

Spectral, Transmittance and Thermal Analysis of Tb³⁺ doped Ytterbium Zinc Lithium Sodalime Cadmium Borogerminate Glasses

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Abstract

Glass sample of Yttrium Zinc Lithium Sodalime Cadmium Borogerminate Glasses (25-x) GeO₂:10ZnO:10Li₂O:10Na₂O:10CaO:10CdO:10Y₂O₃:15B₂O₃:xTb₂O₃ (where x=1,1.5,2 mol%) have been prepared by melt-quenching technique. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. DTA curve was analysed to evaluate the glass transition temperature, crystallization temperature and melting temperature. The absorption and fluorescence spectra of three Tb³⁺ doped yttrium zinc lithium sodalime cadmium borogerminate glasses have been recorded at room temperature. The various interaction parameters like Slater-Condon parameter F₂, Lande' parameter (ζ_{4f}), nephelauxetic ratio (β') and bonding parameter ($b^{1/2}$) have been computed. Judd-Ofelt intensity parameters and laser parameters have also been calculated.

Keywords: YZLSLCBG glasses, Thermal Properties, Optical properties, Transmittance analysis.

I. Introduction

Rare earth doped glasses have attracted much attention, because they have potential applications such as optical amplifiers, waveguide laser, optoelectronic devices, optical waveguide and solid state ionic devices [1-5]. Among many other glasses the germinate glasses find wide spread applications in laser technologies, optical data transmission, laser action and thermionic emission. Germinate glasses doped with rare-earth ions are considered as valuable materials for physical, optical and electrical applications. The high density in germinate glasses is due to high solubility of rare earth ions in germinate network. Glasses contain B₂O₃ have received increased attention due to their application in the field of glass ceramics, thermal sensors, reflecting windows, [6-10]. The addition zinc oxide improves the glass forming ability and to ensure low rates of crystallization in the glass system. Lithium oxide is added in the glass matrix to increase thermal stability and chemical durability of the glasses. Among active rare earth ions Tb³⁺ exhibits high solubility in ceramic glasses, which also possess excellent physical, optical, thermal and luminescent properties [11-15]. Recently germinate based glasses have a wide range of potential applications in laser technology, optical data transmission, sensing, energy solar cell and NIR lasers applications [16-18].

The aim of the present study is to prepare the Tb³⁺ doped yttrium zinc lithium sodalime cadmium borogerminate glass with different Tb₂O₃ concentrations. The absorption, fluorescence and transmittance spectra of Tb³⁺ of the glasses were investigated. The Judd-Ofelt theory has been applied to compute the intensity parameters Ω_{λ} ($\lambda=2, 4, 6$). These intensity parameter have been used to evaluate optical properties such as spontaneous emission probability, branching ratio, radiative life time and stimulated emission cross section. DTA curve was analysed to evaluate the glass transition temperature, crystallization temperature and melting temperature.

II. Experimental Techniques

Preparation of glasses

The following Tb³⁺ doped borogerminate glass samples (25-x) GeO₂:10ZnO:10Li₂O:10Na₂O:10CaO:10CdO:10Y₂O₃:15B₂O₃:xTb₂O₃ (where x=1,1.5, 2) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of GeO₂, ZnO, Li₂O, Na₂O, CaO, CdO, Y₂O₃, B₂O₃ and Tb₂O₃. They were thoroughly mixed by using an agate pestle mortar. then melted at 980°C by an electrical muffle furnace for 2h., After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250°C for 2h to

remove thermal strains and stresses. Every time fine powder of cerium oxide was used for polishing the samples. The glass samples so prepared were of good optical quality and were transparent. The chemical compositions of the glasses with the name of samples are summarized in Table 1.

Table 1

Chemical composition of the glasses

Sample	Glass composition (mol %)
YZLSLCBG (UD)	25 GeO ₂ :10ZnO:10Li ₂ O:10Na ₂ O:10CaO:10CdO:10Y ₂ O ₃ :15B ₂ O ₃
YZLSLCBG TB (1.0)	24 GeO ₂ :10ZnO:10Li ₂ O:10Na ₂ O:10CaO:10CdO:10Y ₂ O ₃ :15B ₂ O ₃ : 1 Tb ₂ O ₃
YZLSLCBG TB (1.5)	23.5 GeO ₂ :10ZnO:10Li ₂ O:10Na ₂ O:10CaO:10CdO:10Y ₂ O ₃ :15B ₂ O ₃ : 1.5 Tb ₂ O ₃
YZLSLCBG TB (2.0)	23 GeO ₂ :10ZnO:10Li ₂ O:10Na ₂ O:10CaO:10CdO:10Y ₂ O ₃ :15B ₂ O ₃ : 2 Tb ₂ O ₃
YZLSLCBG (UD)	-Represents undoped Yttrium Zinc Lithium Sodalime cadmium Borogerminate glass specimen.
YZLSLCBG (TB)	-Represents Tb ³⁺ doped Yttrium Zinc Lithium Sodalime cadmium Borogerminate glass specimens.

III. Theory

3.1 Oscillator Strength

The spectral intensity is expressed in terms of oscillator strengths using the relation [19].

$$f_{\text{expt.}} = 4.318 \times 10^{-9} \int \epsilon(\nu) d\nu \quad (1)$$

Where, $\epsilon(\nu)$ is molar absorption coefficient at a given energy ν (cm⁻¹), to be evaluated from Beer–Lambert law.

Under Gaussian Approximation, using Beer–Lambert law, the observed oscillator strengths of the absorption bands have been experimentally calculated [20], using the modified relation:

$$P_m = 4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_0}{I} \times \Delta\nu_{1/2} \quad (2)$$

Where c is the molar concentration of the absorbing ion per unit volume, l is the optical path length, $\log I_0/I$ is optical density and $\Delta\nu_{1/2}$ is half band width.

3.2. Judd-Ofelt Intensity Parameters

According to Judd [21] and Ofelt [22] theory, independently derived expression for the oscillator strength of the induced forced electric dipole transitions between an initial J manifold $|4f^N(S, L) J\rangle$ level and the terminal J' manifold $|4f^N(S', L') J'\rangle$ is given by:

$$\frac{8\pi^2 m c \bar{\nu}}{3h(2J+1)n} \frac{1}{n} \left[\frac{(n^2+2)^2}{9} \right] \times S(J, J') \quad (3)$$

Where, the line strength $S(J, J')$ is given by the equation

$$S(J, J') = e^2 \sum_{\lambda=2, 4, 6} \Omega_{\lambda} \langle 4f^N(S, L) J || U^{(\lambda)} || 4f^N(S', L') J' \rangle^2$$

In the above equation m is the mass of an electron, c is the velocity of light, $\bar{\nu}$ is the wave number of the transition, h is Planck's constant, n is the refractive index, J and J' are the total angular momentum of the initial and final level respectively, Ω_{λ} ($\lambda = 2, 4, 6$) are known as Judd-Ofelt intensity parameters.

3.3 Radiative Properties

The Ω_{λ} parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability (A) and radiative life time (τ_R), and laser parameters like fluorescence branching ratio (β_R) and stimulated emission cross section (σ_p).

The spontaneous emission probability from initial manifold $|4f^N(S', L') J'\rangle$ to a final manifold $|4f^N(S, L) J\rangle$ is given by:

$$A [(S', L') J'; (S, L) J] = \frac{64 \pi^2 \nu^3}{3h(2J'+1)} \left[\frac{n(n^2+2)^2}{9} \right] \times S(J', \bar{J}) \quad (4)$$

Where, $S (J', J) = e^2 [\Omega_2 \| U^{(2)} \|^2 + \Omega_4 \| U^{(4)} \|^2 + \Omega_6 \| U^{(6)} \|^2]$

The fluorescence branching ratio for the transitions originating from a specific initial manifold $|4f^N (S', L') J\rangle$ to a final many fold $|4f^N (S, L) J\rangle$ is given by

$$\beta[(S', L') J'; (S, L) J] = \sum_{S L J} \frac{A[(S' L)]}{A[(S' L') J'(\bar{S} L)]} \quad (5)$$

Where, the sum is over all terminal manifolds.

The radiative life time is given by

$$\tau_{rad} = \sum_{S L J} A[(S', L') J'; (S, L) J] = A_{Total}^{-1} \quad (6)$$

Where, the sum is over all possible terminal manifolds. The stimulated emission cross-section for a transition from an initial manifold $|4f^N (S', L') J\rangle$ to a final manifold $|4f^N (S, L) J\rangle$ is expressed as

$$\sigma_p(\lambda_p) = \left[\frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}} \right] \times A[(S', L') J'; (\bar{S}, \bar{L}) \bar{J}] \quad (7)$$

Where, λ_p the peak fluorescence wavelength of the emission band and $\Delta\lambda_{eff}$ is the effective fluorescence line width.

3.4 Nephelauxetic Ratio (β) and Bonding Parameter ($b^{1/2}$)

The nature of the R-O bond is known by the Nephelauxetic Ratio (β') and Bonding Parameter ($b^{1/2}$), which are computed by using following formulae [23, 24]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{\nu_g}{\nu_a} \quad (8)$$

where, ν_g and ν_a refer to the energies of the corresponding transition in the glass and free ion, respectively. The values of bonding parameter ($b^{1/2}$) is given by

$$b^{1/2} = \left[\frac{1-\beta'}{2} \right]^{1/2} \quad (9)$$

IV. Result and Discussion

4.1 XRD Measurement

Figure 1 presents the XRD pattern of the sample contain – GeO₂ which is show no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature within the resolution limit of XRD instrument.

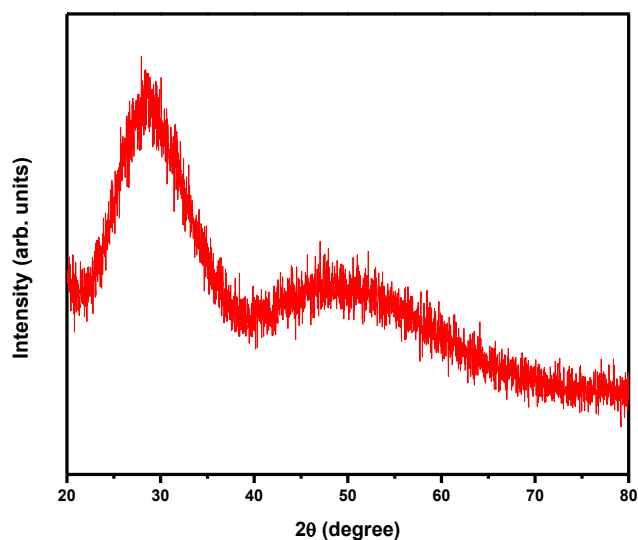


Fig.1: X-ray diffraction pattern of YZSLCBG TB (1.0) glass.

4.2 Transmittance Spectrum

The transmittance spectrum of Tb³⁺ doped in yttrium zinc lithium sodalime cadmium borogermanate glass is shown in Figure 2.

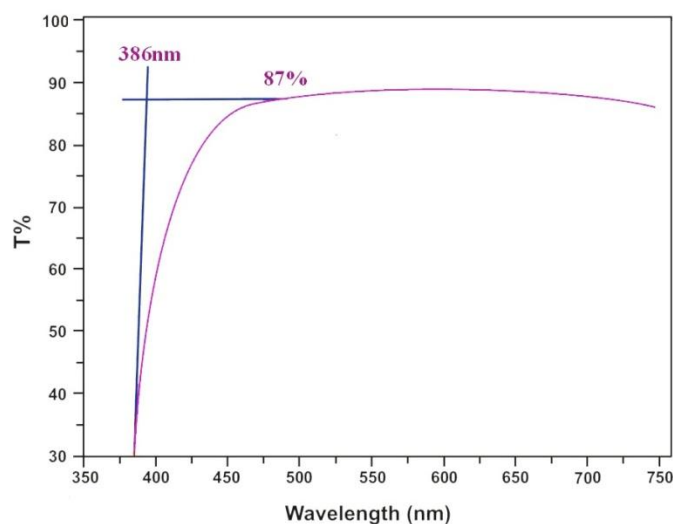


Fig.2: Transmittance spectrum of YZSLCBG TB(1.0) glass.

4.3 Thermal Property

Differential thermal analysis checks the heat absorbed by glass samples during heating or cooling. Fig. 3 depicts the DTA thermogram of powdered YZSLCBG sample. The glass transition temperature (T_g), onset crystallization temperature (T_c), crystallization temperature (T_p), melting temperature (T_m), thermal stability (T_s), Balaji Parameter (B_p), Hurbe's criterion (H_R) and reduced glass transition temperature (T_{rg}) were calculated. Shankar's parameter also calculated by using eq. (14). All the determined thermal parameters are given in table 2.

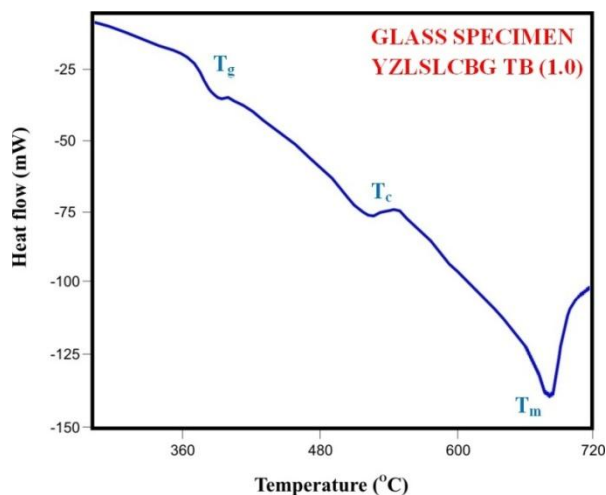


Fig.3: DTA curve of YZLSLCBG DY (1.0) glass.

Table 2. Thermal parameters determined from the DTA traces of YZLSLCBG TB glasses.

Sample Name	T _g (°C)	T _c (°C)	T _p (°C)	T _m (°C)	T _s (°C)	B _p (°C)	H _R (°C)	K _S (°C)	T _{rg} (°C)
YZLSLCBG TB (1.0)	376	510	548	686	134	3.526	0.216	34.379	0.548
YZLSLCBG TB (1.5)	380	511	550	688	131	3.359	0.220	33.702	0.552
YZLSLCBG TB (2.0)	386	512	555	691	126	2.930	0.240	33.640	0.559

The thermal stability of the glass samples can be calculated by difference between onset crystallization temperature and transition temperature [25].

$$\text{Thermal Stability (T}_s\text{)} = T_c - T_g \quad (10)$$

Balaji Parameter can be calculated using [25].

$$\text{Balaji Parameter (B}_p\text{)} = [(T_c - T_g)/(T_p - T_c)] \quad (11)$$

Hruby's criterion is calculated using the Hurby's relation [25].

$$\text{Hruby's criterion (H}_R\text{)} = [(T_p - T_c)/(T_m - T_c)] \quad (12)$$

Reduced glass transition temperature is given as [25].

$$\text{Reduced glass transition temperature (T}_{rg}\text{)} = T_g/T_m \quad (13)$$

Thermal Parameter is given as [25].

$$K_S = [(T_m - T_c)(T_c - T_g)/T_m] \quad (14)$$

4.4 Absorption Spectrum

The absorption spectra of Tb³⁺ doped YZLSLCBG TB (1.0) glass specimen has been presented in Figure 4 in terms of optical density versus wavelength (nm). Five absorption bands have been observed from the ground state ⁷F₆ to excited states ⁵D₄, ⁵D₃, ⁵L₁₀, ⁵D₂ and ⁵L₉ for Tb³⁺ doped YZLSLCBG TB (1.0) glass.

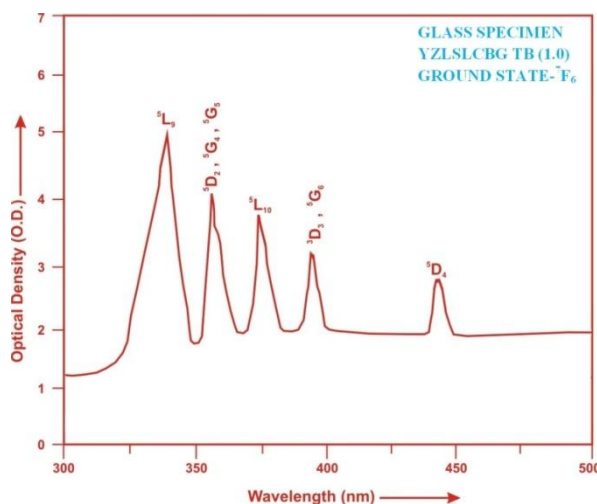


Fig.4: Absorption spectrum of YZLSLCBG TB (1.0) glass.

The experimental and calculated oscillator strengths for Tb³⁺ ions in Yttrium Zinc Lithium Sodalime cadmium Borogermanate glasses are given in Table 3.

Table3: Measured and calculated oscillator strength ($P_m \times 10^{+6}$) of Tb³⁺ ions in YZLSLCBG glasses.

Energy level 7F_0	Glass YZLSLCBG TB(1.0)		Glass YZLSLCBG TB(1.5)		Glass YZLSLCBG TB(2.0)	
	P_{exp}	P_{cal}	P_{exp}	P_{cal}	P_{exp}	P_{cal}
5D_4	0.57	0.06	0.55	0.06	0.53	0.07
5D_3	0.91	0.41	0.89	0.42	0.87	0.44
${}^5L_{10}$	1.61	1.16	1.58	1.16	1.56	1.18
5D_2	1.90	0.58	1.88	0.59	1.86	0.61
5L_9	2.18	1.03	2.15	1.03	2.12	1.04
r.m.s. deviation	0.8704		0.8410		0.8066	

The small value of r.m.s. deviation indicates fairness of fitting between experimental and calculated oscillator strengths.

Computed values of F_2 , Lande' parameter (ξ_{4f}), Nephelauxetic ratio (β') and bonding parameter ($b^{1/2}$) for Tb³⁺ doped YZLSLCBG glass specimen are given in Table 4.

Table 4. F_2 , ξ_{4f} , β' and $b^{1/2}$ parameters for Terbium doped glass specimen.

Glass Specimen	F_2	ξ_{4f}	β'	$b^{1/2}$
Tb ³⁺	400.26	1820.87	0.9703	0.1219

The value of Judd-Ofelt intensity parameters are given in **Table 5**

Table5: Judd-Ofelt intensity parameters for Tb³⁺ doped YZLSLCBG glass specimens.

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	Ω_4/Ω_6	Trend	References
YZLSLCBG TB(1.0)	4.688	3.031	2.675	1.133	$\Omega_2 > \Omega_4 > \Omega_6$	P.W.
YZLSLCBG TB(1.5)	5.719	3.039	2.684	1.132	$\Omega_2 > \Omega_4 > \Omega_6$	P.W.
YZLSLCBG TB(2.0)	7.420	2.797	2.723	1.027	$\Omega_2 > \Omega_4 > \Omega_6$	P.W.
PKAPN (HO)	8.680	3.040	2.370	1.283	$\Omega_2 > \Omega_4 > \Omega_6$	[26]
LLPNBP (SM)	5.152	4.553	4.198	1.085	$\Omega_2 > \Omega_4 > \Omega_6$	[27]
ZLCBP (SM)	4.599	4.096	3.813	1.074	$\Omega_2 > \Omega_4 > \Omega_6$	[28]
ZLCMBP (DY)	2.790	1.715	1.354	1.267	$\Omega_2 > \Omega_4 > \Omega_6$	[29]
	8.580	4.310	2.880	1.497	$\Omega_2 > \Omega_4 > \Omega_6$	[30]

4.5 Excitation Spectrum

Excitation spectra of YZLSLCBG TB (01) glass recorded at the emission wavelength 544 nm is depicted as figure 5. The excitation spectra was recorded in the spectral region 300-550nm fluorescence at 544 nm having different excitation band centered at 305,316,331,345,359,368,377,386 and 484 are attributed to the $^5H_6, ^5H_7, ^5D_1, ^5L_7+^5L_8, ^5L_9, ^5G_5, ^5L_{10}, ^5D_3+^5G_6$ and 5D_4 transitions respectively. The highest absorption level is $^5D_3+^5G_6$ and is at 386 nm. So this is to be chosen for excitation wavelength.

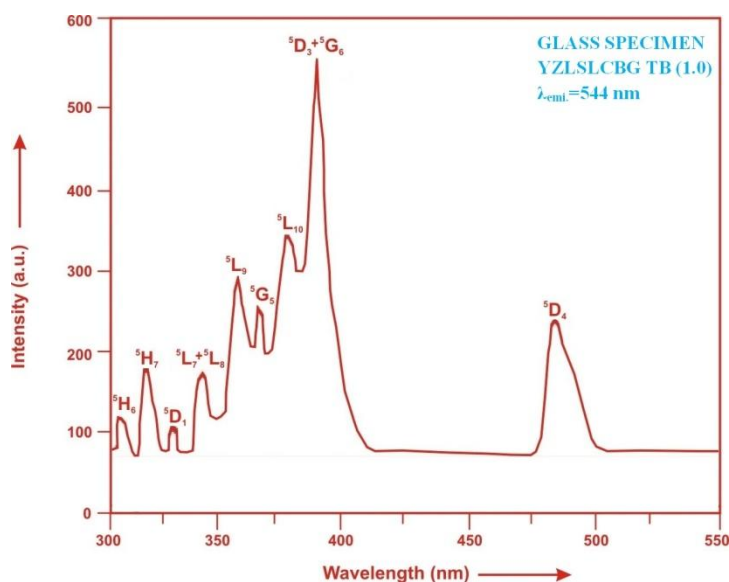


Fig.5: Excitation spectrum of YZLSLCBG TB (1.0) glass.

4.6. Fluorescence Spectrum

The fluorescence spectrum of Tb^{3+} doped in Yttrium Zinc Lithium Sodalime cadmium Borogerminate glass is shown in Figure 6. There are four broad bands observed in the Fluorescence spectrum of Tb^{3+} doped Yttrium Zinc Lithium Sodalime cadmium Borogerminate glass. The wavelengths of these bands along with their assignments are given in Table 5. Fig.(6).Shows the fluorescence spectrum with four peaks ($^5D_4 \rightarrow ^7F_6$), ($^5D_4 \rightarrow ^7F_5$), ($^5D_4 \rightarrow ^7F_4$) and ($^5D_4 \rightarrow ^7F_3$) for glass specimens.

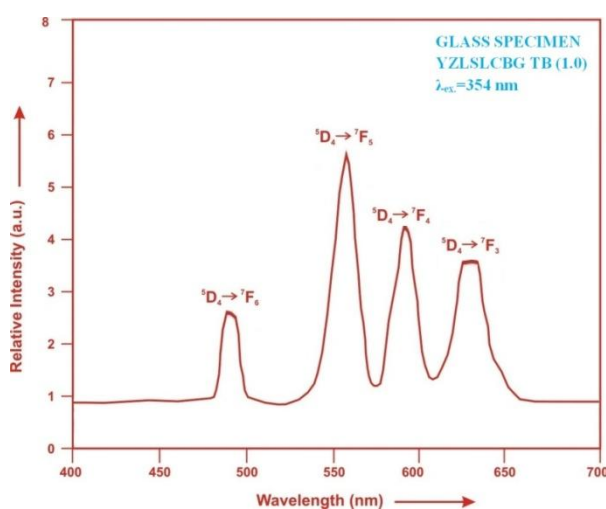


Fig.6: Fluorescence spectrum of YZLSLCBG TB (1.0) glass.

Table 6. Emission peak wave lengths (λ_{\max}), radiative transition probability (A_{rad}), branching ratio (β), stimulated emission cross-section (σ_p) and radiative life time (τ_R) for various transitions in Tb³⁺ doped YZLSLCBG glasses.

Transition	YZLSLCBG TB (1.0)				YZLSLCBG TB (1.5)				YZLSLCBG TB (2.0)				
	λ_{\max} (nm)	$A_{\text{rad}}(\text{s}^{-1})$	β	σ_p (10 ⁻²⁰ cm ²)	$\tau_R(\mu\text{s})$	$A_{\text{rad}}(\text{s}^{-1})$	β	σ (10 ⁻²⁰ cm ²)	$\tau_R(\mu\text{s})$	$A_{\text{rad}}(\text{s}^{-1})$	β	σ_p (10 ⁻²⁰ cm ²)	$\tau_R(\mu\text{s})$
⁵ D ₄ → ⁷ F ₆	488	2210.61	0.1122	0.347	50.75	2426.44	0.1058	0.374	43.61	2742.67	0.0978	0.413	35.66
⁵ D ₄ → ⁷ F ₅	550	12963.30	0.6579	2.253		15476.40	0.6750	2.645		19572.10	0.6979	3.303	
⁵ D ₄ → ⁷ F ₄	582	1985.88	0.1008	0.756		2042.24	0.08907	0.761		2044.33	0.0729	0.746	
⁵ D ₄ → ⁷ F ₃	625	2544.17	0.1291	0.707		2983.59	0.1301	0.819		3686.12	0.1314	0.996	

V. Conclusion

In the present study, the glass samples of composition (25-x) GeO₂: 10ZnO: 10Li₂O: 10Na₂O:10CaO:10CdO:10Y₂O₃:15B₂O₃:xTb₂O₃, (where x=1, 1.5, 2mol %) have been prepared by melt-quenching method. The Judd-Ofelt theory has been applied to calculate the oscillator strength and intensity parameters Ω_λ ($\lambda=2, 4, 6$). The radiative transition rate and the branching ratio are highest for (⁵D₄→⁷F₅) transition and hence it is useful for laser action. The stimulated emission cross section (σ_p) value is also very high for the transition (⁵D₄→⁷F₅). This shows that (⁵D₄→⁷F₅) transition is most probable transition. The large value of Balaji and Shankar parameters indicate that the prepared glass samples have good thermal stability.

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