

INFLUENCE OF ALKALI-EARTH IONS ON 1.06 μm NEAR INFRARED EMISSION OF Nd^{3+} -DOPED LITHIUM ALUMINOPHOSPHATE GLASS FOR LASING APPLICATION

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Abstract

The influence of the alkali-earth ions (Mg^{2+} , Ca^{2+} , Sr^{2+}) on structural (FTIR) and spectroscopic (optical absorption and near infrared emission) properties of Nd^{3+} -doped lithium aluminophosphate glasses were systematically examined and discussed. Under an excitation wavelength of 580 nm, the emission spectra of Nd^{3+} -doped glasses showed three prominent peaks at 900, 1058, and 1327 nm, it was related to the $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{9/2}$, $^4\text{I}_{11/2}$, and $^4\text{I}_{13/2}$ transitions, respectively. It was found that the peak emission intensity occurs at a wavelength of 1058 nm. The Ω_2 , Ω_4 and Ω_6 were calculated by applying Judd-Ofelt theory to the absorption and emission measurements, which resulted in the radiative transition probability and stimulated emission cross-section being calculated. As a result, when compared to Ω_2 , Ω_4 , and Ω_6 , have a larger compositional dependency on ionic radius and modifier content. In addition, it was found that the stimulated emission cross-section of $\sigma_{em}(\text{Sr}) > \sigma_{em}(\text{Ca}) > \sigma_{em}(\text{Mg})$, respectively. The luminescence intensity is greatly increased, and the underlying mechanism is clearly seen when cationic replacements of Sr^{2+} , Ca^{2+} , and Mg^{2+} are carried out. The research proved that spectroscopic properties of trivalent Nd^{3+} depend significantly on the kind of presence of alkali-earth ions in glass host matrices and could be used as a lasing application.

Keywords: Alkali Earth Ion; Aluminophosphate glass; Nd^{3+} doped glass; Near Infrared Emission

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Introduction

Phosphate glasses have captured significant attention in research due to their extensive high-tech applications. Compared to conventional glasses, phosphate glasses exhibit several intriguing and unique physical characteristics, including exceptional corrosion resistance, low melting temperatures, high hardness, transparency at normal temperatures, and the ability to serve as effective laser materials. These glasses are utilized as transmitting optical components and magneto-optic materials in various laser applications (Osman *et al.*, 2022).

Nd³⁺ ions serve as dopants in numerous high-energy, high-power laser phosphate glasses, primarily due to their potent $^4F_{3/2} \rightarrow ^4I_{11/2}$ emission at approximately 1.06 μm . This ion has emerged as one of the most efficient rare-earth ions for solid-state lasers. Investigating the spectroscopic characteristics of Nd³⁺-doped glass lasers, including parameters like lifetime, quantum efficiency, peak wavelengths, linewidths, emission and absorption cross-sections, and fluorescence quenching mechanisms, is imperative for assessing their suitability. Specifically, the extraction and stored energy in the $^4F_{3/2}$ state are determined by the emission cross-section and quantum efficiency. Hence, large emission cross-sections and prolonged fluorescence lifetimes are essential for optimal laser performance of the $^4F_{3/2} \rightarrow ^4I_{11/2}$ emission. Non-radiative processes, such as quenching induced by impurities like OH groups, multi-phonon relaxation, and Nd-Nd concentration quenching, can shorten the laser's lifetime, reducing stored energy and altering its output energy. Dehydroxylating phosphate glasses is crucial to achieving optimal laser performance since non-radiative losses in Nd³⁺-doped glasses are primarily caused by energy transfer to OH vibrational modes (Muñoz-Quinero *et al.*, 2022).

Glass modifiers, especially alkaline earth metal oxides such as MgO, CaO, and SrO (denoted as MO), play a crucial role in influencing the characteristics of glass systems, making them particularly significant (Kuwik *et al.*, 2020). When modifier oxides are incorporated into the glass matrix, they facilitate the depolymerization of lengthy phosphate chains and generate non-bridging oxygen ions. These ions then substitute P-O-P links with more chemically stable bonds, consequently enhancing the chemical durability of phosphate glasses (Prasad *et al.*, 2019). In order to improve both mechanical and chemical stability, alkaline

earth elements are additionally incorporated (Veeramohan Rao *et al.*, 2017). Adding Li₂O helps reduce the hydrophilic nature and enhances the chemical resistance of phosphate glass. Additionally, the addition of Li₂O leads to the formation of non-bonded oxygen (NBOs) within the glass (Jia *et al.*, 2004). While these glasses may initially have limited practical applications due to their relatively poor chemical durability, this drawback can be addressed through the addition of different metal oxides, such as Al₂O₃. These oxides play a crucial role in forming stable M-O-P cross-linked bonds, thereby significantly improving the chemical durability of the glasses (Madhu *et al.*, 2018).

This study investigates the influence of alkaline earth elements, specifically MgO, CaO, and SrO, on glass samples with the chemical composition 58P₂O₅-9Al₂O₃-17Li₂O-1Nd₂O₃-15MO (where MO represents MgO, CaO, and SrO).

This paper is divided into two parts, each discussing essential features of laser glasses. Data is provided wherever possible to assist glass researchers in estimating the important performance characteristics of laser glasses. The concentration of Nd³⁺ ions increase with the increase in the M²⁺ ionic radius of the glass host matrix (where M represents Mg, Ca, Sr). Based on this result, systems with promising optical characteristics can be selected, making them potential candidates for red and near infrared photoluminescent materials in photonic devices, which is significant for future applications.

Experimental Details

Glass Preparation

The glasses were synthesized using the melt-quenching method. They had a stoichiometric ratio of (58P₂O₅-9Al₂O₃-17Li₂O-1Nd₂O₃-15MO, where MO represents MgO, CaO, and SrO) were prepared and coded as MgNd, CaNd, and SrNd. The batch chemicals, including NH₄H₂PO₄, Al₂O₃, MgCO₃, CaCO₃, SrCO₃, and Nd₂O₃, were used as analytical reagent grade raw materials. These components were thoroughly mixed in an aluminum crucible before undergoing a 3-hour melting process at 1,200°C in a prepared electrical furnace. Subsequently, the glasses were annealed at 500°C for three hours, followed by cooling to room temperature and optical polishing for spectroscopic measurements.

Physical and Spectroscopic Measurements

The density of the glass samples was determined through Archimedes' method, where the mass of the glass in both air and water was measured using a 4-digit microbalance (A&D, HR-200). Subsequently, the molar volume was calculated by dividing the total molecular weight by the density of the glass components. The refractive index of the glasses was measured using an Abbe refractometer at a wavelength of 589 nm. Fourier transform infrared measurements were conducted at room temperature over the range of 650–4,000 cm^{-1} using an Agilent Technologies Cary 630 FTIR spectrometer. The absorption spectra from 200 to 2,000 nm were studied using a UV-VIS-NIR spectrometer (UV 3600, Shimadzu). NIR luminescence and decay curves were recorded using a photoluminescence spectrometer (PTI, Quantum Master-300) with an exciting light source. All data were recorded at ambient temperature.

Results and Discussions

Physical Properties

The substitution of P_2O_5 with Nd_2O_3 within the glass network enhances the optical properties of phosphate glass, making it suitable for advanced optical applications. Table 1 shows physical and optical characteristics of Nd^{3+} ions in $\text{P}_2\text{O}_5\text{-Al}_2\text{O}_3\text{-Li}_2\text{O-MO}$ ($\text{M} = \text{Mg, Ca, and Sr}$). The results indicate that the larger atomic radius and higher molecular weight of Mg, Ca, and Sr lead to an increase in both density and molar volume in the sequence of MgNd , CaNd , and SrNd . The densification of the glass is attributed to the elevated molecular weight. However, the larger atomic radius of the alkaline earth atom may result in it occupying more space within the glass system, consequently increasing the molar volume of the glass (Luewarasirikul and Kaewkhao, 2018). When compared to the other two glass samples, the SrNd mixed glass sample exhibited higher density and refractive index values.

It is noted that the refractive index increases with the increase in the cationic radius of the network modifier element. Additionally, it is well-established that non-bridging oxygen (NBO) with increased polarizability can elevate the index of refraction in the glass host. The higher atomic mass of Sr^{2+} compared to other alkali ions contributes to greater compactness in glass structure. This is evident in the density of SrNd glass, which is 2.751 g/cm^3 , owing to the relatively large atomic mass (115.49 g/mol) of strontium. The high ionic radius contributes to greater glass compactness primarily due to its influence on the packing efficiency of atoms and ions within the glass matrix. When an ion with a larger ionic radius, such as Sr^{2+} , is incorporated into the glass structure, it affects the spacing between neighboring ions and atoms.

Specifically, a larger ionic radius allows the ions to occupy more space within the crystal lattice, leading to closer packing of atoms and ions. This results in a more efficient arrangement of the constituent particles, with less vacant space between them. As a consequence, the glass becomes denser, as a greater amount of mass is packed into a given volume. In summary, the high ionic radius of Sr^{2+} promotes a tighter and more orderly packing of atoms and ions within the glass matrix, ultimately leading to greater glass compactness (Kirdsiri *et al.*, 2018).

The polaron radius, denoted as r_p , must exceed the radius of the atom where the electron is localized but remain less than the distance r between these sites. According to Table 1, there is an observable increase in the polaron radius (r_p) as we move through different alkali earth elements. This increase is attributed to providing flexibility to the asymmetry around the Nd^{3+} ion within the host matrices. Notably, in SrNd glass, the polaron radius is observed to be higher compared to other hosts, potentially contributing to the highest emission from this particular host. In contrast, the r_p values for MgNd and CaNd glasses are lower, suggesting a possible congestion of Nd^{3+} ions within the glass

Table 1. Physical and optical characteristics of Nd^{3+} ions in $\text{P}_2\text{O}_5\text{-Al}_2\text{O}_3\text{-Li}_2\text{O-MO}$ ($\text{M} = \text{Mg, Ca, and Sr}$)

Physical properties	MgNd	CaNd	SrNd
Molecular mass (M_r , g/mol)	105.99	108.36	115.49
Density (ρ , g/cm ³)	2.574	2.602	2.751
Molar volume (V_m , cm ³ /mol)	41.172	41.650	41.977
Refractive index – at 589.3 nm (n)	1.5245	1.5325	1.5366
Nd^{3+} ions concentration (N_i , $\times 10^{20}$ ions/cm ³)	2.926	2.892	2.870
Polaron radius of Nd^{3+} (R_p , Å)	6.071	6.094	6.110
Inter-ionic distance of Nd^{3+} (R_i , Å)	15.064	15.122	15.161
Molar fraction (R_m , cm ³ /mol)	12.608	12.916	13.101
Electronic polarizability (α_m , Å ³)	5.005	5.128	5.201
Ionic Radii (Å)	0.72	1.00	1.18
Atomic Mass (g/mol)	24.30	40.07	87.62

host. Table 1 provides clear evidence that glasses doped with Mg²⁺ exhibit lower electronic polarizability compared to those doped with other cations. This suggests a higher level of ionicity in these glass samples, leading to a reduced inter-ionic radius compared to other glass compositions (Kirdsiri *et al.*, 2018).

Absorption Spectra

The optical absorption spectra of Nd³⁺-doped MgNd, CaNd, and SrNd glass systems are presented in Figure 1. Phosphate glass exhibits clear and sharp absorbance peaks due to the unique properties of the doped rare earths. Nd³⁺ ions are characterized by the arrangement of electrons within an unfilled 4f shell (Said Mahraza *et al.*, 2018). The distinctive absorption bands are ascribed to the shifts from the ground level ⁴I_{9/2} to the elevated energy levels of Nd, while the absorption peaks are ascribed to the Nd³⁺ transitions from the ground ⁴I_{9/2} to the excited ⁴D_{1/2} + ⁴D_{3/2}, ²P_{1/2}, ⁴G_{11/2}, ²K_{5/2} + ²G_{9/2} + ²D_{3/2}, ⁴G_{9/2}, ⁴G_{7/2}, ⁴G_{5/2}, ²H_{11/2}, ⁴F_{9/2}, ⁴F_{7/2} + ⁴S_{3/2}, ⁴F_{5/2} + ²H_{9/2}, ⁴F_{3/2} manifolds (Brahmachary *et al.*, 2014; Nayab *et al.*, 2017). Due to its hypersensitivity transition governed by specific selection rules $|\Delta S|=0$, $|\Delta L| \leq 2$ and $|\Delta J| \leq 2$, the transition ⁴I_{9/2} → ⁴G_{5/2} has the highest intensity, which was measured at 580 nm (Prasad *et al.*, 2019). The absorption ability, in order of the peak at 580 nm, is SrNd > CaNd > MgNd, as illustrated in Figure 1.

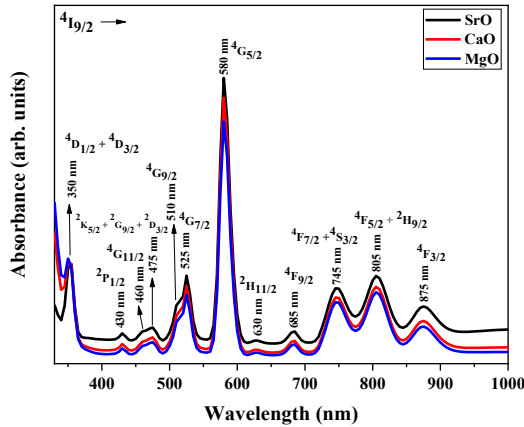


Figure 1. The UV-vis-NIR absorption spectrum of Nd³⁺ ions in P₂O₅-Al₂O₃-Li₂O-MO (M = Mg, Ca, and Sr)

Judd-Ofelt Theory

For quantitative spectroscopic investigation of RE ions imbedded in various settings, the Judd-Ofelt (JO) theory has served as a key foundation. The intensity of an absorption band can be expressed in terms of the “oscillator strength”.

The experimental oscillator strength (f_{exp}) of the absorption transitions was determined using the following relation (Kirdsiri *et al.*, 2018)

$$f_{exp} = 4.32 \times 10^{-9} \int \varepsilon(v) dv \quad (1)$$

where $\varepsilon(v)$ is the molar absorptivity of a band at the mean energy v (cm^{-1}). The intensities of all the bands of absorption detected in the present instance were calculated using the area approach. Assuming that every ground state crystal field split level is evenly populated, the computed oscillator strength for an induced electric dipole and magnetic dipole transition from ground state ψJ to an excited state $\psi' J'$ is given by eq. (2) in accordance with JO theory (Kirdsiri *et al.*, 2018).

$$f_{cal} = \frac{8\pi^2 m c v}{3 h (2J+1)} \frac{(n^2+2)^2}{9n} \sum_{\lambda=2,4,6} \Omega_{\lambda}(\psi J \| U^{\lambda} \| \psi' J')^2 \quad (2)$$

Where n represents the refractive index of the medium, J denotes the ground state total angular momentum, v signifies the energy of the transition in cm^{-1} , Ω_{λ} ($\lambda = 2, 4, 6$) stands for the JO parameters and $\|U^{\lambda}\|^2$ refers to the squared doubly reduced matrix elements of the unit tensor operator with rank $\lambda = 2, 4, 6$. These elements are computed from the intermediate coupling approximation for the transition ψJ to $\psi' J'$ at energy v and remain independent of the host.

Table 2 shows the oscillator strengths of the detected absorption bands of Nd³⁺-doped glass matrix measured (f_{exp}) and calculated (f_{cal}) using the method described in eq. (1) and (2). The f_{exp} and f_{cal} values are maximum at 580 nm, which corresponds to the ⁴I_{9/2} → ⁴G_{5/2} transition, which is consistent with the absorption experiment results. The ⁴I_{9/2} → ⁴G_{5/2} transition is the highest because the ligand is strongly covalent and the surrounding Nd³⁺ ions are less symmetrical than the other absorption bands which atomic environment surrounding the Nd³⁺ ions during the specified transition (⁴I_{9/2} → ⁴G_{5/2}). Essentially, it indicates that the arrangement of ligands or atoms around the Nd³⁺ ions during this transition are not as balanced or uniform compared to other absorption bands. the behavior of molecules or atoms surrounding Nd³⁺ ions, the Franck-Condon theory can be employed to elucidate the concept of asymmetry in their distribution. When atoms disperse with symmetry, the duration of dispersion tends to produce regular oscillations akin to orderly wave patterns. Conversely, asymmetrical dispersion may result in irregular wave patterns. Therefore, in absorption bands characterized by asymmetry, the dispersion of molecules or atoms around Nd³⁺ ions may also be asymmetric. This can lead to

Table 2. The values of the experimental ($f_{exp} \times 10^{-6}$) and calculated ($f_{cal} \times 10^{-6}$) oscillator strengths and root mean square deviation ($\delta_{rms} \times 10^{-6}$) of Nd^{3+} ions in $P_2O_5-Al_2O_3-Li_2O-MO$ (M = Mg, Ca, and Sr)

Transition $^4I_{9/2}$	λ (nm)	MgNd		CaNd		SrNd	
		$f_{exp} \times 10^{-6}$	$f_{cal} \times 10^{-6}$	$f_{exp} \times 10^{-6}$	$f_{cal} \times 10^{-6}$	$f_{exp} \times 10^{-6}$	$f_{cal} \times 10^{-6}$
$^4D_{1/2} + ^4D_{3/2}$	350	0.6582	0.9877	4.2302	2.6817	5.0531	3.0913
$^2P_{1/2}$	430	0.0761	0.1349	0.4408	0.3100	0.493	0.3574
$^4G_{11/2}$	460	0.0393	0.1109	0.3142	0.1267	0.3913	0.1517
$^2K_{5/2} + ^2G_{9/2} + ^2D_{3/2}$	475	0.0785	0.273	0.8077	0.2622	1.0284	0.3123
$^4G_{9/2}$	510	0.2464	0.3775	1.113	0.8461	1.4005	1.0034
$^4G_{7/2}$	525	0.527	0.609	2.0063	1.8898	2.4583	2.2372
$^4G_{5/2}$	580	1.9823	1.9773	7.1413	7.1485	8.6096	8.6232
$^2H_{11/2}$	630	0.0315	0.0297	0.104	0.0965	0.1147	0.1166
$^4F_{9/2}$	685	0.1146	0.0883	0.2722	0.3493	0.3245	0.4227
$^4F_{7/2} + ^4S_{3/2}$	745	0.5301	0.8073	2.5442	1.5526	3.044	1.8924
$^4F_{5/2} + ^2H_{9/2}$	805	1.0866	0.9252	2.9473	3.478	3.5577	4.1698
$^4F_{3/2}$	875	0.3384	0.2812	0.945	1.2579	0.9853	1.4704
δ_{rms}		0.154		0.592		0.741	

Table 3. The JO parameters ($\Omega_\lambda \times 10^{-20} \text{ cm}^2$) of Nd^{3+} ions in $P_2O_5-Al_2O_3-Li_2O-MO$ (M = Mg, Ca, and Sr)

Glass	Ω_2	Ω_4	Ω_6	$\chi = \Omega_4 / \Omega_6$	Ω_λ Trend	References
MgNd	0.55	0.60	1.13	0.53	$\Omega_6 > \Omega_4 > \Omega_2$	PW
CaNd	1.86	2.44	3.28	0.74	$\Omega_6 > \Omega_4 > \Omega_2$	PW
SrNd	2.29	2.81	3.98	0.70	$\Omega_6 > \Omega_4 > \Omega_2$	PW
PMNMg	2.55	3.74	4.12	0.91	$\Omega_6 > \Omega_4 > \Omega_2$	Prasad <i>et al.</i> , 2019
LaBO	0.54	2.31	4.51	0.51	$\Omega_6 > \Omega_4 > \Omega_2$	Jia <i>et al.</i> , 2004
CaNd4	1.09	1.97	3.37	0.58	$\Omega_6 > \Omega_4 > \Omega_2$	Chanthima <i>et al.</i> , 2017
BiPbGaNd	0.95	2.01	4.30	0.47	$\Omega_6 > \Omega_4 > \Omega_2$	Babu <i>et al.</i> , 2006
LG-750	4.6	4.8	5.6	0.86	$\Omega_6 > \Omega_4 > \Omega_2$	Campbell and Suratwala, 2000

absorption bands with higher energy levels or irregular characteristics compared to other absorption bands characterized by greater symmetry in molecular distribution (Luis *et al.*, 2004; Said Mahraza *et al.*, 2018). The Ω_2 , Ω_4 and Ω_6 values were obtained through least-square fitting between f_{exp} and f_{cal} . Radiative transition probabilities (A_R), stimulated emission cross-sections (σ_e), branching ratios (β_R), radiative lifetime (τ_R), and quantum efficiency (η) can all be calculated using the JO parameters. These parameters are utilized to assess the suitability of Nd^{3+} -doped host glass for laser applications (Kirdsiri *et al.*, 2018).

Table 3 presents the parameters of JO and spectroscopic quality factor (χ) of MgNd, CaNd, and SrNd. “ Ω_λ ” are crucial intensity factors to look at the bonding and local structures around rare earth ions. In the glass matrix, the parameter Ω_2 describes the environmental asymmetry surrounding Nd^{3+} ions and the chemical nature bonds between Nd^{3+} to O ions (Said Mahraza *et al.*, 2018). Higher Ω_2 values suggest that there is more asymmetric and covalent bonding between Nd^{3+} ions. The results showed that $\Omega_2(\text{Sr}) > \Omega_2(\text{Ca}) > \Omega_2(\text{Mg})$, indicating that SrNd has a higher disordered glass network modification than CaNd and MgNd, respectively. The higher values of oscillator strengths further demonstrate the stronger asymmetry at the Nd^{3+} ion site in the Sr glass (Prasad *et al.*, 2020).

The parameter Ω_4 is commonly associated with the long-range potential, whereas Ω_6 may be linked to the system’s rigidity. Increasing Ω_4 with increasing suggests potentially far-reaching effects on the host matrix around Nd-O. In simpler terms, the “far-reaching effects” refer to the wide-ranging impact or implications that changes in the Ω_4 parameter can have on the overall structure and properties of the material surrounding the Nd-O bonds (Oliveira Dantas *et al.*, 2013). It is well known that when a system’s mechanical rigidity rises, Ω_6 reduces. From Table 3, the value $\Omega_6(\text{Sr}) > \Omega_6(\text{Ca}) > \Omega_6(\text{Mg})$ indicates that MgNd has a hardness of SrNd and CaNd compared to other glasses in the experiment.

The ratios of Ω_4 and Ω_6 directly influence the parameters A_R and β_R . According to the literature, when the spectroscopic quality factor (χ) is less than 1, the intensity of the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition surpasses that of the $^4F_{3/2} \rightarrow ^4I_{9/2}$ transitions, suggesting potential laser emission at 1.06 μm . A lower χ value indicates a stronger transition for the $^4F_{3/2} \rightarrow ^4I_{11/2}$ laser. The use of the spectroscopic quality factor (χ) allows for the prediction of stimulated emission in the laser medium (Thongyoy *et al.*, 2022). Therefore, the JO parameters serve as indicators of a glass’s potential for laser applications.

Table 4. Stimulated emission cross-section ($\sigma_e \times 10^{-20} \text{ cm}^2$), radiative transition probability (A_R, s^{-1}), calculated and experimental branching ratio (β_{cal} and β_{exp}) of Nd³⁺ doped alkali earth phosphate glasses

Transition		$\sigma_e(\lambda_p)$	A_R	β_R		$\tau_R \text{ (ms)}$
				β_{cal}	β_{exp}	
MgNd	$^4F_{3/2} \rightarrow ^4I_{9/2}$	0.684	172.66	0.3394	0.0157	1.965
	$^4F_{3/2} \rightarrow ^4I_{11/2}$	1.278	273.28	0.5373	0.6781	
	$^4F_{3/2} \rightarrow ^4I_{13/2}$	0.441	59.75	0.1175	0.3061	
CaNd	$^4F_{3/2} \rightarrow ^4I_{9/2}$	2.824	647.07	0.3830	0.0172	0.591
	$^4F_{3/2} \rightarrow ^4I_{11/2}$	3.513	857.03	0.5073	0.6681	
	$^4F_{3/2} \rightarrow ^4I_{13/2}$	0.986	176.42	0.1044	0.3147	
SrNd	$^4F_{3/2} \rightarrow ^4I_{9/2}$	2.850	769.73	0.3754	0.0182	0.493
	$^4F_{3/2} \rightarrow ^4I_{11/2}$	3.574	1,038.78	0.5126	0.6705	
	$^4F_{3/2} \rightarrow ^4I_{13/2}$	1.019	216.27	0.1067	0.3114	

NIR Emission Spectra and Radiative Properties

The $^4F_{3/2} \rightarrow ^4I_{11/2}$ fluorescence spectra of MgNd, CaNd, and SrNd glasses are shown in Figure 2. The wavelength range for MgNd, CaNd, and SrNd glasses is 750–1,500 nm with an excitation wavelength at 808 nm (laser lamp source). Three emission peaks are observed at approximately 899, 1,058, and 1,327 nm, corresponding to $^4F_{3/2} \rightarrow ^4I_{9/2}$, $^4F_{3/2} \rightarrow ^4I_{11/2}$, and $^4F_{3/2} \rightarrow ^4I_{13/2}$ transitions, respectively. The transition potential with the greatest intensity among these three bands is $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition (1,058 nm). The trend of emission intensity observed in Figure 2, with SrNd exhibiting the highest intensity, followed by CaNd and MgNd, is consistent with the stimulated emission cross-section (σ_e) and radiative transition probability (A_R) values presented in Table 4. Specifically, the trend aligns with the relationship where the values of σ_e are highest for SrNd, followed by CaNd and MgNd, $\sigma_e(\text{Sr}) > \sigma_e(\text{Ca}) > \sigma_e(\text{Mg})$, and the radiative transition probability follows a similar order $A_R(\text{Sr}) > A_R(\text{Ca}) > A_R(\text{Mg})$. This alignment suggests that the emission intensity is directly influenced by the stimulated emission cross-section and radiative transition probability. Higher values of these parameters for SrNd indicate a greater likelihood of stimulated emission and a higher efficiency of radiative transitions, leading to the observed higher emission intensity compared to CaNd and MgNd. The trend of emission intensity observed in Figure 2, where SrNd exhibits the highest intensity, followed by CaNd and then MgNd, can be attributed to several factors related to the interaction between the added elements (strontium, calcium, magnesium) and the Nd³⁺ ions in the phosphate glass. Strontium (Sr), being a larger cation with a greater ionic radius compared to calcium (Ca) and magnesium (Mg), is more effective in facilitating energy transfer processes with the Nd³⁺ ions. This enhanced energy transfer mechanism results in a higher population of excited Nd³⁺ ions and subsequently higher emission intensity. The addition of strontium to the phosphate glass

matrix may induce structural changes that favor the incorporation and distribution of Nd³⁺ ions within the glass lattice. This optimized structural environment can enhance the efficiency of energy transfer processes and increase emission intensity. The concentration of dopant ions (Nd³⁺, Sr²⁺, Ca²⁺, Mg²⁺) in the glass composition can significantly influence emission intensity. It is possible that the SrNd-doped glass has a higher concentration of Nd³⁺ ions compared to CaNd and MgNd-doped glasses, leading to increased emission intensity. The energy levels and transition probabilities of the Nd³⁺ ions may vary depending on the specific host lattice and dopant ions. Strontium may induce energy level modifications that favor higher transition probabilities for radiative transitions, resulting in enhanced emission intensity compared to calcium and magnesium (Kirdsiri *et al.*, 2018; Luewarasirikul and Kaewkhao, 2018; Kuwik *et al.*, 2020).

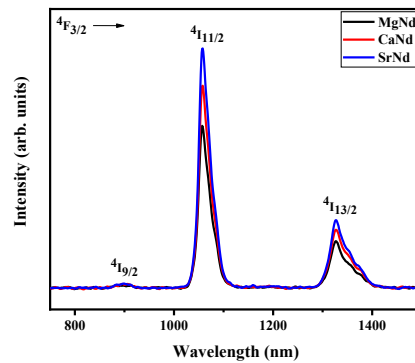
**Figure 2. NIR emission spectra of Nd³⁺ ions in P₂O₅-Al₂O₃-Li₂O-MO (M = Mg, Ca, and Sr)**

Figure 3 displays a partial energy level diagram of the Nd³⁺ ions and describes the mechanism of NIR luminescence in the Nd³⁺ ion doped in alkaline based P₂O₅-Al₂O₃-Li₂O-SrO glasses. Upon excitation at 808 nm, the Nd³⁺ ions were initially stimulated to the $^4F_{5/2}$ level. They then

where all the states at energy less than $\Psi'J'$ are included in the aggregate. The fluorescence branching ratio (β_R), a crucial radiative feature, is computed for distinct emissions with a same starting level based on

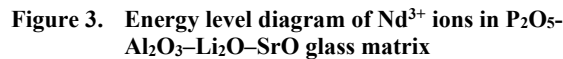
and is employed for predicting the relative strength of lines that come from a certain excited state.

Table 4 displays the radiative properties of the prepared glass system, encompassing radiative transition probabilities (A_R), stimulated emission cross-sections (σ_e), branching ratios (β_R), and radiative lifetimes (τ_R) of the optical transitions for MgNd, CaNd, and SrNd glasses from the emission level ${}^4F_{3/2}$. The findings of the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition revealed that the A_R of MgNd, CaNd, and SrNd are 273.28 s^{-1} , 857.03 s^{-1} , and 1038.78 s^{-1} , respectively. The stimulated-emission cross-section, (σ_e) ($\Psi J, \Psi' J'$), between the states ΨJ and $\Psi' J'$ with the probability of $A(\Psi J, \Psi' J')$, might be computed by:

where λ_p is the wavelength of the emission peak and $\Delta\lambda_{eff}$ is effective line width is determined by,

where I is the fluorescence intensity and $I_{\max}$ is the intensity at maximum. The transition from ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ possesses the highest stimulated emission cross-section (σ_e) value ($\times 10^{-20} \text{ cm}^2$) among the observed transitions, with values of 1.278, 3.513, and 3.574 for MgNd, CaNd, and SrNd, respectively. Upon comparing the near-infrared (NIR) emission at 1058 nm across different glass systems, it becomes evident that the current glass system exhibits a superior stimulated emission cross-section. This discovery suggests that the present glass formulation holds significant potential for applications in NIR lasing devices. Table 5 provides a comprehensive overview of the stimulated emission cross-section (σ_e) for the prepared glass samples.

The decay curves of Nd³⁺ ions in P₂O₅-Al₂O₃-Li₂O-MO (M = Mg, Ca, and Sr) glass system, were measured for excitation at 808 nm (using a laser lamp source) and emission at 1058 nm. These decay curves are depicted in Figure 4. The experimental


$$A(\Psi J, \Psi' J') = A_{ed} + A_{md} \quad (3)$$

where the electric and magnetic-dipole contributions, denoted as A_{ed} and A_{md} , respectively, are computed from

$$A_{ed} = \frac{64\pi^4 v^3}{3h(2J+1)} \frac{n(n^2+2)^2}{9} S_{ed} \quad (4)$$

$$A_{md} = \frac{64\pi^4 v^3}{3h(2J+1)} n^3 S_{md} \quad (5)$$

where the local field correction for electric dipole transitions is represented by $\frac{n(n^2+2)^2}{9}$, and for magnetic dipole transitions by n^3 . The electric and magnetic-dipole line strengths, denoted as S_{ed} and S_{md} , respectively, are determined using the following expressions:

$$S_{ed} = e^2 \sum_{\lambda=2,4,6} \Omega_{\lambda} (\Psi J \| U^{\lambda} \| \Psi' J')^2 \quad (6)$$

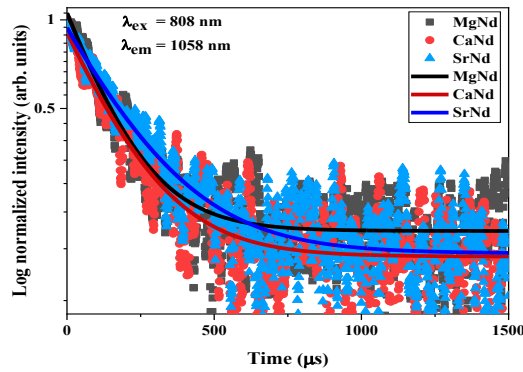
$$S_{md} = \frac{e^2 \hbar^2}{16\pi^2 m^2 c^2} (\Psi J \| L + 2S \| \Psi' J')^2 \quad (7)$$

The total radiative probability (A_T) for the states involved is equal to the sum of $A(\Psi J, \Psi' J')$,

$$A_T(\Psi J) = \sum_{\Psi' J'} A(\Psi J, \Psi' J') \quad (8)$$

Table 5. Comparison of stimulated emission cross-section ($\sigma_e \times 10^{-20} \text{ cm}^2$) of the transition $^4F_{3/2} \rightarrow ^4I_{11/2}$ for prepared glass system

Glass	$\sigma_e \times 10^{-20} \text{ cm}^2$	Reference
MgNd	1.278	PW
CaNd	3.513	PW
SrNd	3.57	PW
LG-770: PAKMg	3.90	Campbell and Suratwala, 2000
PKBALaNd	2.92	Tang <i>et al.</i> , 2021
PSN	2.31	Prasad <i>et al.</i> , 2020
PMN	3.26	Prasad <i>et al.</i> , 2019
PKSFAN	3.29	Vijaya <i>et al.</i> , 2009
PCBSN	3.90	Mazurak <i>et al.</i> , 2012

**Figure 4. Decay curves of the $^4F_{3/2}$ level in Nd³⁺-doped phosphate glasses with varying concentrations of Nd³⁺ ions**

lifetime (τ_{exp}) of the $^4F_{3/2}$ fluorescent level was computed from the exponential decay curves by determining the effective lifetime using the provided formula (Kirdsiri *et al.*, 2018).

$$\tau_{exp} = \frac{\int tI(t)dt}{\int I(t)dt} \quad (12)$$

where $I(t)$ denote the intensity of the emission at time 't'. For these glasses, the experimental lifetimes (τ_{exp}) were calculated by fitting the single exponential fit. The single exponential behavior of these glasses is evident with a Nd₂O₃ content concentration of 1 mol%. The SrNd sample has a larger experimental lifetime (τ_{exp}) in Figure 4 compared to the other samples. The difference in the size of Sr ions, which are larger than Ca and Mg, may lead to a greater ability to scatter light. This increased light scattering can potentially result in a longer decay time. Greater light scattering can help reduce the correlation between particles and energy accumulation, potentially decreasing the likelihood of energy loss and prolonging energy retention. Therefore, having larger Sr ions may increase the likelihood of longer decay times.

The potential substitution of Nd³⁺ ions at a different anionic site might be the cause of the short decay component. In Figure 4, lifetime values for several fluorescence are presented. Since the SrNd level's typical lifetime values were longer than those of the other levels, it is possible that the strontium phosphate host is a good fit for Nd³⁺ doping with minimal non-radiative losses. When compared to SrNd, the lifetimes were shortened by favoring the non-radiative energy transfer mechanisms (Balakrishna *et al.*, 2018).

The JO parameters were also utilized to compute the radiative lifetime (τ_R). The depopulation rate of a level can also be calculated using the emission probabilities that result from that level's starting population, ΨJ . The radiative lifetime, which can be expressed as the reciprocal of the total transition probability and is in accordance with JO theory (Kirdsiri *et al.*, 2018), is:

$$\tau_{rad}(\Psi J) = \frac{1}{\sum_{\Psi'J'} A_R(\Psi J, \Psi'J')} \quad (13)$$

The decay lifetimes are observed to increase with increasing ionic radius of alkaline earth in the phosphate glass matrix. Different lifetimes may result from variations in the ligand fields around the Nd³⁺ ion. This could result from the Nd³⁺ ion's greater asymmetry in the SrNd glass as opposed to the MgNd and CaNd glasses. In comparison to the MgNd and CaNd glasses in Table 2, it is discovered that the τ_{exp} value in the SrNd glass is greater. It is observed that addition of SrO may enhance the lifetime in the Nd³⁺-doped phosphate glasses (Prasad *et al.*, 2020). The lifetimes were found to be 152, 191, and 224 μ s for MgNd, CaNd, and SrNd glasses, respectively.

The quantum efficiency (η) is a critical metric defined as the ratio of radiation released to incident photons absorbed. It serves as a key factor in predicting the suitability of a material as a laser host. This prediction relies on various factors, including

Table 6. Radiative lifetimes, τ_R (μ s), experimental lifetimes, τ_{exp} (μ s), non-radiative energy transfer rate, W_{NR} (s^{-1}), and quantum efficiencies, η (%), gain bandwidth, $\sigma_e \times \Delta\lambda_{eff}$ ($\times 10^{-26} cm^3$), and optical gain, $\sigma_e \times \tau_{exp}$ ($\times 10^{-24} cm^2s$) of the $^4F_{3/2}$ level of MgNd, CaNd, and SrNd glasses

Glass	τ_R	τ_{exp}	W_{NR}	η	$\sigma_e \times \Delta\lambda_{eff}$	$\sigma_e \times \tau_{exp}$	References
MgNd	1965	152	6070	7.74	19.54	1.94	Present work
CaNd	591	191	3544	32.32	60.09	6.71	Present work
SrNd	493	224	2436	45.44	72.84	8.01	Present work
PSN10	451	256	1689	57.00	6.5	-	Prasad <i>et al.</i> , 2020
PKSAN10	319	172	2679	54.00	-	-	Kumar <i>et al.</i> , 2008
PANKB1Nd	990	300	2323	33.00	-	-	Ismail <i>et al.</i> , 2020
TMNd1.0	320	136	4255	42.31	95.2	10.7	Yaacob <i>et al.</i> , 2021
LPBTN1	171	73	7850	43.00	19.2	-	Madhu <i>et al.</i> , 2018

emission cross-sections, radiative transition probabilities, lifetimes of metastable states, concentration of rare-earth ions, and the ligand field effect (Kumar *et al.*, 2008). For Nd^{3+} ion-doped glasses, the ratio of τ_{exp} to τ_R is equivalent for the respective levels ($\eta = \tau_{exp}/\tau_R$) (Prasad *et al.*, 2020). In this experiment, SrNd glass exhibited the highest quantum efficiency among MgNd and CaNd. Specifically, SrNd glass achieved a maximum quantum efficiency of 45.44%, as indicated in Table 6. This low non-radiative relaxation is responsible for this excellent quantum efficiency. Two factors can influence the decay rate of an excited state: radiative decay $W_R = \frac{1}{\tau_R}$ and $W_{NR} = \frac{1}{\tau_{exp}} - \frac{1}{\tau_R}$ rates (Yaacob *et al.*, 2021). The change in the local crystal symmetry of Nd^{3+} is the main reason for the radiative decay rate. The depopulation of excited-state energy is caused by the host glass's local vibrational density of states (DOS). Nd^{3+} decays non-radiatively from its excited state due to the electron-phonon interaction. The $^4F_{3/2} \rightarrow ^4I_{15/2}$ transition for one Nd^{3+} ion and the $^4I_{9/2} \rightarrow ^4I_{15/2}$ transition for other Nd^{3+} ions are the pathways by which this energy resonance occurs. Then, non-radiatively, both ions decayed to the ground state $^4I_{9/2}$. The amplification of the lasing glass is predicted by the values of gain bandwidth $\sigma_e \times \Delta\lambda_{eff}$. Because it facilitates pulse laser activity with brief pulses, glass with a large gain bandwidth is helpful. In the case of continuous wave (CW) laser operations, the saturation intensity, which is dependent on the host material as well as the product of the emission cross-section of the lasing transition and the measured lifetime of the excited level, determines the input pump power needed to reach the threshold. As a result, glass with a higher $\sigma_e \times \tau_{exp}$ product will have a lower laser threshold and saturation intensity (Yaacob *et al.*, 2021). From Table 6, SrNd glass has the highest $\sigma_e \times \tau_{exp}$ value which causes low saturation intensity. For continuous wave laser operation in the near-infrared region, SrNd glass with $^4F_{3/2} \rightarrow ^4I_{15/2}$

transition, which has a large, stimulated emission cross section, low threshold, high gain bandwidth, and high optical gain, is thus a good choice.

Conclusions

This study investigates the effects of Nd^{3+} ions on the optical and physical properties of PALMNd glasses (where M = Mg, Ca, Sr), synthesized via the conventional melt-quenching technique. The concentration of $MO:Nd^{3+}$ (M = Mg, Ca, Sr) is found to significantly influence key physical parameters, including density, molar volume, refractive index, ion concentration, and polaron radius. Among the studied compositions, the SrNd glass sample exhibits the highest density, attributed to the larger atomic mass and ionic radius of Sr^{2+} . UV-Vis-NIR absorption spectra confirm characteristic Nd^{3+} transitions in all glass samples, with the SrNd glass showing the most intense absorption bands. Judd-Ofelt analysis reveals that the SrNd glass possesses higher Ω_2 values compared to its CaNd and MgNd counterparts, indicating stronger asymmetry and greater modification of the local environment around Nd^{3+} ions. The trend observed in the intensity parameter Ω_2 follows the order: $\Omega_2(Sr) > \Omega_2(Ca) > \Omega_2(Mg)$, suggesting a progressively stronger influence of Sr^{2+} on the disordered glass network. Radiative transition probability (A_r) and branching ratio (β_r) for Nd^{3+} fluorescence levels were calculated based on the derived Judd-Ofelt parameters. The near-infrared (NIR) emission spectra exhibit three main bands, with the most intense corresponding to the $^4F_{3/2} \rightarrow ^4I_{15/2}$ transition at 1058 nm. The SrNd glass shows enhanced luminescence intensity, attributed to a more favorable local environment for Nd^{3+} ions and a higher molar fraction of Nd^{3+} , which correlates with increased electronic polarizability. This enhancement is linked to a greater number of polarizable ions interacting with external electric fields. For the metastable $^4F_{3/2}$ level, radiative lifetime (τ_r), branching ratio (β_r), and radiative

transition probability (A_r) were evaluated. The SrNd glass exhibits advantageous laser parameters, including broad gain bandwidth, low saturation intensity, high emission efficiency ($\eta \approx 45.44\%$), and a high stimulated emission cross-section. These properties make SrNd glass a promising candidate for lasing applications at 1,058 nm.

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