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Development of NaCl Salt as a Reusable Gamma Radiation Dosimeter

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Abstract: *NaCl salt is a well-known material that can be used just one time for retrospective dosimetryin radiation accidents. After first time of use, the sensitivity of NaCl drops dramatically and its TL glow curve changes significantly. In the presentresearch, 5 batches of NaCl salt are prepared in forms of chips (each 20 ± 2.5 % g). All batcheswere subjected to heating rate 10°C/min and kept for 1 hour at different annealing temperatures namely, 200, 300, 400, 500 and 600 °C respectively; after annealing, the batcheswere quenched at room temperature. The batches have been sensitized with a dose of 5 Gy using Cs^{137} gamma radiation source and their TL responses were measured. It has been found that TL response was enhanced as the annealing temperature increases showing maximum response at 500 °C and then drops afterwards; enhancement in TL response of 500 °C annealed batch is more than 500% compared to the un-annealed batch. Further investigation showed that both the glow curve and the TL response have no change as the NaCl chips re-irradiated with gamma radiation showing a linear dose response up to 10 Gy. Finally, kinetic parameters have been deduced using Computerized Glow Curve Deconvolution (CGCD). CGCD analysis showed that the glow curve of 500 °C annealed group consists of 5 1st order glow peaks. The results obtained in this study indicate that annealed NaCl salt at 500 °C could be used as a reusable and efficient gamma radiation dosimeter.*

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I. INTRODUCTION

Thermoluminescence characteristics of alkali halides have been studied by many research groups [1,2,3] and sodium chloride in specific has been claimed as suitable candidate for TL retrospective dosimetry. Later, Heywood and Clarke [4,5] used pure NaCl and NaCl:Ca in order to understand the TL properties after exposure to gamma rays. More recently, Some TL studies on alkali halides including those of Purohit and Joshi [6], Davidson et al. [7,8,9], Ortiz et al. [10] have discussed the TL glow curves and the effect of impurities on TL of NaCl crystals.

In case of radiation accidents, NaCl was proposed from different authors [11-15] for rapid retrospective estimation of the absorbed dose to individuals such as general public and first responders who normally do not carry any kind of dose measuring tool; this is because it is an affordable material that can be easily found very close to the potentially exposed people.

Raw household NaCl without any type of thermal treatment exhibits high sensitivity to gamma radiation. There is a Sensible stability of the TL glow curve while in the storage of the salt as well as, linear response of the TL emission with the dose along wide range of dose that can reach up to 20 Gy [16].

However, it has been noticed that in many of previous researches, the glow curve of the NaCl exhibits two prominent peaks, when it was re-read or subjected to previous thermal treatment rather than having one prominent peak associated with first time readout as has been shown [11,17,18].Also, the overall integral TL intensity of the 2nd readout drops dramatically in comparison to the 1st readout. Therefore household NaCl powder can be used as a material for 'one time' TL dosimeters [19]and doesn't show the characteristics of TL dosimeter. The change in the number of the glow peaks, their positions and amplitudes are mainly attributed to the meta-stable traps between valence band and conduction band that were easily dimensioning with thermal treatment and disappeared during 1st readout. This high sensitivity of NaCl against temperature is investigated in more details. The trap concentration and the glow curve configuration in the household NaCl were subjected to different scenarios of thermal treatment.

In this paper, we studied the TL response of NaCl (Portugal Gourmet, Flor do Algarve, Natural see salt, made in Germany), when the salt is kept for 1 h at different annealing temperatures namely 200, 300, 400, 500 and 600 °C taking in consideration that the salt will heated up to these temperatures with slow heating rate 10 °C/min to allow for smooth reconfigurations of the traps in the NaCl crystals. The overall integral TL intensity and the glow curve are recorded for each aforementioned annealing temperature. All data have been investigated thoroughly and a conclusion will be drawn.

II. SAMPLE PREPARATION.

10 g of non-iodized household NaCl has been ground gently in agate mortar and then kept in the oven for 24 h at 50 ^oC to remove the moisture. The dried salt has been prepared in the form of chips of 5 mm diameter and each chip weights 20± 0.5 mg. The 20 mg can be taken as suitable sample weight for TL studies in the lower doses, [20]. 36 chips have been prepared and divided into 6 groups then kept at room temperature in desiccator contains silica gel to protect them from moisture. 5 batches (each 6 chips) have been annealed at 200, 300, 400, 500 and 600 ^oC and kept individually at each temperature for 1h. The chips are heated up to these temperatures with slow heating rate 10 ^oC/min to allow for smooth rearrangements of the traps in the NaCl crystals. The samples have been quenched at room temperature and returned to the desiccator till irradiation process. The 6thbatches have not been subjected to any thermal treatment.

III. INSTRUMENTATION

X-ray diffraction pattern has been recorded for the dried NaCl salt using Shimadzu XRD-6000 diffractometerwith Cu Kα radiation(λ= 1.54056 Å). Irradiation of the salt samples were done using Cs-137 Gamma source. The dose rate and activity of Gamma source are 6.69×10^{-3} Gy/sec. All the samples were read immediately after the exposure to radiation. The TL-reader used in the current study is Harshaw TL Reader model 3500 THERMO FISHER. Samples measurements were registered by the TL reader at a heating rate of 5^{o}C/s and up to a temperature of 350^{o}C in the same manner we followed previously in [15]. Using low heating rate is highly preferable to allow for uniform heating of the whole sample and hence avoid temperature lack that could disturb the glow curve; this in return allows for different glow peaks in the glow curve to be enough distinguished and the kinetic parameters could be easily retrieved from the glow curve [15].

IV. RESULTS AND DISCUSSION

4.1. XRD results

Figure (1) represents the XRD pattern recorded for the household NaCl salt in the 2θ range 25^o-90^o using Bragg Brentano geometry. Several diffracted peaks can be seen with ‹*hkl*› values indicating a complete crystalline structure in a cubic phasewith space group Fm3m. The resultant peaks correspond to values {111}, {200}, {220}, {222}, {400}, {420} and {422} and their relevant positionsat 27.4^o, 31.8^o, 45.5^o, 56.5^o, 66.3^o, 75.3^o, and 84.0^{o}respectively. No abnormal peaks have been noticed in the diffraction pattern which confirms the purity of the household NaCl understudy.

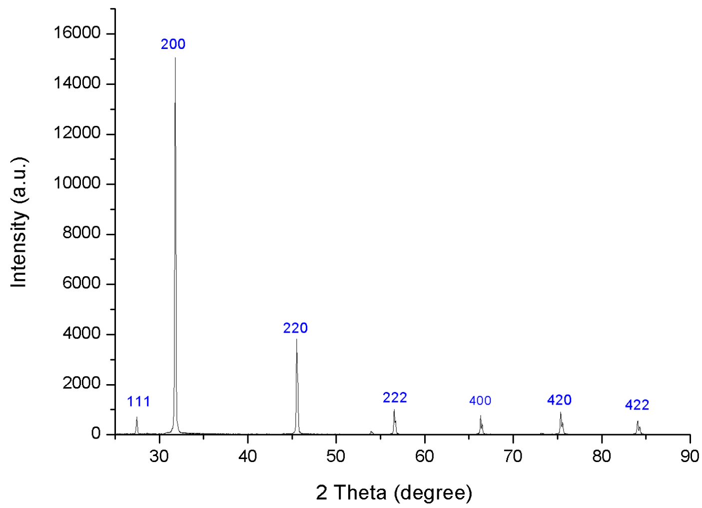
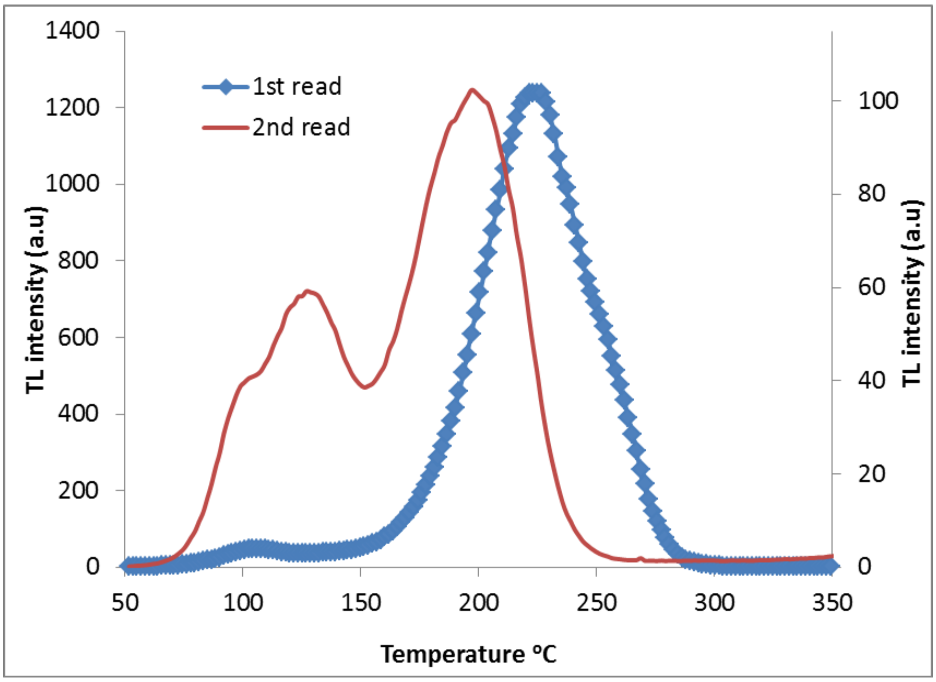


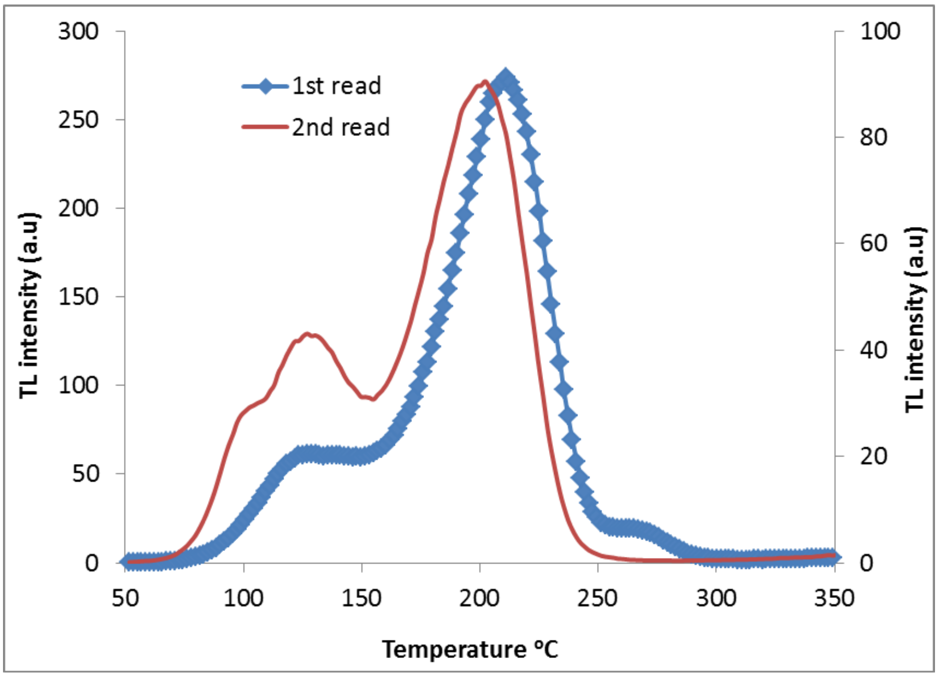
Figure (1): XRD pattern of NaCl salt.

4.2. TL results

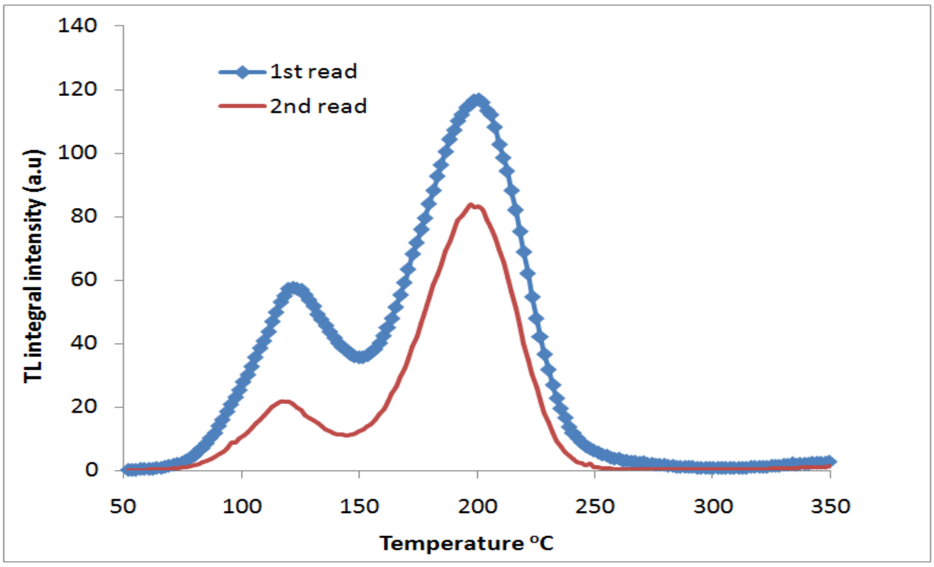
Figure (2) shows the average of the 1st and 2nd readout of all groups. As shown in figure (2-a), the glow curve of the 1st readout of the raw NaCl has one prominent peak while the glow curve of the 2nd readout has two prominent peaks and the maximum intensity is 12 times less compared with the maximum intensity of the un-annealed sample; the overall behavior is in agreement with previous studies [11,17,18]. The positions of resultant two peaks in 2nd readout do not match the position of the original peak of 1st readout. This implies that the original traps responsible for the glow peak in the 1st readout have been disappeared totally and replaced with new types of traps at lower temperatures in case of 2nd readout. At 200 ^oC annealing temperature as shown in figure (2-b), it has been noticed that a new earlier peak in the 1st readout start to appear while the 2nd readout still has the same behavior of the 2nd readout of un-annealed samples. The positions of peaks in 1st and 2nd readout in this case are very close but do not match; the TL response of the 2nd readout still much less that 1st readout, almost 30 times less. At 300 °C annealing temperature as shown in figure (2-c), the glow curve of both 1st and 2nd readout have almost the same behavior and the peak positions in both cases are completely matched; their TL responses get much closer to each other.



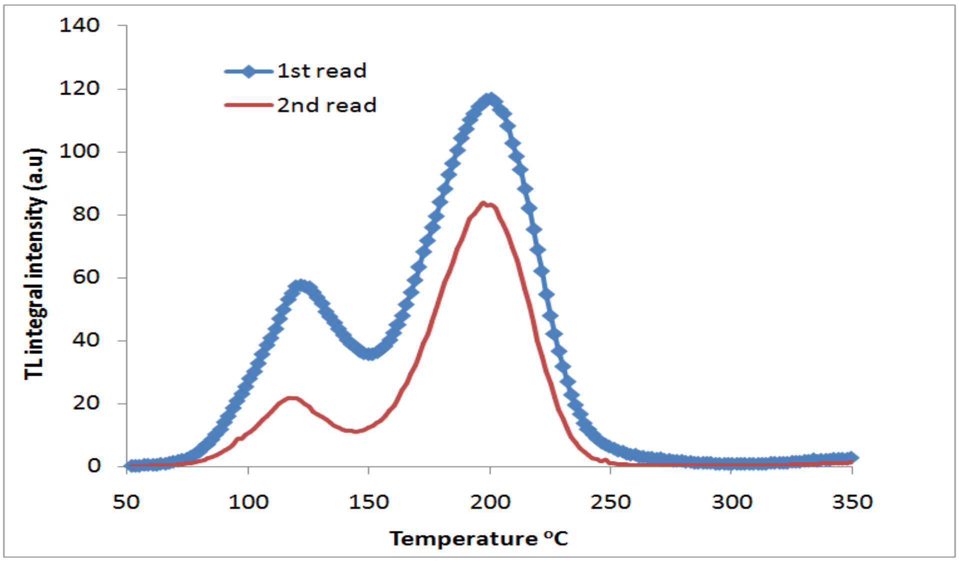
(a)



(b)


140
120
100
80
60
40
20
0
TL integral intensity (a.u)
1st read
2nd read
50
100
150
200
250
300
350
Temperature °C

(c)


140
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TL integral intensity (a.u)
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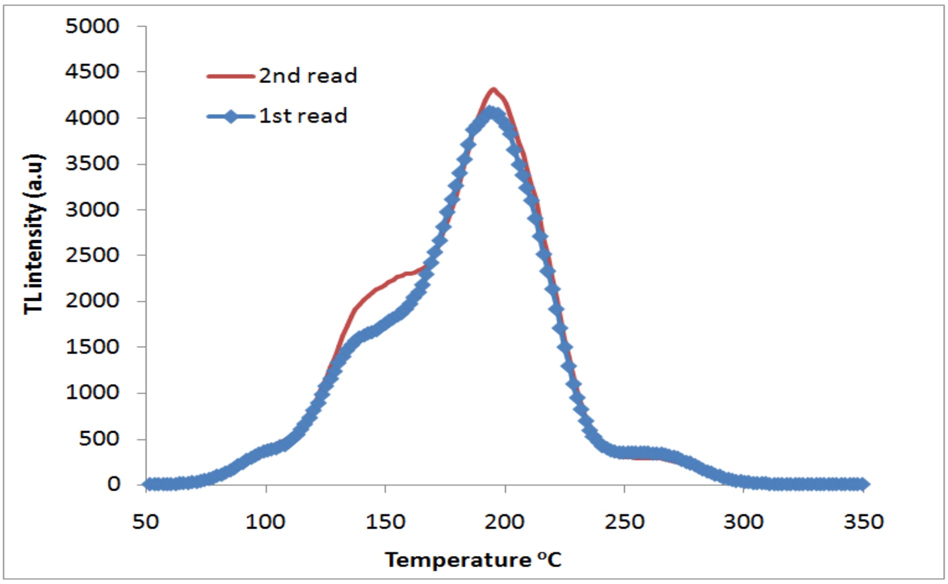
Temperature °C

(d)


5000
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2500
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TL intensity (a.u)
2nd read
1st read
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200
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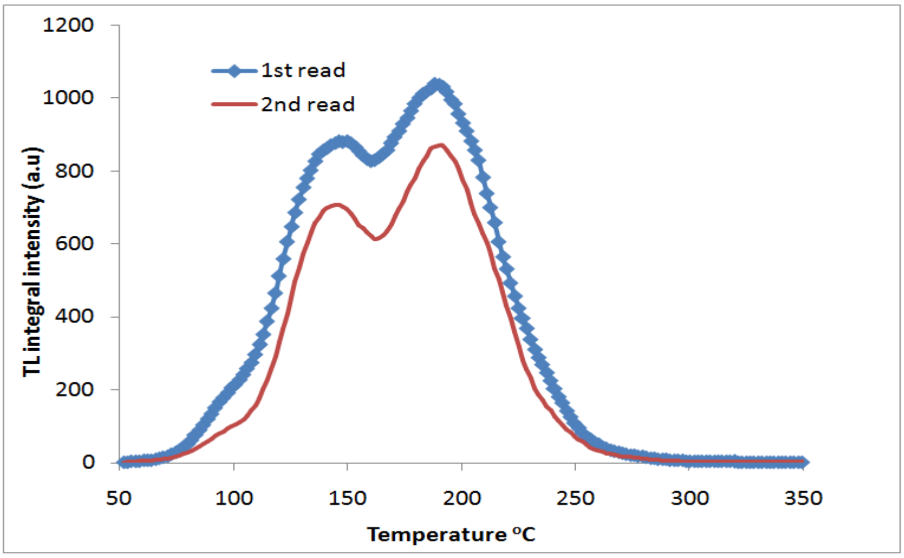
300
350
Temperature °C

(e)



(f)

Figure (2): TL intensity of 1st and 2nd readout of irradiated NaCl salt with 5 Gy as a function of annealing temperature; (a) zero annealing temperature, (b) 200 °C, (c) 300 °C, (d) 400 °C, (e) 500 °C and finally (f) 600 °C.

However the TL response of 1st readout at 300 °C is less than the TL response of the 1st readout at 200 °C. Figure (2-d) represents the 1st and 2nd readout of the samples annealed at 400 °C. As clearly shown, the TL response jumps dramatically for both 1st and 2nd readout and they have almost the same glow curve behavior. The TL response in this case approaches the TL response of the un-annealed samples with the advantages of preserving its sensitivity during the 2nd readout. In figure (2-e), the TL responses of 1st and 2nd readout have a complete match in peak positions and behavior and the TL intensity is almost the same. The overall TL response *increases 500% compared to the 1st readout of un-annealed*. Finally, in figure 2-f at 600 °C annealing temperature, the TL response of the 1st and 2nd readout drops dramatically and approaches the TL response of the 1st readout of un-annealed samples with the advantages of preserving both its sensitivity during the 2nd readout and the glow curve behavior. Also, the peak positions remained unchanged. Figure (3) represents the effect of the 2nd peak position as function of the annealing temperature for the 1st and 2nd readout. It is clearly noticed that the peak position is quite different in case of un-annealed sample and 200 °C annealing temperature. This implies that the traps responsible for that peak in 1st and 2nd readout are totally different in their thoughtful. At higher temperatures starting for 300 °C annealing temperature, the position of the 1st and 2nd peaks are matched for 1st and 2nd readout which in turns means that the traps in the samples responsible for the glow curve remains unchanged with consecutive readouts. What are only changes from annealing temperature to another are the trap concentrations that reaches its maximum at 500 °C. On the other hand, figure (4) represents the TL integral intensity of 1st and 2nd readouts of irradiated NaCl salt as a function of annealing temperature. As noticed, the maximum enhancement in TL response is obtained at 500 °C. Finally, the dose response of the samples annealed at 500 °C has been investigated from 0.25 Gy and up to 10 Gy.

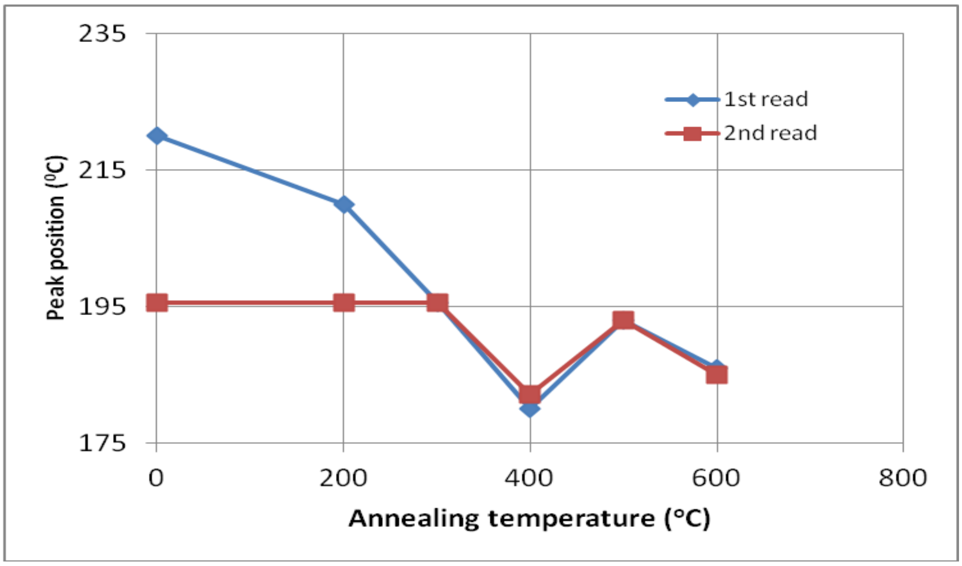


Figure (3): Dosimetric peak position as a function of annealing temperature.

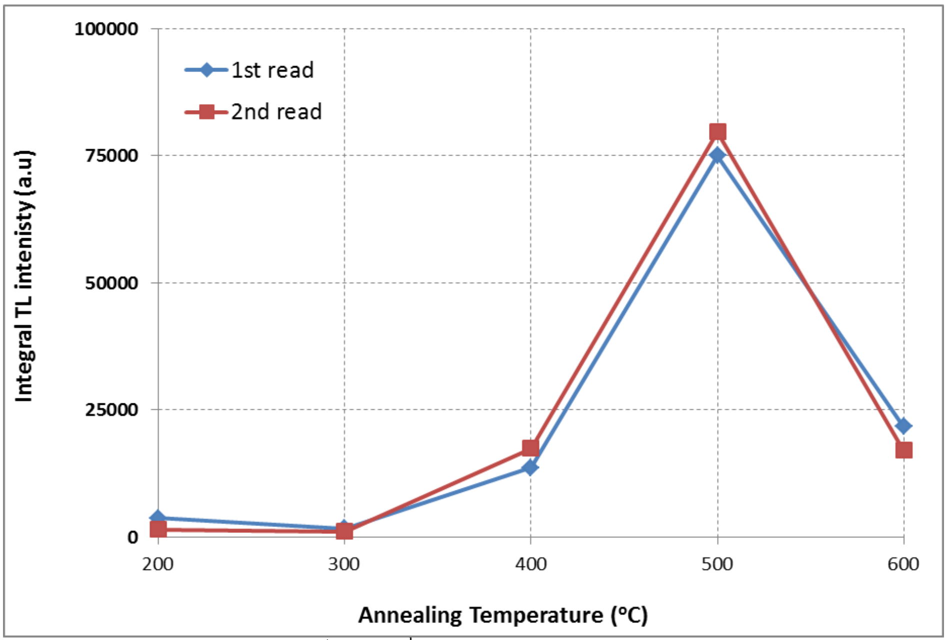


Figure (4): TL integral intensity of 1st and 2nd readout of irradiated NaCl salt as a function of annealing temperature. Maximum enhancement in TL response is obtained at 500 °C annealing temperature.

Figure (5) represents the TL response as a function of gamma dose given to the NaCl annealed at 500 °C. The graph is plotted in log-log scale and fitted to the formula $I_{max}=a\ D^k$ where D is the applied dose, a is proportionality factor and k defines the type of non-linearity; sublinear ($k < 1$) or supralinear ($k > 1$) and obviously ($k = 1$) means a linear dependence, [21,22]. In our case the fit process yields $I_{max}= 12014\ D^{1.213}$ which means that the NaCl salt annealed at 500 °C a supralinear response against gamma dose irradiation.

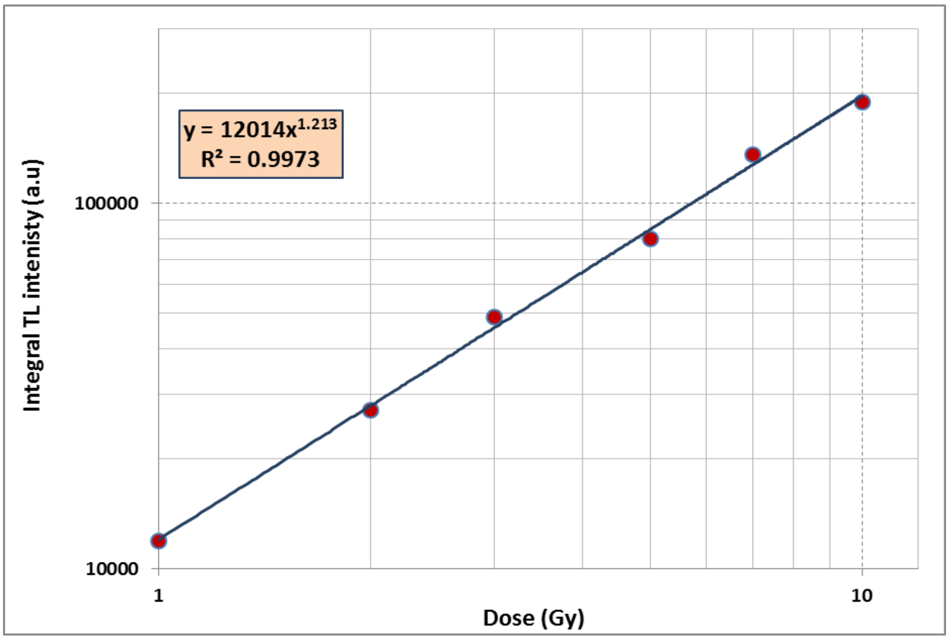


Figure (5): Dose response of NaCl salt annealed at 500 °C.

4.3. Glow Curve Deconvolution

It is very advantageous to investigate the inherent mechanism of the TL phenomenon in any TL material to understand how the TL process takes place inside the material; or in other words how the electrons excited by heat to the conduction band will recombine with the holes above the valence band. Many kinetic models have been proposed in literature by many authors for that purpose. One of such model is taking in consideration the probability of direct recombination is dominating the probability of indirect recombination (re-trapping before recombination), the case is called "first-order" kinetics and such mechanism is proposed by

Randall and Wilkins [23].Kinetic parameters of the 500 ^oC annealed samples was done by means of Glow Curve Deconvolution on the glow curve using the first order kinetics analytical function given by equation (1)[24].

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

where *I(T)* is the intensity at temperature *T* in Kelvin, I_m is the maximum peak intensity, *E* is the activation energy in eV, *K* is the Boltzmann constant in eVK^{-1}, T_m is the maximum temperature at the peak maximum in *K*, □ is $2KT(E)^{-1}$, and □$_m$ is $2KT_m(E)^{-1}$.

The experimental glow curve has been fitted using equations (1). The resultant glow curve deconvolution is shown in figure 6 and the glow curve was de-convoluted into 5 glow peaks each peak is represented by equation (1). Table (1) summarizes the resultant fitting parameters for all peaks.

Table 1: Fitting parameters of 5 glow peaks resulted from glow curve deconvolution using first order kinetic model (equation 1) of 500 ^oC annealed samples irradiated at 5 Gy.

Peak No.	Maximum Intensity I_m(a.u)	Activation Energy *E* (eV)	Temperature Position T_m (K)
1	125	1.602	368.5
2	1103	0.8164	411.7
3	2783	0.9206	459.7
4	2058	1.198	483.2
5	364.1	0.8601	527

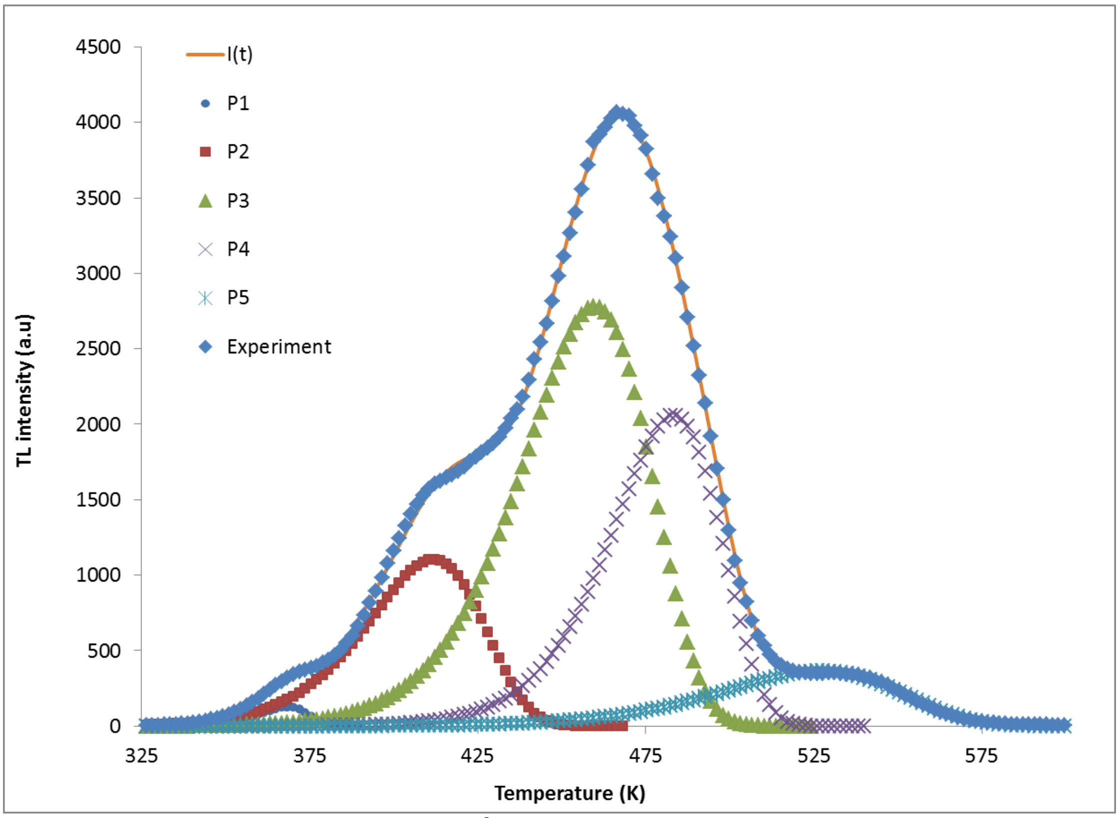


Figure (6):. Glow curve deconvolution of 500 ^oC annealed sample. Dotted, experimental data, red line is the sum of 5 deconvoluted individual peaks of 1st order represented by different colours

V. CONCLUSION

Thermoluminescence characteristics of the thermally treated household NaCl have been investigated thoroughly to check the reliability of using NaCl as reusable TL dosimeter. The salt was subjected to heating rate 10^{o}C/min and kept for 1 hour at different annealing temperatures namely, 200, 300, 400, 500 and 600 ^oC respectively. It has been found that TL response is enhanced as the annealing temperature increases showing maximum response at 500 ^oC and then drops afterwards; enhancement in TL response of 500 ^oC annealed group is more than 500% compared to the un-annealed group. Moreover, the TL glow curve and associated glow peaks remained unchanged from the 1st to the 2nd readout which means that the traps type and concentration are also unchanged. Dose response investigation showed that the NaCl chips annealed at 500 ^oC has a supralinear response with gamma dose up to 10 Gy. The results obtained in this study indicate that annealed NaCl salt at 500 ^{o}C could be used as a reusable and efficient gamma radiation dosimeter. This study introduces a new treatment approach to use affordable material such as household NaCl as a gamma ray dosimeter that can be used many times in contrast to its common known use in retrospective dosimetry where it is used just for one time. Glow curve deconvolution revealed that the glow curve of the newly developed NaCl dosimeter consists of 5 1st order glow peaks; this means that the TL process could be interpreted by the simplest kinetic mechanism in which electrons excited by heat to the conduction band are directly recombined with their relevant holes just above valence band.

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