Non-destructive Micro-charatrazition of Gamma Irradiation Effects on Borosilicate Glass $BaO\text{-}2B_2O_3\text{-}3SiO_2$ by Acoustic Signature

By

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Abstract

In this paper, we investigate gamma-radiated effects on the elastic acoustic parameters(reflection coefficients R(θ), acoustic signatures V(z), critical angles of wave excitation, spatial periods, velocities) of barium lead borosilicate with the different gamma-radiated dose rate of 0 , 3, 6, 9, and 12 . The radiation effects on structural and elastic properties were evaluated by measuring the ultrasonic velocities. It was found that, as Gama radiation dose rate increases, longitudinal velocities vary from 4172 to 4344 m/s whereas Rayleigh velocities vary from 2991 to 3084 m/s. Hence, we first deduce the values of propagating surface acoustic wave velocities, and in the second step we find the values of bulk and surface velocities before radiated and after Gamma-radiated glass samples. The effect of Gamma irradiation dose range from 0 Gy to12 Gy effects on the acoustic signature, V(z) amplitudes and periods and reflection coefficients R(θ) critical angles of wave excitation of these glasses is also reported. The calculated reflectance functions that experience several features near critical angles at which longitudinal, shear and Rayleigh modes are existed depend on Gamma radiation dose. It has been observed that, irradiation of the glasses with the Gamma rays increases the BO_3 groups and the non bridging oxygen which make the network loose.

1 INTRODUCTION

Borate glasses are very interesting amorphous materials considering their specific structure and physical properties. In recent years, research has paying attention on the development of new types and scientifically important materials as Borosilicate glasses that motivate forming system and often used as a dielectric insulating and good chemical resistance material [1]. Organic materials as well as some glassy ones, especially polymers, have been increasingly interesting for different purposes such as thick films technology ink constituents. Therefore, the scientific information of the glass structure before and after irradiation is a requirement for understanding the structural progress of nuclear glasses under continuing irradiation during storage of radioactive wastes or isotopes sources, radiation shielding, radiation detection by using glass dosimeter, etc.[2]. Studies on irradiated glasses have been previously published on simple glass systems such as silicate glasses [3]or on multi-component glasses such as borosilicate glasses[4-6].Several earlier work known that lead borate glass is interest because of the occurrence of the boron anomaly foremost to rich in glass chemistry [7]. Therefore, the structure and the classification of the main structural groups of borosilicate glass has been widely studied by using a number of techniques, as this system has a extensive diversity of technological applications such as; optical lenses, nuclear waste materials, shielding materials and use in the electronics industry [8, 9]. The potentials and inter-atomic forces in the lattice structure are the important parameter related to properties of glass. Thus, changes in the lattice, due to doping and/or radiation can be directly affected [10-14].

In the other hand, the ultrasonic non-destructive pulse-echo technique plays a important role n understanding the structural properties of glass network and evaluating the acoustical parameters, such as longitudinal, shear and Rayleigh ultrasonic wave velocities, elastic moduli [15-18]. These parameters give information about the microstructure as well as the behavior of the network formers and change in the glass [19-21].

Gamma irradiation causes changes in the physical properties of materials. Where, Gamma irradiations scattering and absorption generally related to the density and atomic number of material sample. Conversely, in composite materials, the important is knowledge of the mass attenuation coefficients, as well as the relation to density and effective atomic number, is of prime [22]. Conventional influence due to Gamma irradiation in glass producing defects where can be Clearfield by UV-visible spectroscopic work [22, 23]. Electronic processes change is the principal effect of ionizing radiation. Particularly, these effects take place because electrons are excited to leave their normal positions and move through the glass set of connections. Ultrasonic velocities were measured as a function of composition, from which elastic resistances of the network former and the modifier are obtained on the basis of the theory of elastic internal energy [24].

2 MATERIALS AND COMPUTING PROCEDURE

2.1 MATERIALS

A series of barium lead borosilicate glass samples were choosing consistent with the formula, xBaO-(50-x)PbO– 20B2O3 – 30SiO2 (x = 2 mol. %). The starting materials used in the experimental study were reagent grade BaO, PbO, B2O3 and SiO2. It was mixed together and calculated to give a sample of 50g. The homogeneity of the mixture of chemicals was achieved by repeated grinding using agate mortar. The powder mixture was then put in appropriate clay crucibles and heated in melting furnace at 1250 °C for 4 h. These melts were cast in a stainless steel mold and annealed at 450 °C for 2 h and allowed to cool to room temperature [24].

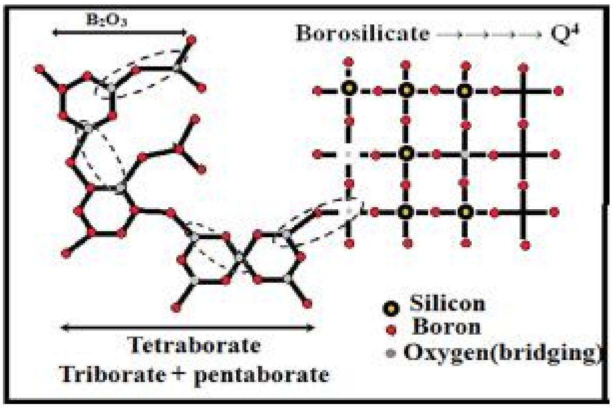

Fig. 1. showed combination of triboratepentaborate units. Filled circles and open circles represent boron and oxygen atoms, respectively. Dashed lines in the structures denote connections to the network, and charges are shown for the non- bridging oxygen (NBO) in the metaborate groups [24].

2.2 GAMMA IRRADIATION

The glass samples were radiated by exposure machine (THERATRON 780C) using 60Co c-ray source at a dose rate of 174.70 cGy min and field size of 30×30 cm^2 at 80 cm from the source at room temperature. The different doses of irradiation were achieved by exposing the sample to the source of different periods. The used Gamma-ray dose rates are 3, 6, 9 and 12 Gy, respectively [24]. It was shown in table 1. that the densities of the glass samples increase with increasing the irradiated by Gamma radiation rates.

Table 1

Experimental data [24] of longitudinal and shear Velocities in BaO-PbO–20B2O3–30SiO2 Doses of Irradiation.

Dose (Gray)	ρ (kg/m3)	V_L (m/s)	V_S (m/s)
0	4212	4200	2186
3	4222	4234	2219
6	4231	4273	2258
9	4241	4318	2303
12	4253	4372	2357

2.3 NONDESTRUCTIVE TESTING AND CALCULATING PROCEDURE

The calculation method, illustrated in detail somewhere else [17-23], which is consists of a number of steps:

1. calculating the reflection coefficient, R(θ) from equation (1) [14]:

$$R(\theta) = \frac{Z_L \cos^2 2\theta_S + Z_S \sin^2 2\theta_S - Z_{L1q}}{Z_L \cos^2 2\theta_S + Z_S \sin^2 2\theta_S + Z_{L1q}} \quad (1)$$

where Z_L liquid longitudinal impedance, Z_S liquid transversal impedance,ρ_{Liq}coupling liquid density and $V_{Liq}$the propagating wave velocity in the liquid which can be calculated from Eq.2.

$$Z_{L1q} = \frac{\rho_{L1q} V_{L1q}}{\cos \theta_i}, Z_L = \frac{\rho V_L}{\cos \theta_l}, Z_S = \frac{\rho V_S}{\cos \theta_s}$$

2. Calculating the acoustic signature V(z) curves of the full specimen-lens system from the angular spectrum model [18] given by the Eq. 3 :

$$V(Z) = \int_0^{2\pi} P(\theta)^2 R(\theta) e^{i2kz \cos\theta} \sin\theta \cos\theta \, d\theta \quad (3)$$

where $P^2(\theta)$is the pupil function, θ is the half-opening angle of the lens, z is the defocusing distance and is the wave number in the coupling liquid, j = √-1.

3. deducing acoustic signature V(z) of the sample by subtracting the response of the lens Vl(z) from V(z),
4. applying fast Fourier transform, FFT, a spectral technique to the obtained signal V(z), in order to determine spatial periods Δz, deducing the phase velocity of the corresponding mode from the following relation (4) [18]:

$$V_R = \frac{V_{Liq}}{\sqrt{1-\left[\frac{V_{Liq}}{2f\Delta z}\right]^2}} \qquad (4)$$

where f is the operating frequency.

2.4 SIMULATION CONDITIONS

Theoretical simulations were carried out in the case of a scanning acoustic microscope in the reflection mode under normal operating conditions: an operating frequency of 140 MHz, a half opening angle of 50° and using the water as the coupling liquid whose density, ρ = 1000 kg/m3 and longitudinal velocity VL = 1500 m/s.

TABLE 2

calculated z_l liquid longitudinal impedance, z_s liquid transverse impedance of barium lead borosilicate glass BaO-PbO–$20B_2O_3$–$30SiO_2$

Reflection mode	θ_L		θ_S	
Dose (Gray)	Z_{Liq}(MKSA)	Z_m(MKSA)	Z_{Liq}(MKSA)	Z_m(MKSA)
0	1.60672	53.5606	2.018	11.963
3	1.60458	39.1565	2.0021	11.990

6	1.60246	39.6686	1.9750	12.278
9	1.60037	23.5326	1.9411	12.787
12	1.59728	42.7682	1.9161	13.009

3 RESULTS AND DISCUSSION

3.1 GAMMA-IRRADIATION EFFECTS ON R (Θ)

It is informative to follow the modify of the reflection coefficient R(θ) over the whole range of incidence angles for the case of a water-barium lead borosilicate glass interface with several irradiation doses of 0, 3, 6, 9 and 12 Gray shown in Fig. 2 . It should be noted that, R(θ) is a complex function that admits an amplitude (modulus) |R(θ)| and phase from Eq. 1. For waves incident on the structure we individually calculate it. At normal incidence, the reflection coefficient value lies between 0 and 1 depending on the acoustic mismatch between the two media. Only the longitudinal wave transmitted and there is no mode conversion, i.e., no shear wave transmitted at normal incidence. As θ increases, longitudinal and shear waves are excited in the solid. R(θ) stays more or less constant until the longitudinal critical angle, at which point it rises sharply to spike at |R(θ)| ≡ 1. At this angle, the longitudinal wave propagates along the surface so no energy propagated into the solid. The shear wave amplitude goes to zero at this angle and there is total reflection as shown in Fig. 2a. As θ further increases, we get at a second critical angle θ_s for shear waves, which now spread along the surface From θ_s out to 90° there is total reflection of the incident wave, |R(θ)| ≡ 1.

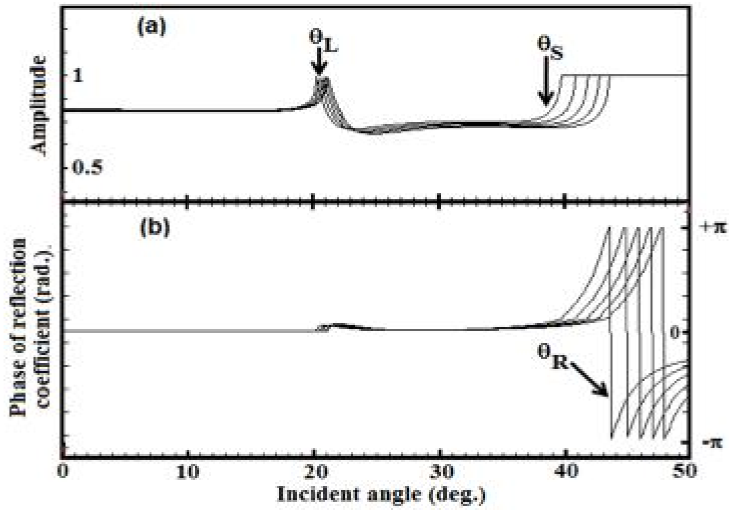

Fig .2.illustrates typical results of the amplitude (Fig. 2a) and the phase (Fig. 2b) as a function of incidence angle.

It is clear that, as the angle of incidence increases we observe important changes in both amplitude and phase. It can readily be deduced that shear modes are excited at θ_S ~ 42°, 41.5°, 40.6°, 39.4° and 38.5° for Gamma irradiated dose rate of 0 , 3, 6, 9, and 12 respectively. In phase, the most important variation, with $\Delta\varphi = 2\pi$, occurs at the critical angle, θ_R, at which Rayleigh modes are excited. Thus, one can simply determine that θ_R ~ 47.5°, 46.7°, 45.7°, 44.6° and 43.4° for Gama irradiated dose rate of 0 , 3, 6, 9, and 12 respectively. Moreover, it can be seen visibly longitudinal modes θ_L variation from 21° to 20.1°. It should be noted that the onset of the 2π phase change corresponds to the shear critical angle θ_S, (as indicated by the arrow in Fig. 2b). The slight shift between
θ_S and θ_R is due to the fact that the Rayleigh velocity varies slightly from 87 to 95 % of the shear velocity [14]. The degree fluctuations in phase and amplitude in R (θ) in the different Gama irradiated dose rate of each critical angle are indicative of the generation efficiency of different modes.

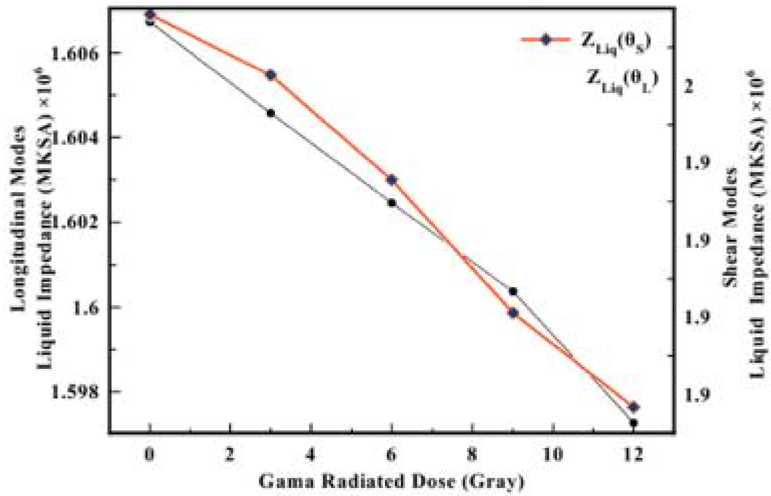

Fig 3 variations of longitudinal and shear liquid impedance with gamma irradiated dose rate.

3.2 GAMMA-IRRADIATION EFFECTS ON ACOUSTIC SIGNATURE V(Z)

Acoustic signatures, or V(z), can be either measured experimentally or determined theoretically. In the present investigation, we considered the latter case via the application of Eq. 5 deduced by the angular spectrum model [18].
In Fig. 4, we illustrate the calculated acoustic signatures for a 50° lens opening angle, a frequency of 140 MHz and water coupling. It can be seen that there are strong oscillations, where a series of periodic maxima and minima occurs, characterized by a period Δ(z). This region is characteristic of the sample's acoustic properties. The patterns vary with the material doping, as do the depths of the minima and the relative magnitude of the maxima which on Ag doping as well.

Acoustic materials signatures, V(z), are the most important parameter that can be experimentally measured by a scanning acoustic microscope. Using equation (1) and previously determined the curves of (Fig. 2a and Fig. 2b), we deduced the acoustic signatures that are illustrated in Fig.4.It can clearly be seen that all the curves exhibit a series of regular periodical oscillations due to constructive and destructive interference between different propagating modes in different Gama irradiated dose rate. However, a close look show that as the Gama irradiated dose rate increases we notice a change in V(z) amplitudes as well as in periods. Moreover, The difference between two successive minima (or maxima) is known as the spatial period, Δz, related to the velocity of the propagating mode byEq. 4. Thus, it is obvious that the spatial period Δz decreases gradually from Gama irradiated dose 47.5 to 43.4 and the amplitude of V(z) output increases. The deduced values reported in Table 3.

Table 3. Calculated of elastic parameters of Barium Lead Borosilicate Glass BaO-PbO–20B2O3–30SiO_2

Mode	Rayleigh			Longitudinal		
Dose (Gray)	θ_R (deg.)	Δz (μm)	V_R (m/s)	θ_L (deg.)	Δz (μm)	V_L (m/s)
0	47.5	39.7	2991	21	80.1	4172
3	46.7	40.8	3029	20.8	81.0	4195
6	45.7	41.1	3039	20.6	82.6	4236
9	44.6	41.8	3063	20.4	85.0	4295
12	43.4	42.4	3084	20.1	87.1	4344

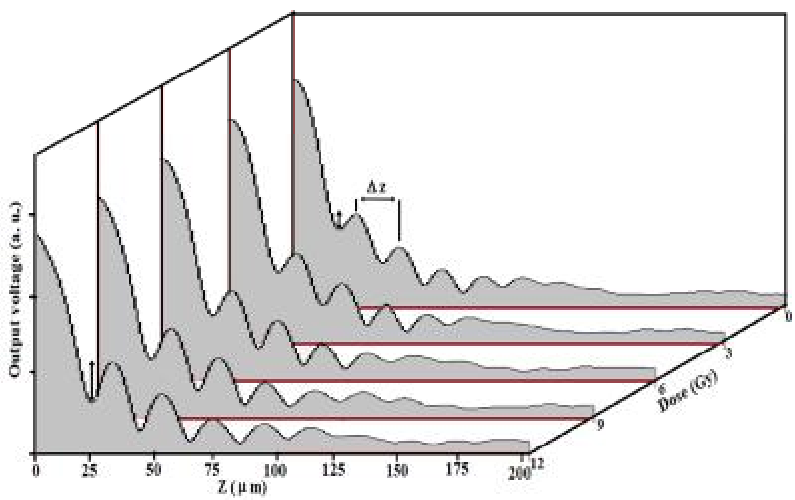

Fig. 4V(z) Acoustic materials signatures of BaO-PbO–20B2O3–30SiO2 at different dose

3.3 GAMMA-IRRADIATION EFFECTS ON WAVE VELOCITIES.

To analyze and quantify acoustic signature of Fig. 4, we first subtract the effect of the acoustic lens signal from these curves to obtain the real material signatures. The variations in Δz is better put into evidence by the spectral treatment of V(z) periodic signals Fig. 4and Fig.5 via fast Fourier transform analysis. Representative obtained results displayed in Fig. 6. Similar behaviors were obtained as those of Rayleigh modes for different Gama irradiated dose and its related attenuation of the propagation mode for different parameters: changes of spatial periods, longitudinal critical angles, changes in positions of FFT principal rays, variations in peak heights and variations in the values of longitudinal velocities Fig. 6. All these was observations reported.

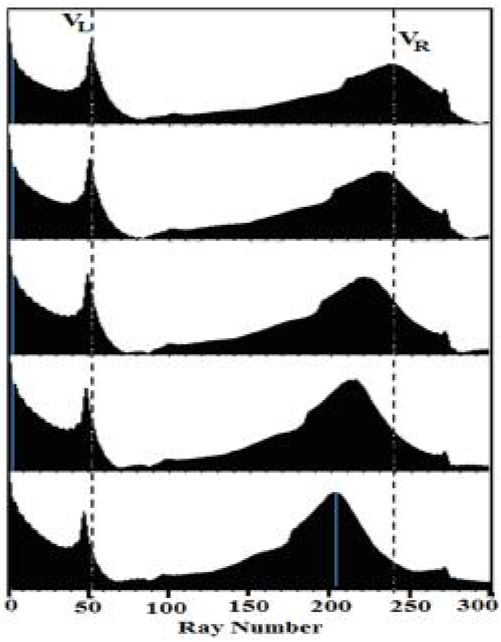

Fig. 5 FFT spectra of BaO-PbO–20B2O3–30SiO2 at different gamma irradiated dose rate

gamma irradiated dose rate

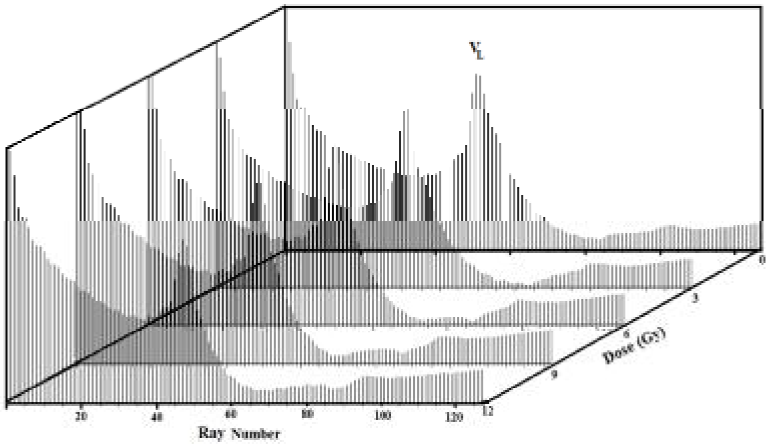

Fig. 6 longitudinal velocities from FFT spectra of BaO-PbO–20B2O3–30SiO2 at different

4 CONCLUSIONS

in conclusion, irradiation affected the structure of the glass samples by damaging and/or breaking of bonds, leading to the increased formation of bao-pbo. the results of elastic properties of the glass samples indicated bond distortion by gamma irradiation. it was found that, as Gama radiation dose rate increases, longitudinal velocities vary from 4172 to 4344 m/s whereas Rayleigh velocities vary from 2991 to 3084 m/s. The increase of the ultrasonic velocity Vl and Vs with increasing BaO content, is attributed to the increase in connectivity of the network structure. The values of the theoretical bond compression model were calculated for asseveration of the obtained results. The agreement between the theoretically calculated and experimental elastic moduli is excellent for the studied sample.

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