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Research Article

$NaLi_2PO_4$: 0.5 mole% Eu^{3+} Phosphor Used for Carbon Dosimetry

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Abstract. New $NaLi_2PO_4$, phosphor doped with 0.5 mole% Eu^{3+} prepared by solid state method were used for carbon dosimetry. XRD of the material matched with the data available in the literature (JCPDS file #80-2110) and confirmed formation of the material in its orthorhombic structure. The TL of the carbon beam irradiated sample recorded on Harshaw TLD-3500 reader. The samples are irradiated for different fluencies from the range of 5×10^9 to 1×10^{13} ions cm^{-2}. TL glow curve consist of single main TL peak around 457 K surrounded by kinks at 408, 504 and 542 K. TL glow curve of the carbon irradiated sample has same shape as that of gamma irradiated one. With increasing dose the intensity is increases. The phosphor is much more sensitive than the standard available standard phosphor. It is about 6.5 and 41.3 times more sensitive than the TLD-100 and TLD-700H. For these range of fluences phosphor show sub-linearity. Efficiency of the for phosphor for C^{6+} with respect to gamma radiation is calculated and is around 0.50754 for the fluence of 5×10^{09} ions cm^{-2}.

Keywords. $NaLi_2PO_4$:0.5 mole %Eu^{3+}, XRD, TL study, Dose response, LET, Supralinearity function and Efficiency

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1. Introduction

Traditionally surgery is performed to remove the tumour. In addition to it chemotherapy, radiotherapy are also used for the treatment. From the last two decades there is rapid improvement not only in photon radiotherapy but also particle therapy with proton, helium ions, carbon ions and other heavy ions. Ions heavier than helium ion are referred as swift heavy ions. Radiotherapy with heavy ions has unique dose deposition profile. Dose rapidly increases up to a maximum depth (called Bragg Peak) after which it decline sharply. At the entrance the dose deposition is very weak due to which its application is more effective than the conventional photon [7].

$NaLi_2PO_4$:Eu^{3+}(0.5mole%), a new highly sensitive low Z TLD phosphor meet many of the properties of a good TLD phosphor. The work on this phosphor using γ ray irradiation has already been reported by Sahare *et al*. [2, 6]. Recently, carbon ion beam is prominently used in radiotherapy [5]. In this paper, we investigated this phosphor for carbon dosimetry for its application in radiotherapy and in other related fields. These are preliminary results and further Studies are in progress.

2. Experimental

2.1 Method of Preparation

The material was prepared by a simple solid state diffusion method using analytical reagent (AR) grade $LiOH.H_2O$ and $NaH_2PO_4.2H_2O$ (CDH India, 99% purity) as starting materials and $EuCl_3$ (Alpha Aesar, 99.99% purity) as a source of impurity. The materials were used as received without further purification. All the ingredients were taken according to the following chemical formula, mixed using agate mortar and pestle and fired in a resistive furnace at 400°C and then at 800 °C for 12 h, respectively. Following is the reaction:

$$NaH_2PO_4.2H_2O + 2LiOH + 2EuCl_3 \rightarrow NaLi_2PO_4 : Eu(2mol\%) + 3H_2O\uparrow + Cl_2\uparrow .$$

The product was crushed to fine powder sieved and collected the particle size between 50 - 150 μm. The obtained powder was further annealed at 700 °C for one hour its pellets of diameter ~ 7.0 mm and thickness 0.5 mm were prepared by using a die in pelletizer machine [2].

2.2 Characterization

XRD patterns were recorded on a high-resolution D8 Discover Bruker X-ray diffractometer using a monochromatic Cu-K_α line obtained through a pair of Göbel mirrors with a scan rate of 1.0 s/step and step size of 0.02 s. The intensity (number of counts) of the diffracted lines was recorded using a point detector (scintillation counter) [1, 2].

The samples in the form of pellets were irradiated by 80 MeV C^{6+} ion beam for different ion fluences in the range 5×10^9 to 1×10^{13} ions/cm^2, using a Van de Graff type electrostatic 16 MV Tandem Accelerator facility (15 UD Pelletron) at the Inter University Accelerator Centre (IUAC), New Delhi, India [3]. The samples were mounted on a copper target ladder with silver paste giving good thermal and electrical conductivity between them. This prevents sample heating during SHI irradiation. The samples were also irradiated to γ rays from ^{60}Co source for comparison. For taking TL the irradiated surface of the pellet was kept facing upwards towards the detector (PMT) of the TLD reader. TL glow curves were recorded using a Harshaw TLD reader (Model 3500).The heating rate was kept at 5 °C/s.

3. Result and Discussion

3.1 XRD Analysis

XRD of the prepared $NaLi_2PO_4:Eu^{3+}$ phosphor is shown in Figure 1. It could be seen from the figure the experimental data is matched very well with the standard data available in JCPDS # 80-2110. No extra peak due to impurity and/or precursor compound(s) is seen in the figure which conform the formation of the phosphor material. This material has orthorhombic crystal structure belonging to space group Pmnb(62) [1, 2].

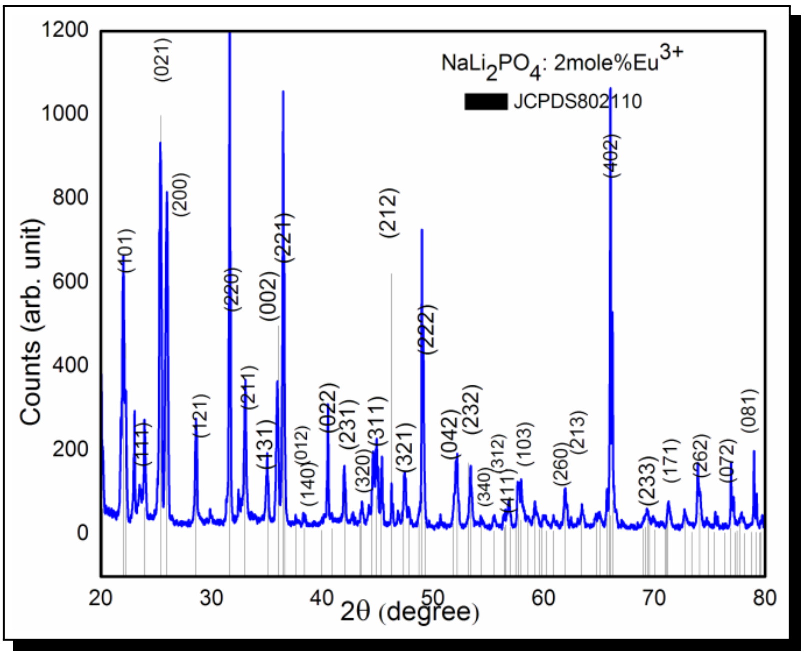

Figure 1. XRD of $NaLi_2PO_4$:0.5 mole% Eu^{3+} matched with JCPDS #802110

3.2 TL Glow Curves

TL glow curve of $NaLi_2PO_4:Eu^{3+}$ irradiated with ϒ irradiation from Co^{60} source for 1 kGy and C^{6+} ion beams is shown in Figure 2. It is clearly seen from the graph that no extra peaks were created by C^{6+} ion beam. But the computerized glow curve deconvolution (CGCD) and thermal cleaning (T_m-T_{stop}) confirm that a kink is formed around 542 K which is not seen for ϒ irradiation [6]. TL glow curve of C^{6+} consist of maximum peak around 453 K surrounded by kinks at 408, 504 and 542 K.

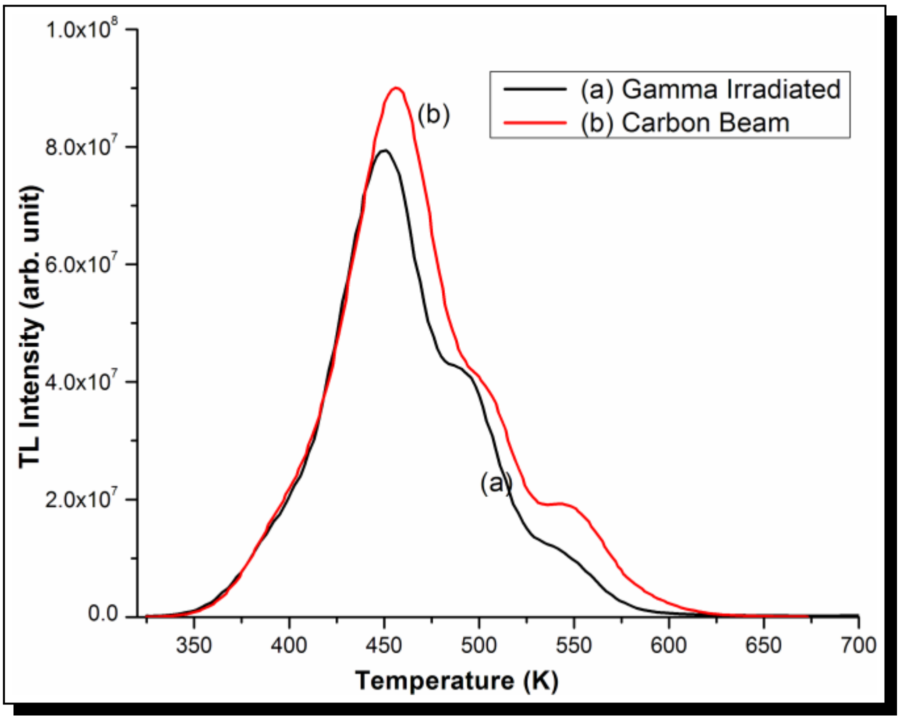

Figure 2. TL of $NaLi_2PO_4$:Eu^{3+} (a) irradiated with 1 kGy of gamma radiation, (b) irradiated with C^{6+} ion beam of fluence 1×10^{12} ions cm^{-2}

Phosphor was irradiated for different fluencies 5×10^{09} to 1×10^{13} ions cm^{-2} of C^{6+} and TL was recorded on Harshaw TLD reader. TL glow curve shown in Figure 3, it is seen that with increase in fluence the TL intensity increases. The broadness and area under the curve of the glow curve also increases with the increasing fluence with keeping the peaks position at the same points. Figure 4 show the CGCD graph of C^{6+} irradiated sample. CGCD and thermal cleaning show that there are four peaks. The main peak is around 453 K and other small peaks are around 408, 504 and 542 K. Singh *et al*. [6] showed that the same phosphor CGCD has only three peaks for γ irradiation and peaks are around 400, 458 and 500 K, respectively. Comparison of the trapping parameters of the glow curves irradiated by γ rays and C^{6+} the ion beam (Table 1), also indicate the generation of new traps [6]. Now, to understand whether the new peak could be attributed to incorporation of carbon ions or due to some other reasons we prepared another sample $NaLi_2PO_4$ doped with both Eu and C following the same procedure. TL recorded for samples irradiated with gamma radiation showed that the extra peak is due to the carbon ion implantation in the phosphor. From the figure it is clear that the extra peak is due to the carbon ion implanted in the phosphor.

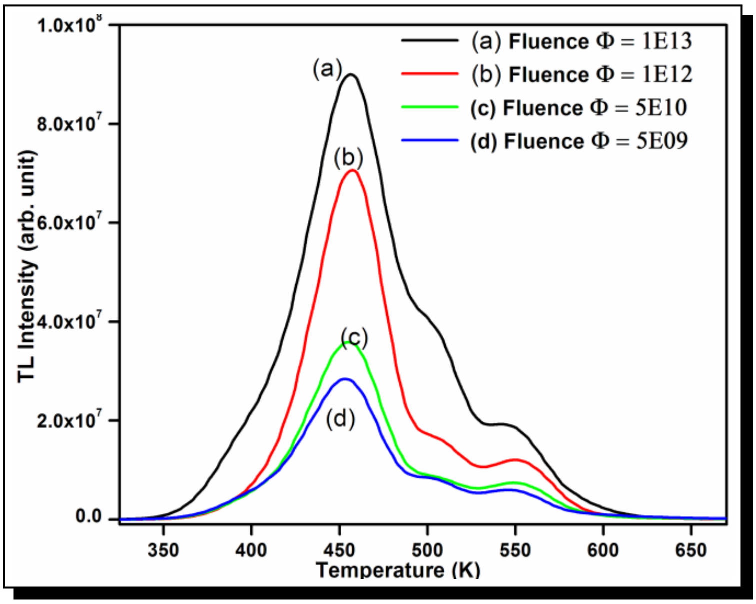

Figure 3. TL Glow curve with varying fluences: (a) 1×10^{13} ions/cm^2, (b) 1×10^{12} ions/cm^2, (c) 5×10^{10} ions/cm^2 and (d) 5×10^9 ions/cm^2

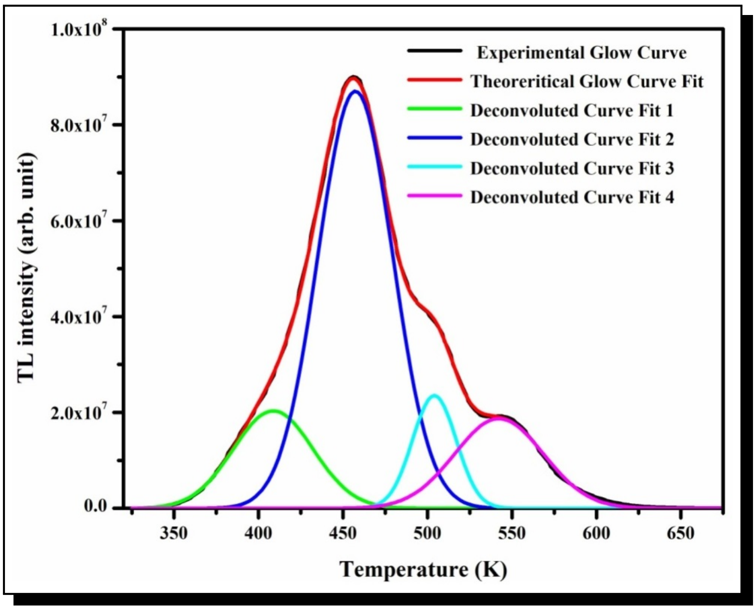

Figure 4. Deconvolution curve of $NaLi_2PO_4$:Eu^{3+} (c, d, e, f) with experimental curve (a) and theoretical curve, (b). The phosphor is irradiated with 1×10^{12} ions/cm^2C^{6+}

Table 1. Trapping parameters of $NaLi_2PO_4$:Eu irradiated with 80 MeV carbon ion beams

Peak	For γ rays			For carbon ion beam		
	Peak Temp (K)	E (eV)	s (s^{-1})	Peak Temp (K)	E (eV)	s (s^{-1})
Peak 1	425	0.6	2.04×10^6	408	0.79417	1.56996E9
Peak 2	459	1.26	5.72×10^{13}	453	1.11683	5.5071E11
Peak 3	503	1.78	2.25×10^{10}	504	2.11493	5.43036E20
Peak 4	—	—	—	542	1.22204	4.45232E10

4. Dose Response

The intensity and area under curve verses fluence are given in Figure 5. Both the curves are exponentially increased with dose. Absorbed dose from the ion fluence is calculated using eq. (6.1).

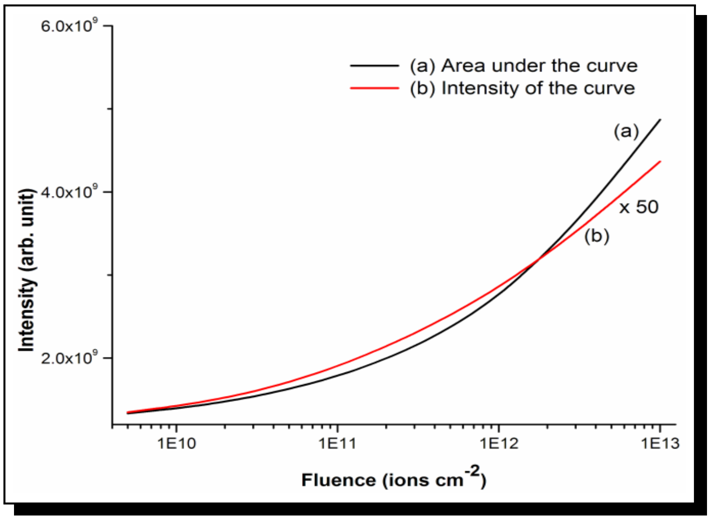

Figure 5. TL fluence response of $NaLi_2PO_4$:Eu^{3+} irradiated by C^{6+} ion beam using (a) area under the curve, (b) Intensity of peak method

5. Comparison with Standard Phosphors

Figure 6 shows the comparison of TL sensitivity of the $NaLi_2PO_4$:Eu^{3+} with that of TLD-100 and TLD-700H irradiated with carbon ion beam of fluence 1×10^{13} ions/cm^2. Sensitivity of $NaLi_2PO_4$:Eu^{3+} is much more (6.5 and 41.3 times, respectively) than the standard phosphors.

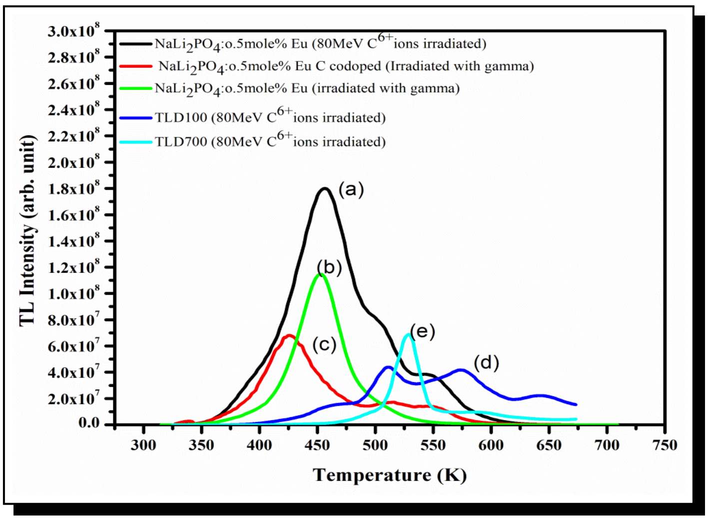

Figure 6. TL glow curve of (a) $NaLi_2PO_4$:Eu^{3+} irradiated with C^{6+} ion of fluence 1×10^{12} ions/cm^2, (b) $NaLi_2PO_4$:Eu C irradiated with gamma, (c) $NaLi_2PO_4$:Eu irradiated with gamma, (d) TLD-100 irradiated with 80 MeV C^{6+} ions of fluence 1×10^{12} ions/cm^2, (e) TLD-700 irradiated with 80 MeV C^{6+} ions of fluence 1×10^{12} ions/cm^2

6. LET and Penetration Depth by SRIM

SRIM code was used to calculate the *linear energy transfer* (LET) dose and the penetration depth. The absorbed *dose* (D) due to ion beam was calculated using equation:

$$D = 1.602 \times 10^{-10} \times \frac{1}{\rho}\left(\frac{dE}{dx}\right) \times n\,, \tag{6.1}$$

where ρ is the density in unit of gcm^{-3} of the phosphor, $\left(\frac{dE}{dx}\right)$ is LET and n is the fluence of the beam. Dose values were given in Table 3 [3, 4].

Penetration depth of the 80 MeV C^{6+} ion beam found to be 232.63 μm. The LET (electronic and nuclear), also range of ion are given in Table 2. Doses (for different fluences calculated using the same code) are given in Table 3.

Table 2. Calculation of energy loss and penetration depth of 80 MeV C^{6+} ion beam using SRIM

Sample	Density (ρ) (gcm^{-3})	$\left(\frac{dE}{d}\right)_e$ (MeV mm^{-1})	$\left(\frac{dE}{dx}\right)_n$ (MeV mm^{-1})	Range (μm)
$NaLi_2PO_4$:Eu	2.58	1.815	9.22×10^{-4}	232.63

Table 3. Calculation of absorbed doses by the sample when exposed to different fluences of 80 MeV C^{6+} ion beam

Fluence (η) ions/cm^2	Dose (D) Gy
5×10^9	3.17
5×10^{10}	31.74
1×10^{12}	6.35×10^2
1×10^{13}	10^3

7. Supralinearity Function

The supralinearity function, $f(n)$ is also plotted with the fluence and is shown in Figure 7. The function is defined as:

$$f(n) = \frac{\frac{F(n)}{n}}{\frac{F(n^*)}{n^*}}\,, \tag{7.1}$$

where $F(n)$ is the TL signal intensity (peak height) for the fluence n and $F(n^*)$ is the TL signal intensity for the very small fluence n^* on the linear region. From the graph it is seen that $f(n) < 1$, so it has sublinearity for this range of fluence/dose [3, 4].

8. Efficiency

Efficiency of the phosphor for C^{6+}ion radiations with respect to the gamma radiation is found out with the help of equation [3]:

$$\eta = \frac{\frac{F(D)}{D}}{\frac{F(D^*)}{D^*}}\,. \tag{8.1}$$

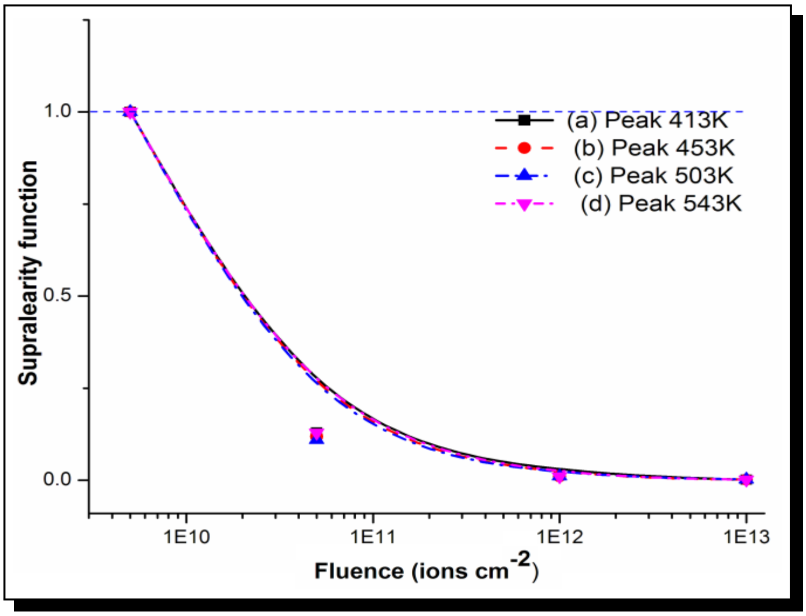

Figure 7. Supralinearity function for the (a) peak 413 K, (b) peak 453 K, (c) peak 503 K, (d) peak 453 K of $NaLi_2PO_4$:Eu irradiated by C^{6+} for the fluence of 1×10^{12} ions/cm^2

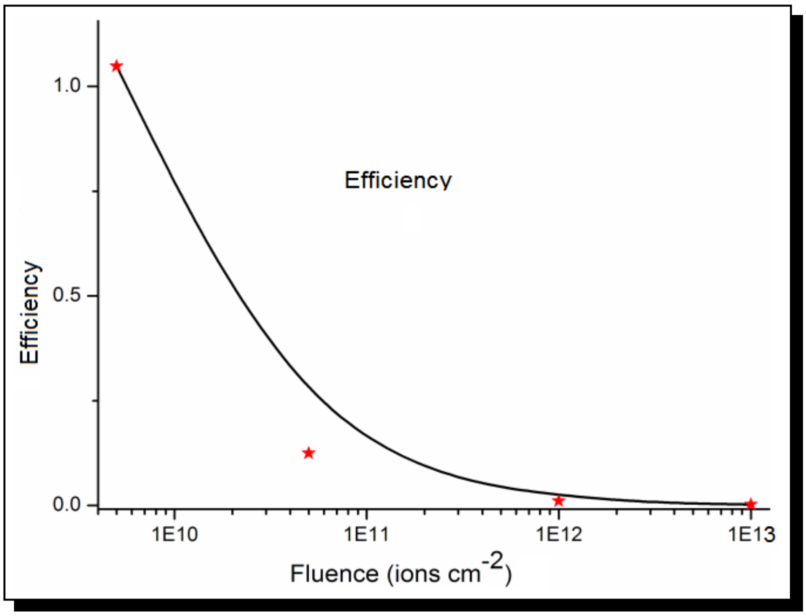

Figure 8. Efficiency of $NaLi_2PO_4$:Eu for C^{6+} with respect to Gamma radiation

Here, $F(D)$ is the TL intensity for dose of D which is absorbed from the fluence of ions calculated using formula given in eq. (6.1) and $F(D^*)$ is the TL intensity for the dose of D^* absorbed from the gamma source. Figure 8 shows that the efficiency declines exponentially with increase in fluence/dose. The efficiency for the fluence of 5×10^{09} ions cm^{-2} is about 0.50754.

9. Conclusion

$NaLi_2PO_4$:0.5 mole %Eu^{3+} is the new phosphor in the field of TLD and OSLD. It has very simple way of preparation and the reactant are very cost effective. For C^{6+} heavy ion beam TL glow curve has main one peak flanked by kinks. Single peaked glow curve is sign of good TLD phosphor. The main point is that the peak position is fixed for all fluences of ions. TL intensity is increase with increasing dose. This phosphor is highly sensitive than the available standard phosphor i.e., 6.5 and 41.3 more sensitive than TLD 100 and TLD 700 H. From TRIM/SRIM calculation the penetration depth is found to be 232.63 μm. Supralinearity function show that it has sub-linearity for the given range of fluence. Efficiency of the phosphor for the C^{6+} is found to be 0.50754 for the fluence of 5×10^{09} ions cm^{-2}. With increase in fluence, the efficiency also decreases.

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Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

All the authors contributed significantly in writing this article. The authors read and approved the final manuscript.

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