

SPECTROSCOPY CHARACTERISTICS OF Sm^{3+} -DOPED SODIUM BARIUM BORATE OXIDE AND OXYFLUORIDE GLASSES FOR LASER MATERIALS

Piyachat Meejitpaisan^{1,3,*}, Yotsakit Ruangtaweep^{2,3}, HongJoo Kim⁴ and Jakrapong Kaewkhao^{2,3}

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Abstract

Sm^{3+} -doped sodium barium borate oxide and oxyfluoride glasses were prepared by melt-quenching technique and investigated through the optical absorption and emission analysis. The optical absorption of Sm^{3+} -doped oxide and oxyfluoride glasses presents the highest peak at 401 nm in visible region while the absorbance of both glasses is found to be similar. The optical absorption has been used to derive the Judd-Ofelt (JO) parameters ($\Omega\lambda$, $\lambda = 2, 4$ and 6). The trend of JO parameter is $\Omega_4 > \Omega_2 > \Omega_6$. Moreover, the JO parameters are used to predict the radiative properties which are significant for the assessment of the suitability of a glass for laser emission. The emission of both glasses comprises 4 peaks at 563, 600, 646 and 706 nm. When compared emission intensity between oxide and oxyfluoride glasses, the oxyfluoride glass is higher than oxide glass due to the reduction of phonon energy. Furthermore, the emission spectra can be calculated the stimulated emission cross section which is one of the most important parameter that characterizes the laser performance of glasses.

Keywords: Borate glass, Judd-Ofelt (JO), Oxyfluoride, Samarium

Introduction

On account of the excellent growth of optic displays, optical data storage and fiber optic technology, for instance, sensors, lasers, 3-D communications, crystals and glasses are

¹ Industrial Physics Program, Faculty of Science and Technology, Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand.

² Physics Program, Faculty of Science and Technology, Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand.

³ Center of Excellence in Glass Technology and Materials Science (CEGM), Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand.

⁴ Department of Physics, Kyungpook National University, Daegu 41566, Republic of Korea.

* Corresponding author

materials for doping rare earth (RE³⁺) ions which have received from scientist. However, glasses are better than crystals owing to inexpensive cost, good thermal stability, flexibility in shape and size, good transparency and high doping capability (Deopa *et al.*, 2018; Mesady and Alawsh, 2018; Steudel *et al.*, 2016). Normally, the doping RE³⁺ ions in glasses could be improved due to the result of high emission efficiencies. It related with f-f electronic transitions which are shielded by the 5s and 5p outer shells (Basavapoornima and Jayasankar, 2014; Meejitpaian *et al.*, 2017). Nonetheless, it is well know that the emission efficiencies are depending on phonon energy that is expected through ions surrounding of host glasses (Lin *et al.*, 2006; Rao and Jayasankar, 2013). Among oxide glasses, the selection of borate glasses as the host for doping RE³⁺ ions due to high optical transparency, hardness, low melting point and good RE³⁺ solubility (Krause *et al.*, 2014; Kaur *et al.*, 2015). Nevertheless, the borate glasses have high phonon energy around 1300 cm⁻¹ because of the stretching vibration of network bonding oxides which is a cause of increase non-radiative transition probability and reduce emission efficiency (Deopa and Rao, 2017; Mohan *et al.*, 2017). This major problem can be enhanced by adding Na₂O and BaO into glass network (Hivrekara *et al.*, 2017). Besides, Na₂O and BaO can also lead to reduce melting temperature of glasses. Another one that enhances emission efficiency is changing oxide to fluoride in glass composition resulting to the reduction of hydroxyl group (OH-group) vibration (Reddy *et al.*, 2017).

Among RE³⁺ ions doped glasses, Sm³⁺ ions doped glasses is one of the most important because of its wide applications e.g. color displays, red and orange emission in solid state lasers, fluorescence devices and undersea communication (Thomas *et al.*, 2013). In general, the Sm³⁺-doped glasses present green, red and orange emission which related with the 4G_{5/2} to 6H_{5/2}, 6H_{7/2} and 6H_{9/2} transition, respectively (Kindrat *et al.*, 2015). Not only emission but also Judd-Ofelt (JO) theory is used to discussed the calculated stimulated emission cross section (σ_{em}). The

spectroscopic properties of Sm³⁺ ions doped glasses have been investigated. The stimulated emission cross section has been reported in various host glasses: Lithium cesium mixed alkali borate glasses (Ratnakaram *et al.*, 2006), KNbSiSm glasses (Mohan *et al.*, 2013), ZLNB glasses (Sailaja *et al.*, 2015) and PKSASm glasses (Vijaya *et al.*, 2013). In this work, Sm₂O₃ concentration doped in sodium barium borate oxide and oxyfluoride glasses is fixed at 1.0 mol% due to its concentration quenching (Ramachari *et al.*, 2013). The physical, optical and emission properties of Sm³⁺-doped glasses have been investigated. Besides, the predicted radiative transition probabilities (A_R), emission cross-section and branching ratio (β_R) for the 4G_{5/2}→6H_{7/2} transition have been analyzed.

Materials and Methods

Glass Preparation

Sm³⁺-doped sodium barium borate oxide (30Na₂O : 15BaO : 54B₂O₃ : 1Sm₂O₃, refer as SmNO) and oxyfluoride glasses (30NaF : 15BaO : 15B₂O₃ : 1Sm₂O₃, refer as SmNF) were prepared via melt-quenching technique. Analytical reagent grade chemicals use in the present study comprises sodium carbonate (Na₂CO₃), sodium fluoride (NaF), barium carbonate (BaCO₃), boric acid (H₃BO₃) and samarium oxide (Sm₂O₃). The mixture was melted at 1200°C for 3 hours and anneal at 500°C for 3 hours before cooled down at room temperature. Finally, the SmNO and SmNF glasses were cut and then finely polished to a dimension of 1.0 cm x 2.0 cm x 0.3 cm.

Characterization

The densities (ρ) were measured by Archimedes's method with using water as immersion liquid at room temperature. After that the molar volumes (V_m) were calculated from relation by

$$V_m = \frac{M_T}{\rho} \quad (1)$$

where MT is total molecular weight of glasses. The optical absorption spectra of

glasses were measured by using UV-VIS-NIR spectrophotometer (Shimadzu, UV3600) in the range of 380-1800 nm at room temperature. The emission spectra were carried out by using a fluorescence spectrophotometer (Cary Eclipse) with xenon lamp as a light source. The experimental (f_{exp}), calculated (f_{cal}) oscillator strengths and JO parameter were considered by absorption results while the predicted radiative transition probabilities (A_R), emission cross-section and branching ratio (β_R) for the 4G_{5/2}→6H_{7/2} transition were determined from JO parameter and luminescence results.

Results and Discussion

Physical Properties

The molecular weights, densities, molar volumes and refractive indexes of SmNO and SmNF glasses were shown in Table 1. The density of SmNO glass was more than SmNF glass because the molecular weight of SmNO glass was higher than SmNF glass. On the other hand, the molar volume of SmNF glass was higher than SmNO glass because the fluoride creates non-bridging atom and F- sites in glass (Rasool *et al.*, 2013). It indicated that the network of glass was loosed because of the substitution of NaF in the place of Na₂O. Besides, the refractive index shows the same trend with densities.

Optical Absorption

The optical absorption of SmNO and SmNF glasses were noted in wavelength from 380 to 1800 nm which were shown in Figure 1. Both glasses show spectra in visible and near infrared (NIR) region. All spectra performed the ⁶H_{5/2} ground state to excited state, namely ⁶P_{3/2} (401 nm), ⁴M_{17/2} (439 nm), ⁴I_{11/2} (473 nm), ⁴F_{3/2} (526 nm), ⁴G_{5/2} (561 nm),

⁶F_{11/2} (944 nm), ⁶F_{9/2} (1077 nm), ⁶F_{7/2} (1227 nm), ⁶F_{5/2} (1373 nm), ⁶F_{3/2} (1474 nm), ⁶H_{15/2} (1529 nm) and ⁶F_{1/2} (1585 nm) transitions (Raddy *et al.*, 2017; Kumar and Jayasankar, 2014). The ⁶H_{5/2}→⁶P_{3/2} transition was observed the highest absorption band. It was hypersensitive transition to the environment around Sm³⁺ by following the selection rules |ΔS| = 0, |ΔL| ≤ 2 and |ΔJ| ≤ 2 (Meejitpaisan *et al.*, 2016). Additionally, the SmNO and SmNF glasses were compared and they show the similar absorbance. It indicated that the fluoride does not affect the optical absorption. From optical absorption results, the experimental (f_{exp}) and calculated (f_{cal}) oscillator strengths of both glasses were evaluated by

$$f - 9 \int \varepsilon(v) dv_{exp} \quad (2)$$

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

where ε(v) is the molar absorptivity at wavenumber v(cm⁻¹), n is the refractive index of medium, J is the total angular momentum of the ground state, Ω_λ (λ = 2, 4 and 6) are Judd-Ofelt (JO) intensity parameters and ||U_λ||² are the doubly reduced matrix elements of the unit tensor operator of the rank λ = 2, 4 and 6. They are calculated from the intermediate coupling approximation for the transition, ψJ → ψ'J' (Basavapoornima and Jayasankar, 2014; Lim *et al.*, 2013). The experiment and oscillator strength of SmNO and SmNF glasses are presented in Table 2 and the JO intensity parameters are demonstrated in Table 3.

From the Table 3, JO intensity parameter made up of Ω₂, Ω₄ and Ω₆ parameter. Normally, the Ω₂ parameter is relates to the asymmetry environment around Sm³⁺ and covalence between Sm³⁺ ions and ligand

Table 1. Molecular weights, densities, molar volumes and refractive indexes of glasses

Glasses	Molecular Weights (g/mol)	Densities (g/cm ³)	Molar Volumes (cm ³ /mol)	Refractive Indexes
SmNO Glass	82.6748	2.8250	29.2653	1.6583
SmNF Glass	76.6775	2.6085	29.3948	1.6368

anions. The Ω_4 and Ω_6 parameters exhibit the viscosity and rigidity of glasses (Ramachari *et al.*, 2013; Chen *et al.*, 2012). The SmNO and SmNF glasses possessed the trend of $\Omega_4 > \Omega_2 > \Omega_6$ that was similar to Sm³⁺-doped other host glasses such as ZABBSm0.3 (Yuliantini *et al.*, 2018) and PKAZFSm10 glasses (Sreedhar *et al.*, 2014). The largest value of Ω_4 parameter mean the higher rigidity of glasses. In any case, the trend of JO intensity parameter is $\Omega_2 > \Omega_4 > \Omega_6$ for ZnLiBiB (SM02) (Meena, 2017) and TNS4 (Mawlut *et al.*, 2017) glasses. The highest value of Ω_2 parameter indicated more covalent and asymmetry environment for Sm³⁺ in these glasses. By the way, the spectroscopic quality factor (χ) is used to characterize the quality of glasses which was a significant predictor for stimulated emission in a laser active medium (Sardar *et al.*, 1993; Kesavulu *et al.*, 2013). In this work, the spectroscopic quality factor is found to be 1.72 and 1.83 for SmNO and SmNF, respectively. Furthermore,

the spectroscopic quality factor of SmNF glass was also higher value than other host glasses as shown in the following Table 3. It indicated that SmNF glass was possibly a good laser active medium.

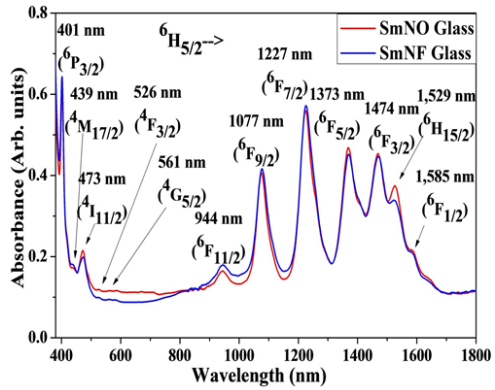


Figure 1. The optical absorption of SmNO and SmNF glasses

Table 2. Energies (cm⁻¹), experiment (f_{exp}) and calculated (f_{cal}) oscillator strengths ($\times 10^{-6}$) for SmNO and SmNF glasses

Level	Energy	SmNO glass		SmNF glass	
		f_{exp}	f_{cal}	f_{exp}	f_{cal}
$^6P_{3/2}$	24937	5.18	5.31	6.00	5.34
$^4M_{17/2}$	22779	0.21	0.17	0.23	0.20
$^4I_{11/2}$	21142	2.20	2.35	2.20	2.42
$^4F_{3/2}$	19011	0.07	0.01	0.08	0.01
$^4G_{5/2}$	17825	0.06	0.04	0.07	0.05
$^6F_{11/2}$	10593	1.37	1.85	1.72	1.83
$^6F_{9/2}$	9285	5.18	5.37	7.26	7.50
$^6F_{7/2}$	8150	8.78	8.77	9.77	10.44
$^6F_{5/2}$	7283	6.86	6.59	8.34	8.53
$^6F_{3/2}$	6784	6.85	6.94	7.10	7.36
$^6H_{15/2}$	6540	4.86	4.00	5.42	5.00
$^6F_{1/2}$	6309	1.42	1.78	1.46	1.77

Table 3. The Judd-Ofelt intensity parameters (Ω_λ , $\times 10^{-20}$ cm²), spectroscopic quality factor (χ) and trend of SmNO and SmNF glasses and for some of the other reported Sm³⁺-doped glasses

Glass ID	Ω_2	Ω_4	Ω_6	$\chi = \Omega_4 / \Omega_6$	Trend	References
SmNO	8.30	10.66	6.07	1.72	$\Omega_4 > \Omega_2 > \Omega_6$	This Work
SmNF	8.43	12.54	6.87	1.83	$\Omega_4 > \Omega_2 > \Omega_6$	This Work
ZABBSm0.3	4.58	5.62	3.17	1.77	$\Omega_4 > \Omega_2 > \Omega_6$	[26]
PKAZFSm10	2.35	4.38	3.46	1.27	$\Omega_4 > \Omega_2 > \Omega_6$	[27]
ZnLiBiB(SM02)	4.12	3.84	3.52	1.09	$\Omega_2 > \Omega_4 > \Omega_6$	[28]
TNS4	0.80	0.72	0.45	1.61	$\Omega_2 > \Omega_4 > \Omega_6$	[29]

Luminescence Properties

The luminescence spectra of SmNO and SmNF glasses have been carried out at wavelength 525 – 750 nm and they were shown in Figure 2. Both glasses exhibited 4 peaks which composed of 563 nm ($^4G_{5/2} \rightarrow ^6H_{5/2}$), 600 nm ($^4G_{5/2} \rightarrow ^6H_{7/2}$), 647 nm ($^4G_{5/2} \rightarrow ^6H_{9/2}$) and 706 nm ($^4G_{5/2} \rightarrow ^6H_{11/2}$) under excitation at 401 nm (Rajaramakrishna *et al.*, 2014). Among of transitions, the $^4G_{5/2} \rightarrow ^6H_{7/2}$ is the strongest transition. Besides, it is magnetic dipole (MD) allowed yet electric dipole nature is more inductive with selection rule $\Delta J = \pm 1$. While the $^4G_{5/2} \rightarrow ^6H_{9/2}$ is purely electric dipole (ED) transition with $\Delta J = \pm 2$. The intensity ratio between the $^4G_{5/2} \rightarrow ^6H_{7/2}$ (MD) and $^4G_{5/2} \rightarrow ^6H_{9/2}$ (ED) give more information about asymmetry and local environment of Sm^{3+} -doped glasses (Parandamaiah *et al.*, 2016; Maheshvaran *et al.*, 2011). In this work, the values of SmNO and SmNF glasses were approximately 2.09 and 2.19, respectively. It specified that the SmNF glass was asymmetry nature more than SmNO glass which was confirmed through the Ω_2 parameter. Furthermore, the emission intensities of SmNO and SmNF were compared. It revealed that the emission intensity of SmNF glass was higher than SmNO glass since the fluoride ions were result of the reduction of phonon energies and increasing of emission efficiency (Mohan *et al.*, 2017). In addition, the emission results can predicted the radiative transition probabilities (A_R), stimulated emission cross-section ($\sigma_{em}(\lambda)$) and branching ratio (β_R) for the $^4G_{5/2} \rightarrow ^6H_{7/2}$ transition which presented in Table 4. Generally, the higher transition probabilities provide larger opportunity to achieve better laser action. The stimulated emission cross-section indicated

rate of energy excitation from the lasing material and the potential laser performance. Meanwhile, the branching ratio was greater or equal to 50% that typifies the lasing power of a transition in glass matrix. In this work, the SmNF glass showed experiment branching ratio values around 0.56 and the higher stimulated emission cross-section when compared to the SmNO glass and other host, viz., PKBFAS (Venkatramu *et al.*, 2007), LiPbFP (Babu *et al.*, 2014), PKAlCaFSm10 (Rasool *et al.*, 2013) and PbFPSm10 (Kesavulu and Jayasankar, 2012) glasses. Hence, it is concluded that the SmNF glass is appropriate for laser materials.

Conclusions

Sm^{3+} -doped sodium barium borate oxide (SmNO) and oxyfluoride (SmNF) glasses were successfully prepared via melt quenching technique. The absorption and photoluminescence

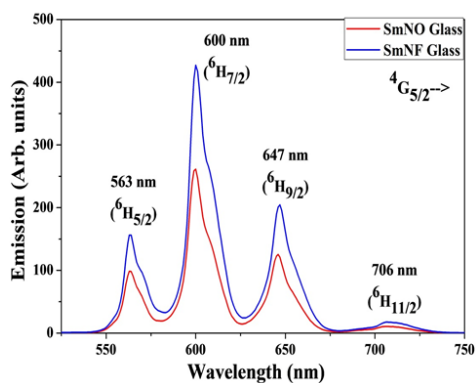


Figure 2. The emission of SmNO and SmNF glasses under excitation at 401 nm

Table 4. The predicted radiative transition probabilities (A_R , s^{-1}), emission cross-section ($\sigma_{em}(\lambda)$ $\times 10^{-21}$ cm^2) and branching ratio (β_R) for the $^4G_{5/2} \rightarrow ^6H_{7/2}$ transition of Sm^{3+} -doped glasses

Glass ID	A_R	$\sigma_{em}(\lambda)$	β_R		References
			Exp.	Cal.	
SmNO	184	12.19	0.53	0.51	This Work
SmNF	181	13.55	0.56	0.53	This Work
PKBFAS	159	9.10	0.39	-	[35]
LiPbFP	177	8.10	0.61	0.34	[36]
PKAlCaFSm10	174	9.30	0.56	0.55	[37]
PbFPSm10	185	8.98	0.57	0.43	[38]

results were determined for comparing between SmNO and SmNF glasses. From absorption results, the JO intensity parameter was evaluated and show same trends of $\Omega_4 > \Omega_2 > \Omega_6$. While, the spectroscopic quality factor value of SmNF glass was higher than SmNO glass. The luminescence results indicated that the emission intensity the SmNF glass was better than SmNO due to the reduction of phonon energy and increasing of quantum efficiency. Furthermore, the stimulated emission cross-section value and branching ratio for the $^4G_{5/2} \rightarrow ^6H_{7/2}$ transition of SmNF glass was higher than SmNO glass. Consequently, from all of results, it is concluded that the SmNF glass is a potential candidate for laser materials.

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