

# An investigation of structural and elastic properties of soda-lime glasses doped with rare earth oxide

N Suebsing\*, N Chutithanapanon, P Juntarat, R Laopaiboon and C Bootjomchai

Glass Technology Excellent Center (GTEC), Department of Physics, Faculty of Science, Ubon Ratchathani University, Ubon Ratchathani, Thailand 34190

E-mail: n.suebsing@gmail.com

**Abstract.** Glass samples in system  $(90-x)\text{RWG}-10\text{Na}_2\text{O}-x\text{Nd}_2\text{O}_3$ . The glass samples were prepared using melt quenching method. The elastic moduli were studied by measurement ultrasonic velocities using pulse echo technique. The structural properties of the glass samples were investigated using infrared absorption spectroscopy. The results illustrate that increase of  $\text{Nd}_2\text{O}_3$  lead to the distorted the glass network, which brings to the decrease of rigidity of glass network.

## 1. Introduction

Glasses are types of materials that often use in our life such as bottles, glasses, window glasses, and mobile phone screen. On the other hand, there are a lot of wastes from glass, which are some product can be recycled and produced the new products such as fiberglass insulation [1]. In this work, the soda-lime wasted glass of window glass used as a raw material because it is a very commercial product, relatively inexpensive, and chemically stable. In addition, there are many research reported on the optical property of glass with doping rare earth oxides. Glasses added rare earth oxides have also used in the optical field such as optical fibers, waveguide lasers, solid state laser, and mid-infrared laser [2–4]. Moreover, these glasses were doped rare earth oxides (ROs) show a good the mechanical strength, chemical, and thermal stability [4]. Neodymium oxide ( $\text{Nd}_2\text{O}_3$ ) is a type of ROs which have been attracting since it has efficient pumping by flash-lamps [5]. Hence, glasses containing  $\text{Nd}_2\text{O}_3$  are one of the interesting in optical applications [2]. However, doping of  $\text{Nd}_2\text{O}_3$  lead to change in glass structure. Thus, the investigation of the structural property related to the change of the network into the glass structure is interesting. The ultrasonic technique is a very significant role in order to understand the structure of the glass network [6]. Therefore, the aims of this investigation are the structure and elastic properties of the glasses by using the ultrasonic technique. Fourier-transform infrared spectroscopy (FTIR) technique was used to confirm the ultrasonic results.

## 2. Experimental details

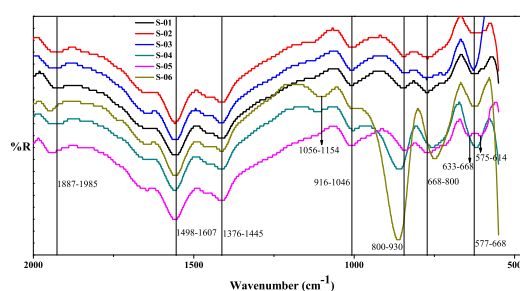
Glass system  $(90-x)\text{RWG}-10\text{Na}_2\text{O}-x\text{Nd}_2\text{O}_3$  (where  $x = 0, 0.1, 0.25, 0.5, 0.75$  and  $1$  in mol% respectively) were prepared by using melt quenching method. The recycling window glass (RWG) was cleaning and crushing into powder. The RWG powder and high purity chemicals ( $\text{Na}_2\text{O}$  and  $\text{Nd}_2\text{O}_3$ ) were weighted by means of an electronic balance. The RWG powders and

chemicals were mixed in the ceramic crucible. The mixtures were melted in an electric furnace at 1250 °C for 2 hours to ensure homogeneously. The density of the glass samples was measured at room temperature using Archimedes principle with n-hexane as the immersion liquid. This experiment was repeated five times for accurate the value of density. The molar volume was calculated for each sample from the expression;  $V_m = M/\rho$ , where M is the molecular weight of the glass samples. Ultrasonic velocities were measured using an ultrasonic flow detector (SONATEST site scan 230) with resonant frequency 4 MHz. The measurements were repeated three times to check the reproducibility of the data. FTIR spectra of the glass samples were recorded in the range of 500-4000  $\text{cm}^{-1}$  using attenuated total reflection (ATR) technique. A recording was revealed the absorption spectra by using Perkin-Elmer spectrum II with the resolution of 4  $\text{cm}^{-1}$ .

### 3. Result and discussion

The densities of glass samples increase with  $\text{Nd}_2\text{O}_3$  was added (table 1). This is due to the molecular weight of  $\text{Nd}_2\text{O}_3$  (336.487  $\text{g}/\text{cm}^3$ ) higher than the host structures of the glass samples ( $\text{SiO}_2$  is 60.0843  $\text{g}/\text{cm}^3$ ). Therefore, adding the  $\text{Nd}_2\text{O}_3$  into network structures lead to increases of the densities. Molar volume is the parameter that describes the volume occupied by the unit mass of a glass, which depends on the ionic radius of the glass modifier oxide [7]. From table 1, the molar volume of the glass samples sharply inclined with adding of  $\text{Nd}_2\text{O}_3$  in mol%. The increase of the molar volume due to adding the lager of ionic radius of  $\text{Nd}^{3+}$  (1.123 Å) into interstitial of host structure (the ionic radius of  $\text{Si}^{4+}$  is 0.400 Å) lead to expansion of glass structure and increasing of the molar volume of the glass samples [6].

The longitudinal and shear velocities decreased with an increase of the concentration of  $\text{Nd}_2\text{O}_3$ . Decreasing of both ultrasonic velocities are related to the amount of non-bridging oxygens (NBOs) in a structure of the glass [8]. These indicate that the network of the glass destroyed, which lead to increase the formation of NBOs with  $\text{Nd}_2\text{O}_3$  was added. Also, dropping of ultrasonic velocities because of  $\text{Nd}^{3+}$  ions were involved in the glass structure as modifiers by breaking the tetrahedral of  $\text{SiO}_4$  units. These results will confirm and discuss by using FTIR technique. The spectra were measured at the range 500-4000  $\text{cm}^{-1}$  using ATR technique. The water groups showed at about frequency bands over 2000  $\text{cm}^{-1}$ . The FTIR spectra of the glass samples showed in figure 1 and the details of the vibrational of the structures listed in table 2.



**Figure 1.** The FTIR spectra of glass samples

From the figure 1, it clear that the increase of stretching of NBOs (about 800-930  $\text{cm}^{-1}$ ) peak with increment of  $\text{Nd}_2\text{O}_3$ , while the Si-O-Si and O-Si-O symmetrical stretching of BOs between tetrahedral (about 668-800  $\text{cm}^{-1}$ ) peak show decrease with increase of the  $\text{Nd}_2\text{O}_3$ . The results reveal that the higher increase of the intensity of the FTIR peak at about 800-930  $\text{cm}^{-1}$ . It is clear that the network structures of the glass samples were transformed the formation of structures from tetrahedral to the trigonal unit, which is the transformation of  $\text{SiO}_4$  form  $\text{Q}_4$  to

$\text{SiO}_4^-$  form  $\text{Q}_3$ . Also, the glass structures occurred the formation of NBOs with adding  $\text{Nd}_2\text{O}_3$ . The FTIR results are good agreement with the results of the ultrasonic technique.

**Table 1.** Glass composition, density ( $\rho$ ), and molar volume ( $V_m$ ) of the glass samples

| Sample | Composition (mol%) |                   |                                | $\rho$<br>(g/cm <sup>3</sup> ) | $V_m$<br>(cm <sup>3</sup> /mol) |
|--------|--------------------|-------------------|--------------------------------|--------------------------------|---------------------------------|
|        | RWG                | Na <sub>2</sub> O | Nd <sub>2</sub> O <sub>3</sub> |                                |                                 |
| S-01   | 90.00              | 10                | 0.00                           | 2.5752                         | 23.2804                         |
| S-02   | 89.90              | 10                | 0.10                           | 2.5828                         | 23.3191                         |
| S-03   | 89.75              | 10                | 0.25                           | 2.5954                         | 23.3658                         |
| S-04   | 89.50              | 10                | 0.50                           | 2.6130                         | 23.4732                         |
| S-05   | 89.25              | 10                | 0.75                           | 2.6369                         | 23.5229                         |
| S-06   | 89.00              | 10                | 1.00                           | 2.6545                         | 23.6275                         |

**Table 2.** The main FTIR spectra of glass samples at the range of 500–2000 cm<sup>-1</sup>

| Wavenumber (cm <sup>-1</sup> ) | Assignment   | References |
|--------------------------------|--|------------|
| <600                           | Vibration of cations such as Na <sup>+</sup>                         | [9]        |
| 668–800                        | Si-O-Si and O-Si-O symmetrical stretching of BOs between tetrahedral | [10]       |
| 800–930                        | Si-O-Si stretching of NBOs   | [10]       |
| 916–1046                       | Si-O-Si anti-stretching of BOs within tetrahedra                     | [11]       |
| 1376–1455                      | Carbonate group  | [11]       |

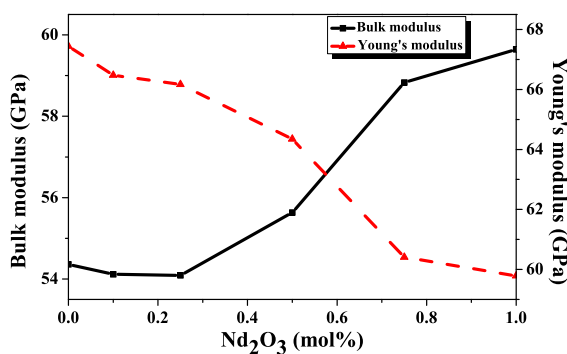
**Table 3.** Longitudinal ( $v_l$ ) and shear ( $v_s$ ) velocities, longitudinal ( $L$ ), shear ( $G$ ), bulk ( $K$ ) and Young's modulus ( $E$ ), micro-hardness and poisson's ratio ( $\sigma$ ) of the glass samples

| Samples | $v_l$ (m/s) | $v_s$ (m/s) | $L$ (GPa) | $G$ (GPa) | $K$ (GPa) | $E$ (GPa) | $H$ (GPa) | $\sigma$ |
|---------|-------------|-------------|-----------|-----------|-----------|-----------|-----------|----------|
| S-01    | 5883        | 3182        | 89.1269   | 26.0742   | 54.3612   | 67.4402   | 3.5942    | 0.2932   |
| S-02    | 5848        | 3152        | 88.3294   | 25.6604   | 54.1156   | 66.4743   | 3.5023    | 0.2932   |
| S-03    | 5827        | 3136        | 88.1240   | 25.5245   | 54.0914   | 66.1660   | 3.4691    | 0.2961   |
| S-04    | 5818        | 3069        | 88.4478   | 24.6112   | 55.6328   | 64.3452   | 3.1628    | 0.3072   |
| S-05    | 5814        | 2936        | 89.1341   | 22.7303   | 58.8270   | 60.4103   | 2.5936    | 0.3288   |
| S-06    | 5808        | 2907        | 89.5439   | 22.4271   | 59.6411   | 59.7873   | 2.4980    | 0.3329   |

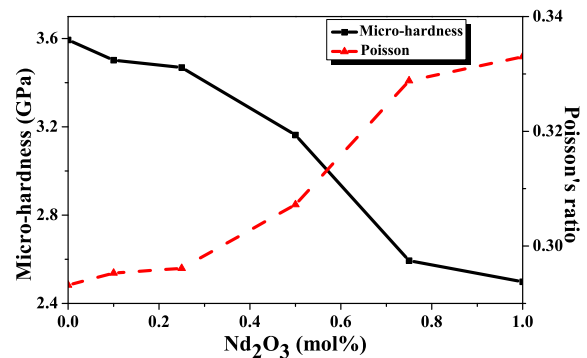
The ultrasonic velocities were used to calculate the elastic moduli. The elastic moduli showed in table 3. The elastic moduli were related to the average strength of the bond between cation-anion [12]. From the result, it found that shear modulus decreased with increasing  $\text{Nd}_2\text{O}_3$  due to increasing the number of NBOs in structures. The longitudinal modulus decreased with increasing of  $\text{Nd}_2\text{O}_3$  in the range of 0-0.25 mol%, then the longitudinal modulus returned to increase with increasing of  $\text{Nd}_2\text{O}_3$  in the range of 0.25-1.0 mol%. The decrease of longitudinal modulus indicates that adding  $\text{Nd}_2\text{O}_3$  lead to the decrease of resistant to an external force of the network structures of the glass samples. However, more adding of  $\text{Nd}_2\text{O}_3$  lead to increase

of densities into the glass samples, which indicate the structures of the glass samples were compactions of interatomic structures lead to the return to increase of longitudinal modulus in this range.

Bulk modulus and Young's modulus displayed in figure 2. The result of Young's modulus indicates the average bond energy of the glass samples decreased due to the increase in the formation of NBOs. However, the bulk modulus of the glass samples was similar trends as the longitudinal modulus. This parameter indicates that the resistant of the glass samples on the external force in all direction is dominant depend on the density. Poisson's ratio climbed steadily with concentration of  $\text{Nd}_2\text{O}_3$  from 0 mol% to 0.25 mol% and then rose dramatically with increasing  $\text{Nd}_2\text{O}_3$  from 0.5 mol% to 1 mol%. This quantity related to the cross-link density [13]. Increasing of Poisson's have been affected by the decrease of cross-link density in glass network because of the influence of  $\text{Nd}_2\text{O}_3$ . The cross-link density of glass structure decrease due to adding of  $\text{Nd}_2\text{O}_3$ . On the other hand, the dropping of cross-link density leads to the decrease of micro-hardness. These variations are defined that the resistance of a material to permanent indentation or penetration. From the figure 3, the micro-hardness dipped with increasing concentration of dopant into the glass network.



**Figure 2.** Variation of bulk and Young's modulus of the glass samples with the difference doping of  $\text{Nd}_2\text{O}_3$



**Figure 3.** Variation of micro-hardness and Poisson's ratio of the glass samples with the difference doping of  $\text{Nd}_2\text{O}_3$

#### 4. Conclusion

This study leads to the understanding of the structural properties of recycled window glass which is the one type of soda lime silicate glass with doped  $\text{Nd}_2\text{O}_3$  by measuring the ultrasonic velocities. The result shows that adding  $\text{Nd}_2\text{O}_3$  content lead to the distorted glass network structure. The elastic property and micro-hardness decrease with the increasing of  $\text{Nd}_2\text{O}_3$  content indicating the decrease of rigidity of glass network.

#### Acknowledgments

The authors would like to thank the Research Professional Development Project Under the Science Achievement Scholarship of Thailand (SAST) for financial support.

#### References

- [1] Poutos K, Alani A, Walden P and Sangha C 2008 *Constr. Build Mater.* **22** 557–65
- [2] Mohan S and Thind K S 2017 *Opt. Laser Technol.* **95** 36–41
- [3] Zhou Y, Zhu C, Zhang M and Liu J 2016 *J. Alloys Compd.* **688** 715–20
- [4] Pawar P, Munishwar S, Gautam S and Gedam R 2017 *J. Lumin.* **183** 79–88

- [5] Mahamuda S, Swapna K, Rao A S, Jayasimhadri M, Sasikala T, Pavani K and Moorthy L R 2013 *J. Phys. Chem. Solids* **74** 1308–15
- [6] Bootjomchai C 2015 *Radiat. Phys. Chem.* **110** 96–104
- [7] Singh H, Singh K, Gerward L, Singh K, Sahota H S and Nathuram R 2003 *Nucl. Instrum. Methods Phys. Res. A* **207** 257–62
- [8] Gaafar M and Marzouk S 2007 *J. Phys. Condens. Matter* **388** 294–302
- [9] Kamitsos E, Patsis A, Karakassides M and Chryssikos G 1990 *J. Non-Cryst. Solids* **126** 52–67
- [10] Zahran R R 1998 *Mater. Lett.* **37** 83–9
- [11] Wang M, Li M, Cheng J and He F 2014 *J. Mol. Struct.* **1063** 139–44
- [12] Higazy A and Bridge B 1985 *J. Non-Cryst. Solids* **72** 81–108
- [13] Laopaiboon R and Bootjomchai C 2014 *Ann. Nucl. Energy* **68** 220–7