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From Rice Husk to Transparent Radiation Protection Material

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Abstract

High amount of silica (SiO_2) content, 99.36% was successfully prepared from Rice Husk Ash (RHA) through the burning of rice husk. Eventually, quaternary glasses were fabricated using melt quenching technique based on chemical compound composition xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA. The final thickness of glass samples prepared was within ~3.41mm. As the mole percentages of Bi_2O_3 increased, the glass density and molar volume was also increased from 4.45 gcm^{-1} to 6.60 gcm^{-1}. The increment is due to larger atomic number of Bi^{3+} compared to the Si^{2+}, B^{3+} and Zn in the glass samples. However, the oxygen packing density decreased as mole percentages of Bi2O3 were increased from 240.44 g-atom/l to 161.24 g-atom/l. The decreasing in oxygen packing density was due to the high non-bridging oxygen in the glass samples. The radiation attenuation parameterization was measured using ^{241}Am as source of gamma ray interactions at energy 59.54 keV. The mass attenuation coefficient increased from $0.53cm^2g^{-1}$ to $5.12cm^2g^{-1}$ relatively towards the increment of Bi_2O_3 mole percentages. The phenomenon is due to the escalating in photoelectric absorption effect and declining in the Compton scattering caused by high atomic number of Bi^{3+} in the glass samples. Using Lorentz-Lorentz formulation, the refractive index obtained for glass samples prepared was within favourable range from 1.97 to 1.73.

Keywords: Rice husk, Glass, Radiation, Attenuation

Introduction

Rice husk ash (RHA) is a creation of the burning of rice husk and has become a major material for research of various materials. Based on previous study, an experimental was held to determine the maximum amount of silica obtained from rice husk. A cause of high grade amorphous silica is from a rice husk ash with a proper profile of temperature and duration. The amount of silica had been observed and it is decreasing above 700°C (Olawale and Oyawale, 2012). From the recent study, the silica is used as major material to produce binary glass and ternary glass. Borosilicate glass have a wide purpose in a range of fields such as optoelectronics, thermo chemical, solar energy technology and nuclear. The physical properties of borosilicate glass consist of excellent transparent materials, low thermal expansion coefficient; high soften temperature, high refractive index with low dispersion and a high resistance to chemical attack properties (Hu *et al*., 2012). Furthermore, the structural network will be perturbed and may lead to the formation of new structural units (Vijaya Prakash *et al.*, 2001; Khaled *et al.*, 1994). Glass forming substances are fall into two categories of inorganic compounds containing bonds which are partially ionic and partially covalent, and, inorganic or organic compounds which form chain structures with covalent bonds within the chains and van der Waals' bonds between the chains. Glasses containing heavy metal oxide (HMO) have recently attracted the attention of several researchers for the excellent infrared transmission compared with conventional glasses. In present study, bismuth oxide is used and has been identified as a appropriate element to replace lead oxide due to its suitability as a shielding material for gamma rays due to its high effective atomic number and strong absorption of gamma rays. In addition, bismuth oxide has low softening point, high refractive index, and high density (Singh and Karmakar 2012). The toxicity of bismuth and its biological effects have been estimated less than lead (Kaewkhao *et al*., 2011). Futhermore, zinc oxide (ZnO) is used in a most ternary and quaternary glasses as stabilizer in the glass network. It will increase the surface tension of the melts and recover the crystallization stability within the content of the existing portion (Abd. Aziz et al., 2013).

Methodology

SiO_2 preparation

Rice husks (RH) were weighted with electronic balance having accuracy of 0.0001g for 50g and washed with distilled water to remove dirt by used conical flask and filter paper. After that, the cleaned RH were mixed with 0.4 M of hydrochloric acid (HCI) in the ratio of 50g of RH towards 500ml of HCI, stirred for 20 minutes and heated at 110ºC for 3 hours. Then, the RH was rinsed with distilled water to remove the HCl acid. The RH was dried by using oven at 110ºC for 3 hours. Once dried, the RH were placed in alumina crucibles and burned at 550ºC for 6 hours to get white Rice Husk Ash (RHA). The RHA were tested using X-Ray Fluorescence (XRF) instrument for elemental analysis and determine the amount of silica.

Glass Fabrication

The quaternary xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass system (x=0.1, 0.2, 0.3, 0.4 and 0.5) were prepared using a conventional melt-quenching method (Sidek *et al.*, 2009). All the glass samples arranged were homogenous, transparent and bubble free. The glasses were prepared by mixing together specific weights of rice husk ash as silica source – SiO_2 (99.34%), bismuth (III) oxide, – Bi_2O_3 (Alfa Aesar, 99%), zinc oxide – (Alfa Aesar, Nano Tek, 40-100nm APS Powder 99.9%) and boron trioxide – B_2O_3 (Alfa Aesar, 97.5%). The chemicals were then thoroughly mixed in an agate mortar and pestle for half an hour and poured into an Alumina crucible. The crucible was transferred to a furnace and heated at 1100 °C for 1 hour 30 minutes to aid the melting process. When the melting process was complete, the molten liquid was cast into a stainless steel cylindrical shape mould and annealed at 250 °C for 1 hour 30 minutes. The furnace was then turned off for cooling process towards the atmospheric temperature (24 hours). The glass samples were polished using Buehler's silica carbide paper at different grit towards preferable thickness of approximately 3.5 mm for required measurements.

Physical Properties

The density of each sample was measured by the Archimedes' principle using distilled water as the immersion fluid applying from Equation (1) (Suparat, Jakrapong *et al.*, 2011).

$$\rho_s = \rho_{dw}\left(\frac{W_{air}}{W_{dw}}\right) \qquad (1)$$

Where ρ_{dw} = density of distilled water W_{air} is a bulk glass was weight in air and W_{dw} weight of glass in distilled water (Hashim, *et al.*, 2011). Molar volume is given by using the relation between density of glass (ρ_s) and molecular weight (M) of glass as Equation (2):

$$\text{Molar volume} = M/\rho \qquad (2)$$

$$\text{Oxygen packing density} = (1000 \times \rho \times O) \qquad (3)$$

Where ρ = glass density, M= molecular weight of glass composition and O = number of oxygen atoms in the composition (Altaf and Chaudhry, 2010).

Gamma rays attenuation

The radiation attenuation parameterization was measured using ^{241}Am as source of gamma ray interactions at energy 59.54 keV.

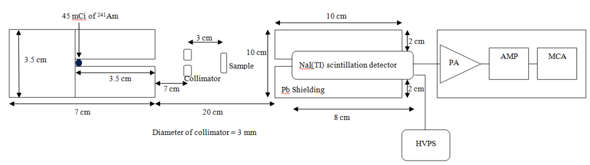

Figure 1: ^{241}Am gamma ray scattering experimental setup.

Refractive indices

The refractive index of glass sample was measured by using UV-Visible NIR Spectrophotometer (model AU12370013 and part number by cary 5000). The UV-Visible NIR spectrophotometer is setup at reflectance mode. The range of wavelength that used was from 0nm to 1500nm. The recording of reflectance percentages recorded from 0 to 100%.

Data analysis, results and discussions

Elemental analysis

After the treatment process using hydrochloric acid (HCl), the size and mass of rice husk decreased from 50g to 28.7g. This is due to the environmental effect during the treatment process. The colour of rice husk transformed from yellow to dark brown. Rice husk had to be burn at certain temperature and time. The RHA was treated at different temperature in order to establish the suitable temperature obtaining elevated amount of SiO_2 by reduction of carbonaceous materials present within the samples (Della *et al.*, 2002). After the burning process, RHA is obtained in white powder appearance at temperature 550ºC for 6 hours. Consequently, the colour of ash was depending on carbon content in the ash. The amount of carbon decrease as the temperature raise (James and Rao, 1986). Therefore, to remove the carbon inside the ash, it takes a long duration and higher temperature (Patel *et al.*, 1987). The Rice Husk Ash (RHA) samples were analyzed by using XRF in order to achieve the element and oxide percentage composition within the samples. X-ray Fluorescence (XRF) spectrometer is an X-ray device used for analysis of rocks, minerals, sediments and fluids. XRF were non-destructive chemical analyses. The percentages of element for the RHA prepared at 550^{0}C are shown in Table 1. The highest amount of silica is 99.36% while other percentages are impurities element in the RHA. The SiO_2 is higher compared to the Della Kühn *et al.*, 2002 which is 94.95% at 700^{0}C for 6 hours.

Table 1 : The Element in Rice Husk Ash (RHA)

Element	Percentages (%)
SiO_2	99.36
P_2O_5	0.16
SO_3	0.10
Na_2O	0.06
CaO	0.15
K_2O	0.05
MgO	0.04
Al_2O_3	0.03
Fe_2O_3	0.03
CI	0.01

Physical properties

The range of the glass quaternary xBi_2O_3-(1-x)ZnO-$0.2B_2O_3$-$0.3(SiO_2)$RHA glass system prepared was around 3.22mm to 3.59mm. The density of glasses were calculated by using Archimedes Principle as Equation (1). The results were showed in the Table 2:

Table 2: Density, molar volume and oxygen packing density of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples.

Mole Percentages of Bi_2O_3 (%)	Density (g/cm^3)	Molar Volume (cm^3)	Oxygen Packing Density (g-atom/l)
10	4.45 ±0.72	24.95±0.23	240.44±0.76
20	5.03±0.32	29.71±0.23	201.96±0.21
30	5.35±0.11	35.12±0.53	170.86±0.23
40	6.10±0.40	37.14±0.63	161.55±0.36
45	6.60±0.74	37.21±0.63	161.24±0.37

The density was increased at range from 4.45kg/cm^3 to 6.60kg/cm^3 when the percentage mole of Bi_2O_3 increase. The density of glass prepared plays an important role of estimating the compactness (Abd. Aziz et al., 2013). Besides that, the molar volume of glass is also increase as density increase at range 24.95cm^3 to 37.21cm^3 as shown in Table 2 and Figure 2. The molar volume and density of glass increased as mole percentages of Bi_2O_3 increased because the molecular weight of Bi_2O_3 is higher than ZnO , B_2O_3 and SiO_2 and atomic number of Bi is higher than Zn, B and Si. Eventually, the increase in molar volume is due to the augmentation in bond length increased in the bond length or an increased inter atomic spacing between atoms (Halimah, Daud *et al.* 2010).

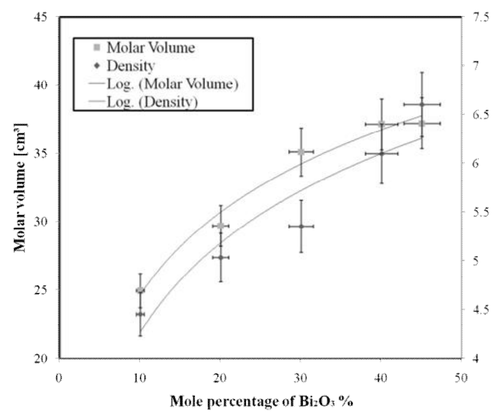

Figure 2: Molar Volume and Density of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples.

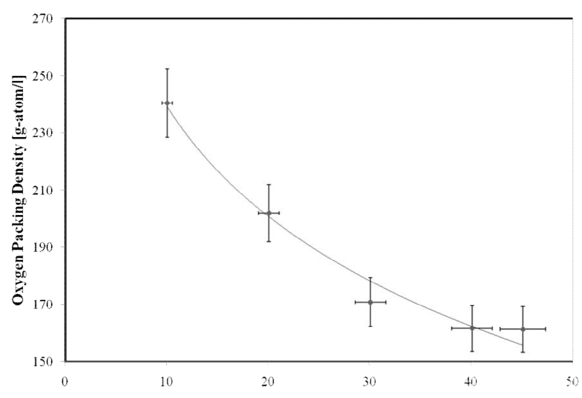

Figure 3: Oxygen Packing Density of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples.

While, the oxygen packing density is decreased when the molar volume and density of glass increased at range 240.44g-atom/l to 161.24g-atom/l as show in Table 2 and Figure 3.

The decreases in oxygen packing density as mole percentages of Bi_2O_3 increased are also discover by the

Saritha *et al.*, 2009. These show that the structure becomes freely packed with increased in mole percentages of Bi_2O_3 (Saritha *et al.*, 2009). It is due to the density which was affected by the structural softening, compactness, change in coordination number and dimension of interstitial spaces of glass (Abd. Aziz et al., 2013). As a result, the non-bridging oxygen atom increased due to the higher atomic number of Bi^{3+} in the glass system. Therefore, the oxygen packing density deceased. The result was also recorded by Rakpanich, Kaewkhao et al. 2013 in X-rays luminescence, optical and physical studies of Bi_2O_3-B_2O_3-Sm_2O_3 glasses system.

Gamma rays attenuation

The linear attenuation coefficient (μ) is the fraction of photos removed from mono-energetic beams of x-rays or gamma rays per unit thickness of materials. The unit of μ is inverse centimeters (cm^{-1}). The μ can be determined by using equation (4):

$$N = N_0 e^{-\mu x} \tag{4}$$

Where N is the number of transmitted photon, N_0 is the number without an absorber, x is the thickness of the sample (Bushberg and Boone, 2011). The thickness of glass is at range 3.15mm to 3.5mm. The result of linear attenuation coefficient (μ) was illustrated in Figure 4.

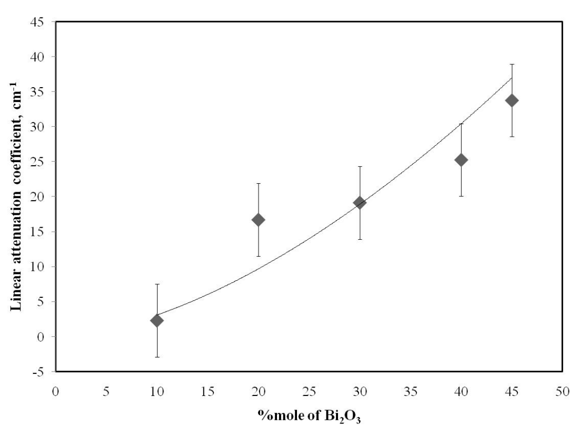

Figure 4: Linear attenuation coefficient of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples.

The mass attenuation coefficient is a measurement of how strongly a chemical species or substance absorbs or scatters light at a given wavelength, per unit mass. The mass attenuation coefficient is a measurement of how strongly a chemical species or substance absorbs or scatters light at a given wavelength, per unit mass. The mass attenuation coefficient is the quantity obtained by dividing the linear attenuation coefficient by the physical density. Mathematically, it is articulated as:

$$\mu_m = \frac{\mu}{\rho} \tag{5}$$

where: μ_m = mass attenuation coefficient, μ = linear attenuation coefficient of a medium and ρ = physical density. The result obtained for mass attenuation coefficient, μ_m is illustrated in Figure 5:

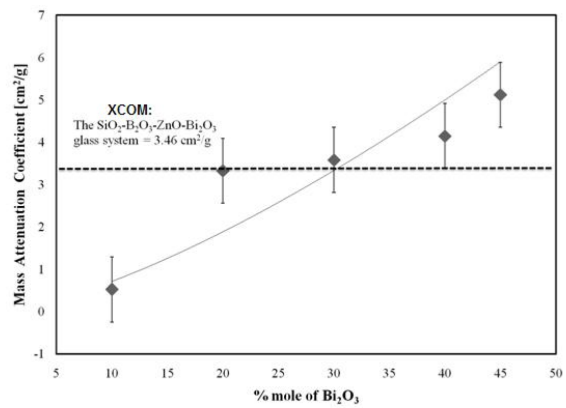

Figure 5: Mass attenuation coefficient of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples compared to given value (3.46 cm^2/g using XCOM prediction program.

From Figure 4 and Figure 5, the linear and mass attenuation coefficient is increasing respectively with addition in mole percentages of Bi_2O_3. Linear attenuation coefficient is increasing from 0.23 cm^{-1} to 3.43 cm^{-1} and mass attenuation coefficient is increasing from 0.052 $cm^2\ g^{-1}$ to 0.520 cm2 g^{-1}. This is due to the rising the value of Bi atom which has higher atomic number as compared to the other element (Sharma *et al.*, 2006). As the percentage mole of Bi_2O_3 increased, it relatively shows a good agreement with prediction value obtained from XCOM program. The increasing in linear and mass attenuation coefficient can be recognized to the growing in photoelectric absorption interaction of glass sample (Singh *et al.*, 2005). The meaning of photoelectric absorption is an x-ray photon interaction with an inner-shell electron. The incident photon is fully absorbed by the ejection of the electrons. Therefore, the photoelectric interaction is more possible to take place with an electron that is more strongly bound in its orbit. Binding energies of the electrons are larger in the high atomic number element evaluate to the low atomic number elements.

The half-value layer (HVL) is defined as different as the thickness of material necessary to reduce the intensity of an X-ray or γ-ray beam to one half of its original value. The HVL of a ray is an indirect measure of the photon energies know as the quality of a beam, previously measured under conditions of narrow-beam geometry. Frequently practical applications of attenuation such as patient imaging occur under broad-beam condition. The HVL was calculated from the linear attenuation coefficient (Bushberg and Boone 2011).

$$HVL = \frac{0.693}{\mu} \tag{6}$$

From Figure 6, the HVL value decreased when the linear and mass attenuation coefficient increased. The decreasing in HVL was from 3.01 cm to 0.20 cm with increased in mole percentages of Bi_2O_3 from 10% to 45%. It is due the increasing of photoelectric absorption. The HVL value is low relative to the XCOM prediction value for lead, Pb with 0.01 cm thickness which indicates that the glass samples were in better gamma radiation shielding materials because of lesser volume requirements of the materials (Sharma *et al.*, 2006).

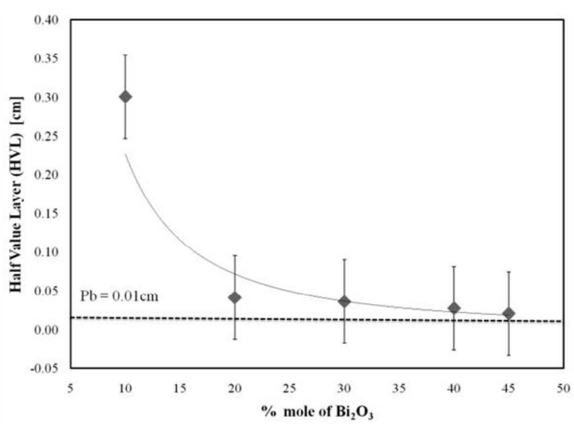

Figure 6: The Half Value Layer (HVL) of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples compared to given value of Pb (0.01cm) using XCOM prediction program.

Refractive indices

The refraction of light means that the bending of light when it passes from one transparent medium into another. the refractive index was calculated by using Lorentz-Lorentz equation. The calculation of refractive index by using Lorentz-Lorentz Equation for glass in Equation (7):

$$R_m = \left[\frac{(n^2-1)}{(n^2)+2}\right] V_m \tag{7}$$

Where R_m is molar refraction of glass , V_m is molar volume of glass and n is refractive index (Dimitrov and Komatsu, 2010).

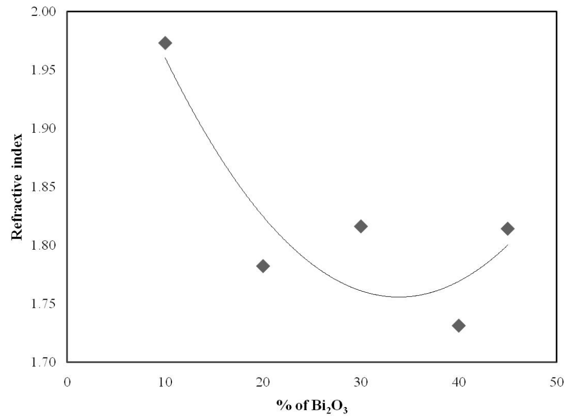

Figure 7: Refractive indices of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples.

As illustrate in Figure 7, the polarization refractive index was comforming decreament from 1.97 at 10% of Bi_2O_3 towards 1.73 at 40% and slight increased at 45% for 1.81. From the data obtained, the refractive index for the glasses sample was not stable due to the oxygen packing and structure in the amorphous Bi^{3+} and ZnO of glass system. Futhermore, the refractive indices of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass samples complies with commercial glass and lead glass refractive indices, range from 1.44 – 1.90.

Conclusions

Heavy metal oxide uaternary glasses were successfully fabricated using SiO_2, 99.36% prepared from Rice Husk Ash (RHA) through the burning of rice husk. As the percentage mole of Bi_2O_3 increased, the experimental value of linear and mass attenuation coefficient relatively shows a good agreement with prediction value obtained from XCOM program. Furthermore, the refractive indices of xBi_2O_3 - (1-x)ZnO - $0.2B_2O_3$ - $0.3(SiO_2)$RHA glass complies with commercial glass and lead glass refractive indices, range from 1.44 – 1.90. Thus, the glasses prepared are considered a good transparent radiation protection material.

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References

Abd. Aziz, Sidek and Shaharuddin, Rosmawati and Zakaria, Azmi and Shaari, Abdul Halim (2013) Effect of ZnO on the thermal properties of tellurite glass. *Advances in Condensed Matter Physics*, 2013 . art. no. 783207. pp. 1-6. ISSN 1687-8108; ESSN: 1687-8124.

Altaf, M. and M. A. Chaudhry (2010). Physical Properties of Lithium Containing Cadmium Phosphate Glasses. *Journal of Modern Physics* 1(4).

Bushberg, J. T. and J. M. Boone (2011). The essential physics of medical imaging, Lippincott Williams & Wilkins p 46-50.

Della, V., I. Kühn and D. Hotza (2002). Rice husk ash as an alternate source for active silica production. *Materials Letters* 57(4): 818-821.

Dimitrov, V. and T. Komatsu (2010). An interpretation of optical properties of oxides and oxide glasses in terms of the electronic ion polarizability and average single bond strength." *J. Univ. Chem. Technol. Metal* 45: 219-250.

Halimah, M., W. Daud, H. Sidek, A. Zaidan and A. Zainal (2010). Optical properties of ternary tellurite glasses. *Materials Science-Poland* 28(1): 173-180.

Hashim, S., H. Sidek, M. Halimah, K. Matori and W. Yusoff (2011) .Physical Properties Of Borotellurite Glass Dopes With Manganase. *Solid State Science and Techology*: Vol.19 No.2: 342-347

Hu, X., G. Guery, J. Boerstler, J. D. Musgraves, D. Vanderveer, P. Wachtel and K. Richardson (2012). Influence of Bi_2O_3 content on the crystallization behavior of TeO_2–Bi_2O_3–ZnO glass system. *Journal of Non-Crystalline Solids* 358(5): 952-958.

James, J. and M. S. Rao (1986).Silica from rice husk through thermal decomposition. *Thermochimica acta* 97: 329-336.

Kaewkhao, J., K. Kirdsiri, P. Limkitjaroenporn, P. Limsuwan, J. Park and H. Kim (2011). Interaction of 662 keV Gamma-rays with Bismuth-based Glass Matrices. *Journal of the Korean Physical Society* 59(2): 661-665.

Khaled, M.A.; Elzahed, H.; Fayek, S.A.; El-Ocker, M.M. Optical absorption, infrared and differential thermal analysis studies of borotellurite glass containing nickel. *Mater. Chem. Phys.* 1994, 37, 329–332.

Olawale, O. and F. A. Oyawale (2012). Characterization of Rice Husk via Atomic Absorption Spectrophotometer for Optimal Silica Production. *International Journal of Science and Technology* 2(4).

Patel, M., A. Karera and P. Prasanna (1987). Effect of thermal and chemical treatments on carbon and silica contents in rice husk. *Journal of Materials Science* 22(7): 2457-2464.

Saritha, D., M. Salagram and G. Bhikshamaiah (2009). Physical and optical properties of Bi_2O_3-B_2O_3 glasses. IOP Conf. Series*: Materials Science and Engineering* 2 012057, doi:10.1088/1757-899X/2/1/012057.

Sharma, G., K. Singh, S. Mohan, H. Singh and S. Bindra (2006). Effects of gamma irradiation on optical and structural properties of PbO–Bi_2O_3–B_2O_3 glasses. *Radiation Physics and Chemistry* 75(9): 959-966.

Sidek, H.A.A.; Rosmawati, S.; Talib, Z.A.; Halimah, M.K.; Daud, W.M. (2009). Synthesis and Optical Properties of ZnO-TeO_2 Glass System. *American Journal of Applied Sciences* 6 (8): 1489-1494. ISSN 1546-9239

Singh, N., R. Singh, K. Singh and K. Singh (2005). X-ray shielding properties of lead and bismuth borosilicate glasses. *Glass Technology-European Journal of Glass Science and Technology Part A* 46(4): 311-314.

Singh, Shiv Prakash and Karmakar, Basudeb (2012) Bismuth Oxide and Bismuth Oxide Doped Glasses For Optical and Photonic Applications. In: Bismuth: Characteristics, Production and Applications. *Materials Science and Technologies*. Nova, Hauppauge, New York, Chapter-9. ISBN 978-1-61470-640-3.

Suparat, T., K. Jakrapong, L. Pichet and C. Weerapong (2011). Development of BaO: B_2O_3: flyash glass system for gamma-rays shielding materials. *Nucl Sci Technol* 1: 110-113.

Vijaya Prakash, G.; Narayana Rao, D.; Bhatnagar, A.K. Linear optical properties of niobium-based tellurite glasses. *Solid State Commun.* 2001, 119, 39–44.