

Spectral, Transmittance and Photoluminescence Analysis of Nd³⁺ Doped Borotellurite Glasses for Optoelectronic Device Applications

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ABSTRACT: Glass of the system $(20-x)\text{TeO}_2: 10\text{ZnO}: 10\text{Li}_2\text{O}: 10\text{K}_2\text{O}: 10\text{Nb}_2\text{O}_5: 10\text{Na}_2\text{O}: 10\text{CaO}: 10\text{Y}_2\text{O}_3: 10\text{B}_2\text{O}_3: x\text{Nd}_2\text{O}_3$. (where $x=1, 1.5, 2$ mol %) have been prepared by melt-quenching method. The amorphous nature of the glasses was confirmed by X-ray diffraction studies. Optical absorption spectra were recorded at room temperature for all glass samples. Slater-Condon parameters F_k ($k=2, 4, 6$), Lande parameter ξ_{4f} and Racah parameters E^k ($k=1, 2, 3$) have been computed. Using these parameters energies and intensities of these bands has been calculated. Judd-Ofelt intensity parameters Ω_λ ($\lambda=2, 4, 6$) are evaluated from the intensities of various absorption bands of optical absorption spectra. Using these intensity parameters various radiative properties like spontaneous emission probability (A), branching ratio (β_R), radiative life time (τ_R) and stimulated emission cross-section (σ_p) of various emission lines have been evaluated.

KEYWORDS: YZLPNSLBT Glasses, Optical Properties, Judd-Ofelt Theory, Photoluminescence Properties.

I. INTRODUCTION

Among different ceramic glasses, scientific interest on borotellurite glasses is attributed to their favorable properties such as, high refractive index, high density, large third order nonlinear optical susceptibility [1-6]. Tellurite glass is an extremely promising material for mechanical sensors, reflecting windows and nonlinear applications in optics. They have low phonon energy, low melting temperature, excellent transparency, high thermal stability, high transparency and low dispersion rates [7-10]. Tellurite glasses are potentially important host materials for developing rare earth doped optical devices. They present superior properties like that high transparency, low melting point, high gain density, high solubility for rare-earth ions and low dispersion. [11,12]. Thus, it can be widely used in visible and infrared laser, fiber amplifier and optical data storage devices. The host matrix composed of ZnO, a glass modifier/glass former a heavy metal oxide along with TeO₂, Li₂O, K₂O, Nb₂O₅, Na₂O, CaO, Y₂O₃ and B₂O₃. The addition of Nb₂O₅ to the glass mixture improves the rare earth ion solubility leading to the possibility of using even higher concentrations of ions [13-15].

The present work reports on the absorption, Transmittance and emission properties of Nd³⁺ doped yttrium zinc lithium potassium niobate sodalime borotellurite glass. The intensities of the transitions for the rare earth ions have been estimated successfully using the Judd-Ofelt theory. The laser parameters such as radiative probabilities (A), branching ratio (β_R), radiative life time (τ_R) and stimulated emission cross section (σ_p) are evaluated using Judd-Ofelt intensity parameters (Ω_λ , $\lambda=2, 4$ and 6).

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Nd³⁺ doped borotellurite glass samples $(20-x)\text{TeO}_2: 10\text{ZnO}: 10\text{Li}_2\text{O}: 10\text{K}_2\text{O}: 10\text{Nb}_2\text{O}_5: 10\text{Na}_2\text{O}: 10\text{CaO}: 10\text{Y}_2\text{O}_3: 10\text{B}_2\text{O}_3: x\text{Nd}_2\text{O}_3$. (where $x = 1, 1.5, 2$) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of TeO₂, ZnO, Li₂O, K₂O, Nb₂O₅, Na₂O, CaO, Y₂O₃, B₂O₃ and Nd₂O₃. They were thoroughly mixed by using an agate pestle mortar. Then melted at 1095 °C by an electrical muffle furnace for 2 hours. After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250 °C for 2 h 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**.

Table1.

Sample	Glass composition (mol %)
YZLPNSLBT (UD)	20TeO ₂ : 10ZnO: 10Li ₂ O: 10K ₂ O: 10Nb ₂ O ₅ : 10Na ₂ O: 10CaO: 10Y ₂ O ₃ :10B ₂ O ₃
YZLPNSLBT ND (1.0)	19TeO ₂ : 10ZnO: 10Li ₂ O: 10K ₂ O: 10Nb ₂ O ₅ : 10Na ₂ O: 10CaO: 10Y ₂ O ₃ :10B ₂ O ₃ :1Nd ₂ O ₃
YZLPNSLBT ND (1.5)	18.5TeO ₂ : 10ZnO: 10Li ₂ O: 10K ₂ O: 10Nb ₂ O ₅ : 10Na ₂ O: 10CaO: 10Y ₂ O ₃ :10B ₂ O ₃ : 1.5Nd ₂ O ₃
YZLPNSLBT ND (2.0)	18TeO ₂ : 10ZnO: 10Li ₂ O: 10K ₂ O: 10Nb ₂ O ₅ : 10Na ₂ O: 10CaO: 10Y ₂ O ₃ :10B ₂ O ₃ : 2Nd ₂ O ₃

YZLPNSLBT (UD) - Represents undoped Yttrium Zinc Lithium Potassiumniobate Sodalime Borotellurite glass specimen.

YZLPNSLBT (ND) - Represents Nd³⁺ doped Yttrium Zinc Lithium Potassiumniobate Sodalime Borotellurite glass specimens.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines is expressed in terms of oscillator strengths using the relation [16].

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

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 [17], 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 [18] and Ofelt [19] 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:

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

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

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

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} ($\lambda=2, 4$ and 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:

$$A [(S', L') J'; (S, L) J] = \frac{64 \pi^2 v^3}{3h(2J'+1)} \left[\frac{n(n^2+2)^2}{9} \right] \times S(J', \bar{J}) \quad (5)$$

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 manifold $|4f^N(S, L) J \rangle$ is given by

$$\beta [(S', L') J'; (S, L) J] = \sum_{S, L, J} \quad (6)$$

The radiative life time is given by

$$\tau_{rad} = \sum_{S, L, J} A[(S', L') J'; (S, L) J] = \quad (7)$$

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

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

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 Parameters ($b^{1/2}$), which are computed by using following formulae [20,21]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{v_g}{v_a} \quad (9)$$

where, v_a and v_g refer to the energies of the corresponding transition in the glass and free ion, respectively. The values of bonding parameter $b^{1/2}$ are given by]

$$b^{1/2} = \left[\frac{1-\beta'}{2} \right]^{1/2} \quad (10)$$

IV. RESULT AND DISCUSSION

4.1. XRD Measurement

Figure 1 presents the XRD pattern of the samples shows 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.

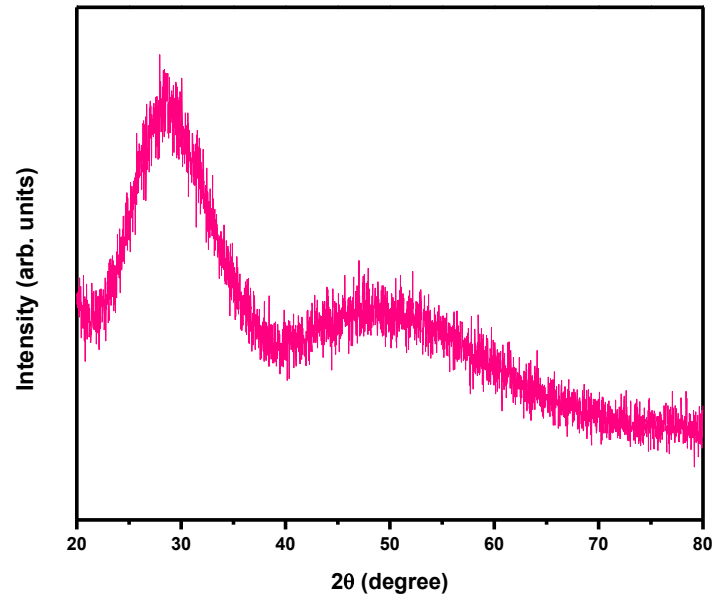


Fig.1: X-ray diffraction pattern of YZLPNSLBT ND(1.0) glass

4.2 Transmittance spectra

The Transmittance spectrum of YZLPNSLBT ND (1.0) glass is shown in Figure 2.

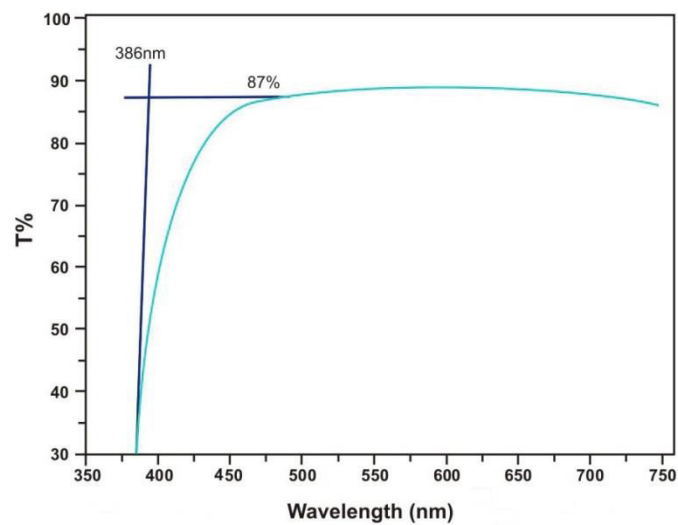


Fig (2) Transmittance spectrum of YZLPNSLBT ND (1.0) glass.

4.3. Absorption spectra

The absorption spectra of YZLPNSLBT ND(1.0) glass, consists of absorption bands corresponding to the absorptions from the ground state $^4I_{9/2}$ of Nd^{3+} ions. Nine absorption bands have been observed from the ground state $^4I_{9/2}$ to excited states $^4F_{3/2}$, $^4F_{5/2}$, $^4F_{7/2}$, $^4F_{9/2}$, $^2H_{11/2}$, $^4G_{5/2}$, $^4G_{7/2}$, $^4G_{9/2}$, and $^2G_{9/2}$ for Nd^{3+} doped YZLPNSLBT ND(1.0) glass.

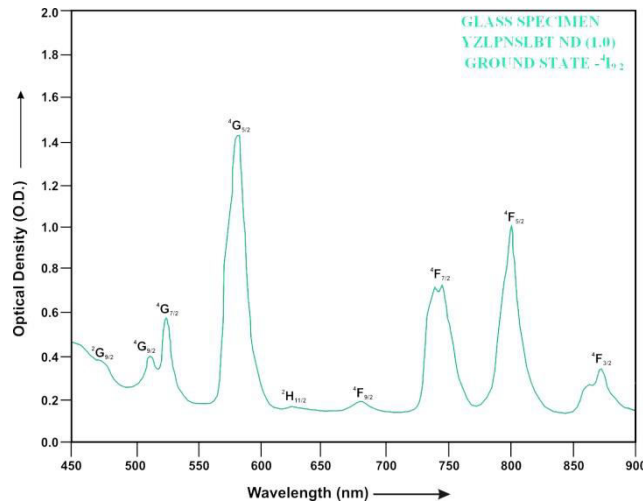


Fig.3: Absorption spectra of YZLPNSLBT ND (1.0) glass.

The experimental and calculated oscillator strength for Nd^{3+} ions in YZLPNSLBT glasses are given in Table 2.

Table2 Measured and calculated oscillator strength ($P^m \times 10^{+6}$) of Nd^{3+} ions in YZLPNSLBT glasses.

Energy level from $^4I_{9/2}$	Glass YZLPNSLBT ND(1.0)		Glass YZLPNSLBT ND(1.5)		Glass YZLPNSLBT ND(2.0)	
	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$	$P_{exp.}$	$P_{cal.}$
$^4F_{3/2}$	3.85	3.69	3.81	3.68	3.77	3.67
$^4F_{5/2}$	8.38	8.54	8.35	8.51	8.30	8.47
$^4F_{7/2}$	9.45	9.71	9.40	9.69	9.34	9.64
$^4F_{9/2}$	0.65	0.52	0.63	0.52	0.61	0.52
$^2H_{11/2}$	0.26	0.15	0.24	0.15	0.21	0.15
$^4G_{5/2}$	25.45	25.74	24.36	24.67	23.26	23.58
$^4G_{7/2}$	4.25	5.23	4.21	5.16	4.18	5.08
$^4G_{9/2}$	2.22	2.28	2.18	2.27	2.15	2.26
$^2G_{9/2}$	0.96	2.92	0.93	2.91	0.91	2.90
r.m.s.deviation	0.7471		0.7504		0.7474	

Table3. Computed values of Slater-Condon, Lande, Racah, nephelauxetic ratio and bonding parameter for Nd³⁺ doped YZLPNSLBT glass specimens

Parameter	Free ion	YZLPNSLBT ND(1.0)	YZLPNSLBT ND(1.5)	YZLPNSLBT ND(2.0)
F ₂ (cm ⁻¹)	331.16	324.58	324.69	324.52
F ₄ (cm ⁻¹)	50.71	50.72	50.70	50.74
F ₆ (cm ⁻¹)	5.154	5.038	5.044	5.038
ξ _{4f} (cm ⁻¹)	884.0	882.73	882.91	882.61
E ¹ (cm ⁻¹)	5024.0	4947.12	4948.45	494.71
E ² (cm ⁻¹)	23.90	23.08	23.10	23.06
E ³ (cm ⁻¹)	497.0	489.58	489.56	489.54
F ₄ /F ₂	0.1531	0.1563	0.1561	0.1564
F ₆ /F ₂	0.0155	0.01552	0.0155	0.0155
E ¹ /E ³	10.1086	10.1048	10.1079	10.1057
E ² /E ³	0.0481	0.0471	0.0472	0.0471
β'		0.9958	0.9961	0.9959
b ^{1/2}		0.04578	0.04431	0.04552

The values of Judd-Ofelt intensity parameters are given in Table 4.

Table4. Judd-Ofelt intensity parameters for Nd³⁺ doped YZLPNSLBT glass specimens.

Glass Specimen	Ω ₂ (pm ²)	Ω ₄ (pm ²)	Ω ₆ (pm ²)	Ω ₄ /Ω ₆	Ref.
YZLPNSLBT ND(1.0)	2.511	7.750	3.453	2.244	P.W.
YZLPNSLBT ND(1.5)	2.156	7.737	3.440	2.249	P.W.
YZLPNSLBT ND(2.0)	1.806	7.714	3.420	2.256	P.W.
SLBB (ND)	7.530	10.070	8.860	1.137	[22].

4.4. Excitation Spectrum

The Excitation spectra of Nd³⁺ doped YZLPNSLBT ND(1.0) glass specimen has been presented in Figure 4 in terms of Excitation Intensity versus wavelength. The excitation spectrum was recorded in the spectral region 700–1000 nm fluorescence at 1065nm having different excitation band centred at 808 nm and 887 nm are attributed to the (⁴I_{9/2}→⁴F_{5/2}) and (⁴I_{9/2}→⁴F_{3/2}) transitions, respectively. The highest absorption level is (⁴I_{9/2}→⁴F_{5/2}) and is at 808 nm. So this is to be chosen for excitation wavelength.

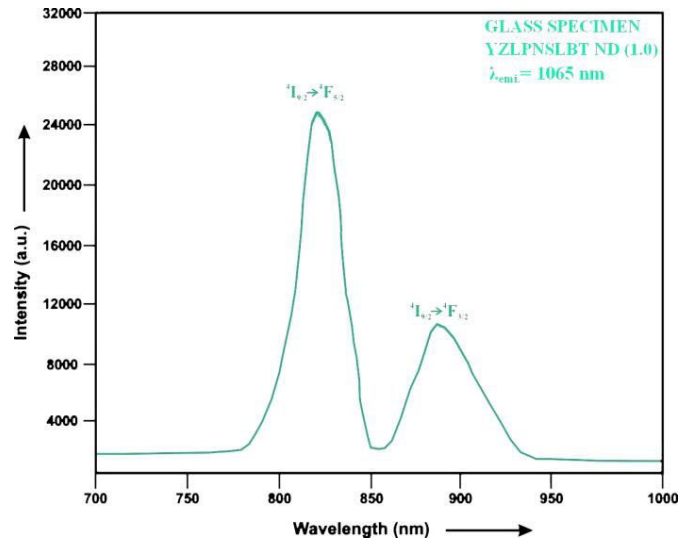


Fig.4: Excitation spectra of YZLPNSLBT ND (1.0) glass.

4.5 Fluorescence Spectrum

The fluorescence spectrum of ND(1.0)doped in yttrium zinc lithium potassiumniobate sodalime borotellurite is shown in Figure 5. There are six broad bands (${}^4G_{7/2} \rightarrow {}^4I_{9/2}$), (${}^4G_{7/2} \rightarrow {}^4I_{11/2}$), (${}^4F_{3/2} \rightarrow {}^4I_{9/2}$), (${}^4F_{3/2} \rightarrow {}^4I_{11/2}$), (${}^4F_{3/2} \rightarrow {}^4I_{13/2}$) and (${}^4F_{3/2} \rightarrow {}^4I_{15/2}$) respectively for glass specimens.

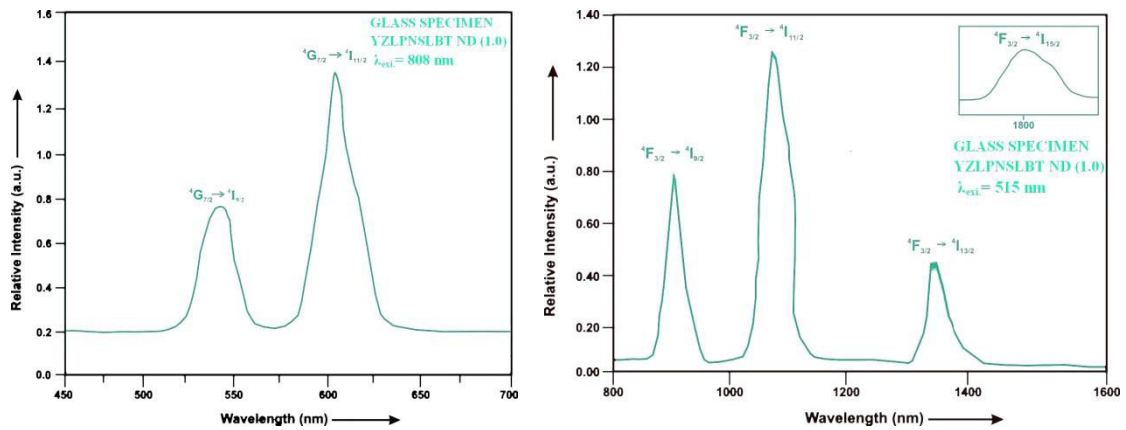


Fig.5: Fluorescence spectrum of YZLPNSLBT ND(1.0) glasses

The wavelengths of these bands along with their assignments are given in Table 5.

Table 5 Emission peak wave lengths (λ_p), radiative transition probability (A_{rad}), branching ratio (β_R), stimulated emission cross section (σ_p), and radiative life time (τ_R) for various transitions in Nd³⁺ doped YZLPNSLBT glasses.

Transition	YZLPNSLBT ND(1.0)				YZLPNSLBT ND(1.5)				YZLPNSLBT ND(2.0)				
	λ_{max} (nm)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (μ s)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (μ s)	A_{rad} (s ⁻¹)	β	σ_p (10 ⁻²⁰ cm ²)	τ_R (10 ⁻²⁰ cm ²)
⁴ G _{7/2} → ⁴ I _{9/2}	53	320	0.42	0.46	131.86	3160.72	0.4	0.480	135.73	3108.32	0.4	0.489	139.96
⁴ G _{7/2} → ⁴ I _{11/2}	59	303	0.40	1.12		2871.53	0.3	1.141		2703.91	0.3	1.118	
⁴ F _{3/2} → ⁴ I _{9/2}	90	798.07	0.10	0.76		798.16	0.1	0.794		797.16	0.1	0.820	
⁴ F _{3/2} → ⁴ I _{11/2}	10	461.34	0.06	2.03		460.96	0.0	2.198		459.79	0.0	2.346	
⁴ F _{3/2} → ⁴ I _{13/2}	13	74.5	0.00	0.41		74.4	0.0	0.432		74.1	0.0	0.437	
⁴ F _{3/2} → ⁴ I _{15/2}	18	1.78	0.00	0.02		1.78	0.0	0.026		1.77	0.0	0.026	
	00	02	02	4									

V. CONCLUSION

In the present study, the glass samples of composition (20-x)TeO₂: 10ZnO: 10Li₂O: 10K₂O: 10Nb₂O₅: 10Na₂O: 10CaO: 10Y₂O₃:10B₂O₃: xNd₂O₃. (where x =1, 1.5, 2 mol %) have been prepared by melt-quenching method. The stimulated emission cross section (σ_p) has highest value for the transition (⁴F_{3/2}→⁴I_{11/2}) in all the glass specimens doped with Nd³⁺ ion. This shows that (⁴F_{3/2}→⁴I_{11/2}) transition is most probable transition. The results show that the Nd³⁺ doped borotellurite glasses could be potential candidates for optoelectronic device Applications.

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