DETERMINATION OF TRAPPING PARAMETERS OF THERMALLY STIMULATED LUMINESCENCE OF DY DOPED BASO4 SAMPLES

M. Deben Singh[*]

Th. Open Singh[**]

N. Bhumeshwar Singh[**]

S. Romesh Singh[***]

Abstract:

$BaSO_4$:Dy samples for different doping concentrations have been synthesized by co-precipitation methods. TL glow curves of $BaSO_4$:Dy samples after irradiation with γ-rays have been recorded. The observed TL glow curves show a complex curve consisting of two sharp peaks at Tm= (385-393) K , (436-445) K and a daughter peak at Tm(405-410) K respectively. Trapping parameters of the TL glow curves are also evaluated using CGCD technique.

Key words: Thermoluminescence , Trapping parameters, Glow curves ,CGCD, Dosimetry.

[*] Department of Physics, Jiri College, Jiribam, Pin-795116

[**] Department of Physics, Standard College, Kongba, Pin-795005

[***] Department of Physics C.I. College, Bishnupur, Pin-795126, Manipur

Introduction:

In the last three decades, many new thermoluminescent (TL) materials have been studied for the application in radiation dosimetry and detection. The commonly used TL materials are $CaSO_4$ and $BaSO_4$ doped with rare-earth elements because of their high thermoluminescent sensitivity and their negligible fading[1&2] Among the TL materials, $BaSO_4$ is well- known material used for TL studies because of the application in personal and environmental dosimetry [3]. Thermally stimulated luminescence (TSL) of $BaSO_4$ both natural and synthetic doped with various impurities has been extensively studied [4]. Some workers reported that $BaSO_4$:Ln(Ln= Sm, Eu, Dy, Tm) have extremely higher TL intensities than that of $CaSO_4$:Dy and $CaSO_4$:Tm, which were used in radiation dosimetric applications [5].

The above investigations influence us to study the thermally stimulated luminescence of Dy doped $BaSO_4$ samples. In the present study, Barium sulphate doped with different concentrations of Dy is prepared by chemical method [6-10]. The kinetic parameters of the recorded TL glow curve of γ-rays irradiated phosphor are calculated. The variation of TL intensity for different concentrations of Dy is reported.

Sample preparation:

The starting chemicals are barium chloride ($BaCl_2.2H_2O$, 99.9%, Aldrich), dysprosium nitrate [$Dy(NO_3)_3.H_2O$, 99.9%, Sigma Aldrich], Concentration sulphuric acids (H_2SO_4, Merck, AR grade). In a typical synthesis of 0.27 mol % of dysprosium, $BaCl_2.2H_2O$ is dissolved in distilled water along with 0.27 mol % dysprosium nitrate. The solution is stirred homogeneously with the help of a magnetic stirrer. Concentrated H_2SO_4 is added drop by drop to the solution until the precipitation is completed. The precipitate ($BaSO_4$:Dy) is collected and washed repeatedly by distilled water till the excess acid is removed. The sample is kept in an oven for 1 hour and then annealed at 870 K for 1 hour and made into powdered form. The sample thus obtained is $BaSO_4$:Dy. In the similar way, the different samples of $BaSO_4$:Dy for different concentrations of Dy at 0.34, 0.20, 0.13 and 0.06 mol % respectively are prepared.

Theory:

Gartia et al [11] and Rasheedy [12] wrote the equation governing the non first order ($b \neq 1$) kinetics including the second order (b=2) kinetics in TL as

$$I(t)= \frac{dn}{dt} = s(n^{b}/N^{b-1})\exp(-E/kT), \qquad (1)$$

Where n is the number of electrons trapped at the trap centre at the time t, N the concentration of trap centre, s the frequency factor, b the order of kinetics, k the Boltzmann constant, T the absolute temperature and E is the activation energy.

Defining a filling factor, $f=n_o/N$ and using linear heating rate β, such that $T=T_o+\beta t$, one obtains the equation for TL intensity I(T) governing the non-first order kinetics including the second order kinetics as

$$I(T) = N f^{b} s \exp(-E/kT) [1+(b-1)f^{b-1}s/\beta \int_{T_0}^{T}\exp(-E/kT')dT']^{-b/b-1}, \qquad (2)$$

Results and Discussions:

Figure 1 shows the XRD patterns of as prepared BaSO4:Dy (0.27 mol% of Dy). The XRD data are analyzed by H'Pert High Score and show that all the diffraction peaks are orthorhombic

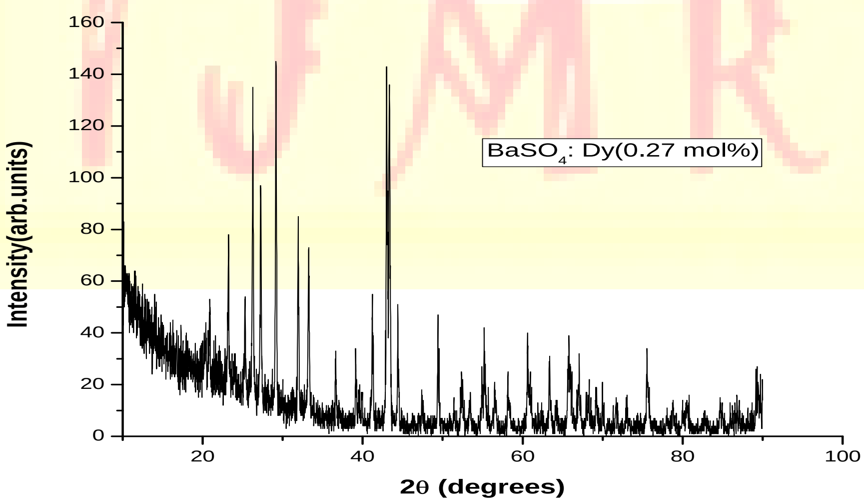

Fig 1 XRD patterns of BaSO4:Dy (0.27 mol% of Dy) Sample

structure (ICDD Ref. No. 00-046-1415). TL glow curves $BaSO_4$:Dy samples for different concentrations of Dy annealed at 873 K for 1 hour are recorded after irradiated with γ-rays for 504 Gy with a linear heating rate of 2.23°C/s as shown in Fig 2. Here, curves a, b, c, d and e show the TL glow curves of $BaSO_4$:Dy for different concentrations 0.06, 0.13, 0.20, 0.27 and 0.34 mol.% of Dy respectively. In each TL glow curves, two prominent peaks at around (385-393) K and (436-445) K respectively are observed. There is also another peak at around 408 K, which is less prominent as compared with the two peaks. The presence of such a shoulder peak can be seen more distinctly for the sample of a particular concentration of Dy (0.27 mol.%) than the other concentrations of Dy. As observed, the TL intensity increases with the increase of the concentration of Dy. This is because of the fact that the number of defect atom in the crystal increases with the increase of the concentration of dopant (Dy) within the saturation condition and consequently increases the number of recombination of the holes and electrons such that maximum TL is observed.

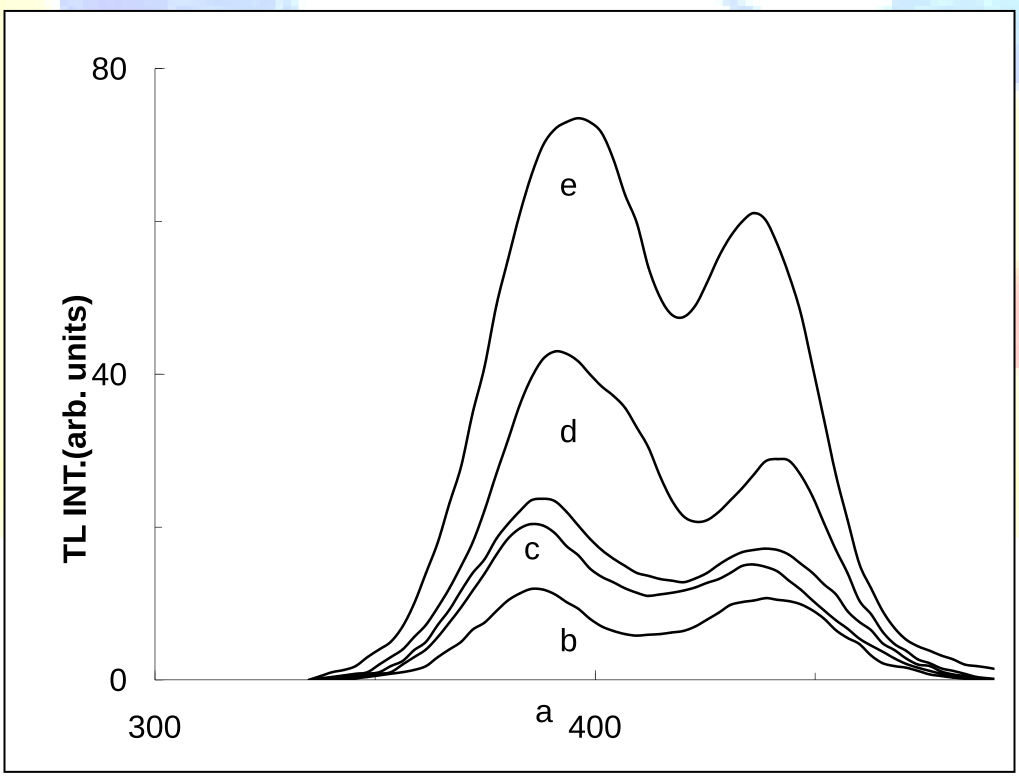

Fig.2. TL glow curves of $BaSO_4$:Dy for different concentrations of Dy and irradiated with γ- rays for 504Gy.

To analysis of the observed TL glow curve, the glow curve of $BaSO_4$:Dy for a particular concentration of Dy (0.27 mol %) irradiated with 504 Gy of γ-rays has been selected and recorded with a constant heating rate of 2.25 ºC/s (Fig.3). There are two prominent peaks at temperatures 388.6 and 439.9 K respectively. And, to identify the number of peaks present in the complex glow curve, thermal cleaning method is applied. In this technique, the sample has been heated upto 400 K and recorded TL glow curve as shown in Fig. 4. It shows a TL glow curve consisting of two peaks with peak temperatures at 410.6 and 440.3 K respectively.

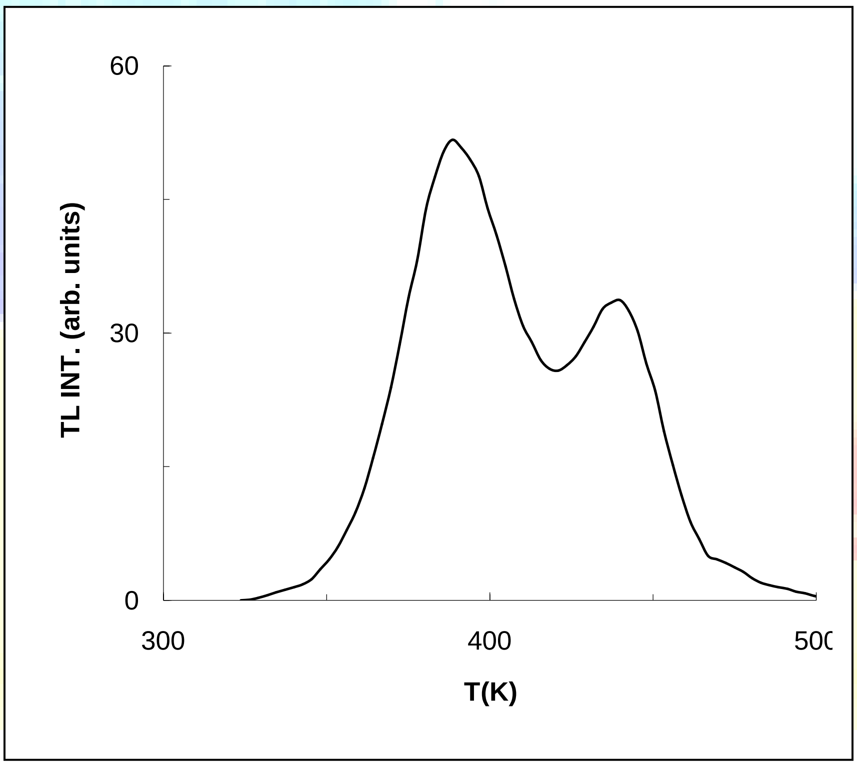

Fig. 3 TL glow curves of $BaSO_4$:Dy irradiated with γ-rays for 504 Gy.

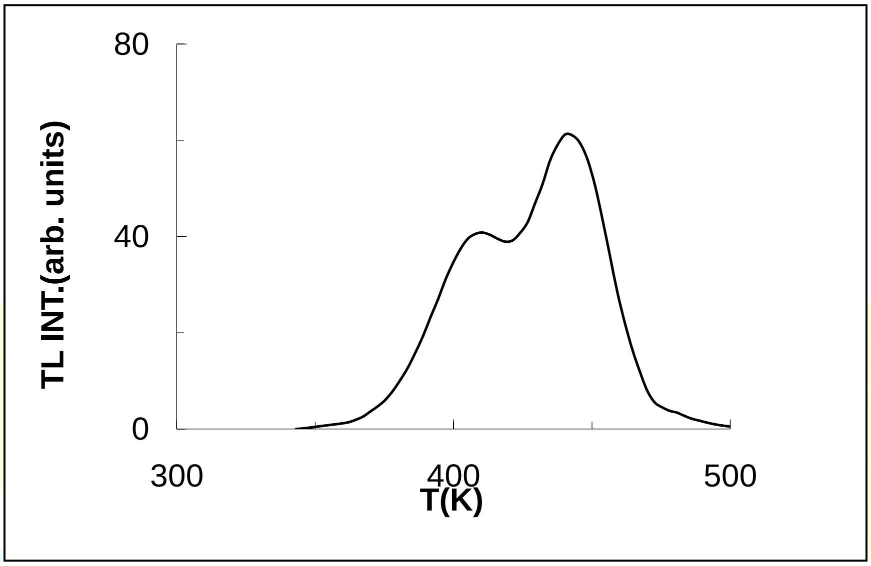

Fig. 4 TL glow curve of $BaSO_4$:Dy irradiated with γ-rays for 504 Gy after thermal cleaning (Tc=400K).

Again, the sample has been thermally cleaned upto 428 K and recorded TL glow curve as shown in Fig. 5. It observes an isolated peak having a peak temperature T_m=442.8 K. Thus, it is able to know the presence of three TL peaks of γ-irradiated $BaSO_4$:Dy in the temperature range (373-473) K. Bhatt et al [6] also reported the presence of three peaks in $BaSO_4$:Eu phosphor.

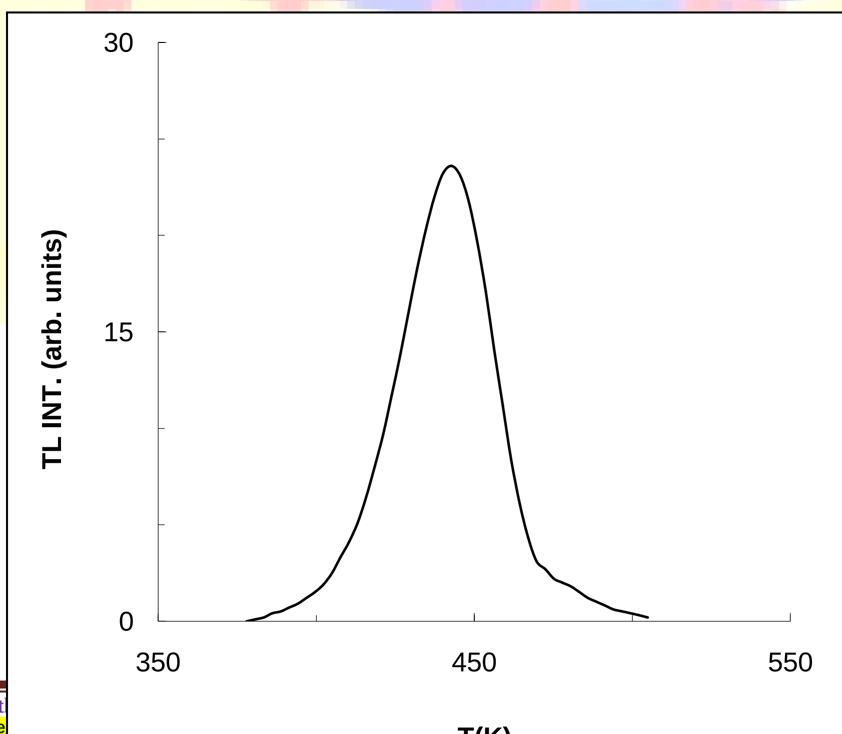

Fig.5 . Isolated TL glow curve of BaSO4:Dy irradiated with γ-rays for 504 Gy (after Tc=428 K) .

To determine the trapping parameters of the TL glow curve of γ-irradiated $BaSO_4$:Dy, the isolated peak with T_m=442.8 K is selected. The half width at the rising portion, (τ) and half width at the falling portion,(δ) of the glow peak are 18.9 and 17.6 ºC respectively. The full width at half maximum intensity (ω) is 36.45ºC and the shape factor ($\mu_g=\delta/\omega$), which indicates the order of kinetics is found to be 0.48. As $\mu_g > 0.42$ but less than 0.52, the TL glow curve is neither first order nor second order but of the non-first order kinetics. Using the peak shape formula

$$E_\alpha = [(A_\alpha k T^2_m)/\alpha] + B_\alpha k T_m \quad (\alpha = \tau, \delta \text{ and } \omega), \quad (3)$$

where,

$A_\tau = 1.51 + 3 (\mu_g - 0.42)$, $B_\tau = -3.16 - 8.4 (\mu_g - 0.42)$

$A_\delta = 0.976 + 7.3 (\mu - 0.42)$, $B_\delta = 0$

$A_\omega = 2.52 + 10.2 (\mu - 0.42)$, $B_\omega = -2$

one can easily determine the activation energy (E) of the observed glow peak. The values of E_τ, E_δ and E_ω of the isolated peak are 1.33,1.33 and 1.32 eV respectively.

By using the activation energy obtained in the above methods as input parameter, the curve fitting technique [8,11 &13] is used to fit the isolated peak as shown in Fig. 5. The curve fitting of the isolated peak is shown in Fig.6 with the kinetic parameters such as activation energy E = 1.32 eV, frequency factor s = $1.8\times10^{14} s^{-1}$ and order of kinetics b=1.3. The overall fitting is good and the misfit in the falling part may be due to the presence another very small peak at the higher temperature side.

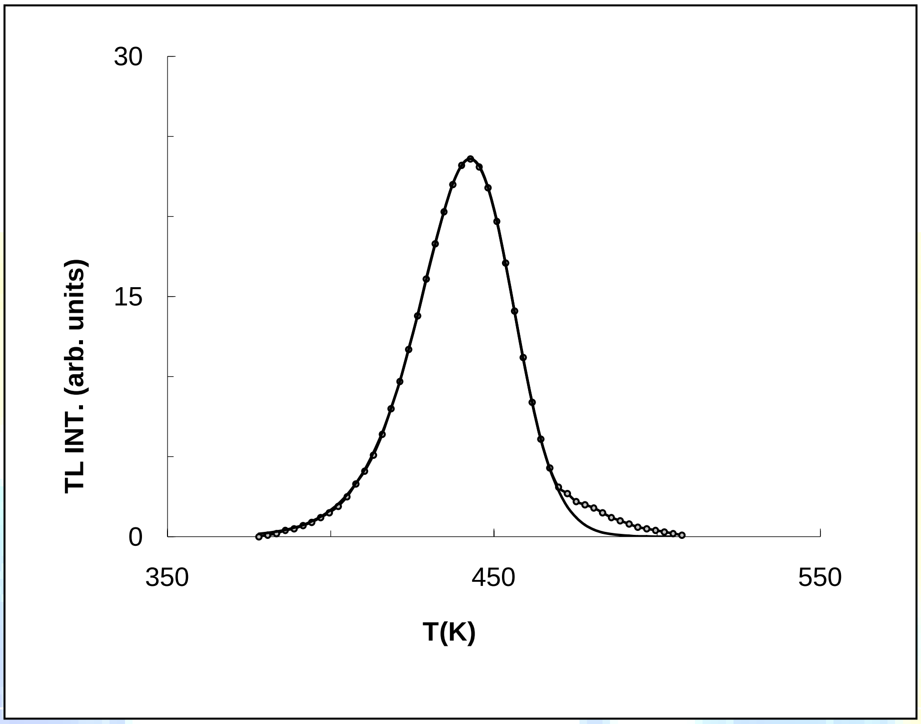

Fig.6 Curve fitting of isolated TL glow curve of $BaSO_4$:Dy irradiated with γ-rays for 504 Gy (after Tc=428 K).

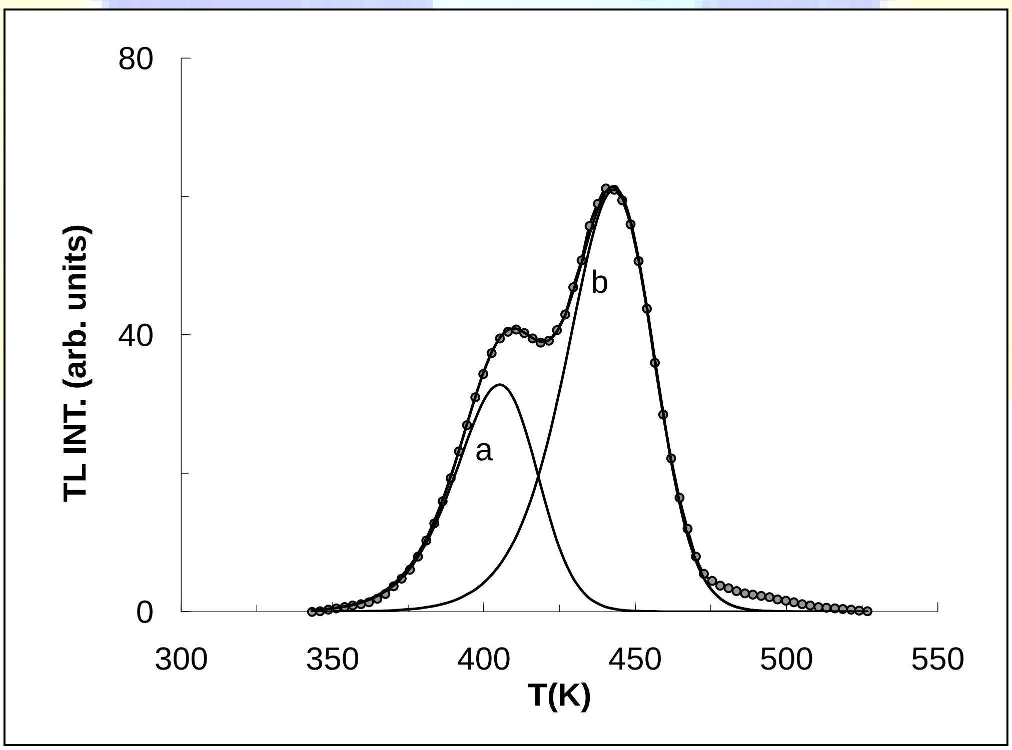

Fig.7 CGCD of TL glow curve (after Tc=400K) of $BaSO_4$:Dy irradiated with γ-rays for 504 Gy.

Computerised glow curve deconvolution (CGCD) technique [8,11 &13] is used to decode the complex glow curve (Fig.4) recorded after thermal cleaning to the sample at Tc = 400 K. The TL glow curve is deconvoluted into two peaks having activation energies 1.17 and 1.32 eV respectively and shown in Fig 7. The order of kinetics of both the peaks is found to be 1.3. The frequency factors of the two peaks are 6.3×10^{13} and 1.7×10^{14} s^{-1} respectively.

Finally Fig. 8 shows the glow curve deconvolution of the glow curve (Fig. 3). The curve can be nicely fitted with three peaks of activation energies 1.06, 1.17 and 1.32 eV respectively.

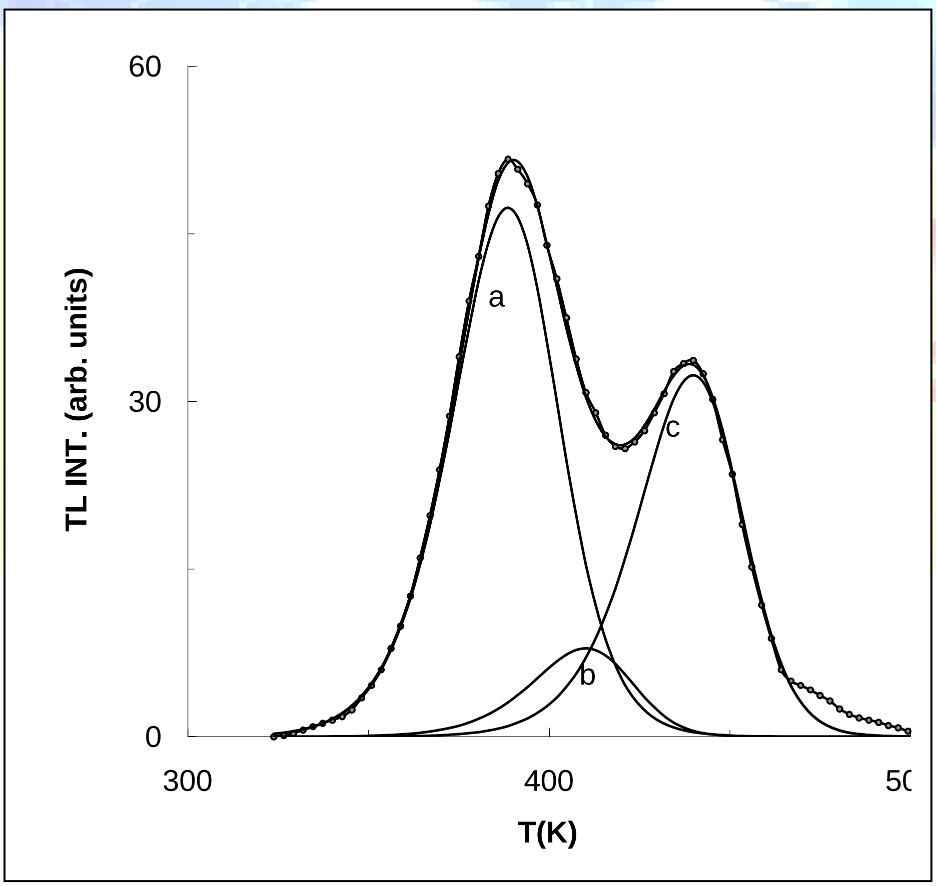

Fig.8 CGCD of TL glow curve of $BaSO_4$:Dy irradiated with 504 Gy of γ-rays.

Conclusions:

TL glow curve of γ-irradiated $BaSO_4$:Dy sample contains three peaks with peak temperatures at around 389, 408 and 440 K respectively. The complex glow curves of $BaSO_4$:Dy can be analysed using the non-first order kinetics model. The TL glow curve can be deconvoluted into three constituent peaks having activation energies 1.06, 1.17 and 1.32 eV and their respective orders of kinetics are found to be 1.5, 1.3 and 1.36 and also the frequency factors of three peaks lie between 9.9 $x10^{12}$ and 2.3 $x10^{14}$ s^{-1}.

Acknowledgements:

One of the authors, M. Deben Singh thanks UGC for major research project grant [F. No. 39-526/2010(SR) dated 7th Jan. 2011]. We acknowledged MU, Canchipur, Manipur for providing TL-recorder and XRD facilities.

References:

1. J. Azorin, G. Gonzalez, A. Gutierrez, R. Salvi, ***Health Phys. 46 (2) (1984) 269***.
2. J. Azorin, A. Gutierrez, ***Health Phys.56(4) (1984) 551***.
3. J. Manam, S. Das, ***Journal of Physics and Chemistry of Solids, 70 (2009) 379-384***.
4. M. S. Atone, S.J. Dhoble and S. V. Moharil***, Radiation effects and defects in Solids, 127 (1993) 225-230***.
5. S. Dorendrajit Singh and M. Deben Singh, ***Proceedings of NCLA, Jan. 18-20 (2007), Bharathiar University, Coimbatore.***
6. Bhatt, B.C., Sanaye, S. S., Shinde, S. S. and Srivastava, J. K., ***Radiation Protection Dosimetry Vol.69.No2.p 105-110 (1997).***
7. Madhusoodanan, U., Jose, M.T.and Lakshmanan, A.R., ***Radiation Measurement 30, 65-72 (1999).***

8. D.Singh,S. and D. Singh,T. ***Proc. of ICLA BARC, Mumbai Feb. 2004, p281-284***.
9. Dwijen Singh, T. and Dorendrajit Singh, S**.,** ***Proceeding of NASLA-2003, p 216***.
10. D. Singh, S. and Deben Singh,M.,***Vol.50 Proc. of the DAE SSP Symp –2005***.
11. Gartia, R. K., Singh, S. D. and Mazumdar***, P. S., Phys. Stat. Sol. 138,319(1993).***
12. M. S. Rasheedy **J. Phys, :Condensed matter phys. 5,633(1993).**
13. McKeever, S. W. S. Moscovitch, M. and Townsend, ***P. D. Thermoluminescence Dosimetry materials : properties and uses, Nuclear Technology Pub.,England 1995***.