissions.

On the other hand, the dosimetric properties of TL materials mainly depend on the kinetic parameters of its glow peak. They are the trap depth (E), the frequency factor (s) and the order of kinetics (b). Using the 238°C glow peak of the common iodised salt (NaCl:I) the activation energy (E), the order of kinetics (b) and the frequency factor (s) values were calculated as 1.0792 eV, 1 and 2.14x10^{10} s^{-1}, respectively, by using the peak shape (PS) method (Murthy et al., 2006). Spooner et al.(2011) applied the method of variation of heating rates (VHR) to measure the electron trap depth E, frequency factor s and thermal quenching behavior of the salt TL peaks observed at 100°C, 200°C and 240°C (5 K s^{-1}). In this study, salt samples were prepared as 180-250µm grains. The principal TL peak at 240°C was found to have E=1.45 eV and s=7.9x10^{13} s^{-1}, giving a lifetime of approximately 4 ka at 20°C. The two other readily identifiable TL peaks, at approximately 100°C and 200°C, were similarly analysed and found to have lifetimes of approximately 7 h and 0.6 ka at 20°C, respectively. In a later study, the TL curves of 1 Gy beta irradiated common salt were also deconvoluted. The activation energy (E) and frequency factor (s) values obtained from the deconvolution of the 220°C dosimetric peak (5°Cs^{-1}) are given as E= 1.46 eV and s= 2.8x10^{14} s^{-1}, respectively (Rodriguez-Lazcano et al., 2012).

Although there have been several studies of the dosimetric properties in household salt, to the best of our knowledge, there have been no systematic studies of the effect of particle size on the TL, kinetic parameters and dosimetric properties of salt. With the above mentioned literature background the present paper reports a study of TL of household table salt (NaCl) crystal powder. The glow curve pattern of zero dose (i.e. not irradiated in the laboratory), beta exposed samples, the effect of particle size on the TL, kinetic analysis, storage stability and dose responses of salts are discussed.
2. Materials and Methods

The common household table salt (commercial name in Turkey is Salina) used in this study has been purchased from regular supermarkets in Ankara (Turkey). The origin of the powdered salt samples is the natural Salt Lake (Tuz gölü in Turkish) which is the second largest lake in Turkey and one of the largest hypersaline lakes in the world. All samples were acquired from supermarkets in which no special care had been taken to avoid exposure to daylight, hence initial processing in the laboratory was also conducted under white light conditions as in the work of Spooner et al. (2012). To obtain require grain size fractions, the purchased salt samples were sieved by using test sieves (Retsch GmbH, Haan, Germany). For the size fraction < 63 µm, the samples were ground by hand with mortar and pestle. The samples produced by these sieving and grinding methods yielded powders with particle size fractions of 180-250 µm, 125-180 µm, 90-125 µm, 63-90 µm and <63 µm. It was not attempted to obtain the smaller grains from 63 µm because of the moisture sensitivity of NaCl. No another sample treatment was performed before TL measurements. All subsequent procedures such as irradiation, storage and TL measurements were performed at normal laboratory conditions (about 20 ± 2°C, relative humidity of 30 %) and in the darkness.

TL measurements were carried out using the automated Risø TL/OSL reader (model TL/OSL -DA-20), equipped with an internal calibrated $^{90}$Sr/$^{90}$Y beta ray source of dose rate 0.137 Gy/s. Light detection was carried out with a photomultiplier tube (PMT) bialkali EMI 9235QA which has an extended UV response with maximum detection efficiency between 300 and 400 nm. To prevent scattered stimulation light from reaching the PMT, a 7.5 mm Hoya U-340 detection filter, which has a peak transmission around 340 (±80) nm, is employed. All the TL measurements were performed using a linear heating rate of 5°C s$^{-1}$ from room temperature up to 500°C in a nitrogen atmosphere. Approximately 5 mg of the sample was spread uniformly on the sample holder to ensure good thermal contact with the sample heater during TL
measurements. In situ $^{90}\text{Sr}/^{90}\text{Y}$ beta source was used for irradiation. The household salt samples were irradiated at approximately 0.4, 0.7, 1.4, 3, 5, 7, 10, 15, 20, 30 and 55 Gy dose values in order to get dose-response curves. The uncertainty in radiation doses was nearly 2%.

In this work, all samples were irradiated and measured only once and hence, sensitivity change associated with multiple regenerations of absorbed dose and heating was of no concern for the experiments (Bernhardsson et.al. 2009). The TL curves were recorded immediately after the irradiation so that there was no significant fading of the peaks at normal laboratory conditions.

3. Results

3.1 Glow Curve Features

Fig. 1 shows the TL curves of zero dose samples with different particle sizes. The TL curves of the zero dose common household salt samples with particle sizes ranging from 63 to 250 μm show native background glow peaks near 300°C. In addition, another native background TL peaks near 220 °C and 420°C were also observed for the size fraction 63-90 μm. As seen from Fig.1, there is not any systematic correlation between the TL peak intensities (or glow peak shapes) and particle sizes.

Zero dose TL signal in salt samples may originate either as a signal-of-formation, as a photo-induced signal due to exposure to daylight, in the case of rock salt, as a result of dose accrual during burial or as a mechanically induced signal due to grinding (Bernhardsson et.al., 2009; Spooner et.al., 2011).

The existence of these background signals in the region of the dosimetric peak may lead to a non-reliable dose assessment, so it is an unwanted situation for a good retrospective dosimetric material. Since the intensities of these background signals were very low, their presence did not obscure the intensity of the radiation-induced TL peaks in the dose range studied (0.4-55 Gy). Even at the lowest applied beta dose (0.4 Gy), the intensity of the
radiation-induced TL peak was a thirty-fold the TL peak intensity of the native background signal except that the size fraction of 63-90 μm (two-fold).

Fig.1. TL glow curves of the zero dose household salt samples for different particle sizes.

Fig.2 shows the characteristic TL glow curves for household salt samples with different particle sizes irradiated with a test dose of 1.4 Gy. As can be seen in Fig.2, the TL glow curves of the samples with particle sizes below 90 μm show glow peaks centered on near 80°C, 110°C, 240°C and 300°C. When the particle sizes are greater than 90 μm, simpler glow curves were obtained. In this case, glow curves of beta irradiated salt samples consist of two main peaks at 110°C and 240°C (Fig.2). Contrary to the results of Ranjbar et al. (1999) on powder quartz samples, as can be seen in Fig.2, the TL intensities of the main peaks centered at about 110°C and 240°C increases with increasing particle size. Similar results were also observed for 1.7 and 3 kGy beta irradiated polymineral (quartz+calcite+ philosilicates) dust samples (Beneitez et al., 1994). The decrease in the response of the finer powders, when compared with the coarser powders, is attributed to lower available number of the NaCl molecules. In other words, the grinding process creates a greater number of defects in the surface molecules of the finer powders so that a lower number of the molecules remain to
undergo radiation interaction and to produce TL signal (Ranjbar et al., 1999; Fukuchi, 1993). Such defects in which the sodium-chloride bond is cut and the electrons are trapped at chloride (anion) vacancy sites on the surfaces of the ground particles.

As can also be seen in Fig.2, the TL glow peak intensities increase with increasing particle size and they are almost the same intensity in the size ranges 125-180 µm and 180-250 µm. The results of preliminary investigations suggest that the household salt can be used effectively for retrospective dosimetry purposes by using its high temperature TL peak at 240°C (measured at 5°Cs⁻¹) (Spooner et al., 2011). Therefore, the TL response of this dosimetric peak at 240°C with different particle size as a function of the exposure to beta radiation was measured from 0.4 Gy to 55 Gy (Fig.3, a, b). In the studied dose range, for all particle sizes, beta radiation caused a significant increase in the intensity of the dosimetric glow peak at 240°C. These dose-response curves could also be fitted with the same linear mathematical functions in the log-log scale, but differed by a proportionality (sensitivity) factor. The differences in the tangents of the four dose-response curves suggest that the salt samples with different particle sizes have different amount of traps. As shown in Fig.3a and 3
b the greatest TL sensitivity (tangent) was obtained for the particle sizes of 125-180 µm and 180-250 µm. In the other hand, there was not a significant difference in TL sensitivities of these two particle sizes. Hence, we suggest that both of these particle sizes can be used in beta dose measurements.

Fig. 3. Dose response curves of TL glow peak at 240°C of household salt sample for different particle sizes. All powdered salt samples were irradiated at beta doses ranging (a) from 0.4 to 5 Gy and (b) from 5 Gy to 55 Gy. Signals were integrated over the temperatures at between 200 °C and 300 °C.
3.2 Effect of Storage Time at Room Temperature on TL Glow Peak Intensity

It is important to know the stabilities of the traps connected with the dosimetric TL peaks, since these reflect the storage capacities of the traps. The knowledge of the fading trend could be useful for application in retrospective dosimetry. In order to determine the thermal fading of the glow peaks, several samples with the particle sizes of 63-90 µm, 90-125 µm, 125-180 µm and 180-250µm were irradiated to 5 Gy beta dose and they were kept out in dark place at room temperature with humidity around 30% RH. The stored samples were read at different interval times within the period of 30 days. Some of the measured glow curves of common household salt (125-180µm) at the end of the planned storage periods are shown in Fig.4. As seen from this figure, the low temperature glow peak at 110°C disappeared after 20 days storage and the intensity of high temperature dosimetric peak reduced by about 20% of its original value after 20 days storage. Almost same glow curve behavior with storage time was also observed for all particle sizes of the samples.

Fig. 4. Glow curves of household salt demonstrating loss of TL signal intensity as a function of storage time in the dark. Samples were irradiated with 5 Gy. The particle sizes of the samples were 125-180 µm.

The fading behavior of the dosimetric glow peak at different particle sizes are given in Fig.5. As can be seen from Fig.5, the glow peak intensity decays at all particle sizes almost in the
same trend over the storage period, moreover, these decays are nearly same extent at all particle sizes. In other words, it was not observed any systematic change in the dosimetric glow peak intensity during the storage period depending on the particle size of samples. Decreases of 43%, 42%, 30% and 30% in the peak intensities of 63-90 µm, 90-125 µm, 125-180 µm and 180-250µm, respectively, were calculated at the end of storage period of 30 days. Based on these decreases in the TL peak intensity, corrections must be performed on the dose values calculated from dose response curves (Fig.3) to get the actual doses received by the samples.

![Normalized TL peak intensity over time](image)

Fig.5. Room temperature variations of the glow peak (at 240°C) intensity with storage time for different particle sizes of household salt samples irradiated with 5 Gy. The signals were integrated over the temperatures at between 180°C and 500°C. TL intensities were normalized over the measurements received immediately after irradiation.

3.3. Kinetic Analysis

The determination of kinetic parameters from the thermoluminescence glow curves is an active area of interest and various techniques have been developed for the determination of kinetic parameters namely activation energy (E), order of kinetics (b) and frequency factor (s).
Analysis of TL glow curves has become more important in view of its applications in dosimetry studies. In this work, three methods were used for the determination of trapping parameters namely initial rise (IR), peak shape (PS) and variable heating rate (VHR) methods. For simplicity, these three methods will be referred to as the IR, PS and VHR methods in the rest of this paper. The detail procedures for these methods given elsewhere (Chen, 1969; McKeever, 1985; Chen and McKeever, 1997; Pagonis et al., 2006). In this work, the trapping parameters of the 240°C glow peak of household salt samples were calculated by using the particle sizes of 180-250 µm, 125-180 µm, 90-125 µm and 63-90 µm.

3.3.1. Initial Rise (IR) Method

Fig. 6 demonstrates radiation-induced TL glow peaks in household salt after a test beta exposure (5 Gy) followed by annealing at 160°C. This pre-heat procedure has minimal effect on the 240°C (5°Cs⁻¹) dosimetric peak of salt, or on the sample sensitivity (Spooner et al., 2011). As shown in Fig.6, the preheating isolates the 240°C glow peak from the neighboring peak at 110°C and then TL was recorded in the initial rise region at a heating rate of 5°Cs⁻¹.

![Fig. 6. Radiation-induced TL glow peaks of household salt samples after 5 Gy beta irradiation followed by pre-heating at 160°C. The particle sizes were 125-180µm.](image-url)
Fig. 7 represents the Arrhenius plots of salt samples with different particle sizes, the graph of \( \ln (I) \) versus \( 1/T \) for the initial rise part of 240°C dosimetric glow peak. A plot of \( \ln (I) \) vs \( 1/T \) would yield a straight line with a slope of \(-E/k\) and a \( y \)-intercept of \( \ln (s/\beta) \), from which \( E \) and \( s \) can be readily calculated. The calculated activation energy and frequency factor values from the slopes and intercepts by the IR method are given in Table 1. It was not observed any significant change in activation energy and frequency factor values depending on particle size of salt samples. It is observed that the values of activation energy and the frequency factors obtained for different particle sizes are agreement with the values obtained by peak shape (PS) method in common iodised salt (Murthy et al., 2006). Nevertheless, the calculated kinetic parameters by IR method are lower than the values obtained by VHR and deconvolution on natural and synthetic sodium chloride (NaCl) (Spooner et al., 2011; Rodriguez-Lazcano et al., 2012).

![Graph showing Arrhenius plots for different particle sizes](image)

Fig 7. Initial rise plots of household salt samples for different particle sizes. All samples were irradiated with 5 Gy.
Table 1. Values of the kinetic parameters of dosimetric peak obtained VHR, IR and PS methods for all particle sizes of house hold salt.

<table>
<thead>
<tr>
<th>Kinetic Method</th>
<th>63-90 µm</th>
<th>90-125 µm</th>
<th>125-180 µm</th>
<th>180-250 µm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$E(\pm \sigma)$ eV</td>
<td>$s (s^{-1})$</td>
<td>$E(\pm \sigma)$ eV</td>
<td>$s (s^{-1})$</td>
</tr>
<tr>
<td><strong>VHR</strong></td>
<td>0.80 ± 0.04</td>
<td>1.00 x 10^8</td>
<td>0.80 ± 0.06</td>
<td>1.00 x 10^8</td>
</tr>
<tr>
<td><strong>IR</strong></td>
<td>1.08 ± 0.01</td>
<td>1.14 x 10^10</td>
<td>1.08 ± 0.02</td>
<td>1.60 x 10^10</td>
</tr>
<tr>
<td><strong>PS</strong></td>
<td>$E_\tau = 1.10 \pm 0.07$</td>
<td>$s_\tau = 2.0 \times 10^{10}$</td>
<td>$E_\tau = 1.19 \pm 0.11$</td>
<td>$s_\tau = 1.8 \times 10^{11}$</td>
</tr>
<tr>
<td></td>
<td>$E_\delta = 1.08 \pm 0.07$</td>
<td>$s_\delta = 1.3 \times 10^{10}$</td>
<td>$E_\delta = 1.20 \pm 0.11$</td>
<td>$s_\delta = 2.3 \times 10^{11}$</td>
</tr>
<tr>
<td></td>
<td>$E_\omega = 1.10 \pm 0.07$</td>
<td>$s_\omega = 2.0 \times 10^{10}$</td>
<td>$E_\omega = 1.20 \pm 0.11$</td>
<td>$s_\omega = 2.3 \times 10^{11}$</td>
</tr>
<tr>
<td>$^*E_{ave} (\pm \sigma)$ eV</td>
<td>$^*s_{ave} (s^{-1})$</td>
<td>$^*E_{ave} (\pm \sigma)$ eV</td>
<td>$^*s_{ave} (s^{-1})$</td>
<td>$^*E_{ave} (\pm \sigma)$ eV</td>
</tr>
<tr>
<td>1.09 ± 0.07</td>
<td>1.76 x 10^10</td>
<td>1.20 ± 0.11</td>
<td>2.13 x 10^10</td>
<td>1.35 ± 0.15</td>
</tr>
</tbody>
</table>

* $E_{ave}$ and $s_{ave}$ are the average values obtained from PS method.
3.3.2. *Peak Shape (PS) Method*

The recorded dosimetric glow curve after preheating at 160°C was also used to determine the kinetic parameters by the PS method (Chen, 1969). From the experimental glow curve the temperature $T_m$ at peak maximum and temperatures at half intensity $T_1$ and $T_2$ are measured.

To determine the kinetic parameters of the dosimetric glow peak at 240°C the following shape parameters are to be determined: the half-width at low temperature side $\tau = T_m - T_1$, the half-width at the fall-off side $\delta = T_2 - T_m$, the full-width at half maximum (FWHM) $\omega = T_2 - T_1$ and the symmetry (geometrical) factor $\mu_g = \delta / \omega$. In the PS method, the kinetic order of TL peak is easily obtained by means of the symmetry factor ($\mu_g$) and it changes with the order of kinetics from $\sim 0.42$ to $\sim 0.52$, where these two limits correspond to first-and second order kinetics, respectively (McKeever, 1985). Another parameter was proposed by Balarin (Balarin, 1975) gives the kinetic order as a function of the parameter $\gamma = \delta / \tau$. This parameter ranges from 0.7 to 0.8 for the first order kinetics and from 1.05 to 1.20 for the second order kinetics.

Firstly, glow curves at different particle sizes are recorded for the beta irradiated (5 Gy) salt samples. The half width parameters $\delta$, $\tau$ and $\omega$ at each particle size were determined from the temperatures $T_1$, $T_2$ and $T_m$. The symmetric factors ($\mu_g$) and Balarin parameters ($\gamma$) calculated for peak at 240°C are listed in Table 2. The values of $\mu_g$ and $\gamma$ for the particle size of 63-90µm were 0.54 and 1.16, respectively and these parameters predicate a second order kinetic property.

Table 2. Symmetry factor ($\mu_g$) and balarin ($\gamma$) parameters of dosimetric TL peak at 240 for different particle sizes.

<table>
<thead>
<tr>
<th></th>
<th>63-90 µm</th>
<th>90-125 µm</th>
<th>125-180 µm</th>
<th>180-250 µm</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Balarin parameter, ( \gamma )</strong></td>
<td>1.16</td>
<td>0.86</td>
<td>0.83</td>
<td>0.85</td>
</tr>
<tr>
<td><strong>Symmetry factor, ( \mu_g )</strong></td>
<td>0.54</td>
<td>0.46</td>
<td>0.45</td>
<td>0.46</td>
</tr>
</tbody>
</table>
For the particle sizes of 90-125 µm, 125-180 µm and 180-250µm, the values of $\mu_g$ and $\gamma$ predicate a first order kinetics. The calculated values of $\mu_g$ and $\gamma$ indicate the domination of the first order process for the 240°C peak of salt samples.

On the other hand, for a constant heating rate, in first order kinetics both peak temperature and peak shape are independent of the initial irradiation dose but in the second order kinetics, the glow peak temperature and peak shape are strongly dependent on initial irradiation dose (Chen, 1969; McKeever, 1985). As can be seen in Figs. 8a and b, the dosimetric peak temperature at all particle sizes did not shift to lower temperatures with increasing beta dose.

![Fig. 8](image-url)

Fig. 8. Change in maximum peak temperature ($T_{\text{max}}$) with increasing dose for particle sizes of a) 63-90 µm b) 180-250 µm.
This means that the peak obeys nearly first order kinetics which contradicts the second order kinetics predicted from the peak shape parameters $\mu_g$ and $\gamma$ for the particle size of 63-90µm. The reason for the contradicting result is essentially due to the complex nature of the dosimetric glow peak. Dosimetric TL peak of salt is not single well isolated peak, but has a small shoulder at lowering side of the glow curve. This shoulder is more pronounced in low particle sizes of samples (Fig.2). We also note that Polymeris et al. (2011), Rodriguez-Lazcano et al. (2012) and Druzhyna et al. (2016) deconvolve the TL of salt into a number of TL peaks with several of these contributing to the observed dosimetric peak. The presence some extra peak components in the dosimetric glow curve region, depending on their position and intensity, may lead to change in half width parameters $\delta$, $\omega$, and $\tau$. So, in the case of complex glow curves the variation of $T_m$ with dose can be used as good indicator to determine the order of kinetics.

The activation energy ($E$) can be calculated by using a general formula for $E$ is given by:

$$E = c_\alpha (kT_m^2/\alpha) - b_\alpha (2kT_m)$$

where $\alpha$ is $\omega$, $\tau$ or $\delta$. The constants $c_\alpha$ and $b_\alpha$ for the first and second order kinetics are given in Table 3.1 of McKeever (1985). Activation energies $E_\tau$, $E_\delta$ and $E_\omega$ of peak at 240°C in salt samples are calculated using equation (1) for different particle sizes. For the calculation, first order kinetic equations ($\mu_g=0.42$) are also applied for the particle size of 63-90 µm. The calculated activation energies ($E_\tau$, $E_\delta$, $E_\omega$) and the mean values of $E_\tau$, $E_\delta$ and $E_\omega$ are shown in Table 1. Activation energies obtained by PS method increased with increasing particle sizes of salt samples.

After obtaining the order of kinetic and activation energy, the frequency factor ($s$) of peak at 240°C can be determined by using the general equation given by Chen (1969):

$$s = \beta E /kT_m^3 \exp (E/kT_m)[1+(b-1)2kT_m/E]^{1/2}$$

(2)
where \( b \) is order of kinetics, \( \beta \) is the linear heating rate \((5^\circ\text{Cs}^{-1})\) and \( k \) is Boltzmann’s constant. Frequency factors \( s_t \), \( s_b \) and \( s_o \) and their mean values for the different particle sizes are tabulated in Table 1. It is seen that as the particle size increases the frequency factor becomes greater. As seen from Table 1, the activation energies and frequency factors obtained for the particle sizes of 125-180µm and 180-250µm are more agreement with the values reported previously (Spooner et al., 2011; Rodriguez-Lazcano et al., 2012). For the low particle sizes, inconsistency with the kinetic parameter values in the literature is probably due to the complexity of the glow peak which is more pronounced at low particle sizes. Moreover, \( E \) and \( s \) found by the PS method are also higher than the values obtained by the IR method in all particle sizes of samples.

3.3.3. Variable Heating Rate (VHR) Method

The 240°C dosimetric glow peak was also examined using variable heating-rate method. In this part of the work, it was not performed any preheating procedure on 5 Gy beta irradiated salt samples. The glow peak maximum temperatures, \( T_m \), corresponding to different heating rates \((1^\circ\text{Cs}^{-1}, 3^\circ\text{Cs}^{-1}, 5^\circ\text{Cs}^{-1}, 7^\circ\text{Cs}^{-1}, 9^\circ\text{Cs}^{-1})\) were recorded by using the different particle sizes of salt samples. This method of analysis consist of a plot of \( \ln \left( \frac{T_m^2}{\beta} \right) \) versus \( 1/T_m \), which is a straight line with slope equal to \( E/k \) and intercept equal to \( \ln \left( E/s k \right) \) (\( \beta \)-heating rate, \( k \)=Boltzmann constant). From the intercept the frequency factor \((s)\) is calculated. Temperature lag may appear in thick samples at especially higher heating rates. To reduce this effect between the heater strip and the sample, small amount (~5 mg) of uniform grain size powder samples were used for the TL measurements. Figs.9 a, b and c show the TL glow curves of salt samples after different heating rates for the particle sizes of 63-90 µm, 90-125 µm and 180-250 µm. As seen from these figures, the overall appearances of glow curves did not change at various heating rates in the same particle size group of samples. It was observed
that main low (at 110°C) and high (at 240°C) temperature glow peak positions, $T_m$, shifted to higher temperatures with an increasing heating rate.

Fig. 9. TL glow curves of salt samples after different heating rates for the particle sizes of a) 63-90 µm b) 90-125 µm c) 180-250 µm.
This shift is in agreement to the theoretical expectation. As can be seen in Figs.9a and b, especially at low heating rates and particle sizes a little shoulder on the high-temperature side of the dosimetric peak can be observed. This small shoulder decays at high heating rates. In other words, dosimetric TL peak becomes simpler with increasing heating rates. Similar shoulders at the high temperature side of TL peak for low heating rates was also observed by Mandowski and Bos (2011) in YPO₄:Ce³⁺, Sm³⁺ phosphors. These extra small shoulders are believed to be the results of a two-stage process of Mandowski (2005), which is a natural generalization of the standard localized transition model. The results of the semi-localized transition (SLT) model show an extra peak for low heating rates, most pronounced seen as shoulder at the high temperature side of main TL peak (Mandowski and Bos, 2011).

Figures 10a and b show both the normalized integrated TL and the maximum TL intensity of the main low temperature TL peak at 110°C. As shown in these figures, the integrated and the maximum TL intensities decrease with increasing linear heating rate for all particle sizes. FWHM of this peak did not significantly change with increasing linear heating rate values for the different particle sizes (Fig.10c). The above mentioned reduction in the intensity of TL peak at 110°C can be attributed to a thermal quenching effect (Montalvo et al., 2004). Contrary to the decrease in the intensity of this main low temperature glow peak, it is interesting to see that as the heating rate increases the dosimetric glow peak at 240°C becomes much higher (Figs.11 a,b and c). For the different particle sizes, the normalized integral area and height of dosimetric peak versus heating rates are plotted in Figs.11a and b. As a general trend, the dosimetric peak intensity increases as a function of increasing heating rate for all particle sizes.

If the TL peak integral or peak height diminishes as a function of heating rate, it was accepted that the thermal quenching is present. However, if TL peak integral is stable then thermal quenching is absent (Chen and McKeever, 1997).
Fig. 10  a) Normalized peak height the main low temperature TL peak at 110°C at different particle sizes as a function of linear heating rates. b) The behavior of normalized peak area at 110 °C at different particle sizes as a function of linear heating rates. c) Full width at half maximum (FWHM) of the main low temperature TL peak at 110°C at different particle sizes as a function of linear heating rates.
So, the increase observed in dosimetric peak intensity does not give any evidence for thermal quenching. The increase in TL peak intensity with increasing heating rate was also observed
in YPO$_4$:Ce$^{3+}$, Sm$^{3+}$ (Bos et al., 2010). The Mandowski model of semi-localized transitions (SLT) was taken as a possible explanation for such observations (Mandowski, 2005; Mandowski and Bos, 2011). Based on this model, as the heating rate is increased, the relative ratio of radiative and non-radiative transitions increases, due to the two-stage process of excitation of charge carriers from traps. In other words, at low heating rates, the probability of non-radiative transitions is higher and as the heating rate increases (when the peak is reached at higher temperatures), the probability of transitions from the localized excited state into the conduction band increases. As the non-radiative transition probability is assumed to be temperature independent, the radiative recombination probability increases as the temperature increases, resulting in a higher TL peak at high heating rates (Delice et al., 2015). Details of SLT model can be found in the literature (Madowski, 2005; Mandowski and Bos, 2011; Pagonis et al., 2013). On the other hand, Spooner et al. (2011) reported that the lack of thermal quenching in common salt samples with heating rates ranging from 0.002 Ks$^{-1}$ to 5 Ks$^{-1}$. Polymeris et al. (2011) also did not observe any thermal quenching in salt samples with heating rates of 1-10 Ks$^{-1}$. Moreover, the behavior of the FWHM for the dosimetric peak as a function of heating rate is shown in Fig 11c. It was observed that as the heating rate increases FWHM values of dosimetric peak did not significantly change for all particle sizes we used. The plots of $Ln \left( \frac{T_m^2}{\beta} \right)$ versus $(1/T_m)$ are shown in Fig.12. Activation energies and frequency factors of dosimetric peak at 240°C were calculated from the slope and intercept of the straight lines. The values of E and s obtained by VHR method are presented in Table 1. It was observed that both activation energy and frequency factor values obtained by VHR and PS methods increase with increasing particle size except ones obtained by IR method. It is seen that the kinetic parameters obtained by PS and VHR methods for the largest particle sizes (180-250µm) are closer to the values obtained in the literature (Spooner et.al. 2011; Rodriguez-Lazcano et al., 2012). For the smaller particle sizes, the kinetic parameters
obtained by PS and VHR methods are much lower than the values obtained in the literature (Spooner et al., 2011; Rodriguez-Lazcano et al., 2012). This reduction in E and s is essentially due to the complex nature of the dosimetric glow curve especially at low particle sizes.

Fig. 12. Plots of ln (Tm²/β) vs. 1/Tm for household salt samples irradiated with 5 Gy. All plots were performed for the different particle sizes of samples.

Based on these results, to obtain more reliable results, it should be avoided from using of particle sizes smaller than 125 µm. Besides, the IR method gave lower values of activation energy and frequency factor than the values obtained by the PS and VHR methods in the case of the largest particle sizes (180-250µm) (Table 1). Due to the thermal quenching effect, (E-W) is obtained instead of E in the IR method (McKeever, 1985).

4. Conclusions

From the data presented in this paper, conclusions can be summarized as follows:

1. There is no systematic relationship between particle size and mechanically induced TL peak strength (or glow curve shape) in zero dose salt samples.
2. Significant mechanically induced TL peak in the region of the dosimetric peak at 240°C may lead to a non-reliable dose assessment. However, no significant mechanically induced TL peak was observed in the vicinity of the dosimetric TL peak. In the dose range studied, intensities of these signals were very low.

3. The number of TL peaks in the glow curves increased with decreasing heating rate and particle size (<125 µm).

4. For a fixed dose, the TL intensities of the radiation-induced main peaks at 110 and 240°C increase with increasing particle size.

5. Beta irradiation caused a significant increase in dosimetric TL peak intensity of salt samples. For all particle sizes, variation of TL peak intensity with absorbed radiation dose can be described well by a linear function (in the log-log scale) in the dose range of 0.4-55 Gy.

6. The beta radiation sensitivity of the radiation-induced TL peak at 240°C increased with increasing particle size. The radiation sensitivity of this peak was found to be nearly equal for the particle sizes of 125-180 and 180-250 µm. So, we can suggest that the particles sizes of 125-180 µm and 180-250 µm can be used as dosimetric salt material. In other words, working with small particle (<125µm) sizes should be avoided for more radiation sensitivity in salt samples.

7. Fading evaluations of dosimetric TL peak of different particle sizes 1-month after irradiation showed that the intensity values decreased by 30-43%. For household salt samples the rate of post-irradiation fade is nearly independent of particle size but dependent upon storage time. Storage tests at different particle sizes conducted over a 1-month period confirmed sufficient stability of the 240°C TL peak for use in retrospective dosimetry.
8. The kinetic parameters obtained by PS and VHR methods increased with increasing particle size, whereas the kinetic parameters obtained by IR method did not change. It may be argued that it is necessary to work with large particles sizes (180-250µm) to obtain more reliable kinetic parameters. The heating rate dependences of the revealed TL peaks of household salt were also studied. An interesting heating rate behavior of the high temperature dosimetric TL peak at 240°C was observed, while the expected behavior was observed for the low temperature TL peak at 110°C. The TL intensity of high temperature dosimetric peak increases with increasing heating rate between 1 and 9°Cs⁻¹. This behavior can be explained by Mandowski model of semi-localized transitions (SLT). This model assumes competing radiative and non-radiative transitions.
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