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# Glass structure responses to gamma irradiation using infrared absorption spectroscopy and ultrasonic techniques: A comparative study between $Co_2O_3$ and $Fe_2O_3$

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## HIGHLIGHTS

- Changes in $BO_3 \rightarrow BO_4$ and $SiO_4^- \rightarrow SiO_4$ due to the effect of radiation.
- Structural changes in $BO_3 \rightarrow BO_4$ and $SiO_4^- \rightarrow SiO_4$ have a more compactness structure.
- FTIR were adequate supporting our discussion in the structural changes.

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## ABSTRACT

The response of glass to radiation was investigated using ultrasonic and FTIR spectroscopy. New materials were prepared from borosilicate-based glass with different cobalt and iron oxide compositions. The results indicate that the glass structures were most responsive to irradiation at 1500 Gy. Moreover, the results show that the radiation effect decreases when the cobalt and iron oxide compositions increase. These results are relevant to studies on high-dose processing, radio-pharmacy and storage.

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## 1. Introduction

Radiation and its application have become indispensable to modern society, through uses such as nuclear power plants and radiotherapy. However, radiation can be dangerous in accidents, such as the Chernobyl disaster in 1986 and, recently, at Fukushima Dai-Ichi in 2011. Therefore, radiation damage in materials must be studied. Understanding the effect of radiation on the structural properties of matter is important in evaluating radiation shielding materials and dosimetric materials (Singh et al., 2008; Han et al., 2009; Abd El-Malak, 2002; Baccaro et al., 2007; Arora et al., 2009; Prado et al., 2001; Sharma et al., 2009; Soliman et al., 2013). Studies on radiation-induced defective centers in glass have been an interesting subject recently because such studies aid in examining the suitability of glass for radiation dosimetry applications. Many studies are available in the literature on the activating or modifying effect of radiation on structural properties due to certain transition metal ions (TMOs), such as $Fe^{3+}$ and $Co^{3+}$, in amorphous materials (Srinivasarao and Veeraiah, 2001; Dance et al., 1986). In addition, glass doped with multivalent ions, such as $Fe^{3+}$ and $Co^{3+}$, have been used for many applications, such as semiconducting glass, active oxide catalysts to oxidize CO and hydrocarbons, and glassy behavior in manganites (Mao et al., 2011; Vasilyeva et al., 2010; Tran et al., 2013; Azianty et al., 2012).

Borosilicate-based glass has certain remarkable features, such as high chemical stability and low thermal expansion coefficients, which render the glass resistant to thermal shock; this glass is also excellent for transmission to visible light. Thus, this glass has been used in application such as coatings, semiconductor microelectronics, optical lenses, scintillation detectors, glass–ceramic cement, and hard nuclear waste materials (Arora et al., 2009; Sharma et al., 2009; Ramkumar et al., 2008; Sawvel et al., 2005). In addition, glass doped with and/or with added transition metal oxide (TMOs) exhibits interesting optical, magnetic, and electrical properties due to possible TMOs ions in two or more valence or coordination states (Shelby, 1997).

The glass properties are closely related to the lattice structure inter-atomic forces and potentials. Therefore, lattice changes due to composition change and/or irradiation can be directly detected.

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The coordination number of the former network and change in oxygen bonds for the network by cation modifiers and/or irradiation are helpful for better understanding the glass structure; these data are generated through ultrasonic techniques and FTIR spectroscopy (Baccaro et al., 2007; Arora et al., 2009). Ultrasonic techniques and FTIR spectroscopy are versatile tools for investigating microstructure change, structural deformation and materials' mechanical properties due to a change in composition and/or irradiation (Sharma et al., 2009; El-Mallawany et al., 1998; Doweidar and Saddeek, 2009).

The aforementioned concerns led to this study, wherein the authors report the effects of TMOs composition ($Co_2O_3$ and $Fe_2O_3$) and side effects from irradiation on structural properties. The results herein were generated to consider the available materials for high-dose processing, radio-pharmacy and storage.

## 2. Experimental details

### 2.1. Glass preparation

The glass samples were prepared in rectangular shapes from the $(Co_2O_3)_x\,(Na_2O)_{20}\,(Al_2O_3)_1\,(B_2O_3)_{13}\,(CaO)_{6.5}\,(PbO)_{1.5}\,(SiO_2)_{58-x}$ and $(Fe_2O_3)_x\,(Na_2O)_{20}\,(Al_2O_3)_1\,(B_2O_3)_{13}\,(CaO)_{6.5}\,(PbO)_{1.5}\,(SiO_2)_{58-x}$ glass systems ($x$ is the mol%) using the melt-quenching method. The oxides used herein were analytical regent grade (more than 99% pure). The raw materials were weighed using an electronic balance with accuracy on order of 0.1 mg; they were mixed and calculated to yield a 50 g sample. Next, the starting materials were mixed carefully in alumina crucibles. To ensure homogeneity, the mixtures were melted at 1250 °C in an electrical furnace that was constructed at the Department of Physics, Faculty of Science, Ubon Ratchathani University. The melted glass was then poured into pre-heated stainless steel molds and annealed at 450 °C. The glass samples were cut and polished using different silicon carbide grades. The sample thicknesses were measured to the micrometer.

### 2.2. Density and molar volume measurements

The density of each sample was measured using Archimedes' principle and n-hexane as the immersion liquid as well as applying the following equation (Marzouk and Gaafar, 2007):

$$\rho = \rho_L\left(\frac{W_a}{W_a - W_b}\right) \tag{1}$$

where $\rho_L$ is the immersion liquid density; $W_a$ and $W_b$ are the sample weights in air and the immersion liquid, respectively. The experiments were repeated three times for an accurate density value. The molar volume ($V_a$) was calculated using the expression $V_a = M/\rho$, where $M$ is the molecular weight of the glass, which was calculated using the following equation: $M = \sum_i x_i M_i$ (Abd El-Malak, 2002), where $x_i$ is the mole fraction of the component oxides $i$ and $M_i$ is its molecular weight.

### 2.3. Gamma-ray irradiation

The glass samples were irradiated with gamma ray using an exposure machine (THERATRON 780C) and Co-60 as the gamma-ray source at the dose rate 1.16 Gy $min^{-1}$ and the field size $30 \times 30$ $cm^2$ at room temperature 30 cm from the source. The samples were irradiated with gamma rays for a sufficiently long time to receive the doses 500, 1000, 1500 and 2000 Gy.

### 2.4. Fourier transform infrared absorption measurements

Infrared absorption spectra for the powdered glass were recorded in the range 400–4000 $cm^{-1}$ using the KBr technique at room temperature. A spectrometer from Perkin-Elmer was used to measure the absorption spectra; the measurements were at a 4 $cm^{-1}$ resolution (Chahine et al., 2004). The FTIR measurements were measured immediately after irradiation by gamma rays (Sharma et al., 2009).

### 2.5. Ultrasonic velocity measurements

To measure the mean ultrasonic velocity of the glass samples, an ultrasonic flaw detector, the SONATEST Sitescan 230, was used (Table 2). The ultrasonic waves were generated from a ceramic transducer at the resonant frequency 4 MHz, which simultaneously acted as a transmitter and receiver. The mean ultrasonic velocity was calculated using the following equation (Marzouk, 2009):

$$v_m = \left[\frac{3v_L^3 v_S^3}{v_L^3 + v_S^3}\right]^{1/3} \tag{2}$$

where $v_L$ and $v_s$ are the longitudinal and shear velocities, respectively. The estimated error in the velocities measurement was $\pm 14.0$ m $s^{-1}$ for the longitudinal velocity and $\pm 10.0$ m $s^{-1}$ for the shear velocity.

## 3. Results and discussion

### 3.1. Density and molar volume

The glass sample density and molar volumes were calculated, and the exact values are listed in Table 1. The density variations with different TMO compositions ($Co_2O_3$ and $Fe_2O_3$) are shown in Fig. 1. The densities increase with increasing TMO concentrations because $SiO_2$ (molecular weight is 60.084 g $mol^{-1}$) is replaced with $Co_2O_3$ and $Fe_2O_3$ (molecular weight are 165.863 and 159.687 g $mol^{-1}$, respectively) (Eraiah, et al., 2010; Laopaiboon and Bootjomchai, 2013; Gaafar and Marzouk, 2007; Veeranna Gowda et al., 2007). Moreover, the density of the glass samples with the added $Co_2O_3$ was higher than the glass samples with the added $Fe_2O_3$ because $Co_2O_3$ has a higher molecular weight than $Fe_2O_3$. However, the glass samples with $Co_2O_3$ and $Fe_2O_3$ added yielded densities with small differences.

**Table 1**
Density ($\rho$) and molar volume ($V_a$) of glass samples with different concentration of $Co_2O_3$ and $Fe_2O_3$, respectively.

| mol% | $Co_2O_3$ | | $Fe_2O_3$ | |
|---|---|---|---|---|
| | $\rho$ (g $cm^{-3}$) | $V_a$ ($cm^3$ $mol^{-1}$) | $\rho$ (g $cm^{-3}$) | $V_a$ ($cm^3$ $mol^{-1}$) |
| 0.00 | $2.6885 \pm 0.0011$ | $24.2183 \pm 0.4990$ | $2.6882 \pm 0.0010$ | $24.2384 \pm 0.6512$ |
| 0.50 | $2.7286 \pm 0.0015$ | $30.3544 \pm 0.7082$ | $2.7019 \pm 0.0012$ | $32.1901 \pm 0.3100$ |
| 0.75 | $2.7324 \pm 0.0012$ | $36.5082 \pm 0.6760$ | $2.7124 \pm 0.0016$ | $37.1804 \pm 0.9983$ |
| 1.00 | $2.7468 \pm 0.0020$ | $54.8988 \pm 0.9683$ | $2.7125 \pm 0.0013$ | $56.2714 \pm 0.8593$ |

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