

# TL GLOW CURVE AND EFFECT OF ANNEALING ANALYSIS ON NATURAL BARITE COLLECTED FROM MANGAMPETA, INDIA

V. Ramaswamy\* & I. Kalaiarasi

Department of Physics, Annamalai University, Annamalainagar, Tamilnadu, India

\*E-mail: srsaranram@rediffmail.com.

## ABSTRACT

The thermostimulated luminescence (TSL) glow curve characteristics of ten barite crystals of Mangampeta mines (Cuddapah District of Andhra Pradesh, India) were analysed. The natural thermoluminescence (NTL) measurements were carried out for all the samples and show three peaks at 80°C, 120°C and 295°C, when recorded with linear heating rate of 10°C/sec. The sample irradiated with a gamma dose of 100 Gy shows a new peak at 215°C with NTL observation, but the absence of 295°C glow peak. The annealed sample also shows the same trend. The sample was annealed in air at the temperatures ranging from 200°C to 1000°C, at an interval of 50°C, for 1 hr duration. Annealing treatment above 400°C increases the sensitivity of 215°C TSL peak but there is no change in other two peaks. On the other hand, annealing at 800°C caused a complete removal of low temperature peaks (80°C and 120°C). The enhancement in TSL sensitivity was found to depend on the annealing temperature. The optimum annealing temperature was fixed for getting maximum TL sensitivity.

**Keywords:** NTL, ATL, Barite, effect of annealing and TSL analysis.

## 1. INTRODUCTION

Thermoluminescent materials are widely used in day-to-day life. Their best-known applications are dosimetry of ionizing radiation, CTV screen phosphors, scintillators, X-ray storage and screens intensifying phosphors, laser materials etc. [1, 2]. Thermostimulated luminescence response of solid state material in natural form such as: quartz, feldspars, calcite, dolomite are reported respectively by many authors [3, 4, 5, 6, 7]. Moreover, information about thermostimulated luminescence of natural barite is still very limited. Meanwhile, thermostimulated luminescence of synthetic BaSO<sub>4</sub> materials with different impurities has been considered in detail [8, 9, 10, 11, 12]. With view of the possibility of employing natural barite as a thermoluminescent radiation dosimeter with high effective atomic number, it is necessary to have detailed knowledge about thermoluminescence processes in this material [13, 14].

The aim of the present investigation is to characterize the naturally occurring well-grown black coloured barite with TSL glow curves (NTL and ATL) and effect of annealing temperature in the range of 200°C-1000°C were carried out in this paper.

## 2. MATERIALS AND METHODS

In the present investigation, several well-grown black coloured barite crystals were collected from Mangampeta mines of Cuddapah District of Andhra Pradesh, India. About 10 samples were collected at different locations from the same rock bed. The crystals were stored in the dark until the TSL measurements were carried out. All these crystals were crushed and ground carefully with a mortar and pestle, and washed for 2min with 1% HCl solution and finally with distilled water to remove any organic material, and then dried in an oven. Grains sizes between 125 - 250µm in diameter were used for the TSL measurements. Magnetic particles were removed using a magnet.

The NTL measurements were first carried out for all the barite crystals and then annealed in an air atmosphere at temperatures ranging from 200°C to 1000°C, at the intervals of 50°C, for 1hr. After annealing the samples were irradiated by a <sup>60</sup>Co gamma source with a dose rate of 680 Gy/hr and the TSL measurement were carried out using a Nucleonix TLD-96 reader at a heating rate of 10°C/sec in nitrogen atmosphere (95% nitrogen and 5% hydrogen). The TSL glow curves were recorded immediately after irradiation to avoid loss due to fading. The data were stored on a computer.

## 3. RESULTS AND DISCUSSION

### 3.1. TL Glow Curve Analysis

TSL glow curves were recorded for all the ten samples with various treatments. The glow curves obtained for all these samples show similar structure with respect to shape, intensity and peak position. A representative glow curve

characteristic of natural, natural plus irradiation induced TSL and together with annealed sample glow curve is illustrated in Fig.1.

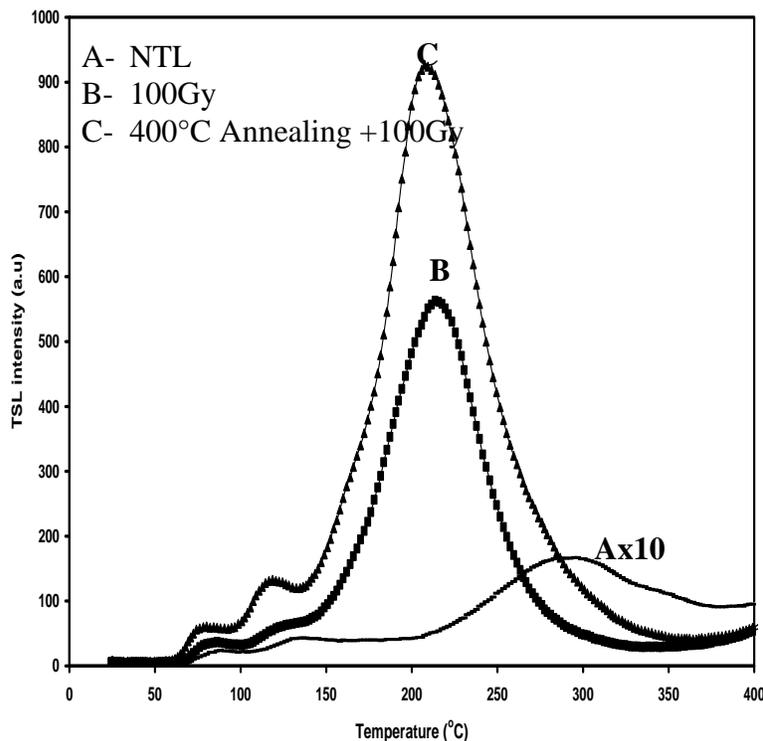


Figure 1. shows the TL glow curves of natural Barite sample.

It is well known that many natural minerals have natural TL due to the natural radiation in the environment, and the natural TL is removed using appropriate annealing temperature, it can affect the results of subsequent irradiation. The barite ( $\text{BaSO}_4$ ) used in this study has natural TL, the glow curve of which is shown in figure 1 (A). The natural TL intensity was obtained from natural  $\text{BaSO}_4$ , which was directly taken from the nature. The background signal was determined making the TL readout again. It is observed that the natural thermoluminescence (NTL) of the sample consists of two low temperature glow peaks appearing at 80°C and 120°C and a high temperature peak is appeared at 295°C (shown in figure 1 (A)). The high temperature natural TL peak is found to be completely removed by annealing the sample at 350°C for 30 min.

When the sample is irradiated by using  $^{60}\text{Co}$ -gamma source in the dose level of 100Gy, it shows a well defined new glow peak at 215°C with the existence of low temperature NTL peaks (80°C and 120°C) as well as the disappearance of high temperature NTL peak (295°C) (“B” in figure 1). The samples were irradiated after annealing at 400°C for 1hr with a dose of 100 Gy, which shows the same three glow peaks as that of 100Gy irradiated sample (without annealing) and there is no change in position and shape of the glow curve except increase in intensity (“C” in figure 1). This type of result may be due to the possibility of changes in charge mobility or removal of certain feasible non-radiative transitions. Increase in intensity of glow peaks on irradiation may due to the increase in the number of traps into the lattice.

The low temperature glow peaks are poorly defined. It was found that the low temperature NTL peaks (80°C and 120°C) are saturated quickly for absorbed doses of 250 Gy, after which no response to laboratory gamma irradiation has been observed (figure 2). This indicates that those peaks are radiation non-sensitive. Another glow peak (215°C) enhances in intensity systematically with dose which shows a radiation sensitive peak. This enhancement of TSL intensity with increase of dose is due to the fact that the number of electrons in the excited state increases with increase of the absorbed radiation dose and hence the possibility of recombination as well as TSL emission.

Previous studies [6] on calcite showed that there was a systematic enhancement in NTL peak (345°C) due to laboratory gamma irradiation upto 150 Gy. They also observed a regeneration effect on this peak. The NTL peak at 345°C was not showing any response to dose even after annealing at 600°C. Hence, the TSL peak at 345°C or above 300°C is found to be non-radiation induced peaks. Studies by Ramasamy et al., (2009) have shown in dolomite a systematic enhancement in intensity of 335°C glow peak with doses (upto  $10^4$  Gy) and there was no observation of

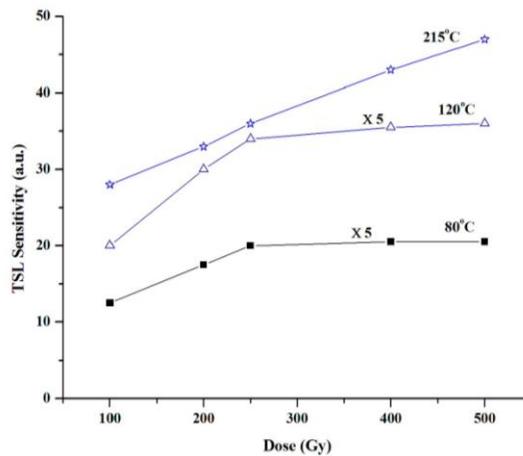


Figure 2. shows the TL sensitivity of the three glow curves versus various doses.

regeneration effect on this peak. Moreover, a systematic increase in TL intensity was also observed with annealing temperature. They concluded that this peak was found to be radiation sensitive. But in barite (present study), the NTL glow peaks shows three different temperatures as 80°C, 120°C and 295°C. The high temperature NTL is found to be completely removed by irradiation as well as annealing the sample at 100 Gy and 350°C for 30 min respectively and the other two peaks (80°C and 120°C) saturated quickly for absorbed doses up to 250 Gy which shows non-radiation induced peaks. Thus, the observed behavior shows a similar trend as that of calcite [6] but there is no observation of regeneration effect in barite.

### 3.2. Effect of Annealing Temperature

Figure 3 shows a plot of TSL sensitivity versus different annealing temperature ranging from 200°C to 1000°C for 1 h at an increment of 50°C. Each annealed sample was irradiated to a test dose of 100 Gy before the TL readout. It is observed that the TL sensitivity of natural barite is significantly changed only after annealing at temperatures above 400°C (below 400 °C there is no change in its intensity is observed).

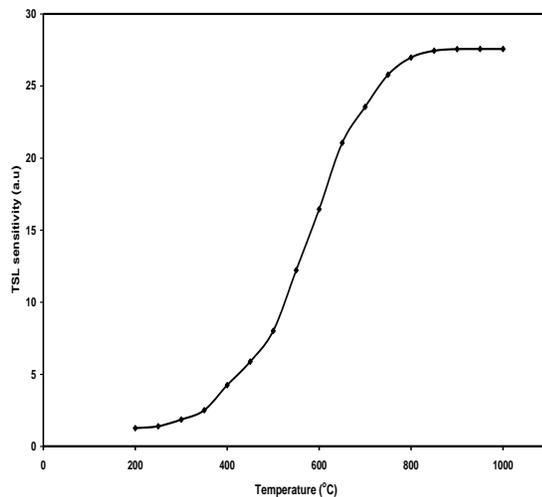


Figure 3. shows the effect of annealing on barite sample (200°C to 1000°C in steps of 50°C) irradiated with 100 Gy.

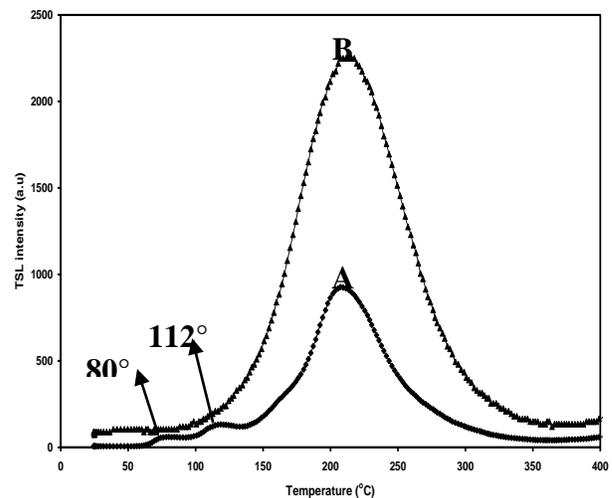


Figure 4 shows the TL glow curves for 400°C (A) and 800°C (B) annealed and irradiated with 100 Gy.

From this, it is observed that the TSL sensitivity of the peak at 215°C increases with increase of annealing temperature up to 900°C and then saturation occurs. But the low temperature glow peaks (80°C and 120°C), which is not showing any response to dose as well as annealing temperatures (upto 700 °C). For this reason, those TL peaks not taken into consideration for all samples. Further, annealing in air for temperature at 800°C caused a

complete removal of low temperature glow peaks and only the appearance of high temperature peak 215 °C is observed (see Figure 4). The low temperature glow peak arise may be due to the presence of certain defects in the lattice. However, at higher temperatures (around 800 °C) these defects are removed.

The samples annealed at a temperature of 900°C for 1hr causes an increase in its sensitivity of the TL peak (215°C) by a factor of 15 times as compared to unannealed samples. Such behaviour of samples annealed in air has been observed by many authors in different materials [6, 7, 15, 16, 17].

#### 4. CONCLUSION

From this study it can be concluded that:

- ❖ The natural barium sulphate sample having three NTL glow peaks, two low temperature peaks at 80°C and 115°C and one high temperature glow peak at 295°C respectively.
- ❖ On annealing and irradiation process, the high temperature glow peak 295°C is disappeared and a new glow peak is appeared at 215°C as well as the disappearance of low temperature peaks is observed after annealing at 800 °C. The low temperature glow peaks are non-sensitive to radiation and annealing temperature. Finally, a single high intense glow peak is appeared at 215°C. The optimum TSL sensitivity is obtained at 900 °C.
- ❖ The high sensitive 215°C peak can be utilized as dosimetric peak. For this, further studies are required to use as dosimeter.

#### 5. ACKNOWLEDGEMENT

We are grateful to Dr. M.T. Jose of RSD, IGCAR and Dr. V. Ponnusamy of Anna University, MIT campus for the valuable help rendered during the TL measurements and constant encouragement.

#### 6. REFERENCES

- [1]. J. Azorin, C. Furetta and A. Scacco, Preparation and properties of thermoluminescence materials, *Phys. Status Solidi A*, 138, 9-46, (1993).
- [2]. S.L. Issler and C.C. Torardi, Solid state chemistry and luminescence of X-ray phosphors, *J. Alloys Compd.* 229, 54–65, (1995).
- [3]. M. A. Gomaa and A. M. Eid, TL dosimetry using natural materials. *Atomkernenergie* 27, 274-276, (1976).
- [4]. M. A. Gomaa and M. M. Morsi, TL dosimetry used fused silica, *Atomkernenergie* 28, 135-137, (1976).
- [5]. S. K. Youssef, L. A. Guirguis and N.A. Shahin, Some aspects of radiation induced effects of TL mechanisms of natural barite for dosimetric utilization, *J. Mater. Sci. Lett.* 12, 1557-1561, (1993).
- [6]. V. Ponnusamy, V. Ramasamy, M. Dheenathayalu and J. Hemalatha, Effect of annealing in thermostimulated luminescence (TSL) on natural blue colour calcite crystal, *Nucl. Instrum. Methods Phys. Res., Sect. B*, B217, 611-620, (2004).
- [7]. V. Ramasamy, V. Ponnusamy, S.S. Gomathi, M.T. Jose, Thermostimulated luminescence characteristics of dolomitic rocks and their use as a gamma ray dosimeter, *Radiat. Meas.* 44 351-358 (2009).
- [8]. R.L. Dixon, K.E. Ekstrand, Thermoluminescence of SrSO<sub>4</sub>: Dy and BaSO<sub>4</sub>: Dy (dosimetric properties), *Phys. Med. Biol.* 19, 196-205, (1974).
- [9]. S.S. Shinde, B.C. Bhatt, J.K. Srivastava, S.S. Sanaye, Development and Characterisation of a BaSO<sub>4</sub>: Eu,P Phosphor as a High Sensitive TL Dosemeter, *Radiat. Prot. Dosim.* 65, 305-308, (1996).
- [10]. U. Madhusoodanan, M T Jose and A.R. Lakshmanan, Development of BaSO<sub>4</sub>:Eu thermoluminescence phosphor, *Radiat. Meas.* 30, 65-72, (1999).
- [11]. Numan Salah, Sami S. Habib, Zishan H. Khan, Salim Al-Hamedi and S.P. Lochab, Nanoparticles of BaSO<sub>4</sub>:Eu for heavy-dose measurements, *J. Lumin.* 129, 192–196, (2009).
- [12]. J. Manam and S. Das, Thermally stimulated luminescence studies of undoped, Cu and Mn doped BaSO<sub>4</sub> compounds, *Indian J. Pure and Appl. Phys.* 47, 435 – 438, (2009).
- [13]. M. Prokic, Thermoluminescent properties of natural barium sulfate, *Int. J.Appl.Rad.Isot.* 25, 545 – 549, (1974).
- [14]. M. Prokic, Mechanism of thermoluminescence in natural barites, *J.Phys. Chem. Solids* 40, 405 – 412, (1979).
- [15]. K. Ninagawa, N. Takahashi, T. Wada, I. Yamato, N. Yamashita and Y. Yamashita, Thermoluminescence measurements of a calcite shell for dating, *Quart. Sci. Rev.* 7, 367-371, (1988).
- [16]. A.D. Franklin, W.F. Hornyak, V. Pagonis and N. Kristianpoller, Thermoluminescence study of annealing a geological calcite, *Nucl. Tracks Rad. Meas.*, 17, 517-523, (1990).
- [17]. V. Pagonis and C. Michael, Annealing effects on the thermoluminescence of synthetic calcite powder, *Radiat. Meas.* 23, 131-142, (1994).