Tailoring Structural and Optical Properties of Composite Glass with Rice Husk Fibre (RHF) as Additive Materials

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Abstract. This paper reports the effect of additional of rice husk fibre (RHF) with various sizes ranging from 63µm, 125µm and 250µm on the structural and optical properties of glasses. A series of glass with composition of 69.50TeO₂ – 20ZnO – 10Na₂O – 0.50Er₂O₃ – (x)0.2%RHF (x = 63µm, 125µm, 250µm) are synthesized by using melt-quenching technique. XRD pattern shows the nonexistence of sharp peak reflect the amorphous nature of glass without RHF content. However, an addition of RHF in glass matrix shows the appearance of sharp peaks signifies the contribution of cristobalite due to the incorporation of rice husk. SEM image of glass containing 125µm RHF illustrates the glass surface morphology. The respective EDX spectra of glass containing 125µm RHF verify the elemental traces of carbon, oxygen, sodium, zinc, tellurium, aluminium, silicon, copper, calcium and erbium. The variation in absorption coefficient of glass with additional of different sizes of RHF are observed due to the lowering value of α.

1. Introduction

Glass is one of the most used materials in our daily life. Glass is hard, brittle, transparent and with no definite melting point [1]. However, crystallization must be avoided in order to form glass [2]. Every type of glass features possess different unique characteristic. Tellurite glass has fragile structure which requires attentive care. Tellurite glass exhibits lower phonon energy, a wider range of infrared light, large solubility of rare earth ion, good chemical stability, excellent optical properties, lower melting temperature, large emission cross section, high refractive index and high nonlinear refractive index [3]. Generally, rice husk can be categorized as biomass which can be used as industrial raw materials, animal feed, ash, brickyard and fuel. Despite, rice husk always discarded in large quantities that require a huge space to accommodate all the waste.

Therefore, researchers come out with a new idea to minimize the abundance of rice husk waste by incorporating rice husk in their glass composition [4]. According to previous research, rice husk has been used in colourless and coloured glass production. In addition, rice husk also can be used as filtration materials [5]. Due to their high potential in many fields, the aim of present work is to fabricate a series of glass samples containing various size of RHF in micron size. The incorporation of RHF in glass matrix has an ability to modify the glass structure and act as filler in between the space in a glass matrix.
2. Materials and Methods

2.1. Extraction of Rice Husk

The extraction of rice husk in this study is based on previous research demonstrated by Low and Lee [6]. The first step involved the winnowing process of rice husk to remove the stones, sand, and dust. Then, the RHF was washed thoroughly by using distilled water and dried under the sunlight. The second step required 20g of RHF treated with 1% of Na$_2$CO$_3$ solution. The mixture was shaken for 45 minutes at room temperature. Then, the rice husk was rinsed by using distilled water for several times and dried under the sunlight. The dried RHF then are blended to attain a fine powder. Lastly, the RHF are subjected to a sieving process to separate the fine powder into the distinctive sizes of 63µm, 125µm and 250µm as shown in Figure 1.

![Figure 1. Distinctive sizes of RHF after sieving process shows fiber with (a) 63µm, (b) 125µm and (c) 250µm.](image)

2.2. Fabrication and Characterization of Glass

The glass composition of 69.5 TeO$_2$ – 20 ZnO – 10 Na$_2$O – 0.5 Er$_2$O$_3$ – (x) 0.2% RHF were synthesized by using melt-quenching technique, where x represent the sizes of RHF with varying size as summarized in Table 1. In current work, 0.2% represents the weight percent of RHF to investigate the effect of RHF as additive materials. The determination of weight percentage of RHF according to the previous work demonstrated by Nevsky and co-workers [7] in which they found the optimum weight percentage of rice husk as additive lies in range of 0.2%. The specific weight of TeO$_2$, ZnO, Na$_2$O, Er$_2$O$_3$ and RHF were mixed thoroughly. The batches were subjected to a milling process for 30 minutes to get homogeneous mixture. A platinum crucible containing the glass constituents was placed in a furnace with temperature at 950 °C for 15 minutes. Then, the molten was poured in a brass and was kept at room temperature for 48 hours. Finally, the samples were ready for structural and mechanical measurements. The X-ray diffraction (XRD) was performed using Philips X’Pert Pro PW3040/60 (at wavelength 1.54056 Å Cu Kα) to verify the nature of glass. EDX analysis was carried out by using Hitachi 3400N VP-SEM for elemental traces. The absorbance spectra were recorded by CARY-60 UV-Visible light spectroscopy in range of 190 nm to 1100 nm. All measurements were carried out under room temperature.

| Glass code            | Glass Composition                     |
|-----------------------|---------------------------------------|
| TZNE                  | 69.5 TeO$_2$ – 20 ZnO – 10 Na$_2$O – 0.5 Er$_2$O$_3$ |
| TZNE - 0.2% 63µm RHF  | 69.5 TeO$_2$ – 20 ZnO – 10 Na$_2$O -0.5 Er$_2$O$_3$ – 0.01g 63µm RHF |
| TZNE - 0.2% 125µm RHF | 69.5 TeO$_2$ – 20 ZnO – 10 Na$_2$O -0.5 Er$_2$O$_3$ – 0.01g 125µm RHF |
| TZNE - 0.2% 250µm RHF | 69.5 TeO$_2$ – 20 ZnO – 10 Na$_2$O -0.5 Er$_2$O$_3$ – 0.01g 250µm RHF |
3. Results and Discussion

3.1. Physical Appearance

Figure 2 shows all prepared glasses are transparent in nature. The physical appearance of glass sample without RHF (refer to Figure 2(a)) appear to be pinkish in color while the glass samples with RHF (refer to Figure 2(b), 2(c), 2(d)) appear to be light pinkish. As the RHF are incorporated into glass matrix, slight changes in color of glass are evidenced. The amount of bubble in glass without RHF (refer to Figure 2(a)) are higher than the glass containing RHF (refer to Figure 2(b), 2(c) and 2(d)). The additional of RHF help to release low amount of carbonaceous gas caused bubbles trapped in RHF glass are fewer, smaller in size and could not be seen clearly [8]. Table 2 shows the average thickness from all the glass samples where the value were 3.67 mm to 3.80 mm in range.

![Figure 2](image)

**Figure 2.** Physical appearance of a) TZNE glass, b) TZNE - 0.2% 63µm RHF glass, c) TZNE - 0.2% 125µm RHF glass, and d) TZNE - 0.2% 250µm RHF glass.

| Glass code          | Reading 1 (mm) | Reading 2 (mm) | Reading 3 (mm) | Average (mm) |
|---------------------|----------------|----------------|----------------|--------------|
| TZNE                | 3.80           | 3.81           | 3.79           | 3.80         |
| TZNE - 0.2% 63µm RHF| 3.68           | 3.66           | 3.69           | 3.68         |
| TZNE - 0.2% 125µm RHF| 3.69         | 3.66           | 3.68           | 3.68         |
| TZNE - 0.2% 250µm RHF| 3.68           | 3.67           | 3.67           | 3.67         |

3.2. Structural Properties

Figure 3(a) shows the XRD pattern for all glass samples. XRD pattern for glass without RHF content shows an appearance of broad hump in range of 25° to 35° signify the amorphous nature of glass [9-10]. However, an appearance of sharp peak in range of 38° are observed with addition of RHF content reflect the crystalline nature of glass containing RHF. The appearance of sharp peak is due to cristobalite formation, in prior to Ca element from RHF content [11]. The formation of crystalline silica phases are precipitated successfully in the glass matrix when the RHF were burned at temperature over 900°C [12-13].
Figure 3. (a) XRD patterns of TZNE, TZNE 63µm, TZNE 125µm and TZNE 250µm. (b) Crystallite size against various size of RHF.

By using Scherrer equation, the crystallite size can be calculated where K is shape factor which taken as 0.9 [14-15], λ is wavelength of radiation, β is full width of half maximum of peak (FWHM) and θ is Bragg angle.

\[ L = \frac{K\lambda}{\beta \cos \theta} \] (1)

Figure 3(b) shows the dependent characteristic of crystallite size with size of RHF ranging from 63µm to 250µm. The variations of FWHM and crystallite size with addition of RHF into glass matrix are summarized in Table 3. It can be observed that the size of crystallite is increases with incorporation of larger size of RHF. The increasing sizes of crystallite due to the fast nucleation originate from the addition of RHF [16].

| Glass code   | Size of RHF | FWHM  | Crystallite size |
|--------------|-------------|-------|------------------|
| TZNE - 0.2% 63µm RHF | 63 µm      | 0.164° | 51.35 nm         |
| TZNE - 0.2% 125µm RHF | 125 µm     | 0.1631° | 52.51 nm        |
| TZNE - 0.2% 250µm RHF | 250 µm    | 0.919° | 91.82 nm         |

Figure 4(a) illustrates the SEM image of glass containing 125 µm RHF. This image reflects the surface morphology and EDX spectra shows the elemental traces for glass containing RHF. The surface morphology shows the appearance of fractured surfaces of the glass indicate more porosity [17]. Figure 4(b) shows the EDX spectra of glass containing 125 µm RHF. The elemental traces show the appearance of carbon (C), oxygen (O), sodium (Na), zinc (Zn), tellurium (Te), aluminium (Al), silicon (Si), copper (Cu), calcium (Ca) and erbium (Er) element in glass matrix. The contribution of metallic elements in prior to Ca, Al, and Cu in glass matrix caused the formation of cristoballite which led to the crystalline nature of glass as verify from XRD spectra [13].
Figure 4. (a) SEM image of TZNE - 0.2% 125µm RHF glass sample. (b) EDX spectra of respective glass sample.

Table 4. The EDX analysis of glass sample with addition of 125µm RHF.

| Element     | Series | Weight (%) | Atomic (%) |
|-------------|--------|------------|------------|
| Carbon (C)  | K-series | 1.12     | 5.84       |
| Oxygen (O)  | K-series | 8.77     | 34.40      |
| Sodium (Na) | K-series | 2.65     | 7.22       |
| Zinc (Zn)   | K-series | 14.41    | 13.83      |
| Tellurium (Te) | L-series | 67.35  | 33.12      |
| Aluminium (Al) | K-series | 0.03   | 0.08       |
| Silicon (Si) | K-series | 0.21    | 0.46       |
| Copper (Cu) | K-series | 0.87    | 0.86       |
| Calcium (Ca) | K-series | 2.07   | 3.24       |
| Erbium (Er) | L-series | 2.53    | 0.95       |
| Total       |         | 100.00   | 100.00     |

3.3. Optical Properties

The optical properties of materials are affected by the technique of sample preparation, surrounding media and element doping. The optical absorption coefficient provides essential information about materials. The absorption coefficient can be determined using the following formula [18-19]:

$$\alpha = 2.303 x \frac{A}{d}$$

(2)

where $\alpha$ is absorption coefficient, $A$ is absorption and $d$ is thickness of sample.

Figure 5 shows the absorption coefficient decreases as the thickness increases. This is due to the inverse relation between transmission and absorption [20]. The value of $\alpha$ decrease with increasing wavelength for all glass samples up to certain wavelength. However, the value of $\alpha$ become invariant beyond the wavelength of 700 nm. It can be deduced that the presence of RHF lowering the value of $\alpha$. Absorption edge of the system is basically determined by the strength of the bridging oxygen’s (BO’s) and non-bridging oxygen’s (NBO’s) in the glass network. The absorption coefficient is decreases due to presence of silica from RHF. The ZnO-SiO$_2$ formed a vast amount of BO’s and NBO’s in the sample [21].
4. Conclusion
Erbium-doped zinc sodium tellurite glass with and without RHF content are prepared using melt quenching method. The effect of varying sizes of RHF is investigated. The XRD spectrum reveals the size of crystallite size increases along the size of RHF. The incorporation of RHF in the glass matrix contributes to the appearance of sharp peak due to the melting temperature over 900°C during glass fabrication. The elemental traces that showed from EDX spectra reveal the contribution of metallic element from RHF caused the formation of cristoballite which led to the crystalline nature of glass. The absorption coefficient decreases as the RHF was added into glass matrix.

5. References
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