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Thermoluminescence of High Sensitive Microcrystalline CaSO4:Dy for High Dose Measurements

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Abstract

*Thermoluminescent (TL) response in the high dose region (0.1 - 10 kGy), isothermal decay, pre-read annealing effect and glow curve analysis of the high sensitive microcrystalline (30-75 µm) $CaSO_4$:Dy phosphor powder developed in our laboratory by a modified acid evaporation route have been studied for the first time by substantially reducing the detector sensitivity. The parameters obtained agree well with those obtained earlier with the 75-210 µm sized $CaSO_4$:Dy phosphor powder made by the conventional re crystallization technique. The results obtained with both these bulk phosphors were explained based on the competition in charge carrier trapping during irradiation by continuous, but narrow distribution of dosimetry traps proposed earlier. The microcrystalline phosphor exhibited a simple TL glow curve whose response increased linearly with dose in the region 0.1 kGy - 7 kGy, beyond which saturation was observed. This was explained on the basis of two opposing effects namely supralinearity and trap saturation which cancel against each other leading to linear TL response. Neither complex glow curve shape nor variation in glow curve shape with dose was observed in the dose range 0.1 - 15 kGy in contrast to the claims made in the literature.***

Key words.
Micro and submicro $CaSO_4$:Dy crystals, high dose response, TL glow curve analysis, competing deep traps

1. INTRODUCTION

Irradiation with ionizing radiation kills harmful bacteria and make the food safer for consumption. Presently used chemical dosimeters [Fe^{2+} (Ferrous) to Fe^{3+} (Ferric ions)] in liquid state are not suitable for sub-ambient dosimetry. Response of electron spin resonance (ESR) based systems (alanine radicals) is reported to be sensitive to humidity and temperature and requires expensive instrumentation based on ESR from alanine **[1],** Thermoluminescent dosimeters (TLD) have been proved to be a reliable passive detectors for the measurement of low levels of radiation exposures encountered in personnel and environmental monitoring. However, measurement of high radiation doses (0.1 - 10 kGy) used for preservation of food has posed some issues since the response of common TLD phosphors saturate at about 1 kGy **[2].** Other issues posed by some phosphors such as LiF in medium and high dose region include post-irradiation fading, nonlinear TL response with dose and irradiation temperature dependence **[3].** Certain food products are preserved in ice and are irradiated in dry ice (-78.5^{0}C) and wet ice (0^0C) conditions **[4].** Therefore the response of dosimeters should not vary with irradiation temperature in this temperature range. $CaSO_4$:Dy TL phosphor is fading free, irradiation temperature independent in the range 77 K - 353 K **[5,6]** but exhibits complex glow curve behavior and super linear behavior below 0.1 kGy and its TL response saturates around 10 kGy **[2].**

Nano $CaSO_4$:Dy made by *(bottom-up approach)* has been reported to exhibit multi peak structure with major peaks at 138, 225, 296 and 379^{0}C which is unfavorable for applications in radiation dosimetry **[7,8].** To overcome the problem of unfavorable TL glow curve structure exhibited by nano $CaSO_4$:Dy, Rahangdale et al. **[9]** have recently made submicron $CaSO_4$:Dy phosphor *(top down approach)* for high dose dosimetry by grinding small quantity of higher sized grains using a high energy planetary ball mill working at 400 rpm with 2 mm diameter balls and grinding bowl of zirconium. $CaSO_4$: (0.1 mol% Dy) synthesized using the well known recrystallization method developed by Yamashita et al. **[10]** which mostly yield grains > 75 µm (called bulk phosphor) was used for this purpose. Both wet and dry grinding were carried out for long duration (12 to 24h). Due to the hammering or collision of zirconium ball on phosphor, all grains acquired less than 1 µm size with random shape. Depending on the grinding condition, the TL sensitivity of the ground phosphor to ^{60}Co γ-rays was 15 to 144 times lower than that of bulk phosphor. The major glow peak temperature of ball milled phosphor was slightly shifted towards the lower temperature (~ 220 to 210^{0}C), along with a slight enhancement in the intensities of low (~ 130 ^{0}C) as well as high temperature (~ 300 ^{0}C) TL peaks as compared to the bulk phosphor. The authors claim that there was no change in the TL glow curve shape of ball milled phosphor for the dose ranging from 0.1 to 15 kGy unlike that of bulk $CaSO_4$:Dy phosphor which shows complex glow curve nature at high doses. But they have not substantiated this claim with a plot of TL glow curves of ball milled phosphor nor bulk phosphor as a function of dose. Neither a comparison of TL characteristics of bulk and ball milled $CaSO_4$:Dy phosphor was made, since the TL response of bulk phosphor, due to its high sensitivity floods and hence will saturate the TLD reader. Moreover, these authors used peak height rather than the customary glow curve area measurements for dosimetry. Their study showed that the height of dosimetric peak at 220^0 C of submicron crystalline $CaSO_4$:Dy powder exhibit a linear behavior up to 6 kGy and then get saturated above 7 kGy dose. They further claim that bulk $CaSO_4$:Dy phosphor show dose linearity up to few Gy only. Thus they conclude that the *top down approach* can be useful as an alternative TL dosimeter in the field of food irradiation dosimetry. No plausible mechanism which purportedly causes the changes in the TL characteristics of bulk and submicron sized $CaSO_4$:Dy powder was offered. The present study cast doubts on the above claims and shows that the dose versus TL response (both peak height and area) of microcrystalline (30-75 µm) sized $CaSO_4$:Dy phosphor exhibit a linear response in the dose range 0.1 - 7 kGy and saturates beyond it similar to the response behavior of submicron sized phosphor reported. Thus except for the reduction in TL sensitivity on grinding, the response behavior of the microcrystalline (bulk) and submicron sized phosphor powders are similar and no change in the TL glow curve shape of bulk phosphor was observed in the high dose range studied (0.1 - 15 kGy) unlike the claim made in literature**.** Hence there appears to be no need to adopt the cumbersome energy consuming ball milling for high dose dosimetry. Existing mechanism based on trap competition during irradiation and trap saturation at high doses proposed for the bulk phosphor powder seems to be equally valid for submicron sized powder.

2. MATERIALS AND METHOD

In this work, detailed studies TL glow curve shape and TL response (both peak height and glow curve area) versus dose in the high dose region, pre-read annealing and isothermal decay characteristics were carried out on high sensitive microcrystalline (also known as bulk) $CaSO_4$:Dy (0.2 mol%) phosphor powder of grain size (30-75 µm) made by a modified acid evaporation technique developed by Lakshmanan et al. **[11-13]** after a pre-irradiation annealing treatment of 700^0C, 1h. In the new preparation, $CaCO_3$ was used instead of gypsum and the quantity of H_2SO_4 to be evaporated was insignificant when compared to that used by Yamashita et al. **[10].** In the new preparation, most of the phosphor grains in as grown condition are 30-75 µm in size and are needle shaped transparent crystals while in the old preparation most of the grains are > 75 µm in size and are transparent crystals of quadrilateral shape which on grinding become irregular opaque crystals due to surface deterioration. The TL sensitivity of the new phosphor used is more than 50% higher than that of the $CaSO_4$:Dy made by conventional recrystalline method. Further the contribution of low temperature peak is negligible**.**

Gamma irradiations were performed in the gamma chamber (dose rate 3.73 kGy.h^{-1}) at IGCAR, Kalpakkam. The dose range covered was 0.1 kGy - 14.93 kGy. Dose versus TL measurements were carried out after a post-irradiation interval of 5d using a Nucleonix PC based TL analyser system type TL1009I having an EMI photomultiplier tube 6095. About 10 mg (carefully weighed using a semi micro balance) of the phosphor powder was spread uniformly on the kanthal planchet for TL measurements so as to make good thermal contact. Dose-response studies were carried out at a linear heating rate of 8^0C/s. Glow curve area (120-400^0C) measurements were used for TL intensity (TL/mg) measurements in this study. Isothermal decay studies were carried out by spreading the irradiated powder (about 10 mg) sample evenly on the heater strip in the TLD reader at 180^0C.

Since the light emitted by the bulk $CaSO_4$:Dy TLD is quite high, two neutral density filters (which together attenuated the light by a factor of 175 as measured experimentally) were inserted in between the sample and the PMT. In addition the EHT voltage applied to the PMT was reduced from the normal value of 900 V to 400V which resulted in the reduction in light collection efficiency by a factor of 1320. Both these factors thus resulted in a net reduction in photo current by a factor of 0.23 million (1320x175 = 231000) which was found essential to prevent photo-current saturation due to intense TL emission from bulk $CaSO_4$:Dy at high doses.

3. RESULTS

3.1. TL glow curves of bulk $CaSO_4$:Dy at high doses

Fig.1 shows the TL glow curves of micro crystalline $CaSO_4$:Dy powder at different doses in the high dose region. They exhibit a single glow curve whose shape remains nearly constant in the dose range (0.1 kGy - 14.93 kGy) studied. There is however, a minor shift in glow peak temperature (T_m) from about 270 to 252^0C with increasing dose. Similar results namely single glow curve and shift in its glow peak to low temperature with increasing gamma dose in the above dose region has been reported earlier with the bulk $CaSO_4$:Dy (0.05 mol%) powder (75-210 µm) made by Yamashita's recrystallization technique **[10].** These two results are in disagreement with the claims made in the literature that bulk phosphor exhibits complex glow curves in the high dose region.

3.2. Dose versus TL response

Fig. 2 shows the glow curve area (130-400^{0}C) values as a function of gamma dose. The TL response increases nearly linearly with dose in the range 0.1-7 kGy beyond which saturation sets in. Similar results are seen with peak height measurements which is very similar to that reported for submicron (ball milled) $CaSO_4$:Dy powder by Rahangdale et al. **[9].** The present result is also in agreement with earlier dose-response characteristics obtained with Yasmashita's recrystalline $CaSO_4$:Dy (0.05 mol%) powder (75-210 µm) in this high dose range by Lakshmanan et al. **[2, 14-16].** Complex glow curve changes and supralinearity have been observed in those studies only below the dose level of 0.1 kGy.

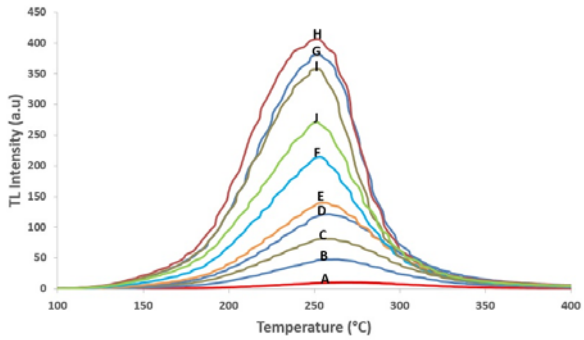

Fig.1. TL glow curves of microcrystalline (bulk) $CaSO_4$:Dy at different gamma dose levels (kGy). A- 0.31, B- 0.62, C- 1.37, D- 1.86, E- 2.80, F- 3.73, G- 7.46, H- 9.32, I- 11.20, J- 14.92 A single glow peak exhibiting a downward shift in T_m from about 270 to 252^{0}C with increasing dose is seen.

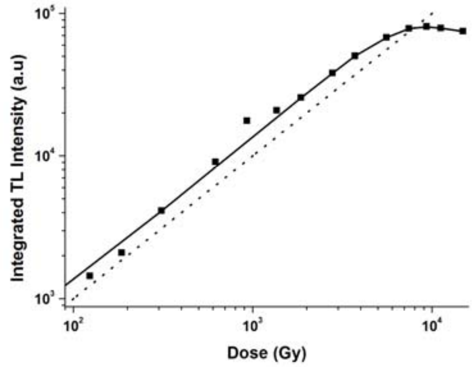

Fig.2. Integrated TL response (130-400^{0}C) of bulk $CaSO_4$:Dy as a function of gamma dose. Line of linearity (-------) is also shown

Thus the claim in literature that bulk $CaSO_4$:Dy shows complex glow curve nature at high doses and exhibits high temperature peaks (350^0 C) even at high dose unlike ball milled phosphor and hence bulk $CaSO_4$:Dy powder cannot be used for high dose dosimetry is untenable. Experimental results of this study and those in literature show that in the limited dose range 0.1- 7 kGy, bulk $CaSO_4$:Dy shows a simple glow curve structure which exhibits near linear response similar to ball milled phosphor. The reason for this peculiar behavior arises because of two competing but mutually opposing effects. One involves supralinear increase in TL response with dose beyond about 3 Gy which continues till saturation dose (~ 10 kGy) as a result of continued elimination of competing deep traps which increases sublinearly with dose in the high dose region **[2,14-16].** The other effect involves the onset of saturation of dosimetry traps at high doses which bring down the TL response with dose. As a result of these two opposing effects, the TL response shows a near linear response. A closer scrutiny of **Fig.2** showed a slowly increasing trend in TL response in the 10^2 -10^3 Gy range due to supralinearity and a slowly decreasing trend in the 10^3 -10^4 Gy which results in scattering of experimental values from the line of linearity. The scattering was found to be relatively more with peak height measurements.

3.3. Shift in T_m with pre-read annealing temperature

A low temperature shift in T_m with dose could occur either due to second order kinetics or due to saturation of high energy traps at higher doses. In first order kinetics involving a single set of traps such as $CaSO_4$:Mn, no such shift occur. To verify this aspect, irradiated bulk samples (0.31 kGy) were annealed for 5 min. at different temperatures in the

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TLD reader itself in the temperature range 150^0C - 210^0C and then the glow curves were recorded. The results of this study shown in **Fig.3** indicate a high temperature shift of glow peak maximum (T_m) from 264^0C - 297^0C accompanied with a decease in TL sensitivity with increase in annealing temperature. This is a clear indication that the continuous distribution of TL traps are active even in the high dose region. Similar shift in T_m has been observed earlier with recrsytallized bulk $CaSO_4$:Tm powder at the low dose level of 0.06 Gy **[17].** In contrast, the T_m in $CaSO_4$:Mn did not shift with increase in annealing temperature. Only a reduction in TL sensitivity was seen. This is a consequence of single trap level obeying first order kinetics in $CaSO_4$:Mn. In contrast $CaSO_4$:Dy exhibits continuous but narrow multi-Gaussian trap distribution with a peak trap depth (central energy), E at 1.38 eV and FWHM of 0.1 eV and frequency factor, s = $3x10^{12}$ s^{-1} which obey first order kinetics as elaborated by Oliveri et al. **[18].**

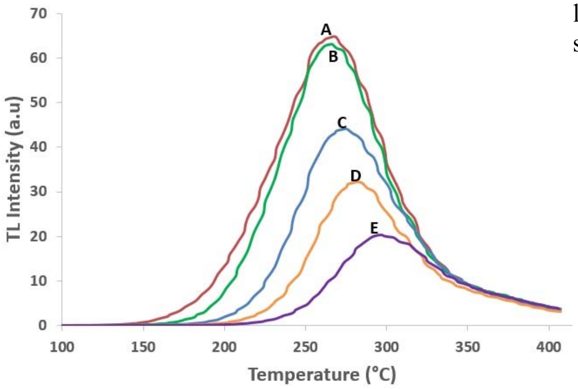

Fig.3. TL glow curves of bulk $CaSO_4$:Dy irradiated with a ^{60}Co γ-dose of 0.31 kGy and pre-read annealed at different temperatures for 5 min duration. A- No anneal, B- 150°C, C- 175°C, D-185°C, E- 210°C. The upward shift in T_m from about 252 to 270^0C coupled with a reduction in TL sensitivity on annealing has been explained on the basis of continuous but narrow multi-Gaussian trap distribution **[18,19].**

3.4. Isothermal decay

Isothermal decay curves of gamma irradiated bulk $CaSO_4$:Dy at 185^0C for various high doses is plotted in a semi-log graph and shown in **Fig.4.** A straight line in this graph would indicate a first-order kinetics from a single trap structure. But the decay curves in **Fig.4** deviate from exponential decay in conformity with continuous trap distribution. Similar deviation has been observed earlier with bulk $CaSO_4$:Tm made by Yamashita's re crystallization technique at the low dose level of 0.06 Gy **[17].**

Interestingly, **Fig.4** shows that the decay rate increases with dose in conformity with the observed low temperature shift in T_m with increase in dose as a result of saturation of high energy traps at higher doses. Sivastava and Supe **[19]** have proposed a continuous distribution traps but with decrease in their population with trap depth, as a result of which the deeper traps get saturated early with dose resulting in the observed low temperature shift in T_m with increase in dose.

3.5. Peak shape analysis

The analysis based on the shape of the TL glow peak shown in **Fig.5** employs three points on the glow curve, i.e., the maximum peak temperature, T_m and two temperatures on either side of T_m noted as T_1 and T_2 corresponding to half the peak intensity **[20]**. The following parameters are then defined, the full width at half maximum (FWHM), $\omega = T_2 - T_1$, the high temperature half-width, $\delta = T_2 - T_m$ and the low temperature half width, $\tau = T_m - T_1$. The symmetry factor $\mu_g = \delta/\omega$.

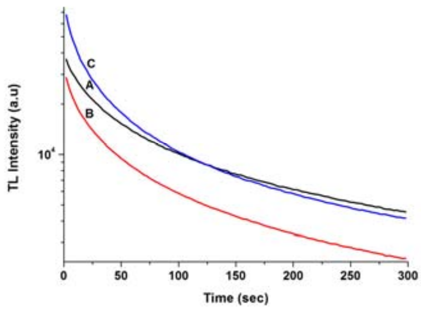

Fig 4. Isothermal Decay of bulk $CaSO_4$:Dy at 180^0C. A- 0.932 KGy, B- 2.795 KGy, C- 14.92 KGy

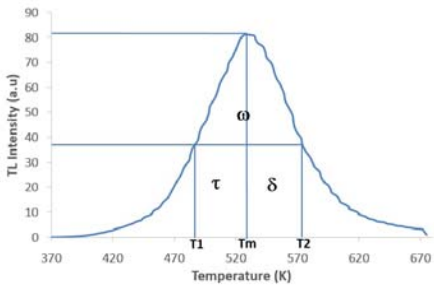

Fig.5. The geometrical shape of the TL glow peak at a dose of 1.37 kGy

Another factor defined in this regard is $\gamma = \delta/\tau$

[21]. The values of μ_g for first and second order kinetics are 0.42 and 0.52, respectively, provided the trap has single depth. Similarly, the parameter, γ ranges from 0.7 to 0.8 for first-order kinetics and from 1.05 to 1.20 for second order kinetics. However, present case deals with continuous trap distribution and hence the above simple technique cannot be used for determining order of kinetics or trap parameters E and s. However, it can be inferred that the reduction in μ_g and γ values with increasing dose seen in **Table 1** indicates a reduction in high temperature half-width, δ with dose as a result of saturation (or sublinear increase) of high energy traps at high doses.

Table 1. The experimentally observed glow curve parameters as a function of gamma dose.

Dose (KGy)	T1 K	Tm K	T2 K	$\mu g= \delta/\omega=$ (T2–Tm)/(T2–T1)	$\gamma= \delta/\tau=$ (T2–Tm)/(Tm–T1)
0.124	502	544	585	0.4939	0.9761
0.187	495	546	607	0.5000	1.0000
0.311	500	544	592	0.5217	1.0909
0.621	485	539	585	0.4600	0.8518
0.932	495	530	558	0.4444	0.8000
1.366	489	531	573	0.5000	1.0000
1.863	490	534	572	0.4634	0.8636
2.795	484	532	570	0.4418	0.7916
3.730	483	527	558	0.4133	0.7045
5.593	481	527	558	0.4025	0.6739
7.460	476	527	560	0.3928	0.6470
9.323	479	525	554	0.3866	0.6304
11.190	478	525	554	0.3815	0.6170
14.920	474	525	562	0.4204	0.7254

4. DISCUSSION

The experimental data obtained in this study show that: (i) similar to Yamashita's **[10]** recrystallized bulk (75- 210 μm) $CaSO_4$:Dy phosphor powder, the micro crystalline (30-75 μm) $CaSO_4$:Dy made by Lakshmanan et al. **[14-16]** does not show complex glow curve shape nor variation in glow curve shape with dose in the dose range 0.1 - 15 kGy in contrast to the claims made in literature **[9]**. Literature data on $CaSO_4$:Dy made by recrystallization **[10]** shows such changes only in the dose range below 0.1 kGy **[2,15],** ii) the glow curve area of the microcrystalline phosphor shows near linearity in the dose range from 0.1 kGy up to about 7 kGy before saturation sets in, similar to the results obtained with submicron sized phosphor by Rahangdale et al. **[9]** when measured with sufficient light attenuation (by a factor of 0.23 million) using neutral density filters and a drastic reduction in applied voltage to the photomultiplier in the TLD reader. Thus there appears to be no need to adopt the cumbersome energy consuming ball milling for the application of $CaSO_4$:Dy TLD for high dose dosimetry, iii) Furthermore, the peak height measurements is not acceptable for dosimetric applications for various reasons. Only glow curve area should be used. The bulk phosphor exhibits a single glow peak in the high dose region studied. In contrast, ball milled phosphor shows enhanced contribution of low and high temperature TL peaks. The contribution of these two unwanted peaks to the total TL intensity could vary with gamma dose which make one wonder if this is the reason why these authors adopted peak height rather than area measurements for dose-response plot.

The high temperature shift of T_m accompanied with a decrease in TL sensitivity in the newly developed high sensitive bulk $CaSO_4$:Dy with increase in annealing temperature is a clear indication that the continuous distribution of TL traps are active even in the high dose region. Similar shift in T_m has been observed earlier with re crystallized bulk $CaSO_4$:Tm at the low dose level of 0.06 Gy**.** Isothermal decay studies at 185^0C after various high dose irradiation with bulk phosphor confirm a non exponential decay of TL with time in conformity with continuous trap distribution. An analysis of glow curve shape shows a reduction in high temperature half-width, δ with dose as a result of saturation (or sublinear increase) of high energy traps at high doses which is consistent with the competing deep trap model proposed earlier **[14-16]**

A drawback of the study by Rahangdale et al. **[9]** which led to their erroneous conclusion on the differences between bulk and submicron sized phosphor powder is that they did not study or analyze in detail literature data **[11,12]** on the TL characteristics of bulk powder in the high dose region. The earlier data covering a wide dose range of 0.01 Gy - 100 kGy showed supralinear TL response accompanied with drastic changes in TL glow curve shape in the dose region 1 Gy - 0.1 kGy as a result of the influence of deep traps competing during gamma irradiation in the trapping process of charge carriers with dosimetry traps. No change in the glow curve shape was reported above 0.1 kGy. In the dose range 0.1 KGy - 10 kGy, two competing but opposing effects lead to the observed linear TL response as explained earlier. An increase in saturation dose, by the incorporation of high Dy concentration in $CaSO_4$ host would perhaps increase the linear dose rage further **[22].**

A closer scrutiny of **Fig.2** shows a slowly increasing trend in TL response in the 10^2 -10^3 Gy dose range due to supralinearity and a slowly decreasing trend in the 10^3 -10^4 Gy dose range due to saturation which results in scattering of experimental values from the line of linearity. This is an inherent drawback of this technique which will limit its accuracy in high dose dosimetry. In addition, the TL response characteristics of $CaSO_4$:Dy, such as supralinearity, high temperature TL response versus dose etc are reported to vary from batch to batch. This means each batch of $CaSO_4$:Dy has to be carefully calibrated before it is chosen for its application for high dose dosimetry.

The TL sensitivity of submicron sized $CaSO_4$:Dy is reported to be less by a maximum factor of 145 as compared to bulk phosphor **[9].** It is therefore easily possible to record its TL glow curves and test the TL versus dose response of this material in the dose region 0.1-30 kGy, where TL glow curve shape change with dose and TL supralinearity with dose have been reported. Such a study would have revealed if there are any basic differences between the characteristics of submicron sized and bulk phosphors apart from the reduction in sensitivity as claimed. In the absence of such a study, the claim that the dose response characteristics of submicron sized $CaSO_4$:Dy are different from those of bulk $CaSO_4$:Dy is not acceptable.

5. CONCLUSION

The TL response characteristics of the high sensitive microcrystalline (also called bulk) $CaSO_4$:Dy developed by the authors' group has been investigated in detail in the high dose region 0.1 - 10 kGy and compared with those reported recently for the submicron sized $CaSO_4$:Dy obtained after extensive ball milling the bulk phosphor.

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References:

[1] Techniques for high dose dosimetry in industry, agriculture and medicine. Proceedings of a symposium held in Vienna, 2-5 November 1998, IAEA-TECDOC-1070.
[2] A.R. Lakshmanan, B. Chandra and R.C. Bhatt. *Nucl. Instrum. Methods* 153, 581 (1978)
[3] A. Kovacs, M. Baranyai ,L. Wojnarovits, S. Miller, M. Murphy, W.L. McLaughlin, I. Slezsak , A.I. Kovacs. *Radiation Physics and Chemistry* 63 (2002) 777–780.
[4] B. Sanyal, V. Natarajan; S.P. Chawla; A. Sharma**.** *Radiation Meas.* 45, 899–905, 2010.
[5] J.K. Lu, C.J. Tung and P.C. Hsu. *Rad. Eff. Lett.* 76, 215 (1983)
[6] T.C. Chen, C.L. Tseng and P.S. Weng. *Rad. Eff. Lett.* 85, 45 (1984)
[7] N. Salah, P.D. Sahare, S.P. Lochab and P. Kumar. *Rad. Meas.* 41(1) Jan 2006, 40-47
[8] N. Salah, S.S. Habib, Z.H. Khan and S.P. Lochab. Intl. Symp. On the Peaceful Applications of Nuclear Technology.in Jeddah, Session 12/NP.2, 2008
[9] S. R. Rahangdale, S. P. Wankhede, Sonal Kadam , B. S. Dhabekar, U. A. Palikundwar and S. V. Moharil. *Int. Jl. of Lum. and Applications*. Vol. l5(4) Dec., 2015, pp 486-488
[10] T. Yamashita, N. Nada, H. Onishi and S. Kitamura. Calcium Sulfate. *Health Phys.* 21, 295-300 (1971)
[11] A.R. Lakshmanan. *phys. stat. soli. (a)* 186, 153 (2001)
[12] A R Lakshmanan, M T Jose, V Ponnusamy and P R Vivek kumar. *J. Phys. D: Appl. Phys.* 35, 386 (2002)
[13] A.R. Lakshmanan and A. Tomita. *Radiat. Prot. Dosim.* 100, 1-4, 235 (2002)
[14] A.R. Lakshmanan and R.C. Bhatt. *Nucl. Instrum. Meth.* 164, 215 (1979)
[15] A.R. Lakshmanan, R.C. Bhatt and S.J. Supe. *J. Phys.D: Appl. Phys. 14*, 1683 (1981)
[16] A.R. Lakshmanan. *J. Phys.D: Appl. Phys.* 15, L189 (1982)
[17] T. Yamashita and M. Takenaga, *Japan. J. Appl. Phys.* 16, 529, 1977
[18] E. Oliveri, O. Fiorella and M. Mangia. *Nucl. Instrum. Meth.* 154, 203, 1978
[19] J.K Srivastava and S.J. Supe. *J. Phys.D: Appl. Phys.* 13, 2337 (1980)
[20] R. Chen. *J. Appl. Phys.* 49, 570 (1969)
[21] M. Balarin. *Phys. Stat. Sol.(a)* 31, K111 (1975)
[22] Bhuwan Chandra and R.C. Bhatt. *Nucl. Instrum Meth.* 164, 571 (1979)