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Spectral Analysis of Pr (III) Doped Metal Borax Glasses

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Abstract

In this paper Pr(III) doped metal borax glasses are fabricated with the following compositions using conventional melt quenching technique. The compositions of the glass systems are 10.ZnO + 35. $Na_2B_4O_7$ + (50-x) PbO + (x+10) CaO + 4.Al_2O_3 + 1Pr_2O_3 where (x = 0,1,2,3, 5 and 7 wt %.). Certain physical properties of these glasses have been evaluated and reported. Spectral data for all these glasses were recorded for X-ray diffraction, Optical absorption and Fluorescence properties. The Judd-Ofelt intensity parameters Ω_λ (λ = 2,4,6) were evaluated from the spectral data and in turn employed to evaluate the lasing parameters of Pr(III) metal glass systems such as radiative transition probabilities (A), radiative life-times (τ_R), branching ratios (β_R) absorption cross-sections (σa) and Stimulated emission cross-sections (σ_e). The experimental and calculated branching ratios (β_R) are found to be in good agreement in the present work.

KEY WORDS: Metal glasses, Optical absorption, Praseodymium, XRD

INTRODUCTION - Glasses doped with rare earth (RE) ions have emerged as a significant category of solid state luminescent material and are finding ever-increasing applications as compact visible and NIR lasers, broad band amplifiers, light-emitting devices, color display panels, optical data storage, sensors, optical communications, etc. In particular, the rising demand in visible laser sources has provoked significant exploitation of RE ions like Eu(III), Sm(III), Dy(III), Tm(III) and Pr(III) within various disordered matrices (1,2). Laser action in visible region has been reported in literature for the transition $^1D_2$ ->$^3H_4$ [3-5] of the Pr(III) ion. The potential Laser transition of $^1G_4$->$^3H_5$ at ~ 1.3μm of Pr(III) find its applications for the development of fiber amplifier for communication purpose in the telecom window. The Judd–Ofelt [6, 7] theory has been proved to be a powerful tool to analyses optical spectra of RE ions in various media. The spectral studies give valuable information about the structure and bonding of

The system along with their lasing properties. Therefore such systems are required as tailor made devices to develop optical devices for laser action.

The present work reports on the preparation and characterization of RE doped metal borax glasses for lasing materials. The host matrix composes of PbO, a glass modifier/ glass farmer a heavy metal oxide along with ZnO, B_2O_3, Al_2O_3 and CaO as their constituents. The conventional Judd-Ofelt theory is reported in literature to analyze the absorption spectra of Pr(III) ions in different host glass matrices. The lasing parameters such as radiative transition probabilities (A), branching ratio (β), , radiative life time (τ_R) absorption cross section (σa) and stimulated emission cross section (σe) are evaluated using J.O intensity parameters (Ω_λ , λ = 2,4,6). The metal such as PbO in the glass composition increases the thermal stability and decreases the maximum phonon energy of the host in which it is present. Thus it provides an effective way to tailor the lasing properties of the system and therefore it indicates us to carry out of the present study.

EXPERIMENTAL- The Metal borax glasses with the wt% compositions of 10 ZnO + 35 $Na_2B_4O_7$ + (50-x) PbO + (x+10) CaO + 4 Al_2O_3 + 1Pr_2O_3 with (x=1, 2, 3, 5 and 7wt%) were prepared using Melt quenching technique. 10g batch of each chemical composition were weight accurately, mixed and grinded in an agate mortar and then transferred into a silica porcelain crucible. It is kept at 450^{o}C for 30 min to remove impurities in the base material and melted in muffle furnace in the temperature range of 700-800 ^{o}C for 1 hour. The melt is then poured on to a preheated borax glass plate and air quenched to get a good optical quality glasses. The samples are annealed at about 400oC for 5 hours to remove thermal strains and then polished before measuring their optical properties. The amorphous nature of the samples was confirmed by XRD spectra obtained by using Shimadzu-XD3A Diffractometer. XRD spectra were recorded for each sample with different chemical compositions to confirm their amorphous nature. PerkinElmer Lambda 3B UV-Vis- IR spectrophotometer is used to record the absorption spectra at room temperature in the wave length range of 400-750 nm at room temperature. Refractive indices (n) were evaluated using conventional methods. Densities were measured by the Archimedes method using Xylem as an immersion liquid.

RESULTS & DISCUSSION-

(1) **Physical Properties-** Various physical properties such as density (ρ), average molecular weight (g), ion concentration molar refractivity (Rμ), Reflection loss (R), Electric susceptibility (χe), Numerical aperture (NA), polaron radius (rP), inter-ionic distance (ri), and field strength (F) are require for computation of radial properties of the glasses are evaluated following the expressions available in the literature [8-10] and are given in the Table 1. All these glasses exhibit good optical efficiencies.

Table-1- Various physical properties of Pr (III): Metal Borax Glasses 1-7 Wt% glasses

SN	Parameters	1 wt%	2 wt%	3 wt%	5 wt%	7wt%
1	Refractive index, (n)	1.623	1.640	1.690	1.761	1.791
2	Density d (gm/cm3)	3.667	3.967	4.167	4.191	4.001
3	Average molecular weight (g)	160.11	180.21	199.28	200.2	210.7
4	Molecular volume(VM) (cm^3)	59.21	53.12	51.31	52.32	53.12
5	Pr3+ conc.(10^{20} ions /cc)	2.507	2.674	2.710	2.622	2.601
6	Optical dielectric constant, (ε)	2.514	2.721	2.916	3.211	3.331
7	Molar refractivity Rμ (cm^3)	24.104	24.401	24.821	25.721	26.011
8	Inter-ionic distance ri (Å)	15.403	15.551	15.755	15.80	15.901
9	Field strength F (10^{15} cm^{-2})	2.694	2.771	2.861	2.869	2.900
10	Electric susceptibility (χ_e)	0.135	0.145	0.150	0.155	0.170
11	Numerical aperture (NA)	0.25	0.25	0.27	0.25	0.27

2. Analysis of Absorption Spectra- The absorption spectra of all the three samples Pr(III): doped metal borax glasses are shown in Fig (1)

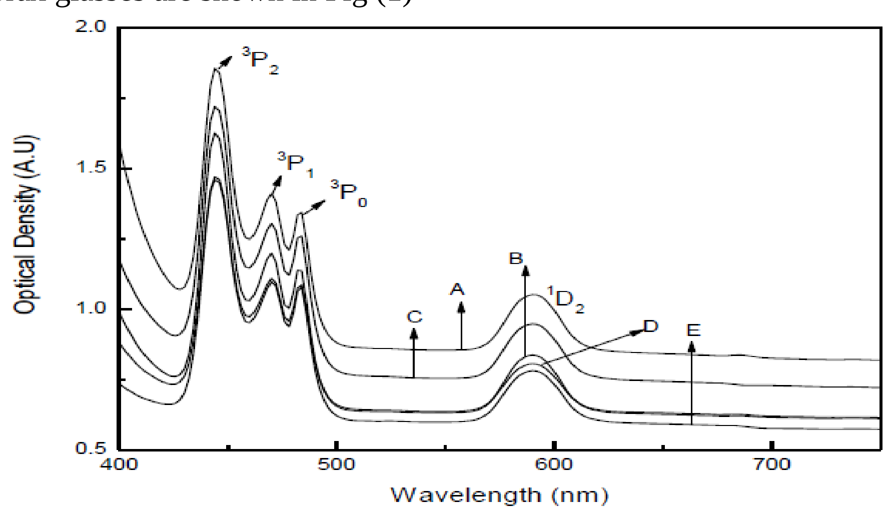


Fig-(1) Optical absorption spectra of Pr(III) Doped metal Glasses.

Table-2. Experimental and calculated energies of Pr(III) doped Metal Borax Glasses.

Transition from $^3H_4$	1wt%		2wt%		3wt%		5wt%		7wt%	
	E_{exp}	E_{cal}	E_{exp}	E_{cal}	E_{exp}	E_{cal}	E_{exp}	E_{cal}	E_{exp}	E_{cal}
$^3P_2$	22470	22455	22475	22509	22500	22515	22515	22521	22521	22519
$^3P_1$	21190	21239	21186	21239	21219	21253	21245	21243	21243	21249
$^3P_0$	20630	20589	20635	20690	20735	20780	20771	20713	20713	20725
$^1D_2$	16898	17110	16873	16919	16948	16952	17131	17129	17129	17103

Four absorption peaks were observed approximately at 16963, 20693, 21268 and 22499 cm^{-1} and their assigned as them as $1D_2$, $3P_0$, $3P_1$ and $3P_2$ levels respectively. The theoretical energies are evaluated [11,12] by applying the Taylor series expansion as and reported in Table (2).

$$E_J = E_{0J} + (\Sigma\, dE_0/dPK)\, \Delta\, PK \text{ ---(1)}$$

Where E_{0J} is the zero order energy of the chosen band and Δ PK are the changes to be effected in the free ion Racah (E1, E2 and E3), Spin-orbit coupling (ξ4f), and Configuration interaction (α, β, γ) parameters were evaluated using the partial derivatives reported in literature [13-15]. The experimental energy values (Eexpt) are satisfactorily correlated with the theoretical energies (Ecal) and are given in Table-2. The rms deviation have been evaluated formed to

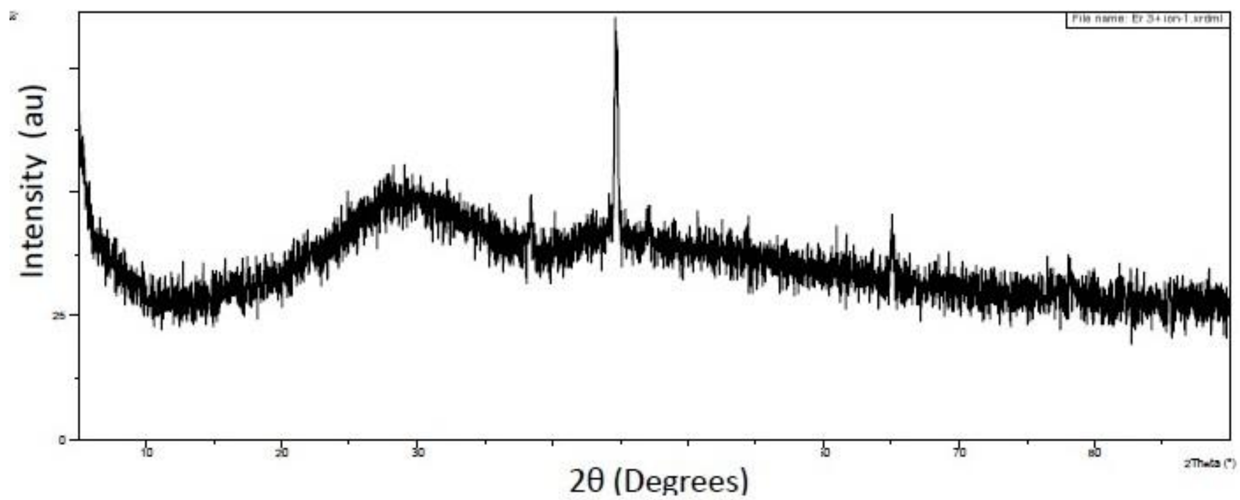
$$\sigma_{rms} = [\, \Sigma\, \Delta\, E^2/ N\,]^{1/2} \text{ -- (2)}$$

Where Δ E is the difference between the values of measured and calculated energies and N is the number of energy levels. The Racah (E1, E2 and E3), Spin-orbit coupling (ξ4f) parameters along with other spectroscopic parameters are given in Table (3). From the table it is observed that that the ratios of Racah parameters E1/E3 and E2/E3 do not vary much from the hydrogenic ratios.

Table(3)-Spectroscopic parameters of Pr(III) doped Borax glasses

Parameter	**1 wt%**	**2 wt%**	**3 wt%**	**5 wt%**	**7 wt%**
E1(cm^{-1})	4991.12	5155.22	6033.87	5823.32	5622.32
E2(cm^{-1})	23.11	22.12	25.23	25.10	24.11
E3(cm^{-1})	510.25	528.11	615.22	591.33	524.31
E1/E3	9.78	9.76	9.86	9.84	10.72
E2/E3	0.04	0.04	0.04	0.04	0.04
ξ4f (cm^{-1})	760.52	765.21	770.25	772.14	779.21

3. XRD Analysis- From the XRD studies, no change in the borax phase related to the addition of rare earth elements was observed. However, peaks of those elements were also become apparent. The XRD pattern of Pr(III) doped Metal Borax glass systems has confirmed their amorphous nature and shown in Fig.(2).



Fig(2):- XRD pattern of Pr(III): doped Borax glass system.

Conclusions:- The Pr(III) doped Metal borax glass systems have been fabricated to study their laser and other radiative and optical properties. J-O theory is successfully implemented for the analysis of optical absorption spectra. The branching ratios (β_R) estimated through the optical absorption data are in good agreement with the experimental results obtained in the optical studies. The $^3P_0 \longrightarrow {}^3H_4$ lasing transition appears to be potential in all the five glass system studied.

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