EXPERIMENTAL AND VALIDATION OF GLASS-CERAMIC COMPOSITE PROPERTIES DERIVED FROM WASTE MATERIALS AT ELEVATED SINTERING TEMPERATURE

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ABSTRACT: The glass-ceramic composite (GCC) from waste material has limited performance especially strength properties that can be improved by reinforced with filler under sintering temperature controlled which commonly used in two steps sintering process. In this study, the influence of elevated sintering temperature using single step or direct sintering process on mechanical properties and microstructural analysis of GCC at various filler loadings were investigated and assessed. The GCC was prepared using eggshell (ES), spent bleach earth (SBE) and soda lime silicate glass (SLSG). Eggshell acted as filler was calcined at temperature of 1000 °C before sintered to direct sintering process. This combination was aimed to improve the performance of GCC by controlling the sintering temperature. The sample was sintered at four types of sintering temperatures at 750, 800, 850 and 900 °C using 2 °C/min heating rate with constant holding time. The GCC was formed with the inclusion of eggshell at 0, 5, 10, and 15 wt% as filler. The performance of GCC was analysed through mechanical testing while microstructure of the samples was characterized by using Field emission scanning electron microscopy (FESEM). The results revealed the average of Vickers Micro-hardness and flexural strength was a function of sintering temperature and filler loading. The analyses from these results were indicating that it is possible to produce a good GCC using local waste contributed by 15% of eggshell with the highest value of micro-hardness (1011.64 HV) and with highest flexural strength (54.82 MPa) obtained. Validation via fractography analysis indicated, at high content of filler, the microstructure was more densified as number of pores was minimized. These findings concluded that the suitable sintering temperature can be controlled for alternative materials for structural applications.

KEYWORDS: *soda lime silicate glass; spent bleach earth; eggshell, sintering; flexural strength*

1.0 INTRODUCTION

In terms of waste materials, scarcity of landfill sites leads to worldwide work improving recycling technologies and reducing as much as possible the solid waste. Malaysia's landfill is indeed in need of a solution because of the dramatic increase in waste [1]. Recycling other waste materials can save the world from rubbish heap to overcome this problem [2] . Different researchers examined the use of waste glass in particular in the clay industry [3]. Soda lime silica waste formed by these substances, which contains 70% silica (SiO_2), 12-15% sodium oxide (Na_2O), 10%-15% calcium oxide (CaO), potassium oxide (K_2O), and aluminum oxide (Al_2O_3) [4]. SLS glass waste can recycle and synthesize new material based on benefits softened due to its softening. Introducing combination of domestic waste for instance eggshell and spent bleach earth, SLSGW will be renewable resources into useful products. The method applied to recovers waste from glass is through 2 steps heat-treatment process. Nevertheless, the time taken for the procedure is extensive and involves approximately and beyond 1000 °C which developed to extravagant. Due to the experience to be exposed at high temperature this will leads to development of uncontrolled porosity and with long procedure are not efficient for natural wastes. Higher strength of sintered glass ceramic composite (GCC) possible to be attained with condition that this green glass composites filled with percentage composition of SBE and ES were well compacted using a direct heat-treatment controlled.

The primary technological justification is the preliminary dissolution of waste into glasses, which allows for the almost permanent retention of pollutants, while the primary uses in the building sectors are as floor and roof tiles or road surface panels [5]. The interest in SLSG waste in Malaysia is due to its large volume and its composition. Some earlier articles claim that reusing SLS glass to create glass-ceramic is a cost-effective and environmentally friendly solution to substitute silicon dioxide (SiO_2) resources [6]. SLSG waste has a specific nature that can be reused several times in various techniques and manufacturing forms without loosening its strength and adaptability. SLSG motivated many researchers to study composites of green glass ceramics composite (GCC) as they include quartz composites.

Asia country mainly Malaysia was known as major palm oil manufacturers. Application of oil palm biomass and palm oil for the production of environmentally friendly biofuels has been of continued interest [7]. SBE waste may be described as domestic waste from industry that produced largely from the extraction of palm oil for useable oil. This waste is commonly predisposed of without pre-treatment in depot area, and generally encompasses approximately up to 40 wt% percent of oil. During the refining process, 0.5-1.0 % of the activated clay or fresh bleaching earth (BE) is used where all BE is converted into waste as SBE and, an estimated 600,000 tons of SBE waste are generated around the world. For this purpose, it is potential to be used for biofuel production as a sustainable raw material.

Eggshell (ES), which in its composition contains around 94% of calcium carbonate ($CaCO_3$) [8]. Although the ES is not considered a harmful waste, improper landfill waste due to the large numbers of eggs produced will result in significant environmental liabilities. This element of $CaCO_3$ is converted into CaO through process calcination, greatly significant for constructural field because it has ascribed an increase in thermal stability and structural strength [9]. As this has shown improvement in properties such as non-hazardous waste and has shown low firing shrinkage with 1.14% as a purely structural aspect as it can alter the physical properties as well as enhance the mechanical properties of glass composite. According to [10], using eggshell as filler has a comparatively lower density than mineral calcium carbonate and has a significantly higher crystallinity than mineral calcium carbonate ($CaCO_3$) composites. Eggshell waste as a filler with a high content of calcium carbonate (94%) can be an significant element for structural applications because it contributes to thermal stability improvement [11].

In terms of the analytical assessment of optimization of composition, characteristics, and fabrication method employing direct sintering, no previous work has explored the production of glass-ceramic composite integrated with spent bleach earth with eggshells. Hence, the present study aims to investigate the relationship between materials (fillers and recycled glass) and their performance (Vickers micro-hardness, flexural strength and microstructure) by measuring mechanical and microstructural parameters. This new combination composition from waste materials is expected to be seen as a potential attempt to minimize sintering rates from 2 steps to direct sintering while greatly improving the properties of the glass-ceramic composite.

2.0 METHODOLOGY

This section details the methodology of the studies. Phase 1 involved design of experiment which were preparation of raw materials and sample preparation.

Phase 2 involved fabrication of glass-ceramic composite by introducing direct sintering process at elevated sintering temperature prior to mechanical testing. The final phase involved verification of results by using microstructural analysis.

2.1 Powder preparation

As part of the preparation process for obtaining CaO powder and SLSW powder, the raw materials of soda-lime-silica glass (SiO_2) waste (SLSW) and eggshell ($CaCO_3$) waste (ESW) was cleaned and dried. SLSW were gathered from household waste. SLSW in crushed form with a size of less than 45 μm was prepared by crushing and sieving. SBE waste was supplied by Mewah Oil Sdn Bhd, Port Klang. The sonication or extration process was used to extract oil from the supplied SBE. Chemical solvent, Hexane was applied as solvent in the extraction process. The SBE powder mixed with hexane in a beaker and sonicated in an ultrasonic machine for half an hour for 5 times. Oil was removed between the time pause. Then the ESW were divided for the calcination process. The eggshells were processed CaO by heating temperature is 1000°C and heating rate 2°C /min for 1 hour. The particle size distribution for SLSG, SBE and, ESW were determined using a particle size analyzer, the Mastersizer 2000.

2.2 Sample preparation

It has been found that the addition SBE of 45 wt% leads to good viscous flow and sintering at 850 °C able to seal the open pores and reduce the porosity [12]. Work form [13], reported that the high degree crystallisation of glass ceramic made from natural raw material and industrial waste was produced at 950 °C and achieved increased density via good viscous flow of glasses. However, at 1000 °C sintering temperature, the sintered sample lead to bloat showing an excessive of viscous flow of glasses which will produce very porous properties of glass ceramic [14]. Based on previous study, 750, 800, 850, and 900 °C were selected as sintering temperature in this study. Firstly, the batch of powder with 3.2 gram (for square mold) was flattened at 2.5 tonnes using a uniaxial die pressing machine and 10 gram (for rectangular mold) at 8.0 tonnes. Tables 1 shows the batch formula of the green bodies of composites. This process followed with heat-treatment via for 1 hour at fixed rate of heating (2 °C/min).

Table 1: Batch formula of sintered green glass ceramic composite (GCC)

Sample code	SLSG (wt%)	Eggshell (wt%)	SBE (wt%)
GCC-1	55	0	45
GCC-2	50	5	45
GCC-3	45	10	45
GCC-4	40	15	45

2.3 Mechanical testing and microstructure analysis

Vickers Micro-hardness test was conducted by using ASTM C1327. Five square samples (18mm × 18mm × 4mm) for each batch were polished by using 400, 600, 800 and 1200 grit sandpaper to obtain mirror surface. Diamond paste applied was at 1 and 3µm. The test was performed by applying 1kg load for 15 s. To get accurate result, the test was repeated five times on each samples. Flexural strength was conducted by using ASTM C1161. Five rectangular samples (65mm× 15mm × 4mm) were prepared for each batch. This testing was conducted using Universal Tensile Machine (3-point bending test). Speed used for was at 0.50mm/min. Therefore, modulus of rupture and break load were identified. Field Emission Scanning Electron Microscopy – Energy Dispersive X-ray (FESEM – EDX) was used to analyse microstructure of the fractured samples and validate the correlation with mechanical properties.

3.0 RESULT AND DISCUSSION

3.1 Effect of Heat-treatment Temperature

Vickers micro-hardness and flexural strength of sintered GCC are important characteristics that impact the material's durability. Figure 1 shows average of Vickers micro-hardness value of sintered GCC at 750°C to 900 °C on various filler loading (0 wt.% – 15 wt.%). To obtain the average, each samples was indented at five times. Figure 1 shows an interesting pattern where an increment temperature of heat-treatment has promoting the hardness value to a promising result. The result revealed that GCC samples with sintering temperature 900°C at 15 wt.% of filler loading recorded the highest micro-hardness value (1011.64 HV) meanwhile samples with sintering temperature 750°C at 0 wt.% of filler loading recorded the lowest micro-hardness value (330.74 HV).

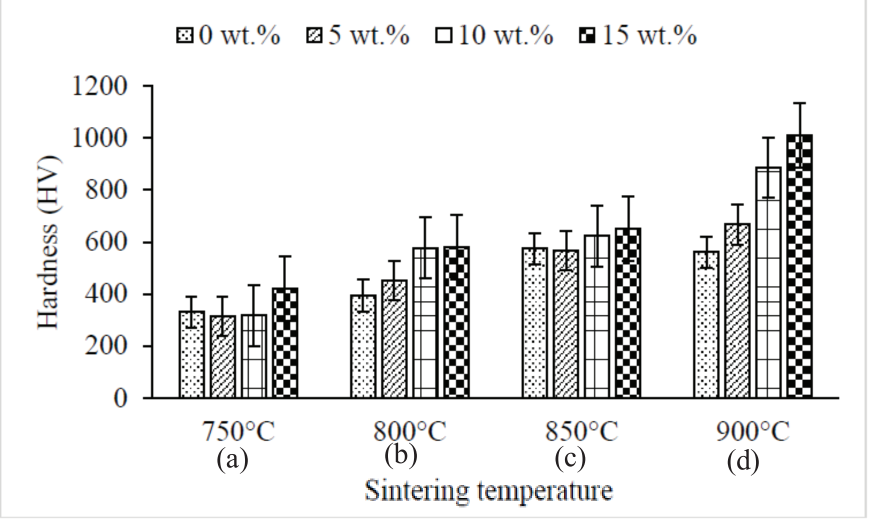

Figure 1: Hardness value of SLSW reinforced different percentage filler at different heat-treatment (sintering) temperatures (a) 750°C, (b) 800°C, (c) 850°C and (d) 900°C.

The physical property namely density of glass-ceramic improved as the sintering temperature increased because this type of glass-ceramic structure has sedimentation and densification of crystal. Densification of the sample occurs as the pressure increases due to the temperature increase. Therefore, the pores at high temperatures cause the sample to be denser and this event is linked with finding by [4] as this alteration affects the performance of the glasses.

Figure 2 shows average of flexural strength value of sintered GCC at 750°C to 900 °C on various filler loading (0 wt.% – 15 wt.%). Based on Figure 2, as sintering temperature increased, the flexural strength increased. A significant change in sample GCC-4 (15 wt.% of filler), the highest flexural strength was observed at sintering temperature 900°C

(54.82 MPa), whereas the lowest flexural strength was observed at sintering temperature of 750°C (32.58 MPa).

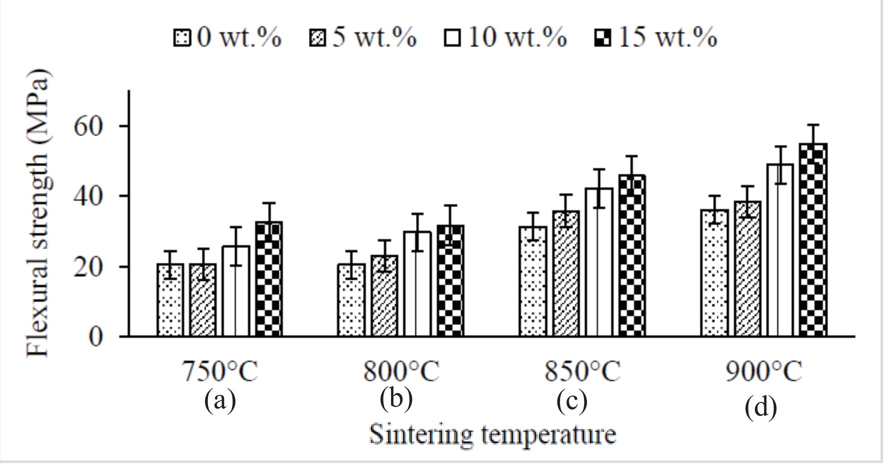

Figure 2: Flexural strength of sintered glass ceramic at different filler loading various sintering temperatures (a) 750°C, (b) 800°C, (c) 850°C and (d) 900°C.

Upon heating up to 750°C, the viscous flow of glassy phase was initiated, followed by the onset of densification. As the temperature was gradually adjusted to 800°C and 900°C, there was less formation of crystals. This led to the formation of a denser crystal skeleton in the glassy matrix, which hindered viscous flow of liquid phase. Thereafter, the densification process was improved and less pores were created [15]. Besides that, particle size of filler applied in this study is < 40μm. This is consistent with the findings of [16], who reported that mechanical properties could be enhanced by increasing the filler fineness.

3.2 Microstructural Evaluation

Figure 3 showed sample with highest and lowest flexural strength value (GCC-4 and GCC-1) that were selected for microstructural analysis. The microstructure revealed that the higher sintering temperature (900 °C) and filler loading (15 wt.%) contributed to smoother fracture surface of the sintered sample. The sintered samples (GCC-1) at lowest sintering temperature (750 °C) shows a larger pores and rough surface.

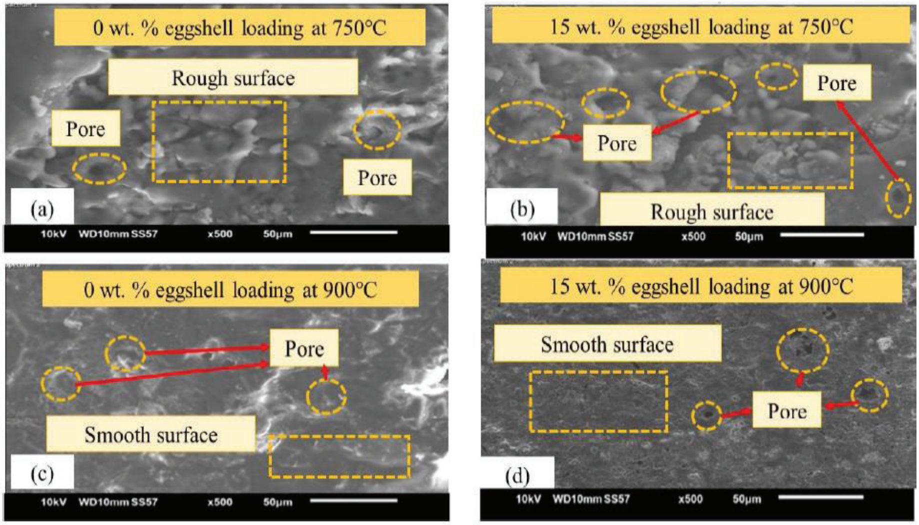

Figure 3: Fractography of fractured surface of sintered glass ceramic composite (GCC) at (a) 750 °C with 0 wt.% eggshell loading, (b) 750 °C at 15 wt.% eggshell loading, (c) 900 °C at 0 wt.% eggshell loading and (d) 900 °C at 15 wt.% eggshell loading at 500x magnification.

Based on this study, the larger pores and rough surface is due to production of gas bubbles of carbon dioxide (CaO_2) during excessive viscous flow. Whereas, sintered sample (GCC-4) produced smoother fracture surface and densified microstructure. It can be deduced that, microstructural attributes are resulted by viscous sintering through its densification process which this finding align with previous research [17] . Therefore, densification and uniformity of sintered GCC was influenced by higher sintering temperature and filler loading. The sintered sample (GCC-4) was selected as an optimum sintered GCC and was believed to have good mechanical properties.

3.3 Sintering Model for Sintered Glass Ceramic Composite (GCC)

Figure 4 shows the mechanism of sintering. Type of sintering applied is direct sintering. Before sintering process, large pores present in the structure as shown is figure above. When the amount of small pores increases dramatically, the small pores are connected along with the large pores, and then the larger pores are created, resulting in the pore sizes become increased which align with previous research [18]. After sintering temperature is increased, grains of the particle become bigger and the structure becomes more densified.

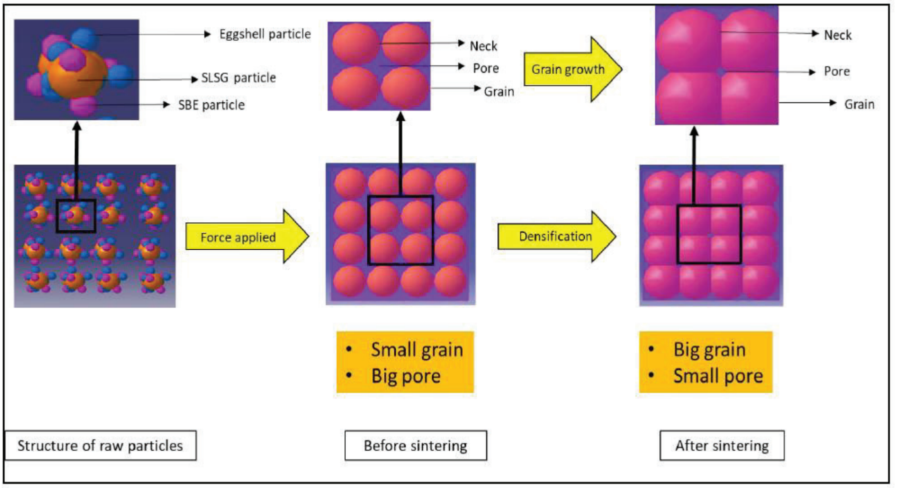

Figure 4: Illustration of model sintering process

Given that the greater of temperature caused the elimination of CO_2 throughout calcination process of eggshell as filler and has decreased the size of pores. However, the larger pores are attained by the lumps of greater grain size formed by sintering process. The findings are consistent with earlier research [19]. The size of the grains increases as the temperature increases while the porosity reduces. This also indicates the efficiency of the heat-treatment process especially on the selection temperature for sintering. As introducing the temperature to the glass-ceramic samples, the bonding between particles is formed, mainly contributed by a diffusion process [4].

4.0 CONCLUSION

The investigation on the relationship between materials (fillers and recycled glass) and their performance (Vickers Micro-hardness, flexural strength and microstructure) by measuring mechanical and microstructural parameters was successfully achieved. Samples with higher sintering temperatures and filler loading contribute to higher hardness-value and strength property that tested under flexure testing. Smoother fracture surface was obtained using highest sintering temperature. The findings concluded that the characteristic in terms of physico-mechanical properties of this SLSG composites can be enhanced by controlling the direct sintering temperature.

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AUTHOR CONTRIBUTIONS

M. Mesri: Methodology, Writing- Original Draft Preparation; Z. Shamsudin: Data Curation, Validation, Supervision, Writing-Reviewing and Editing; AMH. Dom and R. Hassan: Validation; M. Mulyadi: Writing-Reviewing and Editing.

CONFLICTS OF INTEREST

The manuscript has not been published elsewhere and is not under consideration by other journals. All authors have approved the review, agree with its submission, and declare no conflict of interest on the manuscript.

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