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Effect of Chemical Composition on Radio-Photoluminescence Glass Dosimeter Properties

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Abstract.

The radio-photoluminescent glass dosimeter is applicable for measurement of gamma radiation dose using silver-doped phosphate glass. Therefore, investigations were carried out to establish the optimal composition range of phosphate glasses for radiophotoluminescence glass dosimeter. The basic physical and optical properties of the glasses, such as hardness, transmission and absorbance values have been measured and radiophotoluminescence analyses have been carried out. From the light absorption characteristics of the glasses it was established that they possess a very well expressed transmittance in the UV and visible optical regions. A correlation between the properties investigated and glass compositions were discussed.

Keywords: Glass, optical properties, physical properties & dosimeter

INTRODUCTION

Radiophotoluminescence glass dosimeter (RPLGD) is a prospective tool intended for dosimetric control. They possess linear dose sensitivity in a wide gamma and X-ray spectral region. For that reason, phosphate based glass was chosen due to its ability to accommodate large concentrations of active ions without losing their useful properties of the material [1]. Basically, when the glass is exposed to ionizing radiation, electron-hole pairs are produced and will be trapped within forbidden energy level to form luminescence centers in the phosphate glass [2]. Once trapped, the centers of luminescence will never disappear unless the glasses are annealed at high temperatures of about 400 °C. Therefore, this product gives some excellent features such as repeatable measurement [3].

Improving dosimetric sensitivity and quality of glass by improving the design of base and activator composition in phosphate oxide glass was the main objective in this study. Therefore, phosphate glass dosimeters were prepared from reagent powders of P_2O_5, Li_2CO_3, $Al(OH)_3$, $NaNO_3$, H_3BO_3 and $AgNO_3$ using wet melting method. Finally, the physical, optical properties and their correlation with the glass composition were investigated.

EXPERIMENTAL

The phosphate glass for RPLGD were prepared from reagent grade powders of anhydrous P_2O_5, $AgNO_3$, $Al(OH)_3$, Li_2CO_3, H_3BO_3 and $NaNO_3$. Phosphate glasses of an appropriate composition as given in Table 1 were synthesized. Systematically, the batch powders were mixed with deionized water and the well-mixed liquid batch was preheated at 120 ºC for 24 h in an alumina crucible to evaporate water. After that, the liquid batch was melted in electrical heated furnace in atmosphere at 1250 ºC and poured into a preheated graphite plate for annealing treatment at 400 ºC for 12 h to avoid internal mechanical stresses developed during solidification process. For dosimetric measurement, the samples were irradiated with γ-radiation, delivered at 1.37 kGy/h using Gamma-cell (Gamma-excell-220, UKM). Photoluminescence measurements of each sample were performed at room temperature using FLS-920 spectrophotometer (excitation: 350 nm). The structural and hardness of glasses were characterised using X-ray diffractometer (Bruker D8 Advance) and Rockwell Time TH320. Light absorption and transmittance of the glasses was measured in the UV-Vis region using a UV-Vis spectrometer (Perkin-Elmer Lambda 35).

Table 1: Glass composition of RPLGD

No	Chemical content	Nominal composition range (wt.%)
1	P_2O_5	60.0-90.0
2	$Al(OH)_3$	4.5-9.5
3	Li_2CO_3	2.0-16.0
4	H_3BO_3	0-3.0
5	$NaNO_3$	0-8.0
6	$AgNO_3$	2.0-7.0

RESULTS AND DISCUSSION

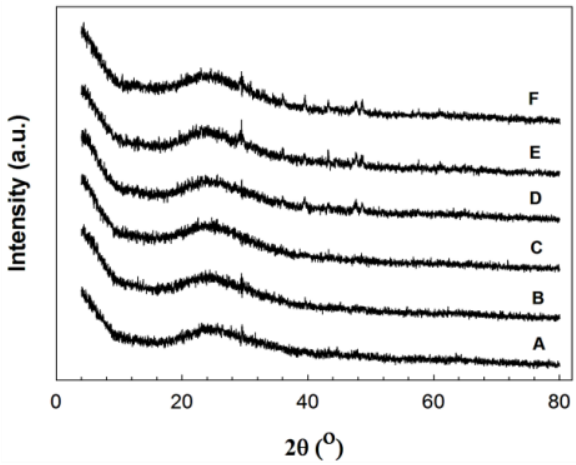
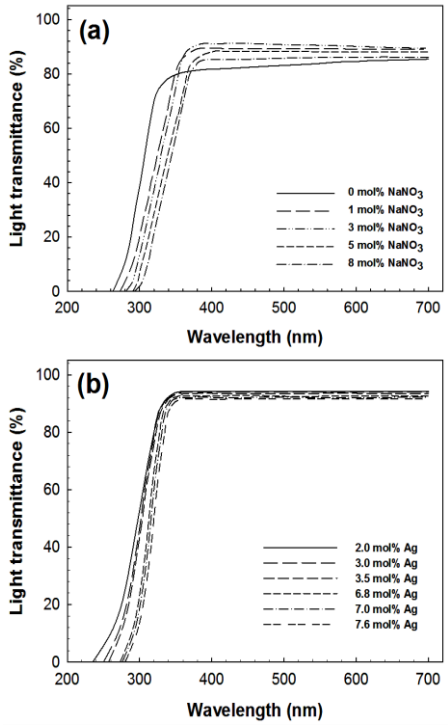


Figure 1: X-ray diffraction patterns of the RPLGD

Figure 1 shows XRD pattern observed for all samples pattern of the silver-activated phosphate glasses. Powder X-ray diffraction patterns for all the glass samples showed a scattering peak at 25° which is due to P-O-P network, characteristic of the glassy phase of RPLGDs, similarly with XRD results obtained from Irman et. al [4]. As the weight chemical content were increasing, no remarkable change was exhibited in the XRD profiles of all samples indicating these glasses achieved metastable phase [5]. Besides, several small sharp peaks were observed in both pattern caused by detector noise forms during the detection and readout process. Similarly, XRD patterns matched the d-spacing corresponding to the monoclinic polymorph.



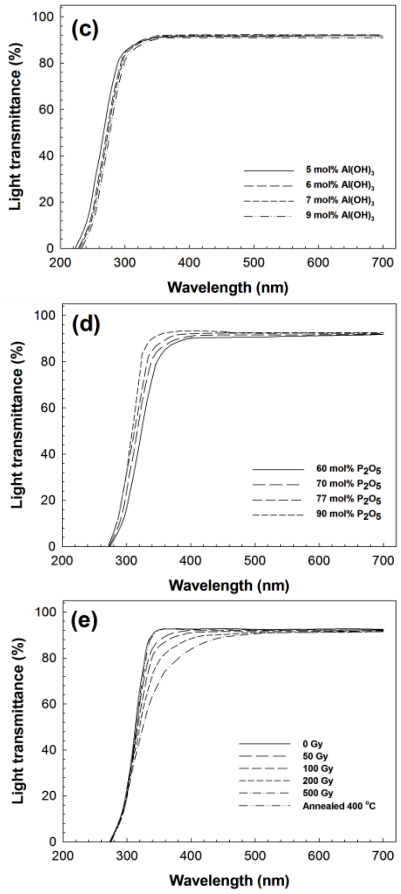


Figure 2: Penetration spectrum of glass system with different wt.% of (a) sodium nitrate, (b) silver metal compound, (c) aluminium hydroxide and (d) phosphorus pentoxide. (e) The irradiated dosage of glasses was from 0 to 500 Gy.

Figure 2 shows the penetration capability of glass in the UV-Vis regions with different wt.% of chemical content. Fig. 2(a) shows that excess Na compound decrease the penetration capability of UV light. However, Na is important for mobility of activator ions in glass matrix. Thus, the optimum wt.% of Na was important in making a good RPLGD with high penetration rate, as well as color center forming in the glass matrix. Therefore, the curve with 3.0wt.% of $NaNO_3$ was chosen as the best result due to the highest penetration rate of the glass system. Besides, Fig. 2(b) shows the light transmittances were decreasing with increasing wt.% of Ag. Hence, higher concentration of Ag results in weaker penetration ability for ultraviolet spectrum [6]. Similarly, excess Al compound also decreased the penetration capability of UV light as shown in Fig. 2(c). In addition, the observed variation in the penetration rate values can be explained based on the conversion of bridging oxygen (BO) to non-bridging oxygen (NBO) atoms brought about by the Al_2O_3 incorporation in the glass. Hence, for glasses containing low wt.% of $Al(OH)_3$, the relative concentration of NBO atoms are high and then their penetration rate values are higher, when compared to the glass with high wt.% of $Al(OH)_3$. Above all, it can be inferred that at higher concentration of $Al(OH)_3$, Al^{3+}

mainly goes to the network to form BO rather than further modifying the network and increasing the concentration of NBO atoms. Additionally, the penetration capability of UV light was enhanced after the wt.% of P_2O_5 were increased due to higher NBO content in the glass system Fig. 2(d). However, different penetration curve was observed in the pattern with different irradiated dose in the same chemical compounds due to formation of neutral Ag atoms within the forbidden band of glass matrix during ionising radiation exposure, as shown in Fig. 2(e). Also, it is evident that glass detectors which have the best transmittance have the highest RPL sensitivity.

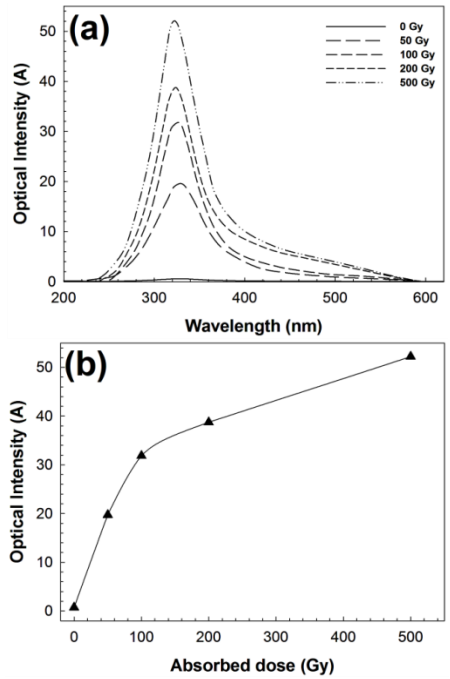


Figure 3: Absorption spectra of RPLGD with 3.5 mol% of silver activator concentrations irradiated with the different doses by Co-60 source. The irradiated dosages of glass were from 50 to 500 Gy with 0 Gy as a control.

Figure 3 shows the absorption spectra of RPLGD with 3.5wt.% of Ag activator concentration with different irradiation doses from 50 to 500 Gy. All the absorption curves are characterized by a broad onset of absorption edge over the region of 200-600 nm as shown in Fig. 3(a). Different irradiation doses affected the absorption spectrum of the irradiated glass. The intensity peak of absorption spectra was increased when irradiation dose increased indicating the formation of significant amount of luminescence centers within the glass network, therefore increasing the absorption intensity peaks as shown in Fig. 3(b). However, the wavelength of absorption spectra peak has not changed for the same synthesised compounds. Thus, the amounts of luminescence centers depend on radiation dosages and optical intensity was directly proportional to the radiation dose absorbed by the RPLGD.

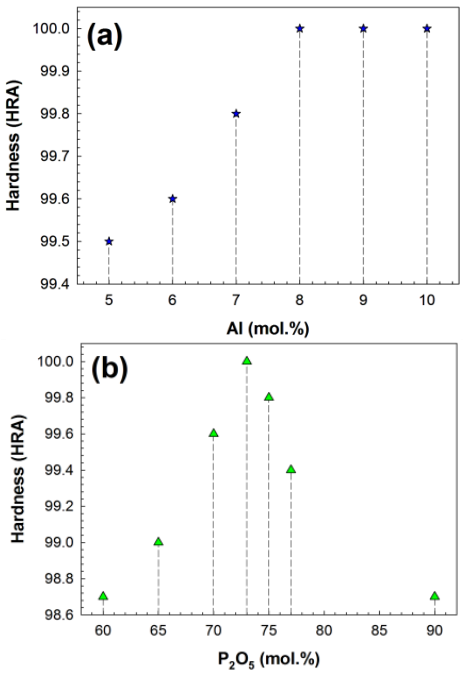
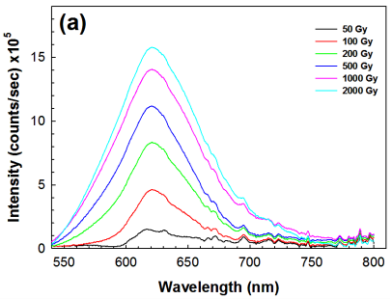


Figure 4: Distribution of Rockwell hardness of RPLGD with different wt.% of aluminium and phosphorus pentoxide contents

The variations of hardness versus RPLGD with different chemical contents are shown in Fig. 4. Knoop hardness numbers were taken, but they had a large standard deviation. Ten hardness measurements were taken with a Rockwell indenter over the polished surface to obtain average hardness numbers glasses. Generally, the Rockwell values of glasses investigated vary from 98 to 100 HRA, which are dependent on the glass composition. From the Fig. 4(a), it is observed that increasing the wt.% of Al results in increasing the hardness values. Al is known to improve durability, but it is also known to raise the glass transition temperature. Therefore, the optimum wt.% of Al for RPLGD system emerges to be in the range of 8-9wt.%. Besides, with other elements fixed, increasing the wt.% of P_2O_5 up to 73% results in increasing the hardness values and decreasing thereafter with increasing wt.% up to 90% as shown in Fig. 4(b). Hence, optimum P_2O_5 percentage for this RPLGD appears to be approximately 73%.



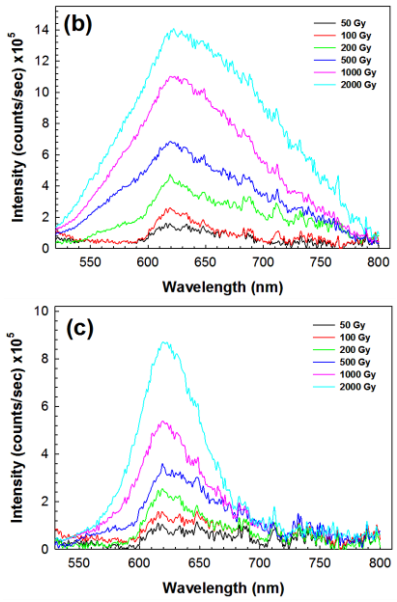


Figure 5: Emission spectrum of RPLGD samples (a) 2.0wt.%, (b) 4.0wt.% and (c) 8.0wt.% of silver contents at different irradiation doses. The samples were excited at 325 nm.

In order to determine the dosimetric sensitivity of RPLGD toward irradiation dose, the glass samples were irradiated with γ-irradiation at varying doses from 50 Gy to 2 kGy, respectively. The RPL spectra were measured to investigate the emission of silver ions corresponding to the dipolar electric transition $4d^{10} \leftrightarrow 4d^{9}5s^{1}$ [7]. Therefore, glass samples with 2.0wt.% in Fig. 5(a) shows the highest intensity of RPL peak followed by 4.0wt.% (Fig. 5(b)) and 8.0wt.% (Fig. 5(c)). The intensity peaks were increasing proportionally with absorbed dose due to the color centers forming within the glass [8]. We suggest that the interpretation of emission spectra is affected by variation in site symmetries from site to site of the color centers, which are the color centers forming gradually with absorbed dose and thus, increase in the PL-intensity peak at visible region around 620 nm band. Moreover, the increase of PL-intensity after a definite absorbed dose values may be explained in terms of build-up in the color centers of Ag°, Ag^{+} and Ag^{2+} within the forbidden band [8].

CONCLUSION:

In summary, the glass made by the wet method had better chemical compounds distribution. The hardness test (Rockwell) indicated that the HRA values increase with increasing Al content. It was established that the glasses possess good optical transmittance (above 80%) for the region above 350 nm. Moreover, we found out that the irradiated glass wavelength of absorption spectra maxima typically occurs in the range of 300-350 nm. Radiation dosage did not affect the peak position of absorption spectra; however, it could affect the optical intensity in the same compounds. Furthermore, different synthesised compounds could also affect the radiation sensitivity. RPLGD series have the highest sensitivity between 2.0 to 4.0 wt.% of silver. Apart from that, the RPL spectra of glass samples were also investigated in this study. RPL emission peaked at 620 nm due to orange RPL at connected to the 325 nm bands of the excitation spectrum. These bands were attributed to the Ag° and Ag^{2+} centres, respectively. The development of a high sensitivity RPLGD is continuously being investigated in our laboratory.

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REFERENCES:

[1] Andreeva, N.Z., Vil'chinskaya, N.N., Dmitryuk, A.V., Perminov, A.S., Petrovskii, G.T. & Savvina, O.C. 1985. Influence of activator concentration on the dosimetric properties of radiophotoluminescent glasses. *Atom. Énergiya*. 58(2): 132-135.

[2] Yokota, R. & Muto, Y., 1971. Silver-activated phosphate dosimeter glasses with low energy dependence and higher sensitivity. *Health Phys*. 20: 662-663.

[3] Ahmed, I., Abou Neel, Valappil, S.P., Nazhat, S.N., Pickup, D.M., Carta, D., Carroll, D.L., Smith, M.E. & Knowles, J.C. 2007. The structure and properties of silver-doped phosphate-based glasses. *J. Mater. Sci. Mater. Med*. 42: 9827-9835.

[4] I. Abdul Rahman, M.T.M. Ayob, H.M.K. Mohd, A.F. Ahmad, S. Sharin, F. Mohamed, S. Ab. Aziz & S. Radiman. 2016. Effect of silver nanoparticle addition on the structure and characteristics of radio-photoluminescence glass Dosimeter. *Malay. J. Anal. Sci*. 20(1): 64-72.

[5] Marta S., Lidia Z., Joanna P., Tomasz G. & Wojciech A.P. 2015. Selective oxide modifiers M_2O_3 (M=Al, Ga) as crystallizing agents in Er^{3+}-doped lead phosphate glass host. *Ceramics International*. 41: 4334-4339.

[6] Hsu, S.M., Yang, H.W., Huang, D.Y.C., Hsu, W.L., Lu, C.C. & Chen, W.L. 2008. Development and physical characteristics of a novel compound radiophotoluminescent glass dosimeter. *Radiat. Meas*. 43: 538-541.

[7] Kurobori, T., Zheng, W., Miyamoto, Y., Nanto, H. & Yamamoto, T. 2010. The role of silver in the radiophotoluminescent properties in silver-activated phosphate glass and sodium chloride crystal. *Opt.*

Mater. 32: 1231-1236.

[8] Maurel, C., Cardinal, T., Bellec, M., Canioni, L., Bousquet, B., Treguer, M., Videau, J.J. & Richardson, M. 2009. Luminescence properties of silver zinc phosphate glasses following different irradiations. *J. Lumin*. 129(12): 1514-1518.