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DOI: 10.21776/ub.jpacr.2016.005.02.265 J. Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal homepage www.jpacr.ub.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 This is an open access article distributed under the terms of the Creative Commons Attribution-NonCommercial 4.0 International which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited. (<http://creativecommons.org/licenses/by-nc/4.0/>) Spectroscopic and Radiative Properties of Several Nd³⁺ Ions in Borate Glass System J. Rajagukguk^{1,*}, M. Djamal², Suprijadi², A. Aminuddin³, J.

Kaewkhao⁴ 1 Faculty of Mathematics and Natural Sciences, Universitas Negeri Medan, Indonesia 2 Faculty of Mathematics and Natural Sciences, Institut Teknologi Bandung, Indonesia 40132 3Department of Physics, Universitas Pendidikan Indonesia, Indonesia 4Center of Excellence in Glass Technology and Materials Science, Nakhon Pathom Rajabhat University, Thailand *Corresponding e-mail : juniastel@yahoo.com Received 20 May 2016; Revised 24 September 2016; Accepted 26 September 2016 ABSTRACT Radiative properties and spectroscopic studies of several Nd³⁺ doped borate glass system have been reported.

Judd-Ofelt intensity parameter and other parameters like oscillator strength (f), effective bandwidth (λ_{eff}), radiative transition probabilities (A_R), stimulated emission cross section (σ), branching ratio (β_R), radiative lifetime (τ_R) and experimental lifetime (τ_{exp}) for the hypersensitive Nd³⁺ doped Borate Glass are listed and discussed. The variation of β_2 values for the different host matrix are expressed their covalency among Nd³⁺ ions in the glass matrix.

In this study, reported that the hypersensitive transition achieved at 4I9/2 \rightarrow 4G5/2,

2G7/2 centered at 580 – 585 nm range. Keywords: Borate glass, Judd-Ofelt, radiative
INTRODUCTION In the several years and recently, laser gain medium based on Nd³⁺
doped glasses have been attracted much attention from researchers in the field of
photonic and laser.

The above related to Nd³⁺ laser application such as optical amplifier, laser pumping,
optical communication, optical waveguide, storage data optically, radar and medical
instrumentation [1-5]. Medium gain laser characteristics for commercial laser required
were must satisfied sharpness fluorescent lines, strong absorption bands and sensible
for high quantum efficiency in accordance with the needed transition photon [6]. The
above requirements have been obtained by a small amount of concentration Nd³⁺
ions-doped glass material, since Nd³⁺ ions were able to produce population inversion
for result stimulated emission in the visible range (such as emission transition at 4G7/2 ?
4I9/2, 4G7/2 ? 4I11/2, 4G7/2 ? 4I13/2) [7] and the NIR range (lasing transition 4F3/2 ?
4I9/2, 4F3/2 ? 4I11/2, 4F3/2 ? 4I13/2) [1].

Improved laser performance is strongly influenced by the composition of the host glass
matrix and the concentration of doped ions, since the stimulated emission quality
depends on host matrix in which the ions are incorporated [7,8]. Several types of
commercial glass are generally used as a laser host matrix, i.e silicate, phosphate, borate
glasses and several heavy metal oxide glasses [1,2,7,9].

Some of the results showed that the silicate glasses has its advantages as well as high
chemical stability, high transparency for UV, low thermal expansion coefficient leading
to strong thermal resistance, a small nonlinear refractive index, high surface damage
threshold, J. Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal
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tensile fracture strength and good durability [10-12].

Those advantages made this type of glass are suitable for application as optical fiber or
waveguide lasers [13]. Phosphate glass appropriately used as a host matrix for Nd³⁺
ions owned properties are low thermal-optical constants, low melting point, low glass
transition temperature and high thermal expansion coefficient [14].

Moreover, the borate glass properties have been reported as well high transparency,
high density, appropriate bandwidth, suitable for infrared transmission, high mechanical
stability, corrosion resistivity and inexpensive [2,15]. In addition, the other types of
glasses and crystals have been reported as well as a host of Nd³⁺ ion laser such as
Nd³⁺ doped alkali niobium zinc tellurite glasses [16], Nd³⁺ doped barium titanium
silicate glasses [17], Nd³⁺ ions in fluoro-phosphate glasses [18], Nd³⁺ doped

lanthanum calcium borate glasses [19] and Nd³⁺ doped alkali boro germanate glasses [20].

In this paper discussed some of the parameters needed in the determination of the glass composition doped Nd³⁺ ion to be used as a laser gain medium. This discussion is limited to the composition of glasses based on borate glass former. As for the parameters that were examined in a laser medium are absorption spectrum, intensity analysis, emission spectrum, energy level, emission cross section, radiative lifetime, etc.

THEORY Optical Spectroscopic The optical spectra of the laser medium affected by the concentration of Nd³⁺ and glass structure as host matrix which the ion in the host matrix have the transition energy level vary with the Nd³⁺ concentration. The energy level depends on covalency and asymmetry of Nd³⁺-O bond in the host matrix [10,21].

For the Nd³⁺ ions doped some different host material such as glass material can be observed the energy level E_j by a Taylor series expansion [22]. (1) Where E_j is the energy levels of the Nd³⁺ ion in the host matrix from the measurement of absorption spectra, E_{0j} is the zero order Energy level of the Nd³⁺ ion, π_i are the partial derivatives and Δ_i are the variations of the interactions with the host matrix [23].

The experimental energy levels observed, and then obtained the deviation of the levels energy by the equation: (2) Where M is the number of absorption bands and J expresses the number of parameters on the theoretical energy values. Judd-Ofelt Parameters The intensity of absorption bands determined by the experimental oscillator strength (f_{exp}) that related to particular transition.

Oscillator strengths were determined according to Judd-Ofelt theory [2,24]. (3) Where ϵ is the molar absorption coefficient, ν is energy (cm⁻¹). The area of the absorption curve utilized for calculating the right side of integral, concentration of the Nd³⁺ ion in J . Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal homepage www.jpacr.ub.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 118 mole/lit and the optical length of glass in cm.

The optical density or absorptivity according to Ber's adioved $y(\nu) = (1/cl) \log (I_0/I)$ where c is the concentration of Nd³⁺ ion (mole/lit), l is the optical path in the medium. The theoretical oscillator strength of an induced electric-dipole transition from ground state J to an excited state J' is given by (4) Where $(2J + 1)$ is the multiplicity of the lower states, m is the mass of the electron, ν is the setting of absorption peak, ν^2 ($\nu = 2,4,6$) are JO intensity parameters and the $U^{\lambda}(\nu)$ are the doubly reduced unit tensor operations calculated in the intermediate coupling approximation [25,26].

The equation of the root mean square deviation (σ_{rms}) clarifies the quality of the fit known is given by (5) where f_{exp} and f_{cal} are the experimental and calculated oscillator strengths, respectively, M is the number of absorption bands used in account, i and f refers the total number of levels included in the fit. Radiative Properties Radiative transition of the laser transition $4F3/2 \rightarrow 4I11/2$ covered by the transition probability (AR), radiative lifetime (τ_R), stimulated emission cross section (σ_{em}), Branching ratio (β_R) are calculated by using of the Judd-Ofelt parameters (Ω_λ).

The relation of the initial level J to J' the transition probability (AR) ves [27] (6) Where S_{ed} and S_{md} are the electric dipole and magnetic dipole line strengths given as and (7) The quality of the laser transition $4F3/2 \rightarrow 4I11/2$ observed by size stimulated emission cross-section and calculated with using of Fuchtbabauer-Ladenburg method.

(8) Where λ_p is the peak wavelength of the emission peak, c is the speed of light, n is the refractive index, $A(\lambda)$ is the intensity of the emission peak and $\Delta\lambda_{eff}$ is an effective line width. The area under of emission peak is used for calculation of full-width at half-maximum. The effective line width of the emission given as (9) Where I_{max} is the maximum intensity at fluorescence emission peaks.

Then the radiative life time (τ_R) is given with reverse of the sum $AR(\lambda, J, J')$ (10) J. Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal homepage www.jpacr.ub.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 119 DISCUSSIONS Judd-Ofelt analysis The oscillator strength (f_{exp}) determined experimentally by substituting of the absorption spectra obtained from the measurement of results. Judd-Ofelt parameters obtained from the absorption spectra have been measured at room temperature.

These values used for calculating oscillator strength each of the absorption bands in accordance with eq. (3). The experimental and theoretical oscillator strength of some Nd^{3+} : doped borate glasses former showed in Table 1. The value of energy levels, especially at $4I9/2 \rightarrow 4G5/2, 2G7/2$ transitions for some materials with various concentrations also presented in Table 1.

In this paper selected Nd^{3+} concentrations that have the best potential for the emission spectra of each reference. Table 1. Hypersensitive absorption transition (from the ground state, $4I9/2 \rightarrow 4G5/2, 2G7/2$) for Nd^{3+} : borate glass Glasses structure λ_p (nm) xNd^{3+} (mole%) E_{xpt} (cm⁻¹) E_{cal} (cm⁻¹) $f_{exp} \times 10^{-6}$ $f_{cal} \times 10^{-6}$
 75B2O3.13PbO.5Bi2O3.5Al2O3 [2] 580 2.0 17.212 17.199 20.70 20.86 99Bi2ZnOB2O6
 [27] 585 1.0 17.094 - 20.63 - 25LiO;25GeO2;49B2O3 [20] 0.5 19.477 - - -

20CdO;15Bi₂O₃-79.5B₂O₃ [28] 582 0.5 - - 5.12 5.11 72.75B₂O₃;4.5CaO-22.37La₂O₃ [29]
583 1.0 - - 1.1649 1.1642 20NaF;30PbO;49.5B₂O₃ [22] 582-585 0.5 17.153 17.123 22.41
22.42 20NaCl;30PbO;49.5B₂O₃ [22] 582-585 0.5 17.185 17.220 22.31 22.29
35Bi₂O₃;30Na₂O;34B₂O₃[30] 582-585 1.0 17.065 - 16.0 16.2

35PbO;30Na₂O;34B₂O₃[30] 582-585 1.0 17.605 - 15.0 15.3
49.5PbO;30B₂O₃;10TiO₂-10AlF₃ [31] 585 0.5 17.094 17.300 16.090 16.033 67
B₂O₃;12Li₂O;20Na₂O [32] 582-585 1.0 17.118 - - - 67 B₂O₃;12Li₂O;20K₂O [32] 582-585
1.0 17.089 - - - 49.5 B₂O₃;49.5Na₂O [33] 580 1.0 - - 26.37 26.31
49.5B₂O₃;24.75Na₂O;24.75NaF [33] 580 1.0 - - 19.90 19.90 20B₂O₃;69.5Bi₂O₃;10SiO₂
[34] 586 0.5 - - 5.53 5.20 Figure 1. Transmission spectra of 1.0

mole% Nd³⁺ doped borate glass system Table 1 reflected the hypersensitive absorption band located in the 580-586 nm wavelength range with the Nd³⁺ concentrations to be potential as a laser medium are 0.5; 1.0 and 2,0 mole%. Absorption band due to the content of the composition of Nd³⁺ ions in the glass material, this affects the shape, peak position and intensity of the transition.

The difference of hypersensitive wavelength position of several glass mediums are attributed to J. Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal homepage www.jpacr.uib.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 120 differences in the crystal field asymmetry which makes the peak position split.

In addition, the intensity of the absorption peak of hypersensitivity split was also caused by changes in the content of the composition glass. On the other hand, some medium glasses have the same absorption peak, which splits disappearances have occurred by expansion of homogeneity [33]. The oscillator strength values are strongly influenced by the type and composition of other metal compounds as modifiers of the glass network structure.

The high content of the borate not guarantee can increase the value of the oscillator strength, but becomes interesting that the current composition of 49.5 mole% boric produced high oscillator strength values. The transmission spectra of 1.0 mole% Nd³⁺ doped borate glass system in the wavelength range 300-1000 nm is shown in Figure 1.

The transmission spectra shows ten sensitive bands derived from the state of 4I_{9/2} to various excited states. The transmission peaks centered at 354 nm, 430 nm, 474 nm, 524 nm, 580 nm, 625 nm, 679 nm, 744 nm, 802 nm and 871 nm were attributed to 2D_{5/2}, 2P_{1/2}, 2G_{9/2}, 4G_{7/2}, 2G_{7/2}+4G_{5/2}, 2H_{11/2}, 4F_{9/2}, 4F_{7/2}, 4F_{5/2}, 4F_{3/2} transitions respectively Table 2.

Judd-Ofelt parameters ($\times 10^{-20}$) and spectroscopic quality factor (Ω_4/Ω_6) of the excellent concentration of Nd³⁺ (x) : doped borate glasses based Glasses xNd³⁺ (mole%) Parameter Ω_2 Ω_4 Ω_6 2ZnO;20B₂O₃ [27] 1.0 2.67 3.31 3.98 0.83 25LiO;25GeO₂;49B₂O₃ [20] 0.5 4.84 5.97 4.59 1.22 25NaO;25GeO₂;49B₂O₃ [20] 0.5 5.75 3.44 3.73 0.92 25KO;25GeO₂;49B₂O₃ [20] 0.5 5.89 3.95 2.85 1.38 25RbO;25GeO₂;49B₂O₃ [20] 0.5 6.18 3.63 2.45 1.48 20NaF;30PbO;49.5B₂O₃ [22] 0.5 4.69 5.09 6.50 0.78 20NaCl;30PbO;49.5B₂O₃ [22] 0.5 4.84 5.31 6.32 0.84 75B₂O₃;13PbO;5Bi₂O₃;3.5Al₂O₃ [2] 2.0 4.53 4.17 6.44 0.65 20CdO;15Bi₂O₃;79.5B₂O₃ [28] 0.5 1.44 3.42 2.89 1.18 35Bi₂O₃;30Na₂O;34B₂O₃ [30] 1.0 4.72 2.12 3.93 0.54 35PbO;30Na₂O;34B₂O₃ [30] 1.0 4.81 1.97 3.94 0.50 49.5PbO;30B₂O₃;10TiO₂;10AlF₃ [31] 0.5

5.82 1.88 4.74 0.21 67 B₂O₃;12Li₂O;20Na₂O [32] 1.0 5.95 7.82 9.84 0.79 67 B₂O₃;12Li₂O;20K₂O [32] 1.0 10.83 7.73 9.04 0.85 49.5B₂O₃;49.5Na₂O [33] 1.0 7.79 3.03 2.80 1.055 49.5B₂O₃;24.75Na₂O;24.75NaF [33] 1.0 5.33 2.84 4.90 0.579 20B₂O₃;69.5Bi₂O₃;10SiO₂ [34] 0.5 3.52 4.19 3.86 1.01 The oscillator strength values as shown in Table 1 were used to determine of Judd-Ofelt parameters, Ω_2 , Ω_4 , and Ω_6 by using of least-square fitting method in eq. (4).

Judd-Ofelt parameters of several Nd³⁺ doped glasses for hypersensitive transition given in Table 2. There is one material [28] shown that intensity parameters $\Omega_2 < \Omega_4$, this condition represent the high covalent bonding that means there was broadening asymmetry around the environments glass [18].

Table 2 showed generally was observed that $\Omega_2 > \Omega_4, \Omega_6$ [2,20,30,31,31,33] indicate the higher covalency of the ion-ligand bond lower symmetry of the Nd³⁺ ion site. This result also explained that the higher intensity of hypersensitive transition and the nephelauxetic effect possessed by these glass [22]. The higher values of Ω_4 and Ω_6 [20,22,28,32,34] for the glasses indicated their mechanics have higher rigidity. Then, the spectroscopic quality factor J.

Pure App. Chem. Res., 2016, 5 (2), 116-124 26 August 2016 X The journal homepage www.jpacr.ub.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 121 known from $\Omega_2 = (\Omega_4/\Omega_6)$ for to find the channel through which the excited metastable state to the ground state [2].

Radiative Properties The Judd-Ofelt parameters ($\Omega_2, \Omega_4, \Omega_6$) used for determining of various spectroscopic parameters such as effective bandwidth (Ω_{eff}), fluorescence lifetime (τ_f), radiative transition probabilities (AR), stimulated emission cross section (σ_{se}) and branching ratio (β) as given in Table 3. These parameters are calculated using the eq.

(6) – (10) for Nd^{3+} : borate glass of $4F_{3/2} \rightarrow 4I_{11/2}$ level transition. Refer to the reported by Q.

Nie et al [35] that the materials suitable to be used as a laser medium is supposed to have some requirements such as large effective bandwidth, long fluorescence lifetime, high stimulated emission cross-section and high branching ratio. In Table 3 shown that the value of these parameters is almost equal to each other of the glass material then stated that borate glasses which contained Nd^{3+} potentially be used as a laser gain medium. Table 3.

Excitation wavelength (λ_{exc}), wavelength peak (λ_p), effective bandwidth ($\Delta\lambda_{eff}$), radiative transition probabilities (AR), stimulated emission cross section (σ), branching ratio (β), radiative lifetime (τ_R) and experimental lifetime (τ_{exp}) for the hypersensitive Nd^{3+} doped Borate Glass and emission transition $4F_{3/2} \rightarrow 4I_{11/2}$ Glasses structure λ_{exc} (nm) λ_p (nm) $\Delta\lambda_{eff}$ (nm) A R (s^{-1}) σ (cm^2) β (τ_{exp}) (μs)

99Bi ₂ ZnOB ₂ O ₆ [27]	808	1063	29.00	3407	4.33	0.04	145	62.0	
75B ₂ O ₃ .13PbO.5Bi ₂ O ₃ .5Al ₂ O ₃ [2]	580	1065	18.81	7.89	0.74	264	-	-	
20CdO.15Bi ₂ O ₃ -79.5B ₂ O ₃ [28]	1064	-	1696	-	0.543	-	20NaF;30PbO;49.5B ₂ O ₃ [22]	1063	
- 2077 - 0.494	238	-	20NaCl;30PbO;49.5B ₂ O ₃ [22]	1063	-	2070	-	0.487	235
35Bi ₂ O ₃ ;30Na ₂ O;34B ₂ O ₃ [30]	808	1067	43	1581	2.0	0.552	349	-	-
35PbO;30Na ₂ O;34B ₂ O ₃ [30]	808	1065	43	1285	1.8	0.548	426	-	-
49.5PbO;30B ₂ O ₃ ;10TiO ₂ -10AlF ₃ [31]	805	1070	34	1184	2.6	0.52	470	230	67
B ₂ O ₃ ;12Li ₂ O;20Na ₂ O [32]	514	1069	3092	6.16	0.499	26	-	67	B ₂ O ₃ ;12Li ₂ O;20K ₂ O [32]
514	1050	4110	8.84	0.512	21	-	49.5	B ₂ O ₃ ;49.5Na ₂ O [33]	800
153	49.5B ₂ O ₃ ;24.75Na ₂ O;24.75NaF [33]	800	1062	17.84	4.18	0.378	347	151	-
20B ₂ O ₃ ;69.5Bi ₂ O ₃ ;10SiO ₂ [34]	800	1075	1130	2.20	0.536	-	-	-	-

The composition of glass structure in the Table 3 is the optimum value selected from each reference.

So the quality of the materials is also influenced by the amount of the glass composition in the host matrix. Kumar K et al [2] report that the fluorescence radiative lifetime optimum obtained when 75 mole% of B₂O₃ (BINLAB2), the higher emission cross section of $4F_{3/2} \rightarrow 4I_{11/2}$ level obtained at 8 mole% PbO (BINLAB3).

The difference shown by branching ratio, which were the greatest results given by BINLAB2, followed BINLAB3 and the next BINLAB1. The intensity of the Nd^{3+} ion at $4F_{3/2} \rightarrow 4I_{11/2}$ transition level so important to consider in a laser medium therefore the addition of SiO₂ content could be increased of the intensity and lifetime level [35].

The highest of the radiative transition probabilities at $4F_{3/2} \rightarrow 4I_{11/2}$ level transition in Table 3 are given by Nd^{3+} doped 67B₂O₃;12Li₂O;20K₂O and J. Pure App. Chem. Res.,

It is claimed the radiative lifetime both glasses are lower than others and according to reported of Y.Chen et.al [36] that the high radiative transition probability and low radiative lifetime are considered to improve the radiative quantum efficiency of the laser medium. Emission spectrum The shapes and position emission spectra were recorded for the NIR range transition shown in Figure 2.

According to the energy level diagram of Nd³⁺ in glass, the NIR emission located at around 940-946, 1060-1070 and 1335-1346 nm that are attributed to the 4F_{3/2} → 4I_{9/2}, 4I_{13/2}, and 4I_{11/2} transition respectively. Furthermore, the shape and peak position emission spectra of Nd³⁺ doped glass borate by exciting at 582 nm and 0.50 mole% Nd³⁺ content shown in Figure 3.

Figure 2 Energy level diagram of Nd³⁺ Figure 3 NIR Emission spectra of Nd³⁺ The position of emission peak wavelength of the 4F_{3/2} → 4I_{11/2} transition shifted a few nanometers due the electric dipole transition of Nd³⁺ ions are very sensitive to the surrounding. This transition assigned as hypersensitive effect and can used to figure out of Nd³⁺-O covalency bond [30].

The peak wavelength shifted when the Nd³⁺ doped into the host glass due the nephelauxetic effect [37-38], this occurs due to electron orbitals with 4f configuration disabled in the host ligand field [39]. The intensity of the 4I_{9/2} → 4G_{5/2}+ 2G_{7/2} level transition for Nd³⁺ is higher than the other transition and this is in accordance with the hypersensitive regulations of the transitions such as J → 2; L → 2 and S = 0 [38].

The energy level structure of Nd³⁺ ion will be contracted with the increase in the intersection of the both oxygen and 4f orbitals leading to the wavelength shift. From the emission spectra transition in Table 3 observed that the peak emission of the 20B₂O₃;69.5Bi₂O₃;10SiO₂ glasses[34] higher than others glass because this glass has a higher polarizability of the Bi-O bond.

The emission spectra depends on 4 and 6 parameters because have related to the rigidity of the host glass. In Table 3 there are three types of glass composition which have the signifies a higher stimulated emission cross section [2,32] and the consequence is the increase of the branching ratio percentage. CONCLUSIONS It has been observed the performance of the Nd³⁺ doped borate glass former.

This paper focused to discuss of optical properties such as absorption spectra, oscillator

strength f , Judd-Ofelt parameters $\Omega_2, \Omega_4, \Omega_6$, branching ratio β , radiative transition probabilities A_{R} , emission spectra and radiative lifetime τ_R for special of the hypersensitive transition Nd^{3+} from $4F_3/2$ *J. Pure App. Chem. Res.*, 2016, 5 (2), 116-124
26 August 2016 X The journal homepage www.jpacr.ub.ac.id p-ISSN : 2302 – 4690 | e-ISSN : 2541 – 0733 123 level to 4111/2 level.

The highest of the oscillator strength obtained when the current of 49,5 mol% boric nevertheless the high content of borate not guarantee can increase the value of the oscillator strength. The quality of the materials is also influenced by the amount of the glass composition in the host matrix, but for to obtain the high of radiative transition, such as τ_{em} , A_{R} and τ_R of the laser medium proposed to use of borate-lithium-potassium oxide glass.

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