Neutron Shielding and Elastic Moduli Properties of Dysprosium Doped Sodium-based Oxide and Oxyfluoride Glass Systems

Sunantasak Ravangvong¹*, Punsak Glumglomchit², Kittisak Sriwongsa^{3,4}

and Chumphon Khobkham⁵

¹Division of Science and Technology, Faculty of Science and Technology, Phetchaburi Rajabhat University, Phetchaburi, 76000, Thailand

²Huahin Vitthayalai School, Hua-Hin, Prachuap Khiri Khan, 77110, Thailand

³Lecturers responsible for Bachelor of Education Program in Physics, Faculty of Education, Silpakorn University, Nakhon Pathom, 73000, Thailand

⁴The demonstration school of Silpakorn University, Nakhon Pathom, 73000, Thailand

⁵Faculty of Engineering, Thonburi University, Bangkok, 10160, Thailand

* Corresponding author. E-mail address: sunantasak.rav@mail.pbru.ac.th

Received: 10 July 2020; Revised: 1 September 2021; Accepted: 8 September 2021; Available online: 28 October 2021

Abstract

The dysprosium doped sodium–based oxide and oxyfluoride glass systems were studied for their neutron shielding and elastic modulus properties. Fast neutron removal cross–section (Σ_{R} , cm⁻¹) was determined by partial density theory. The elastic moduli of glass systems were estimated theoretically by Makishima–Mackenzie (M–M) model. The fast neutron removal cross–section value of dysprosium doped sodium–based oxyfluoride had higher than sodium–based oxide glass systems that indicated oxyfluoride glass system had superior shielding properties than oxide glass system. Moreover, the values of elastic moduli properties for oxyfluoride had higher than also.

Keywords: Oxyfluoride glass, Fast neutron, Elastic moduli

Introduction

The rapid growing used in different sectors of nuclear technology such as nuclear power plants, medical fields, material identification, agriculture, scientific and space technologies. These technologies come with harmful not only for man health but include animals, natural and laboratory instruments. Therefore, nuclear radiations were continuous problem (Azeez, Ahmad, Al-Dulaijan, Maslehuddin, Naqvi, 2019; Kaçal, Akman, Sayyed, & Akman, 2019). The common nuclear radiations are gamma ray (γ -rays), neutron, alpha (α) and beta (β). γ -rays and neutron are neutral unlike α and β are carried charge. Normally, the neutron attenuation was discussed by neutron interaction with matter which these interactions were scattering (inelastic or elastic) or absorption. For fast neutrons (E > 0.1 MeV), the probability of neutron interaction with matter can explained in cross section and neutron removal cross section with shielding medium is probability of neutron reaction per unit path length of penetrate medium (Kaçal et al., 2019; Singh, Kaur, & Kaundal, 2014; Thomas et al., 2019).

Glass is suitable material used for neutrons shielding because of easy to change composition, high transparency to visible light and highly effective neutron absorbers (Issa, Ahmad, Tekin, Saddeek, & Sayyed, 2019). Phosphate glasses are glass systems which can be used for shielding materials because of their properties such as softening temperatures, low melting point, high thermal expansion coefficients and high transparency to visible light which make these glasses are very popular to researches. However, this glass system has limited

properties compared with other glass systems whereas these limited properties can be solved by mixture of suitable oxides into formation (El-bashir, Sayyed, Zaid, & Matori, 2017).

Dysprosium (III) ion (Dy³⁺: 4f⁹), once of popular rare earth ions for doping in glasses. It is a metallic gloss and found in many minerals. Dysprosium is used for backup application because of high magnetic sensitivity and absorber for high heat neutrons in the control rods in nuclear reactors. Salts of soluble dysprosium are slightly toxic while insoluble salts are non-toxic (Huy, Seo, Lim, & Lee, 2011; Sathiyapriya, Marimuthu, Sayyed, Askin, & Agar, 2019; Kaewjaeng et al., 2019).

In this work, dysprosium doped sodium-based oxide and oxyfluoride glass systems had been determined for their neutron shielding and elastic modulus properties for using neutron shielding materials.

Methods and Materials

In this work, chemical composition of dysprosium doped sodium-based oxide and oxyfluoride glass systems were exhibited in Table 1. and Figure 1. These samples were prepared according (Ravangvong, Chanthima, Rajaramakrishna, Kim, Sangwaranatee, & Kaewkhao, 2019).

Table 1 Composition of chemical and density (g cm⁻³) of glass systems (Ravangvong et al., 2019)

Code	Wt.% of elements						Density
	Na	Р	Gd	Dy	0	F	(g cm ⁻³)
NaO	0.1261	0.2837	0.1475	0.0087	0.4340	-	3.2719
NaF	0.0931	0.2837	0.1475	0.0087	0.3901	0.0769	3.2690



Figure 1 Photograph of glass systems

1. Fast neutron removal cross-section ($\Sigma_{\rm R}$)

 $\Sigma_{\rm R}$ mean value to discusses possibility of interaction between neutron and medium that a fission or fast energy neutron travels on main collision that changes it from group of penetrating un-collision neutron. This value of glass systems was estimated by (Kilicoglu et al., 2019):

$$\sum_{R} = \sum_{i} \rho_{i} \left(\sum_{R} / \rho \right)_{i}$$
(1)

here ρ_i and Σ_R / ρ are density (g cm⁻³) and removal cross-section (cm² g⁻¹) of ith element.

2. Physical, structural and elastic properties

The molar volume (V_m) , oxygen molar volume (V_0) and oxygen packing density (OPD) for glass systems have been evaluated using following Eq. (2-4), respectively:

$$V_{\rm m} = \sum \frac{x_i M_i}{\rho} \tag{2}$$

$$V_{o} = V_{m} \left(\frac{1}{\sum x_{i} n_{i}} \right)$$
(3)

$$OPD=1000N\left(\frac{\rho}{M}\right)$$
(4)

here x_i , M_i and n_i are molar fraction, molecular weight of ith component and oxygen atom numbers in each constituent oxide, respectively. ρ , M, and N are density, molecular weight of glass systems and oxygen atom numbers per each composition, respectively (Issa, Kumar, Sayyed, Dong, & Elmahroug, 2018).

Structural parameters of glass systems like bond numbers per unit volume and average crosslink density were evaluated by Eq. (5) and (6):

$$n_{b} = \frac{N_{A}}{V_{m}} \sum_{i} (n_{f} x)_{i}$$

$$\overline{n}_{c} = \frac{\sum_{i} x_{i} (n_{c})_{i} (N_{c})_{i}}{\sum_{i}}$$
(6)

here N_A is constant value of Avogadro, n_f is cations coordination number, x is molar fraction of each component oxide and i is component oxide. x_i is molar fraction of each component, n_c is crosslink per cation and N_c is cation numbers per formula unit for glass system (Ersundu, Ersundu, Sayyed, Lakshminarayana, & Aydin, 2017).

 $\sum_{i} x_i (N_c)_i$

Makishima and Mackenzie (M–M) was introduced theoretical model for estimate Young's modulus of oxide glass. The M–M model which depends on chemical mixture, packing density (V_t) and dissociation energy per unit volume (G_t) of glass was used estimated Young's (E), bulk (B), shear (G), longitudinal (L) modulus, Poisson's ratio (σ), fractal bond connectivity (d) and hardness (H) as following (Issa, Saddeek, Tekin, Sayyed, & Shaaban, 2018; Issa et al., 2018):

$$V_{t} = \left(\frac{1}{V_{m}}\right) \sum V_{i} x_{i}$$
(7)

$$V_{i} = N_{A} \left(\frac{4\pi}{3}\right) \left(xR_{A}^{3} + yR_{O}^{3}\right)$$
(8)

here RA and RO are ionic radius of metal and oxygen, respectively.

$$E=8.36V_{t}G_{t}$$
 (9)

$$B=10V_t^2G_t$$
(10)

$$G = \frac{30V_t^2 G_t}{(10.2V_t - 1)}$$
(11)

$$L=K+\left(\frac{4}{3}\right)G$$
(12)

$$\sigma = 0.5 - \left(\frac{1}{7.2 V_t}\right) \tag{13}$$

$$d=4\left(\frac{G}{K}\right) \tag{14}$$

$$H = \frac{(1-2\sigma)E}{6(1+\sigma)}$$
(15)

Results and Discussion

1. Fast neutron removal cross-section $(\Sigma_{\rm R})$

The determination $\Sigma_{\rm R}$ value was shown in Figure 2. The $\Sigma_{\rm R}$ values of NaO and NaF were 0.1022 and 0.1034 cm⁻¹, respectively. The NaF glass system in this work had $\Sigma_{\rm R}$ values higher than NaO which may be because of NaF had higher atomic number than NaO (Issa, Sayyed, Zaid, & Matori, 2017). From this result, it indicated that NaF glass system was excellent candidate for neutron shielding materials.





2. Physical, formation and elastic properties

The physical, formation and elastic properties for dysprosium doped sodium-based oxide and oxyfluoride glass systems have been exhibited in Table 2 and 3. From Table 2, NaO glass system had value of V_m , V_o , OPD, n_b and $\overline{n_c}$ higher than NaF glass system. The mechanism of V_m perhaps due to ionic radius of O (1.52 Å) was higher than F (1.47 Å). V_o value of NaO higher than NaF; this means that NaO glass system structure had more compact and denser than NaF glass system. The OPD value indicates that NaO glass structure had more tightly bound than NaF.

Code	$V_{m} (cm^{3} mol^{-1})$	$V_0 (cm^3 mol^{-1})$	OPD (mol L^{-1})	$n_{b} imes 10^{23} \text{ cm}^{-3}$	$\overline{n_c}$
NaO	51.3942	0.0771	77.0523	0.7028	2.3586
NaF	50.3991	0.0752	75.1989	0.6760	2.2850

Table 2 $\mathrm{V_{m}},\,\mathrm{V_{o}},\,\mathrm{OPD},\,n_{\scriptscriptstyle b}$ and $\overline{n_{c}}$ of glass samples

The high n_b volume of NaO indicated that NaO had more linked glass network than NaF while low $\overline{n_c}$ value of NaF indicated that NaF glass structure had low network connectivity by creating nonbridging oxygen sites.

Code	V _t	E (GPa)	B (GPa)	G (GPa)	L (GPa)	σ	d	H (GPa)
NaO	0.5694	12.2251	8.3265	5.1955	15.2539	0.2561	2.4959	0.7913
NaF	0.5683	14.2197	9.6663	6.0457	17.7272	0.2556	2.5017	0.9226

Table 3 V, E, B, G, L, O, d and H of glass samples

The estimation of elastic moduli properties by Makishima–Mackenzie (M-M) model as shown in Table 3. The elastic modulus values: longitudinal (L), shear (G), Young's (E), and bulk (B) have been exhibited in Figure 2.

Poisson's ratio (σ) value is parameter discuss the bonds structure of glass system. From Table 3, NaO glass system had σ value higher than NaF glass system which indicates that NaO structure had tighten and rigidity more than NaF glass system.

From Table 3 and Figure 3, the computation elastic moduli (L, G, E, B and H) of glass systems. The results exhibited that NaF glass system had higher elastic moduli than NaO glass system. This result due to the elastic moduli depend on G_t which Na–F bond had G_t higher Na–O bond (Issa et al., 2017).

The fractal bond connectivity (d) parameter used to explained glass formations. The d is 1, 2, 3 mean that chain, layer formation and 3D networks of tetrahedral coordination polyhedral, respectively. From Table 3, d values of glass systems are nearly 2. The result indicated that glass network had 2D layer formation (Issa et al., 2018; Sayyed, Issa, Tekin, & Saddeek, 2018).



Figure 3 Elastic moduli of glass systems

Conclusion and Suggestions

In this study, two glass systems $17\text{NaO} - 17\text{Gd}_2\text{O}_3 - 65\text{P}_2\text{O}_5 - 1\text{Dy}_2\text{O}_3$ and $17\text{NaF} - 17\text{Gd}_2\text{O}_3 - 65\text{P}_2\text{O}_5 - 1\text{Dy}_2\text{O}_3$ were comparative studied neutron shielding and elastic moduli properties. The results found that fast neutron removal cross-section of oxyfluoride glass system had higher values than oxide glass system. Moreover,

the elastic moduli properties of oxyfluoride glass system had higher values than oxide glass system, also. It indicated that oxyfluoride glass system was excellent candidate for neutron shielding materials.

Acknowledgments

The authors thank you to Phetchaburi Rajabhat University, Thailand for support this article.

References

- Azeez, M. O., Ahmad, S., Al-Dulaijan, S. U., Maslehuddin, M., & Naqvi, A. A. (2019). Radiation shielding performance of heavy-weight concrete mixtures. *Construction and Building Materials*, 224, 284–291.
- El-bashir, B. O., Sayyed, M. I., Zaid, M. H. M., & Matori, K. A. (2017). Comprehensive study on physical, elastic and shielding properties of ternary BaO-Bi₂O₃-P₂O₅ glasses as a potent radiation shielding material. *Journal of Non-Crystalline Solids*, 468, 92–99.
- Ersundu, M. C., Ersundu, A. E., Sayyed, M. I., Lakshminarayana, G., & Aydin, S. (2017). Evaluation of physical, structural properties and shielding parameters for K₂O-WO₃-TeO₂ glasses for gamma ray shielding applications. *Journal of Alloys and Compounds*, 714, 278–286.
- Huy, B. T., Seo, M. O., Lim, J. M., & Lee, Y. I. (2011). Application of the Judd–Ofelt Theory to Dy³⁺– Doped Fluoroborate/Sulphate Glasses. *Journal of the Korean Physical Society*, 59(5), 3300–3307.
- Issa, S. A. M., Ahmad, M., Tekin, H. O., Saddeek, Y. B., & Sayyed, M. I. (2019). Effect of Bi₂O₃ content on mechanical and nuclear radiation shielding properties of Bi₂O₃-MoO₃-B₂O₃-SiO₂-Na₂O-Fe₂O₃ glass system. *Results in Physics*, 13, 102165.
- Issa, S. A. M., Kumar, A., Sayyed, M. I., Dong, M. G., & Elmahroug, Y. (2018). Mechanical and gammaray shielding properties of TeO₂–ZnO–NiO glasses. *Materials Chemistry and Physics*, 212, 12–20.
- Issa, S. A. M., Saddeek, Y. B., Tekin, H. O., Sayyed, M. I., & Shaaban, K. S. (2018). Investigations of radiation shielding using Monte Carlo method and elastic properties of PbO-SiO₂-B₂O₃-Na₂O glasses. *Current Applied Physics*, 18, 717–727.
- Issa, S. A. M., Sayyed, M. I., Zaid, M. H. M., Matori, & K. A. (2017). A comprehensive study on gamma rays and fast neutron sensing properties of GAGOC and CMO scintillators for shielding radiation applications. *Journal of Spectroscopy*, 1–9.
- Kaçal, M. R., Akman, F., Sayyed, M. I., & Akman, F. (2019). Evaluation of gamma-ray and neutron attenuation properties of some polymers. *Nuclear Engineering and Technology*, 51, 818–824.
- Kaewjaeng, S., Jumpee, C., Kothan, S., Kaewkhao, J., Srisittipokakun, N., & Kim, H. J. (2019). White emission from NaO-BaO-Bi₂O₃-SiO₂ glass system doped with Dy³⁺. *Materials Today: Proceedings*, 17, 1774-1779.
- Kilicoglu, O., Altunsoy, E. E., Agar, O., Kamislioglu, M., Sayyed, M. I., Tekin, H. O., & Tarhan, N. (2019).
 Synergistic effect of La₂O₃ on mass stopping power (MSP)/projected range (PR) and nuclear radiation shielding abilities of silicate glasses. *Results in Physics*, 14, 102424.



- Ravangvong, S., Chanthima, N., Rajaramakrishna, R., Kim, H. J., Sangwaranatee, N., & Kaewkhao, J. (2019).
 Dy³⁺ ions doped (Na₂O/NaF)-Gd₂O₃-P₂O₅ glasses for solid state lighting material applications. *Solid State Sciences, 97*, 105972.
- Sathiyapriya, G., Marimuthu, K., Sayyed, M. I., Askin, A., & Agar, O. (2019). An investigation on physical, structural and gamma ray shielding features of Dy³⁺ ions doped Telluroborate glasses. *Journal of Non-Crystalline Solids*, *522*, 119574.
- Sayyed, M. I., Issa, S. A. M., Tekin, H. O., & Saddeek, Y. B. (2018). Comparative study of gamma-ray shielding and elastic properties of BaO-Bi₂O₃-B₂O₃ and ZnO-Bi₂O₃-B₂O₃ glass systems. *Materials Chemistry and Physics, 217*, 11-22.
- Singh, K. J., Kaur, S., & Kaundal, R. S. (2014). Comparative study of gamma ray shielding and some properties of PbO-SiO₂-Al₂O₃ and Bi₂O₃-SiO₂-Al₂O₃ glass systems. *Radiation Physics and Chemistry*, 96, 153-157.
- Thomas, C., Rico, J., Tamayo, P., Setién, J., Ballester, F., & Polanco, J. A. (2019). Neutron shielding concrete incorporating B₄C and PVA fibers exposed to high temperatures. *Journal of Building Engineering, 26*, 100859.

