AN EXPERIMENTAL DETERMINATION OF PERFORMANCE ECO-WASTE CERAMIC COMPOSITE UNDER SINTERING CONTROLLED: STABILITY, HARDNESS AND ACOUSTIC MEASUREMENT

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ABSTRACT: The filler loading input plays a key role on physico-mechanical of eco-waste ceramic composite (ECC) under controlled heat treatment. In this study, the addition cockleshell (CS) in soda lime silicate glass (SLSG) was fabricated to investigate the effect of filler loading at different sintering temperature. The selected temperatures were in range between 700 and 850 °C with 2 °C/min heating rate at constant holding time. The ECC was formed with the inclusion of CS as filler between 0 to 50 wt%. The performance of ECCs were subjected to stability test, hardness test while the modulus was measured via acoustic measurement. The results showed that addition 30 wt % of CS filler had outstanding physical and mechanical qualities at 850 °C where apparent porosity and water absorption reduced, which leads to bulk density increased. This indicated that the difference in properties acquired by different heat treatments and filler loading. This suggested that sintering controlled, and filler loading were the result of the performance of ECC as a potential material for structural enhancement in the building application.

KEYWORDS: *Eco-Waste Ceramic Composite; Sintering Controlled; Stability Measurement; Hardness Measurement; Acoustic Measurement*

1.0 INTRODUCTION

One of many ways to retain the steady-state in environment is to reuse the waste from sustainable or eco-materials [1-2] such as soda-lime silicate glass (SLSG) and cockle shell (CS). The aquaculture industry produces significant volumes of seashell waste, including cockle, oyster, mussel, and scallop shells, among others, that are usually disposed or end up in landfills since they have no other value [3]. The untreated and abandoned shells may cause the environment to unpleasant odour [4]. This situation can be avoided by recycling the CS as a filler where the CS is made up of 96% calcium carbonate ($CaCO_3$) [3, 5-6]. A work by George et al. [7] has stated that the rich calcium carbonate ($CaCO_3$)-based shells of the cockle provide an appealing substitute material that is inexpensive, readily available, and abundant that can serve in useful product. Othman et al. [5] reported that filler with 5% to 10% of CS as replacement in cement proved to be an efficient new concrete material. However, Othman et al. [8] studied on the CS powders to minimize manufacturing cost of concrete and reduce waste of shells discovered that higher CS ash in concrete decreasing in compressive and tensile strength. Moreover, the potential of CS also has been pre-explored with different binder for protective material base [9]. Recent studies as mentioned above widely reported due to their advantageous aspect in recyclability. Very little work has been done concerning the fabrication method of the glass-ceramic composite incorporated with CS and analytical assessment of sintering temperature. Therefore, there is a gap to maximise the usage of CS as an eco-friendly alternative material where in this research will be conducted as a filler in waste glass matrices.

In the manufacturing of glass-ceramic composites, soda-lime silicate glass (SLSG), an environmentally friendly material waste produced by rapid industrial development, is used extensively due to unique characteristic that contributed by smooth and non-reactive surfaces [10-12]. It also accompanied with greater physico-mechanical properties such as strength and fracture toughness than the parent glass under conventional heat treatment that is two-steps heat treatment controlled [13]. However, the procedure is time-consuming and expensive because of the high temperature required. Long processes are not appropriate for natural wastes since they are impacted by exposure to high temperatures, which might result in high porosity. Moreover, high-degree crystallisation of glass-ceramic created at 950 °C from natural raw materials and industrial waste led to improved density through good viscous flow of glasses [14].

However, the sintered sample showed bloat at a sintering temperature of 1000 °C, indicating an excessively viscous flow of glasses that would result in extremely porous glass-ceramic characteristics [13, 17]. In this investigation, the sintering temperatures of 700 °C, 750 °C, 800 °C and 850 °C were chosen based on previous research [18].

The risk of high porosity can be reduced if the glass-ceramic composites with combination of filler from waste materials are well densified via a one-step heat treatment controlled or low-cost route sintering and sometimes called as direct sintering process [15]. Therefore, very limited studies have been reported on the performance of CS as main filler embedded in glass matrices using low-cost route sintering process compared to polymer matrices due to the high temperature required and risk of porosity despite of the performance. Moreover, the high calcium carbonate ($CaCO_3$) content of CS, which can potentially be revealed, was chosen as one of the trial materials because there have not been many efforts to find novel materials for giving building materials covering. Hence, the present study aims to investigate the performance of eco-waste ceramic composite using direct sintering controlled. Several sintering processes were tried and the mechanical properties which were evaluated to correlate the effect of sintering temperature ranging between 700 °C and 850 °C and filler loading. The findings from this work may largely explore the knowledge regarding the forming process especially in developing heat treatment profile for eco-waste materials and implementation of eco-waste-based ceramic in composite applications.

2.0 METHODOLOGY

2.1 Materials

Glass made of soda-lime silicate (SLSG) and cockleshell (CS) were gathered from domestic waste. An apparatus called a disc crusher (Model Retsch) was used to crush recycled SLSG before milled for 60 min using planetary ball milling to convert into powder form. The powder is then sieved using a vibratory sieve shaker to obtain an average particle size of approximately 45 μm. Waste from CS was cleaned and dried in an oven for a full day. The CSs were then ground for 30 min in a planetary ball mill. Additionally, the CS powder was sieved to approximately 45 μm. Prior to the calcination process, the CS was then stored in the glass storage with silica gel. The temperature utilised for calcination was fixed to 1000 °C at 2 °C /min heating rate with holding time of 1 h.

2.2 Preparation Sample

A uniaxial die pressing machine was used to compress the 3.2 g of powder at 2.5 tonnes (for a square mould). The final composites' composition is listed out in Tables 1. The temperature range of sintering was established using the Hitachi STA 7300 (Hitachi, Japan) differential thermal analysis (DTA) model. By using a Carbolite (1300 model) laboratory electric furnace, the green bodies were sintered for 60 min at fixed heating rates of 2 °C/min at 700 °C, 750 °C, 800 °C, and 850 °C. Visual observation physical appearance before and after the sintering process was recorded.

Table 1: Composition of CS/SLSG composite in wt%

Batch	Code	Composition (wt%)	Material	Sintering temperature (°C)	Heating rate (°C/min)	Dwelling time (min)
Batch 1	A1	100	SLSG	700	2	60
	A2	70:30	SLSG:CS			
	A3	60:40				
	A4	50:50				
Batch 2	B1	100	SLSG	750		
	B2	70:30	SLSG:CS			
	B3	60:40				
	B4	50:50				
Batch 3	C1	100	SLSG	800		
	C2	70:30	SLSG:CS			
	C3	60:40				
	C4	50:50				
Batch 4	D1	100	SLSG	850		
	D2	70:30	SLSG:CS			
	D3	60:40				
	D4	50:50				

2.3 Physical Testing

Physical testing included analyses of apparent porosity, water absorption, and bulk density which accordance with ASTM C373. Samples of a square form were boiled in distilled water for 8 h at a fixed temperature, then soaked for 24 h. The sample was weighed using an electronic densimeter (model MR3005) to get the final weight. The filler loading (CS) range is 0 wt% to 50 wt%, while the sintering temperature is from 700 °C to 850 °C. The following formulae were used to determine the apparent porosity and water absorption for each sample. Given, 1 cm^3 of water equivalent to 1 g. The exterior volume, (V) was obtained using Equation (1) such as

$$V = M - S \tag{1}$$

where, V is stand for exterior volume (cm³), M is a saturated mass (g) and S is a suspended mass (g). The percentage of apparent porosity, (AP) was calculated using

$$AP(\%) = \frac{M - D}{V} \times 100\% \tag{2}$$

where, AP is apparent porosity (%), M is a saturated mass (g), D is a dry mass (g) and V is an exterior volume (cm³). Percentage of the water absorption, (WA) was calculated using the following equation:

$$WA(\%) = \frac{M - D}{D} \times 100\% \tag{3}$$

where, WA is a water absorption (%), M is a saturated mass (g) and D is a dry mass (g).

2.4 Mechanical Testing

Vickers hardness test results were used to determine the SLSG/CS samples' hardness (HM-200 Series). The square samples' mirror surfaces underwent preparation and testing in accordance with ASTM C1327. A diamond indenter was used for the micro-hardness test, and a load of 0.5 kg was applied for 5 s. Each sample was subjected to five indentation loads, and the average reading was recorded. The longitudinal sound wave velocities were used to determine the Young's modulus for each sample using Olympus Ultrasonic Flaw detector (EPOCH 650 series). Depending on the resonance curve's breadth, the frequency range used for modulus measurement spans from 0.1 MHz to 20 MHz. The test standard ATSM E 494-95's guidelines were followed for conducting the acoustic measurements. The calculation of the longitudinal velocity, v_l, is based on the transit time across the thickness of the samples such as

$$v_l = \frac{2 \times d(m)}{t(s)} \tag{4}$$

where v_l, d, and t represent the longitudinal wave velocity, sample thickness, and time-of-flight, respectively. Equation (5) was used to calculate the Young's modulus from the measurement [22].

$$E = \rho v_l^2 \tag{5}$$

where ρ denotes as the density of the tested sample and v_l denotes as the longitudinal velocity of each sample tested.

3.0 RESULTS AND DISCUSSION

3.1 Physical Properties

A visual inspection showed the recycled SLSG in samples change in colour to opaque light grey indicating that the sample transformed from glass material to glass-ceramic materials. The typical colour of the 100% of SLSG sample before sintering was whitish as shown in Figure 1 and the sample of the 100wt.% CS formulation was light milky cream (dotted line).

Figure 1: The green SLSG/CS composite before the sintering process

The visible changes seen in the treated samples are summarized in Table 2. The sample of SLSG 100wt.% was seen as brownish colour associated with the thermal effect at the onset of the softening temperature (Figure 2) where molecules start to rearrange the glass structure suggesting the crystallization was initially promoted which is in agreement with 14]. Moreover, the present of the TiO_2 in the recycled SLSG as in [11] has slightly shifted to light grey colour starting at 750°C without wt% CS fillers. On the other hand, it was founded that the sample with 30wt.% CS, appeared in shaded grey when sintered at 700°C and 750°C as compared to filler loading of 40 and 50wt.% changed to light grey which according to Bharatham et al. [17], samples had high levels of calcium present vividly coloured.

Table 2: Effect of heat treatment on colour of SLSG/CS glass-ceramic composite

Formulation of Batch (wt.%)	Sintering Temperature (°C)			
	700	750	800	850
100	 Dark Brown	 Dark Brown	 Grey	 Grey
70:30	 Dark Grey	 Dark Grey	 Light Grey	 Light Grey
60:40	 Dark Grey	 Light Grey	 Light Grey	 Light Grey
50:50	 Grey	 Light Grey	 Light Grey	 Light Grey

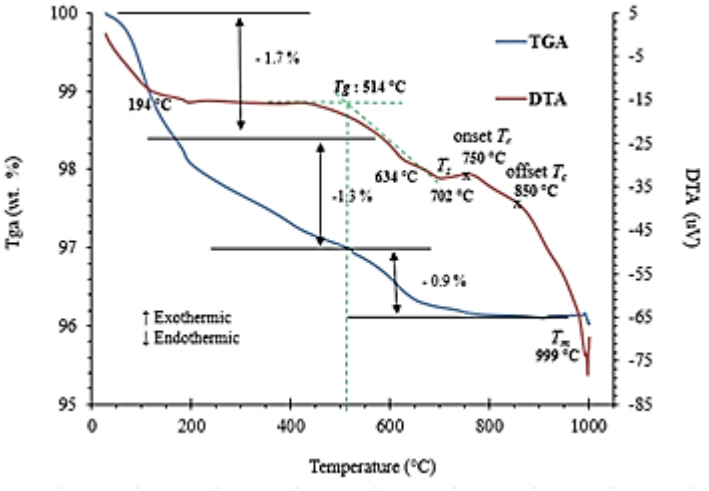
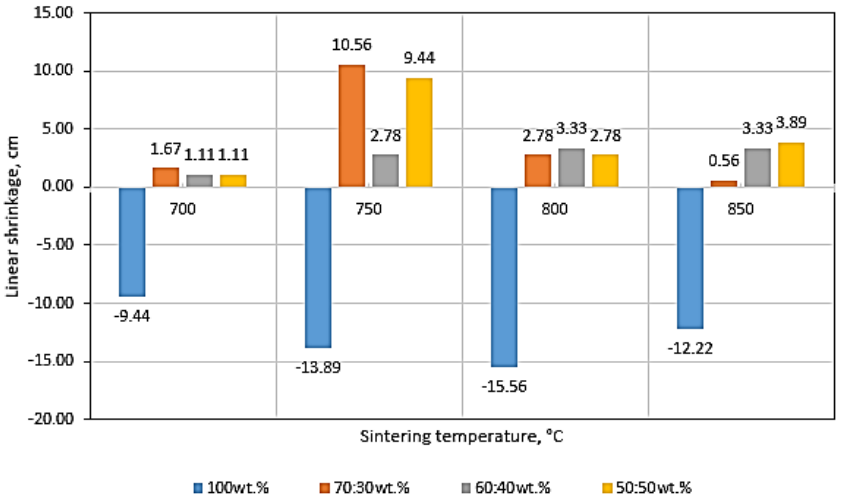
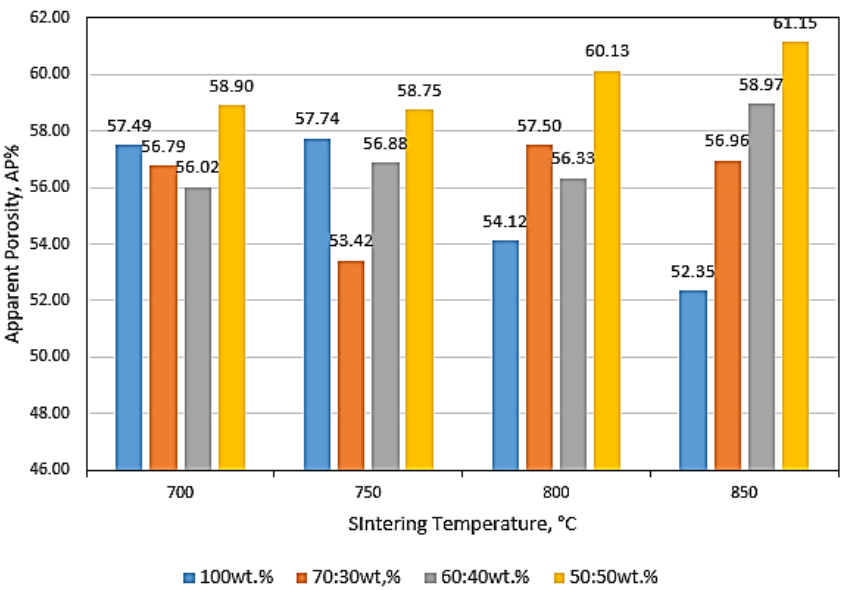


Figure 2: Thermal properties of recycled SLSG

Figure 3 (a) shows the linear shrinkage of all samples SLSG/CS at various sintering temperatures. For all formulations, the initial sample size was 18 mm x 18 mm x 4 mm. Sample with 100% SLSG depicted that the dimension has shrunk at 700 °C, 750 °C, 800 °C, and 850°C which represented by negative values. Positive values imply that the sample has expanded, whereas negative values suggest that it has shrunk. On the other hand, all samples that were sintered at 850°C with 30 wt%, 40 wt%, and 50 wt% CS filler loads show a dimension expansion. At sintering temperature of 750°C and 30wt% CS filler load, the sample expands to its maximum of 10.56 mm.



(a)



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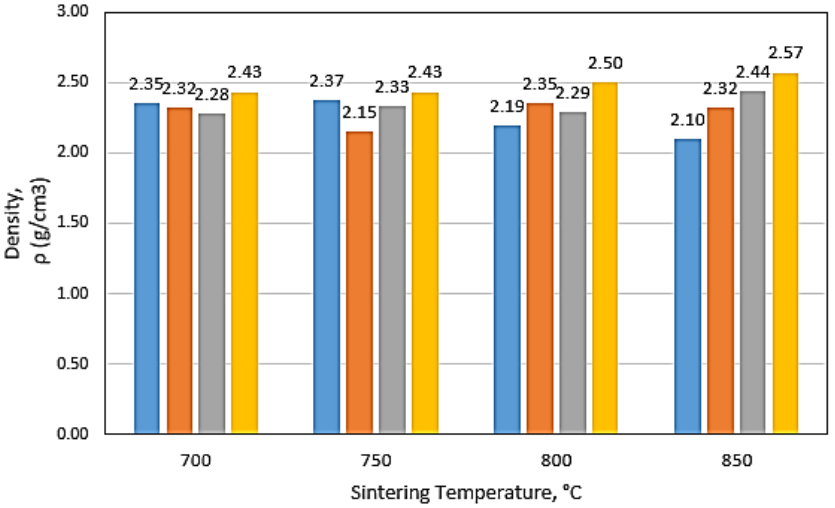

(c)

Figure 3: Physical properties of samples of SLSG with various CS filler loading sintered: (a) linear shrinkage, (b) apparent porosity and (c) bulk density

Referring to Table 2 and Figure 2, it can be related, due to the beginning of the crystallization and the sample started to densify which is similar to the results presented in [18]. Significant differences were observed on apparent porosity where all filler loadings of CS that sintered at 850 °C exhibit a larger percentage of porosity than those sintered at 700 °C. The samples with a 30 wt.% CS filler load had a lower percentage of porosity at 850 °C than the samples at 800 °C where the sample develops porosity because of the cockle shell's incomplete fusion with the SLSG. The bulk densities of SLSG/CS glass as a function of heat treatment temperatures and filler loadings as shown in Figures 3 (b) and (c), respectively. Due to the particle dispersion and the size of particles used in this study were similar (approximately 45µm) within the composite, the graph of the bulk density result displays an inconsistent trend. For samples sintered at 850 °C show an increasing trend for 30, 40, and 50wt.% of CS filler loading. The samples with 50wt.% of CS filler loading shows a linear relationship between the bulk density and the apparent porosity.

The density profile of all samples is similar as work done by Ahmad and Zaidan [19] where the density increased with temperature. Even though the temperature used in their study above 1000 °C for Sayong ball clay materials, no major differences were observed in linear shrinkage as well. This may be explained by the range sintering temperature and filler loading selected for SLSG which control the flow

glass to miscible together with CS at the onset of 850 °C (Figure 2) where it is believed that the viscosity of SLSG started to considerably be decreased. This is supported by research by Yesilay [20], which showed that the inclusion of glass waste into the microstructure improved the dissolution of quartz by lowering viscosity and facilitated the formation of liquid phase.

3.2 Mechanical Properties of SLSG/CS Composite

3.2.1 Hardness Properties

In this study, the ASTM C 1327-99 standard test protocol was used on samples that had no visible faults (shattered or deformation). Selected samples were tested for microhardness using Vickers indentation hardness. Results of hardness test in Table 3 show variation in values for SLSG at different CS wt.%. The sample with 30% of CS filler load has the maximum microhardness value at 850 °C, which is 1004.86 Hv meanwhile samples sintered at 700 °C had a hardness that was over 38% higher than those sintered at 850 °C. Optimal mechanical properties were a result of the filler load of 30wt.% CS in the formulation, highlighting the formulation's strong physical capabilities. The ratio with SLSG incorporated with CS at the second onset of the crystallisation temperature (850° C) as shown in Figure 2 that resulted in the microhardness with the highest value. A study by Leenakul et al. [21] reported that crystal precipitation in glass which occurred at crystallisation temperature affects the composite's overall density which in this study the crystal precipitated started at 850 °C. This value shows to be slightly different to that of sample treated at 700 °C. It is expected that the average reading result will increase as the sintering temperature increases. The lower average reading is the result of the indentation procedure. A large pore may be directly beneath the newly formed indentation. Additionally, one of the factors that affected the microhardness test was how clean the indenter was. In addition, to maximise reading accuracy, the sample's surface should be smooth and ground.

Table 3: Average hardness on selected of sintered CS/SLSG composite

Sintering temperature (°C)	Composition of ECC (wt %)	Average reading (Hv)
700	100	477.90
	70:30	249.76
750	100	508.10
850	100	613.58
	70:30	1004.86

3.2.2 Young's Modulus of SLSG/CS Composite

The Young's modulus was further measured by wave generated ultrasound. The results are shown in Table 4. The glass-ceramic composite samples were discovered to have a Young's modulus of less than 100 GPa. It was found that the samples with a 30 % CS filler load have a Young's modulus of 80.74 MPa at 850 °C of sintering temperature. The modulus at 850 °C with 100% SLSG is lower than the modulus at 700 °C with the same material. The lower Young's modulus value implied that the glass samples did not tolerate the strain. This may be directly related to the chemical composition where Zaid et al. [22] reported that the addition of element with oxygen will fill up interstitial site within SLSG site which led to increase Young's modulus and density. Initially, the element of CS powder was $CaCO_3$ and converted into CaO through the calcination process. It was expected that this CaO has decreased the effect of acidic on the solubility of the glass and it also increases the hardness and durability of the glass.

Table 4: Value of Young's Modulus at different sintering temperature of eco-waste ceramic composite

Sintering temperature (°C)	Composition of ECC (wt %)	Young modulus (GPa)
700	100	0.083
	70:30	0.080
750	100	0.082
850	100	0.073
	70:30	0.080

4.0 CONCLUSION

Correlations between properties and various filler loading as well as the sintering temperatures (700-850 °C) were clearly demonstrated by enhanced in hardness, Young's modulus and stability where the results showed by addition of CS at higher sintering temperature. According to this study, apparent porosity and the proportion of linear shrinkage were related and led to the enhancement in density of eco-waste glass ceramic composite. These findings inspired the future research using eco-waste materials as potential matrices and filler for material structural application.

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