

ND³⁺-DOPED BORATE GLASSES DEVELOPMENT FOR EFFICIENT NIR LASER MEDIUM MATERIAL

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Abstract

This study aims to determine the effect of neodymium concentration on barium sodium borate glasses. The glass samples were fabricated using the melt-quenching technique for analyzing the physical properties, absorption spectra, and NIR emission spectra. The twelve absorption bands increased with the increase in Nd³⁺ concentration. The most intense absorption band is centered at 583 nm (corresponding to the ⁴I_{9/2}→⁴G_{5/2} transition), which is chosen to investigate the NIR emission spectra. The NIR emission spectra exhibited the most intense emission band at 1,077 nm under 583 nm excitation wavelength, corresponding to the ⁴F_{3/2}→⁴I_{11/2} transition, respectively. The maximum NIR emission intensity was observed at the glass sample with 2 mol% of Nd³⁺. The NIR emission spectra and the results from Judd-Ofelt theory confirm that 2 mol% of Nd³⁺-doped Ba-Na-B glasses with a 1,077 nm emission in this work might be used as a NIR laser medium material.

Keywords: Borate glass; Judd-Ofelt theory; neodymium; NIR emission

Introduction

The development of lasers has become increasingly important in various fields, including telecommunications, medical sciences and material processing (Venkata Rao *et al.*, 2020; Nayab Rasool *et al.*, 2022; Ullah *et al.*, 2022; Kotb *et al.*, 2023). The doping substance used to deliver active ions into the laser medium is one of the crucial elements that affect how well a laser device performs. Among various

doping substances, rare-earth ions have gained significant attention due to their unique optical properties, which make them highly desirable for many applications. The rare-earth ions are located in the lanthanide series, that they have incompletely filled 4f electronic shells. The 4f electrons are well-shielded from the outer electrons, resulting in narrow energy levels and sharp optical transitions

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that are highly sensitive to their local environment. This sensitivity to the environment allows rare-earth ions to exhibit a range of optical properties, such as strong absorption and emission bands, long lifetimes, and narrow emission linewidths. These properties make rare-earth ions ideal for use in optical devices, such as lasers, amplifiers, and sensors, as well as in various other applications, such as telecommunication, lighting, and bioimaging (Yua *et al.*, 2018; Shoaib *et al.*, 2019; Venkata Rao *et al.*, 2020; Lin *et al.*, 2022; Nayab Rasool *et al.*, 2022; Ullah *et al.*, 2022; Kotb *et al.*, 2023).

Among the rare-earth, Nd³⁺ ions are suitable ions for use as an active medium in solid-state laser because of their unique optical properties. When excited by an external energy source, such as a flashlamp or a diode laser, the Nd³⁺ ions absorb energy and transition to an excited state. As these excited ions return to their ground state, they emit light in the form of photons. This process, known as stimulated emission, creates a beam of coherent light that is amplified, resulting in a powerful laser beam. One of the key advantages of Nd³⁺ ions is their broad NIR emission band that is useful in various applications. One important application is in telecommunications, where Nd³⁺ doped fibers are used as amplifiers in optical communication systems. The broad NIR emission band of Nd³⁺ ions allows for amplification of multiple channels simultaneously, making it possible to transmit large amounts of data over long distances. In addition to telecommunications, Nd³⁺ doped materials are also used in other fields, such as laser technology. Nd³⁺ doped materials can be used as gain media in lasers, where the broad NIR emission band allows for tunable output wavelengths (Yua *et al.*, 2018; Shoaib *et al.*, 2019; Venkata Rao *et al.*, 2020; Lin *et al.*, 2022; Nayab Rasool *et al.*, 2022; Ullah *et al.*, 2022; Kotb *et al.*, 2023). By understanding the luminescence properties of rare-earth ions, researchers can design materials with specific properties, such as high quantum efficiency, long lifetime and specific emission bands.

Nd:YAG laser is a type of solid-state laser that uses a neodymium-doped yttrium aluminum garnet (Nd:Y₃Al₅O₁₂) crystal as the gain medium. It emits a near-infrared laser beam, which is widely used in

various applications. Nd:YAG laser is known for its high laser power, reliability, long life and high beam quality, making it is a popular choice for many applications. However, the fabrication of crystal is complex and requires specialized equipment. The process typically involves growing the Nd:YAG crystal using a technique such as the Czochralski method, followed by cutting, polishing and doping the crystal with neodymium ions. The fabrication process is more time-consuming and requires higher purity materials, which can make Nd:YAG laser is expensive. Glass, with a more straightforward fabrication process, lower cost and greater flexibility in the design, is introduced for using as Nd³⁺-doped glass laser. In general, Nd³⁺-doped glass laser is less expensive and easier to prepare compared to Nd:YAG lasers. But when compared to Nd³⁺-doped glass laser, Nd:YAG laser has a higher gain coefficient, which means they can produce higher power and higher energy density laser beams (Nayab Rasool *et al.*, 2022; Kotb *et al.*, 2023).

The objective of this work is to develop Nd³⁺-doped glass with better laser efficiency. Barium sodium borate glass is chosen as a host material for incorporating Nd³⁺ ions. In fact, borate glass is not suitable for using as a RE³⁺-doped host glass because it provides very low mechanical and chemical durability. Barium and sodium adding to the borate glass, causing the glass to have better mechanical and chemical durability and also has a low melting temperature (Venkata Rao *et al.*, 2020; Nayab Rasool *et al.*, 2022; Ullah *et al.*, 2022; Kotb *et al.*, 2023; Ahmad *et al.*, 2019). These properties make it possible to incorporate the Nd³⁺ ions into the Ba-Na-B glass for using as an efficient glass laser.

Experimental Details

The Ba-Na-B glasses doping with varying concentrations of Nd³⁺ from 0 to 2.5 mol% were fabricated by the melt-quenching technique, with the chemical compositions as showed in Table 1 as (65-x)Ba₂O₃-25Na₂O-10BaO:xNd, when x = 0, 0.5, 1.0, 1.5, 2.0, and 2.5 mol%. The fabrication of the glass samples typically involves melting and casting

Table 1. Glass compositions

Glass name	B ₂ O ₃ (mol%)	Na ₂ O (mol%)	BaO (mol%)	Nd ₂ O ₃ (mol%)
BaNaB:0Nd	65	25	10	0
BaNaB:0.5Nd	64.5	25	10	0.5
BaNaB:1.0Nd	64	25	10	1
BaNaB:1.5Nd	63.5	25	10	1.5
BaNaB:2.0Nd	63	25	10	2
BaNaB:2.5Nd	62.5	25	10	2.5

the glasses in a furnace at 900°C for 3 h, followed by thermal treatment by annealing the glasses at 500°C for another 3 h. Then the glasses were polished to produce the final laser material with the dimension of 1.0×1.5×0.3 cm³ for better optical properties and suitable operation in the instruments. The density of the glass sample (ρ) was calculated by the Archimedes' method, as the following equation:

$$\rho = (m_a / m_a - m_w) \rho_w$$

where m_a and m_w are the mass of a glass measured in air and water, respectively, and ρ_w is the density of water (1 g/cm³). The molar volume can also vary depending on the composition and dopant concentration. One way to estimate the molar volume is to use the density and molecular weight of the glass. The molecular weight can be calculated from the chemical formula of the glass, considering the dopant concentration. The molar volume (V_m) can be calculated by:

$$V_m = M / \rho$$

where M is the molecular weight. The Abbe refractometer is a commonly used instrument for measuring the refractive index of a glass with the sodium vapor lamp that emits a very specific wavelength of light at 589.3 nm. Shimadzu

UV-3,600 UV-Vis-NIR spectrophotometer is a type of spectrophotometer that can measure the absorption spectra over a wide range of UV, visible and near-infrared wavelengths for measure the absorption spectra. The emission spectra in the near-infrared region of the electromagnetic spectrum were obtained by HORIBA PTI QuantaMaster 300 Spectrofluorometer. The glass samples are typically excited with a suitable light source and the resulting NIR luminescence was recorded by a sensitive photomultiplier tube. The Judd-Ofelt theory, used for analyzing the radiative properties of rare-earth ions in materials, was also employed in this work.

Results and Discussions

As the Nd³⁺ concentration increased from 0 to 2.5 mol%, the density, molar volume and refractive index all increase from 2.6721-2.7450 g/cm³, 28.4722-29.7972 cm³/mol and 1.5405-1.5601, respectively, as showed in Table 2. These results suggest that the incorporation of Nd³⁺ ions in the glass causes the increasing of all three parameters. The increasing of density is likely due to a higher molecular weight of Nd₂O₃ than B₂O₃. The addition of Nd³⁺ ions to the glass matrix results in the increasing of the average interatomic distance between the glass ions. While the refractive index is increased related to the increasing of the density (Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Thongyoy *et al.*, 2022; Ullah *et al.*, 2022; Venugopal *et al.*, 2022).

The absorption spectra of the glasses with different concentrations of Nd³⁺ ions exhibit the series of absorption bands, which correspond to electronic transitions of Nd³⁺ ions between energy levels according to the transitions from ⁴I_{9/2} to ²P_{1/2} (431 nm), ⁴G_{11/2} (459 nm), ²K_{15/2} (476 nm), ⁴G_{9/2} (511 nm), ⁴G_{7/2} (529 nm), ⁴G_{5/2} (583 nm), ²H_{11/2} (626 nm), ⁴F_{9/2} (683 nm), ⁴F_{7/2} (743 nm), ⁴F_{5/2} (808 nm) and ⁴F_{3/2} (878 nm) (Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Thongyoy *et al.*, 2022; Venugopal *et al.*, 2022), as showed in Figure 1. The absorption intensity increases correlated with the increasing of Nd³⁺ concentration.

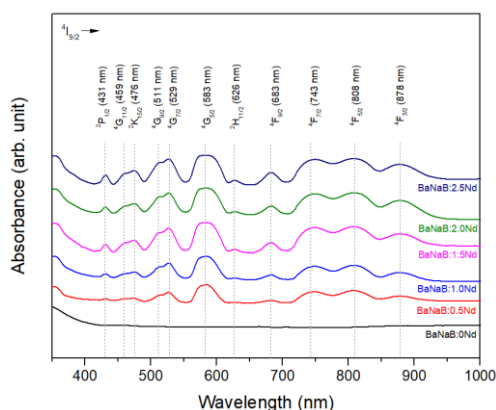


Figure 1. The absorption spectra

Table 2. Density, molar volume and refractive index

Glass name	Density (g/cm ³)	Molar volume (cm ³ /mol)	Refractive index
BaNaB:0Nd	2.6721	28.4722	1.5405
BaNaB:0.5Nd	2.6899	28.7798	1.5487
BaNaB:1.0Nd	2.7087	29.0727	1.5513
BaNaB:1.5Nd	2.7198	29.4446	1.5579
BaNaB:2.0Nd	2.7324	29.7972	1.5582
BaNaB:2.5Nd	2.7450	30.1465	1.5601

The Judd-Ofelt theory involves calculating a set of parameters that describe the strength of the interaction between the rare-earth ion and its ligands. These parameters, derived from the analysis of the absorption and emission spectra of the material, are important in the development of photonic materials for various technologies, including lighting, lasers and telecommunications (Ahmad *et al.*, 2019; Venkata Rao *et al.*, 2020; Nayab Rasool *et al.*, 2022; Kotb *et al.*, 2023). The Judd-Ofelt can be used for determining the experimentally or theoretically based oscillator strength, a dimensionless quantity that relates to the probability of an electronic transition between energy states of the electronic transition, from the absorption spectra. The experimental oscillator strength (f_{exp}) can be evaluated from the molar absorptivity of each absorption band. The calculated or theoretical oscillator strength (f_{cal}) can be calculated by the Judd-Ofelt theory (Zhou *et al.*, 2019; Lalla *et al.*, 2020; Venkata Rao *et al.*, 2020; Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Nayab Rasool *et al.*, 2022; Venugopal *et al.*, 2022; Kotb *et al.*, 2023). Table 3 shows the oscillator strengths of the BaNaB:2.0Nd glass sample. The $^4I_{9/2} \rightarrow ^4G_{5/2}$

transition (583 nm) provides the highest both the experimental and calculated oscillator strengths.

The J-O parameters (Ω_2 , Ω_4 and Ω_6) can be obtained by fitting f_{exp} to f_{cal} from Judd-Ofelt theory model with least-square fitting approach. The value of Ω_2 directly depends on the local asymmetry around the rare-earth ion, as well as the nature of the ligands. A higher value of Ω_2 indicates a stronger interaction between the rare-earth ion and its ligands, which may lead to more intense emission efficiency (Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Venugopal *et al.*, 2022). While the ratio of Ω_4 and Ω_6 can be used to calculate the strengths spectroscopic quality factor (Ahmad *et al.*, 2019). The J-O parameters are found to be $\Omega_2 = 1.92 \times 10^{-20} \text{ cm}^2$, $\Omega_4 = 1.23 \times 10^{-20} \text{ cm}^2$ and $\Omega_6 = 0.96 \times 10^{-20} \text{ cm}^2$, following the $\Omega_2 > \Omega_4 > \Omega_6$ trend which indicates the good lasing ability of this host glass (Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Venugopal *et al.*, 2022). The strengths spectroscopic quality factor (Ω_4/Ω_6) is 1.28. The root-mean-square deviation (RMSD) evaluated from the fitting between f_{exp} and f_{cal} via least-square fitting of the BaNaB:2.0Nd glass is 0.127. The RMSD measures how well the fitted curve matches the experimental or theoretical data (Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Venugopal *et al.*, 2022). A small value of RMSD at 0.127 of the BaNaB:2.0Nd glass indicates that the two sets of fitted data are very close. In other words, the Judd-Ofelt theory can be evaluated in this work.

The NIR emission spectra of Nd³⁺-doped glass with varying Nd³⁺ concentrations were shown in Figure 2. When excited with 808 nm, the spectra showed the emission band centered 937, 1,077, and 1,346 nm, which is attributed to the $^4F_{3/2} \rightarrow ^4I_{9/2}$, $^4I_{11/2}$ and $^4I_{13/2}$ transition of Nd³⁺ ions, respectively (Venkata Rao *et al.*, 2020; Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Nayab Rasool *et al.*, 2022;

Table 3. The oscillator strengths of the BaNaB:2.0Nd glass

Transitions	λ (nm)	f_{exp} ($\times 10^{-6}$)	f_{cal} ($\times 10^{-6}$)
$^4I_{9/2} \rightarrow ^2P_{1/2}$	431	0.3438	0.1895
$^4I_{9/2} \rightarrow ^4G_{11/2}$	459	0.8765	1.0944
$^4I_{9/2} \rightarrow ^2K_{15/2}$	476	0.3537	0.1958
$^4I_{9/2} \rightarrow ^4G_{9/2}$	511	0.9785	0.6389
$^4I_{9/2} \rightarrow ^4G_{7/2}$	529	1.1253	1.4133
$^4I_{9/2} \rightarrow ^4G_{5/2}$	583	6.0944	6.4566
$^4I_{9/2} \rightarrow ^2H_{11/2}$	626	0.0984	0.1211
$^4I_{9/2} \rightarrow ^4F_{9/2}$	683	0.3149	0.4035
$^4I_{9/2} \rightarrow ^4F_{7/2}$	743	2.5833	1.9438
$^4I_{9/2} \rightarrow ^4F_{5/2}$	808	2.6147	3.2685
$^4I_{9/2} \rightarrow ^4F_{3/2}$	878	0.9435	0.6745

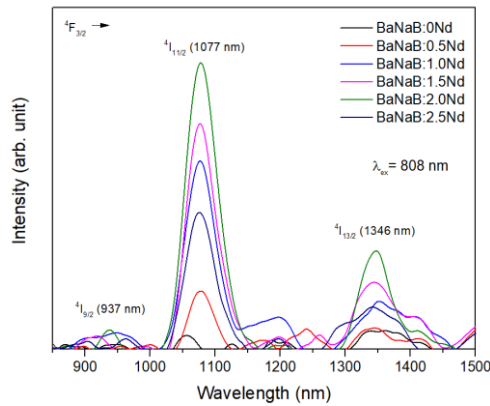


Figure 2. The emission spectra

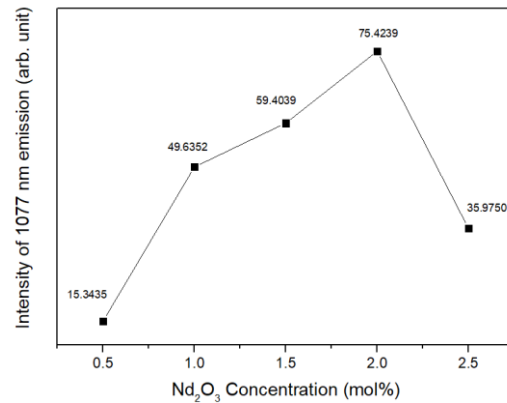


Figure 3. The emission intensity at 1,077 nm

Thongyoy *et al.*, 2022; Ullah *et al.*, 2022; Venugopal *et al.*, 2022; Kotb *et al.*, 2023). The greatest emission yield belongs to the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition at 1,077 nm. The intensity of this emission band increased with increasing Nd^{3+} concentration up to 2 mol% and then decreased at higher concentrations. The maximum NIR emission intensity was observed at the BaNaB:2.0Nd glass (Figure 3). The observed variation in NIR emission intensity can be attributed to the emission of Nd^{3+} ions. At low Nd^{3+} concentrations, there are fewer Nd^{3+} ions to emit the photon, resulting in a weaker emission band. As the concentration increases, more Nd^{3+} ions are present, leading to an increase in emission intensity. However, as the concentration of Nd^{3+} increases, the probability of energy transfer between neighboring ions also increases. This energy transfer process is a nonradiative process, that may lead to a decreasing in emission intensity. So the concentration quenching effect occurs when the energy transfer processes become dominant at high rare-earth ion concentrations, leading to a decrease in the luminescent efficiency.

The radiative transition possibility (A_R), stimulated emission cross section (σ) and branching ratio (B_R) are important parameters in luminescent materials that can be calculated from the emission spectra by the Judd-Ofelt theory. The radiative lifetime (τ_R) can be calculated from the inverse of the sum of radiative transition possibility values from all emission transitions (Venkata Rao *et al.*, 2020; Borisut *et al.*, 2022; Kaewnuam *et al.*, 2022; Nayab Rasool *et al.*, 2022; Venugopal *et al.*, 2022; Kotb *et al.*, 2023). Lasing gain, which provides the amount of amplification that occurs in the gain medium of a laser, can be calculated by the product of σ and τ_R . Bandwidth or gain bandwidth, which shows the range of wavelengths over which the gain medium of the laser can amplify light, can be calculated by the product of σ and FWHM (full width at half maximum) (Kaewnuam *et al.*, 2022; Venugopal *et al.*, 2022). The radiative lifetime of the BaNaB:2.0Nd glass is 865 ns. Table 4 shows the radiative parameters of the BaNaB:2.0Nd glass. The A_R (which presents the transition between two energy levels), σ (which presents the ability of a material to amplify light through the emission) and B_R (which presents the relative contribution of a specific transition to the total emission) of the

$^4F_{3/2} \rightarrow ^4I_{11/2}$ emission transition are all higher than other transitions. So, the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition is suitable for using as the pumping in NIR laser. Furthermore, the efficient NIR laser medium material should have higher values of the lasing gain and gain bandwidth. For the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition, the lasing gain is found to be $1.89 \times 10^{-25} \text{ cm}^2\text{s}$ and the gain bandwidth is $124.26 \times 10^{-29} \text{ cm}^3$, that are all higher than other transitions. This result confirms the suitability of this transition for using as the pumping in NIR laser. These radiative properties indicate the possibility of the Ba-Na-B:2.0 glass with 2 mol% of Nd^{3+} ions in this work for using as an efficient NIR laser medium material.

Conclusions

In this work, The Ba-Na-B glasses doped with the varying of Nd^{3+} concentration of 0, 0.5, 1.0, 1.5, 2.0, and 2.5 mol% were fabricated. When excited with 808 nm, the major broad emission band was centered around 1,077 nm ($^4F_{3/2} \rightarrow ^4I_{11/2}$). The J-O parameters following the $\Omega_2 > \Omega_4 > \Omega_6$ trend, which confirms the good lasing ability of this host glass. The maximum NIR emission intensity was observed at the glass sample with 2 mol% of Nd (BaNaB:2.0Nd). The radiative lifetime of the BaNaB:2.0Nd glass is 865 ns. The A_R (832 s^{-1}), σ ($2.18 \times 10^{-22} \text{ cm}^2$), B_R (0.72), lasing gain ($1.89 \times 10^{-25} \text{ cm}^2\text{s}$) and gain bandwidth ($124.26 \times 10^{-29} \text{ cm}^3$) of the $^4F_{3/2} \rightarrow ^4I_{11/2}$ emission transition are all higher than other transitions. So, the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition is suitable for using as the pumping in NIR laser. The results show that Ba-Na-B:2.0 glass with 2 mol% of Nd^{3+} ions in this work has the performance and efficiency for using as NIR laser medium material.

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Table 4. The radiative properties of the BaNaB:2.0Nd glass

Transitions	λ (nm)	A_R (s^{-1})	σ ($\times 10^{-22} \text{ cm}^2$)	B_R	FWHM (nm)	Lasing gain ($\times 10^{-25} \text{ cm}^2\text{s}$)	Bandwidth ($\times 10^{-29} \text{ cm}^3$)
$^4F_{3/2} \rightarrow ^4I_{13/2}$	937	109	0.18	0.02	27.69	0.16	4.98
$^4F_{3/2} \rightarrow ^4I_{11/2}$	1077	832	2.18	0.72	57.00	1.89	124.26
$^4F_{3/2} \rightarrow ^4I_{9/2}$	1346	215	0.94	0.26	54.58	0.81	51.31

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