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ICAPMA\_2017 Physical and optical investigation of lutetium-sodium- borate glasses Punsak Glumglomchita,b, Juniastel Rajagukgukc, Jakrapong Kaewkhaoa,b, Keerati Kirdsiria,b,\* aPhysics Program, Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand bCenter of Excellence in Glass Technology and Materials Science (CEGM), Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand cDepartment of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Medan, 20221, Indonesia Abstract Physical and optical properties of lutetium sodium borate glasses have been examined.

A series of glass samples with the different chemical composition  $x\text{Lu}_2\text{O}_3:20\text{Na}_2\text{O}:(80-x)\text{B}_2\text{O}_3$  (where  $x = 5, 10, 15$  and  $20$  mol%) were successfully fabricated by a melt quenching technique and their properties were investigated through various techniques include density, absorption, and X-ray diffraction. The amorphous nature was confirmed by the presence of a broad hump in the XRD pattern.

The relevance parameters related physical and optical properties have been calculated and discussed. © 2018 Elsevier Ltd. All rights reserved. Selection and/or Peer-review under responsibility of 3rd International Conference on Applied Physics and Materials Applications. Keywords: Borate glass; Lutetium; Physical properties; Optical properties 1.

Introduction Nowadays, many researchers have been studying about optical and

luminescence of many glass series for developing application device like white emitting diodes (LEDs), solid state lasers and plasma display panels (PDP) [1]. Most glass series were compound from network formers that have short range order such as boron, or network intermediate, other network modifiers such as sodium that have variable coordination numbers (CNs) with less \* Corresponding author. Tel.: +6-684-700-0850; fax: +6-634-261-065. E-mail address: keyrati@hotmail.com P.

Glumglomchita et al./ Materials Today: Proceedings 5 (2018) 15054–15060 15055 coordination geometry [2,3]. Among glass materials, borate glasses were interesting since 1967 due to good transparency, thermal stability, high chemical durability and low melting point [4]. The mixture of network modifiers result the change of formation coordination of boron by oxygen atom from trigonal to tetrahedral [5].

Lutetium ( $\text{Lu}^{3+}$ ) is one of heavy substances of lanthanide families, which generally can be found in common such as in the energy-saving lamps and glasses [6]. Lutetium has been studied especially in the aspect of applications. It is used as based compounds in the oxidic form because of its good quality matrix for lanthanide ions to break-in.

Besides, its physical properties are dominant like high melting point, phase stability, high band gap between the valence and conduction bands (5.2-5.5 eV.) and high- $\epsilon$  dielectric constant (about  $\epsilon = 11-13$ ). In addition, it is also used as a solvent in a single crystal, bioimaging probe, up-conversion phosphor, luminescent phosphors, glass and transparent film [7,8,9].

The  $\text{Lu}^{3+}$  ion possesses mere one free crystallographic site with six oxygen neighbors [10]. This study aims to prepare the  $\text{Lu}_2\text{O}_3\text{-Na}_2\text{O-B}_2\text{O}_3$  glass series and to study the effect of different  $\text{Lu}_2\text{O}_3$  concentration on the density, molar volume, ion concentration, absorption spectra and optical absorption. 2. Experimental details 2.1.

Glass composition Four samples of  $\text{Lu}_2\text{O}_3\text{-Na}_2\text{O-B}_2\text{O}_3$  glass series were prepared with chemical composition  $x\text{Lu}_2\text{O}_3:20\text{Na}_2\text{O}:(80-x)\text{B}_2\text{O}_3$  (where  $x = 5, 10, 15$  and  $20$  mol%). The raw materials  $\text{H}_3\text{BO}_3$ ,  $\text{Lu}_2\text{O}_3$  and  $\text{Na}_2\text{CO}_3$  were mixed (10 g for each batch) and were melted in electric furnace by melt quenching technique at  $1500^\circ\text{C}$  for 3 h. After that, quickly poured into stainless steel block and annealed for 3 h at  $500^\circ\text{C}$ . At the end, cut and polished glass samples as shown in Fig.

1, the glass series are good transparent. Fig. 1. Photograph of glass series with different the  $\text{Lu}_2\text{O}_3$  concentration. 2.2. Instrumentation and parameters Archimedes method was applied to determine the density of glass samples. A 4-digit sensitive microbalance (A&D, HR-200) was used to investigate the weights of glasses in air and water, by using

the formula  $\rho \text{ (g/cm}^3\text{)} = (W_a - W_b)/(V_a - V_b)$ . The molar volume ( $V_M$ ) of glass series are computed by using the formula relation  $V_M \text{ (cm}^3\text{/mol)} = M/\rho$ .

While  $W_a$  and  $W_b$  are the weight of glass series in air and water respectively, and  $\rho_b$  is referred to density of water at the room temperature (25 °C) that was found to be 1.000 g/cm<sup>3</sup> and  $M$  is total molecular weight of mixture component in glass series. The recording optical absorption spectra were used UV-Vis-NIR spectrophotometer (Shimadzu, UV-3600) at room temperature, made at 200-2100 nm.

An X-ray diffractometer (Shimadzu, XRD-6100), was used to investigate amorphous nature for glass series. The refractive index were investigated by using Abbe refractometer (ATAGO) with a light source of a sodium vapor lamp that have wavelength 589.3 nm (D line) and coated by monobromonaphthalene (C<sub>10</sub>H<sub>7</sub>Br) between sample and prism of refractometer at room temperature.

$x = 5 \text{ mol\%}$   $x = 10 \text{ mol\%}$   $x = 15 \text{ mol\%}$   $x = 20 \text{ mol\%}$  15056 P. Glumglomchita et al./ Materials Today: Proceedings 5 (2018) 15054–15060 3. Results and discussion 3.1. Density and molar volume Density is main and foundational physical property of glass series. The density of glass was indicated the change of geometrical configurations, adjust compactness in degree, coordination of formation and differentiation of dimensions of interstitial holes [11,12]. The density of glass series as shown in Table 1, it increased with increased Lu<sub>2</sub>O<sub>3</sub> content.

The increasing of Lu<sub>2</sub>O<sub>3</sub> content that made molecular mass of Lu<sup>3+</sup> increased which has effect to compactness of glass series structure. That may be due to the presence of Lu<sup>3+</sup> ions in glass structure that will made borate glass network adjust by converting more [BO<sub>3</sub>]<sup>-3</sup> triangles to BO<sub>4</sub> tetrahedral. An increasing density of glass series with increase of Lu<sup>3+</sup> concentration that influence the molar volume, refractive index and physical properties of glass series.

The calculated molar volumes of glass series found that it increased because of non-bridging oxygen (NBOs) number increased which is the result of the bond damage so it loose compacted formation. Moreover, as listed in Table 1, the results show that the refractive index of glass series increased with increased Lu<sub>2</sub>O<sub>3</sub> content. The density of glass series may be affected to a packed formation that balks activation of light motivation high refractive index. 3.2.

Ion concentration and other parameters The Lu<sup>3+</sup> ion concentration ( $N$ ), of glass series were computed by using equation [11]  $N \text{ (ion/cm}^3\text{)} = (\text{mol\% of dopant})(N_A)(\rho)/M$  (1) where  $N_A$ ,  $\rho$  and  $M$  are Avogadro's number, density and average molecular weight of

glass, respectively. Table 1. The physical and optical parameters for glass series (mol%). Physical properties Lu<sub>2</sub>O<sub>3</sub> content (mol%) 5 10 15 20 Density,  $\rho$  (g/cm<sup>3</sup>) 2.7090  $\pm$  0.0015 3.1593  $\pm$  0.0034 3.3981  $\pm$  0.0012 3.7946  $\pm$  0.0023 Thickness (cm) 0.4117 0.3733 0.3683 0.4167 Refractive index, n 1.5450  $\pm$  0.0002 1.5777  $\pm$  0.0001 1.5933  $\pm$  0.0002 1.6218  $\pm$  0.0002 Molecular weight, M (g/mol) 84.5075 100.9231 117.3387 133.7543 Molar volume, VM (cm<sup>3</sup>/mol) 31.1951 31.9448 34.5309 35.2483 Ion concentration (N  $\times$  10<sup>20</sup> ions/cm<sup>3</sup>) 9.6489 18.8450 26.1505 34.1577 Polaron radius (rp  $\times$  10<sup>-8</sup> Å) 4.0788 3.2631 2.9255 2.6763 Inter-nuclear distance (ri  $\times$  10<sup>-8</sup>Å) 10.1198 8.0960 7.2584 6.6400 Field strength, F  $\times$  10<sup>15</sup> (cm<sup>2</sup>) 2.4043 3.7567 4.6737 5.5847 Molar refraction, Rm (cm<sup>-3</sup>) 9.8628 10.5967 11.7061 12.4104 Molar polarize ability,  $\chi_m$  (Å<sup>3</sup>) 3.9132 4.2044 4.6446 4.9240 Metallization criteria (M) 0.6838 0.6683 0.6610 0.6479 Optical energy gap (Eg opt) 3.0794 3.0759 3.0611 3.0555 Table 1 showed that the Lu 3+ ion concentration increases with increase Lu 3+ content in the glass series network that may be because of the distributed uniformly of Lu 3+ ions in glass network. To make sure for the compactness of P. Glumglomchita et al./

Materials Today: Proceedings 5 (2018) 15054–15060 15057 glass series, using ion concentration to determine the important parameters that can also determine by calculating equation, Polaron radius,  $r_p$  (Å) = (1/2)( $\rho$  /6N)<sup>1/3</sup> (2) Inter nuclear distance,  $r_i$  (Å) = (1/N)<sup>1/3</sup> (3) From Table 1, the  $r_p$  and  $r_i$  decreased with increased Lu 3+ content. The result can explained that the strength of Lu-O bond increased, that occur intense field around Lu<sup>3+</sup> ions which the field strength can measure by the Eq.

4, Field strength,  $F$  (cm<sup>2</sup>) =  $Z / r_p^2$  (4) where  $Z$  is atomic mass of Lu 3+ ion, the molar refraction ( $R_m$ ) is linked to formation of glass network, that can determine from the relationship of refractive index ( $n$ ), density ( $\rho$ ) and molecular mass as given by,  $R_m = (n^2 - 1)(n^2 + 2)(M / \rho)$  (5) The molar electronic polarizability of material ( $\chi_m$ ), related to molar volume ( $V_m$ ) and molar refraction ( $R_m$ ) to refractive index ( $n$ ) of glass series, were computed by using Lorentz-Lorenz formula [13],  $\chi_m = (3/4 \rho NA)(R_m)$  (6) where the value  $3/(4 \rho NA)$  is Lorentz function.

From Table 1, the result noted that the polarizability of oxide ions increased with molar refractivity and refractive index. This increasing is assumed that the B-O bonds were breached when Lu<sup>3+</sup> ions were sprayed into network place and from non-bridging bonds. Dimitrov and Komatsu explained the metallization criterion on the basic of optical band gap and static refractive index ( $M > 1$  for metallic and  $M < 1$  for non-metallic) by using Lorentz-Lorenz equation [14],  $M = 1 - (R_m / V_m)$  (7) From Table 1, the investigating values of metallization criterion confirm the nature of glass series.

It indicated they are non-metallic. 3.3. Absorption spectra The recording optical absorp

tion spectra of a glass series were made at wavelength range 200–2100 nm. The results were shown in Fig. 2, the optical absorption spectra of glass series were broad band absorption edge, which is nature of glassy status. Considering the basic absorption edge (cut off wavelength,  $\lambda_c$  cutoff), it shifts to higher wavelength side from 210 to 400 nm but the optical energy band gap ( $E_{g, opt}$ ) values decreased with increased of Lu<sub>2</sub>O<sub>3</sub> concentration.

The absorption edge of glass series is broad band which do not happen in crystalline chemical mixture because of the disorder in host glass matrix. 3.4. XRD spectral analysis The typical XRD pattern of glass series for 20 mol% of Lu<sub>2</sub>O<sub>3</sub> content showed in Fig. 3. It has two broad band peaks around 25–35° and 40–55° as result of NaBO<sub>3</sub> structure and this is the characteristic of amorphous nature of glass series [12,17].

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600 900 1200 1500 1800 2100 Wavelength (nm) Intensity (arb. unit)  
xLu<sub>2</sub>O<sub>3</sub>:20Na<sub>2</sub>O:(80-x)B<sub>2</sub>O<sub>3</sub> x = 20 mol% x = 15 mol% x = 10 mol% x = 5 mol% 10 20  
30 40 50 60 70 80 0.0 0.2 0.4 0.6 0.8 1.0 Intensity (arb. units)  $2\theta$  in degree  
20Lu<sub>2</sub>O<sub>3</sub>:20Na<sub>2</sub>O:60B<sub>2</sub>O<sub>3</sub> Fig. 2. Optical absorption spectra of Lu<sub>2</sub>O<sub>3</sub> based glasses series. Fig. 3. Typical XRD pattern for 20 mol% of Lu<sub>2</sub>O<sub>3</sub> concentration.

3.5. XRD spectral analysis The typical XRD pattern of glass series for 20 mol% of Lu<sub>2</sub>O<sub>3</sub> content showed in Fig. 3. It has two broad band peaks around 25–35° and 40–55° as result of NaBO<sub>3</sub> structure and this is the characteristic of amorphous nature of glass series [12,17]. 3.6.

Optical energy band gap In 1970, for amorphous materials, Mott and Davis were modified and simplified the relation between optical absorption coefficient  $\alpha(\lambda)$  and energy of photon ( $h\nu$ ) for indirect and direct optical transition that were investigated by formula [15,16,17]  $\alpha(\lambda) = B_0(h\nu - E_{g, opt})^n/h\nu$  (8) where  $B_0$  is a constant called parameter of band tailing,  $E_{g, opt}$  is optical energy band gap between conduction and valence band,  $n$  is the index number which  $n = 2$  for allowed indirect,  $n = 3$  for indirect forbidden,  $n = 1/2$  for allowed direct and  $n = 3/2$  for direct forbidden transitions.

For amorphous materials,  $n = 2$  is used to the experiment [18], so it was applied in this work. In Fig. 4, the plotting absorption coefficient to zero absorption between  $(\alpha h\nu)^{1/2}$  versus photon energy and Fig. 5 showed a relationship between  $E_{g, opt}$  and Lu<sup>3+</sup> content. The results showed that the optical energy band gap values were decreased from 3.0794 to 3.0555 eV with increase Lu<sup>3+</sup> content in glass network.

That indicating, the formation arrangement and chemical composition of glass network

were impact to the optical energy band gap which made it decreased [11]. The increasing of Lu 3+ in glass network has caused the worn out of a linkage B 2O3 network and that made the increasing number of non-bridging oxygen atoms (NBOs) [11,16] also. The bridges between the structural units in the glass network by oxygen ions are known bridging oxygen atoms (BOs).

Therefore, the increasing of network modifiers oxide, the bridges between oxygen will destroy and NBOs will produce. This optical energy band gap ( $E_g$  opt) were decreased, it may be because of disorder in glass matrix were increased that results in more extension of localized states within optical energy band gap which is as proposed by Mott and Davis [11]. P. Glumglomchita et al./ Materials Today: Proceedings 5 (2018) 15054–15060 15059 1.0

1.5 2.0 2.5 3.0 3.5 4.0 4.5 0.0 0.1 0.2 0.3 0.4 0.5 (  $\frac{1}{2} h \nu$  )  $\frac{1}{2} h \nu$  (eV) 5 10 15 20 3.055 3.060 3.065 3.070 3.075 3.080 Optical energy gap  $E_g$  opt Concentration of Lu 2O3 (mol%) Fig. 4. The plotting between (  $\frac{1}{2} h \nu$  )  $\frac{1}{2}$  and  $h \nu$  (eV) for 20 mol% of Lu2O3. Fig. 5. The plotting between optical energy gap and Lu 3+ concentration in glass series. 4. Conclusion A series of glass samples were prepared by melt quenching technique.

From the experiment, it is found out that glass series depends on formations and optical properties . The density of glass series increased as a result of increasing of the content of Lu 2O3. According to the experiment, when the content of Lu 2O3 increased, both molecular mass of lutetium atom and the atomic distance of the element would increase as well. The molar volumes of glass series increased because of an increasing number of NBOs that brought about to the bond damage in loose compacted formation.

The absorption edge of glass series due to applying allowed indirect ( $n = 2$ ). The optical energy band gap decrease from 3.0794 to 3.0555 eV owing to increase of Lu 2O3 content. This led linkage B 2O3 networks untied. Consequently, NBOs numbers were increased. Acknowledgements Among those to be acknowledged for individual contributions through this research project are the National Research Council of Thailand (NRCT) for the funding and the Research and Development Institute at Nakhon Pathom Rajabhat University for the services and facilities. References [1] K. Swapna, Sk.

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