www.vbripress.com/aml, DOI: 10.5185/amlett.2016.6160 Published online by the VBRI Press in 2016

Investigation of dosimetric features of beta – irradiated Er^{3+} doped strontium pyrophosphate

Nimesh P. Patel[1*], M. Srinivas[1], Vishwnath Verma[1], Dhaval Modi[1], K. V. R. Murthy[2]

[1]*Department of Physics, Faculty of Science, M. S. University of Baroda, Vadodara 390002, India*
[2]*Department of Applied Physics, Faculty of Technology and Engineering, The M. S. University of Baroda, Vadodara 390003, India*

[*]Corresponding author. Tel: (+91) 85-11122925; E-mail: nimesh.0112@gmail.com

Received: 12 September 2015, Revised: 05 December 2015 and Accepted: 10 May 2016

ABSTRACT

Thermoluminescence (TL) of $Sr_2P_2O_7$: Er^{3+} phosphors had been studied in order to investigate the nature of the trapping centers created due to doping ions. The effect of beta – irradiation was analyzed for various doses given to the samples. The measurements of TL glow curves have been done at the heating rate 6 K/s. The TL measurements done for the study of the dosimetric properties of samples showed that the β-dose response is linear from 5 Gy to about 50 Gy. The experimental glow curves had been analyzed by glow curve fitting technique which revealed that the glow curve is consist of three curves with peak temperature 400 K, 430 K and 464 K. The activation energy 'E_a' of the peaks had been calculated to be 0.90 ± 0.03 eV, 1.10 ± 0.04 eV and 1.05 ± 0.03 eV, respectively. The geometrical factor 'μ_g' having values 0.51, 0.52 and 0.48, imply that the glow curve having second order kinetics. The linear dose response, fading effect after exposed and reusability of samples are very reliable for dosimetry applications. Copyright © 2016 VBRI Press.

Keywords: Strontium pyrophosphate; thermoluminescence (TL); peak shape method; activation energy.

Introduction

TLDs are effective tools to assess personal radiation exposure from substances or equipment that emits radiation. To achieve accurate measurement of the absorbed dose, the dosimeter material should have a similar response as the medium being irradiated. The performance of TLD is evaluated by taking into account properties such as linearity, dose range, energy response, reproducibility and stability of stored information (fading effect) [**1**]. To developing efficient TLD phosphor is good research area, because it has large applications in radiation dosimetry, personal dosimetry and environmental dosimetry. Rare earth doped pyrophosphates potential phosphors for TLD application because of its favorable semiconducting and luminescence properties [**2-5**]. Thermally stimulated luminescence (TSL) of europium-ion doped strontium pyrophosphate was reported by V. Natarajan, *et al.* [**2**]. Thermoluminescent properties of $Sr_2P_2O_7$ doped with copper and some rare earth elements had been investigated by A. N. Yazici, *et al.* [**3**]. Thermoluminescence approach of Ce doped α-$Sr_2P_2O_7$ and Dy doped α-$Sr_2P_2O_7$ were reported by our group [**4, 5**]. Energy migration and characterization of trap level distribution was reported by S. Som, *et al.* [**6**].

The motivation of this research is regarding to the less work had been done on this phosphor in radiation dosimetry. Only few research groups had been reported radiation dosimetry approach of the alkali earth pyrophosphate phosphors on the basis of gamma and x-ray irradiation, while in this report we are proposing the phosphor for beta irradiation dosimetry. The effect of beta dose on the phosphor shows it potential application in dosimetry.

In these paper, Er^{3+} doped $Sr_2P_2O_7$ phosphor was synthesized by combustion method and TL property of samples was investigate in order to understand the role of doping ions on the TL properties to make useful the phosphor for TLD applications. We have investigated some of the necessities for TLD phosphor application such as dose response, fading effect and reusability of phosphor.

Experimental

Material Synthesis

Er^{3+} doped $Sr_2P_2O_7$ (Strontium pyrophosphate) phosphors were synthesized by combustion synthesis method at 900 ºC by varying doping concentrations (0.5, 1.0, 1.5, 2.0, 2.5 and 5.0 mol %). The reactants $SrCO_3$ (A.R.), $(NH_4)_2HPO_4$ (A.R.) and Er_2O_3 (99.99 %) (A.R.) were taken in a stoichiometric ratio as per the chemical equation balancing. All reactants were grinded in an agate mortar for half hour to make homogeneous mixture. The homogeneous mixture of reactants with urea (A.R.; 15 % of the mass of the mixture) taken as a flux was grinded. The mixture was heated at 900 °C in alumina crucibles for 3h in a muffle furnace and allowed to cool down naturally up to room temperature. Pure white samples were again grind to obtain fine powder [**4, 5**].

Characterizations

All samples were irradiated with Sr^{90} beta source having average energy radiation rate 10 ± 0.05 Gy per minute and received various doses 5, 10, 20, 30, 50 Gy for TL study. The TL glow curve was measured using a NUCLEONIX TL analyzer type TL1009 at heating rate 6 K/s for the range 325 K to 550 K to evaluate dosimetric features of the prepared phosphors. TL of all samples was recorded for five consecutive days to carryout of the fading effect and reusability of samples. Calculation of trapping parameter was completed by glow curve deconvolution (GCD) technique reported by D. Afouxenidis [7].

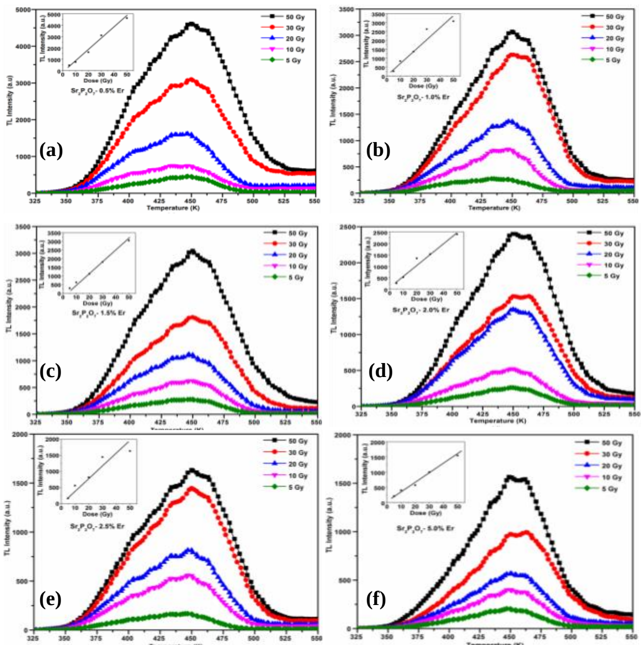

Fig. 1. TL glow cures of β-irradiated samples (a)$Sr_2P_2O_7$: 0.5%Er; (b)$Sr_2P_2O_7$: 1.0%Er; (c)$Sr_2P_2O_7$: 1.5%Er; (d)$Sr_2P_2O_7$: 2.0%Er; (e)$Sr_2P_2O_7$: 2.5%Er; (f)$Sr_2P_2O_7$: 5.0%Er.

Results and discussion

It is very important to create suitable trap depth and large number of trap centers to prepare long lasting phosphors (LLPs) for TLD applications, which can be achieve by proper doping of impurity ion with particular concentration in host for that TL analysis of Er^{3+} doped $Sr_2P_2O_7$ had been carried out.

Fig. 1 shows TL glow curves measured at 6 K/s from the samples of Er^{3+} doped $Sr_2P_2O_7$ after β-irradiation and inserted graph shows the dose response of each samples. To reveal the existence of trap centers generated in energy gap by doping ions, a method of glow curve deconvolution of TL glow curve was carried out.

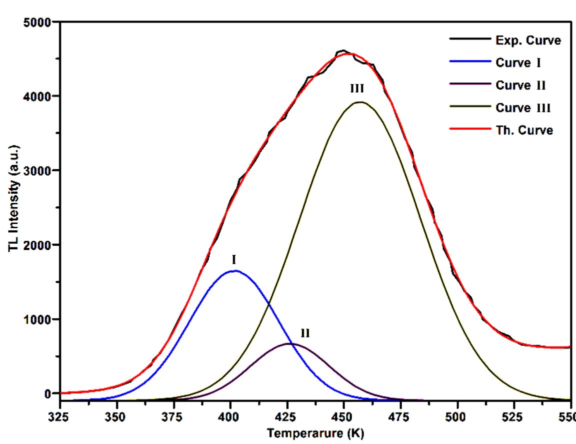

Fig. 2. Glow curve fitting of $Sr_2P_2O_7$: 0.5 % Er phosphor irradiated with 50 Gy beta dose.

Fig. 2 shows the curve fitting for sample $Sr_2P_2O_7$: 0.5%Er irradiated with 50 Gy beta dose with best fitting parameter [**7**]. The curve fitting analyses of TL glow curves revealed that three peaks are present in glow curve at different temperatures located at 400 K, 430 K and 464 K corresponds to the three trap centers formed at different depths. The complex nature of the TL glow curves could be attributed because of the overlapping of various bands present at a close distribution of their trap depths [**8**].

TL analysis gives the vital information about the mechanism behind the formation of trap centers, types of the traps, and the trap depth. All fitted curves of TL glow curve were analyzed by Peak shape method alternatively called Chen's method to find out the kinetic parameters in order to understand the basic mechanisms involved in the emission. In this method, three equations for first- and second-order kinetics are derived correlated to the trap depth: a full width of the peak at half maximum '$\omega = T_2 - T_1$', a low-temperature half width '$\tau = T_m - T_1$' and a high-temperature half width '$\delta = T_2 - T_m$' [**9**].

The general equation for evaluating activation energy 'E_a' is given as:

$$E_a(\gamma) = c_\gamma\left(\frac{kT_m^2}{\gamma}\right) - b_\gamma(2kT_m)$$

where, T_m is the peak temperature at the maximum intensity, k is the Boltzmann's constant, and γ is τ, δ, ω. μ_g is geometric factor which is calculated from the relation,

$$\mu_g = \frac{\delta}{\omega}$$

The average activation energy 'E_a' calculated to be 0.90±0.03 eV, 1.10±0.04 eV and 1.05±0.03 eV for the peak I, peak II and peak III, respectively. The frequency factors 's' for various depths are 1.5×10^{11} s^{-1}, 9.5×10^{12} s^{-1} and 1.1×10^{11} s^{-1}, respectively. The order of kinetics 'b' was determined from the geometric shape factor 'μ_g'. The suggested values of geometric factor are 'μ_g=0.42 and 0.52 for first and second order kinetics, respectively. It is found that geometric factor for different peaks were nearer to 0.52 which revealed that the glow curve having second order kinetics.

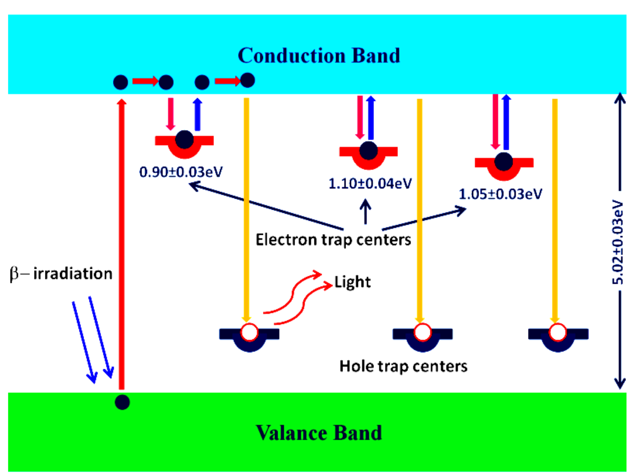

Fig. 3. TL mechanism and energy level diagram of Er^{3+} doped $Sr_2P_2O_7$ phosphor.

Activation energy of the glow curve gives the information about the trap depth located in energy gap. **Fig. 3** shows the TL mechanism of Er^{3+} doped $Sr_2P_2O_7$ phosphor. The TL mechanism explains the electron excitation with β–irradiation, Electron trapping and recombination of electron by emitting light at time of heating.

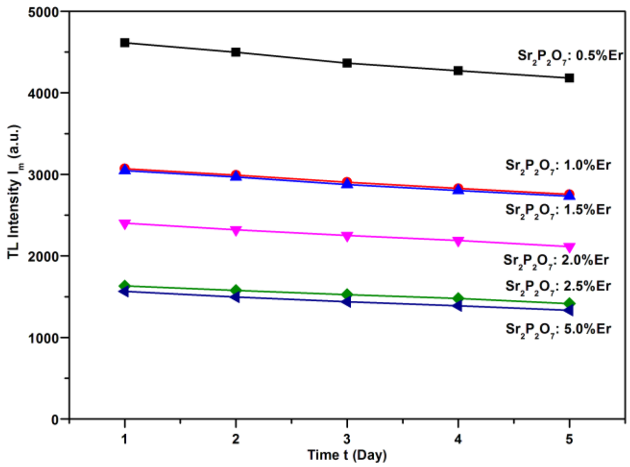

Fig. 4. Graph of TL peak Intensity - Fading time for different concentrations of Er^{3+} in $Sr_2P_2O_7$ phosphor.

Fig. 4 shows the fading of TL intensity of all samples measured for five days successively. Fading effect illustrate the decrease in the TL intensity of the glow curve after irradiation when TL recorded after few days.

It has been observed that the TL intensity of maximum temperature was fading with rate of 3-5% per day to day observed for five day. The fading effect in TL intensity was occurring because of the less precautions were taken to store the sample after irradiation. The low fading effect can be suggests that the phosphor have good stability to store the exposure. Reusability of samples was investigated by taking TL response from the same sample after irradiating it several times (10 times) and the result explain that the TL response for each use was nearly similar to that of first observation. This implies the phosphors having high stability and reusability [**10**].

TL observed from the Er^{+3} doped $Sr_2P_2O_7$ phosphors is mainly due to the defect centers present in lattice as a reason of doping ions and formation of defects also depends upon the synthesis method. In combustion method reaction has been taken place at low temperature around 300° C to 400° C because of flux. But the high temperature annealing is responsible for creating more defects as well as the substitution of host ions by doping ions. The doping ions could be substitute the host Sr^{2+} ions site and create metastable states at certain depth in energy gap. TL sensitivity of Er^{3+} of $Sr_2P_2O_7$ phosphor is mainly depending on Er^{3+} concentration which is observed from **Fig. 1**. In **Fig. 1**, the inserted graphs shows the linear response of TL intensity for various concentrations of Er^{3+} and the peak intensity of the experimental glove curve is around 455 K (182° C).

The electron traps formed at different depth may be produce due to oxygen vacancy which could be decreases with concentration of doping ions and the lowering of trap density is the reason behind lowering the TL intensity with concentration of Er^{3+}. The kinetic parameters calculated from above mentioned method are given in **Table 1**. The analysis of the distribution of energy level distribution between forbidden gap is based on the assumption that the different energy levels are independent of each other. There is no retrapping or redistribution of the electrons takes place between the energy levels. This arbitrary assumption simplifies course of the data analysis. In general one should consider the effects of retrapping and redistribution of charge carriers between conduction band and metastable energy levels [**11**]. The dose response, dosimetry peak temperature, concentration dependency of phosphor, fading effect of TL intensity and reusability are the favorable characteristics of phosphor for radiation dosimetry which suggest it for TLD applications.

Table 1. "Activation energy", "frequency factor" and "order of kinetics" of the TL glow curve of sample $Sr_2P_2O_7$: 0.5%Er after deconvolution curve fitting technique.

Peak	Tm (K)	τ (K)	δ (K)	ω (K)	μ_g	Activation Energy 'E_a'			Frequency factor 's' (s^{-1})	Order of Kinetics 'b'
						E(τ) eV	E(ω) eV	E(δ) eV		
Peak I	400	24	25	49	0.51	0.89±0.03	0.91±0.04	0.90±0.03	1.5×10^{11}	2
Peak II	430	23	25	48	0.52	1.10±0.05	1.11±0.04	1.10±0.03	9.5×10^{12}	2
Peak III	464	27	25	52	0.49	1.05±0.03	1.05±0.03	1.04±0.04	1.1×10^{11}	≈2

Conclusion

This paper gives the description of TL investigations done to characterize dosimetric features of Er^{3+} doped $Sr_2P_2O_7$ phosphor. It is found that the TL sensitivity of the phosphor irradiated by beta radiation is very good for TLD. The β-dose response of intensity for peak at 451 K is linear for dose range 5 Gy to 50 Gy. The TL sensitivity of this phosphor is highly depends on the concentration of doping ion Er^{3+}. The comparatively low fading of TL intensity and multiple time reusability of phosphors gives its potential properties for TLD phosphor. Activation energy and the frequency factor calculated from the analysis are favorable for proposing this phosphor for dosimetric applications.

Acknowledgements

The authors wish thank to UGC-DAE, Consortium for Scientific Research, Indore for the experimental measurements and UGC-BSR for providing the research fellowship.

Author's contributions

Conceived the plan: Nimesh P. Patel, M. Srinivas; Performed the expeirments: Nimesh P. Patel, Vishwnath Verma, Dhaval Modi; Data analysis: Nimesh P. Patel, M. Srinivas; Wrote the paper: Nimesh P. Patel, M. Srinivas, K. V. R. Murthy.

Reference

1. McKeever S. W. S.; Thermoluminescence of solids, Edited by R. W. Cahn, E. A. Davis, I. M. Ward, (Cambridge University Press) **1985**, pp.206.
DOI: 10.1017/CBO9780511564994
2. Natarajan V.; Bhide M. K.; Dhobale A. R.; Godbole S. V.; Seshagiri T. K.; Page A. G.; Lu C. H.; *Mate. Res. Bull.*, **2004**, *39(13)*, 2065.
DOI: 10.1016/j.materresbull.2004.07.009
3. Yazici A. N.; Seyyidoglu S.; Toktamis H.; Yilmaz A.; *J. Luminescence.* **2010**, *130(10)*, 1744.
DOI: 10.1016/j.jlumin.2010.04.003
4. Patel N. P.; Srinivas M.; Modi d.; Verma V.; Murthy K. V. R.; *Luminescence*. **2015**, *30(4)*, 472.
DOI: 10.1002/bio.2762
5. Patel N. P.; Srinivas M.; Modi d.; Verma V.; Murthy K. V. R.; International Conference on Materials and Characterization Techniques. *Int. J. ChemTech Res. vol 6(3)*, 1708, **2014**.

DOI: http://sphinxsai.com/2014/vol6pt3/6/(1708-1711)ICMCT14.pdf
6. Som S,; Dutta S,; Kumar V,; Pandey A,; Kumar V,; Kunti A.K.,; Priya J.,; Sharma S. K.,; Terblans J. J.,; Swart H.C.; *J. Allo. and Comp.*, **2015**, *622*, 1068.
DOI: 10.1016/j.jallcom.2014.11.001
7. Afouxenidis D.; Polymeris G. S.; Tsirliganis N.C.; Kitis G.; *Rad. Prot. Dosimetry*, **2012**, *149(4)*, 363.
DOI: 10.1093/rpd/ncr315
8. Jin Y.; Hu Y.; Chen L.; Wang X.; Mou Z.; Ju G.; Liang F.; *Mate. Sci. & Eng. B*., **2013**, *178*, 1205.
DOI: 10.1016/j.mseb.2013.07.009
9. Chen R.; *J. Electrochem Soc.*, **1969**, *116(9)*, 1254.
DOI: 10.1149/1.2412291
10. Chithambo M. L.; *J. Phys. D: Appl. Phys.*, **2012**, *45*, 345301.
DOI: 10.1088/0022-3727/45/34/345301
11. Pagonis V.; Morthekai P.; Kitis G.; *Geochronometria*, **2014**, *41(2)*, 168.
DOI: 10.2478/s13386-013-0148-z

