Faculty of Manufacturing Engineering

THE EFFECT OF HEATING RATE AND SINTERING TEMPERATURE ON GLASS CERAMIC COMPOSITE AT DIFFERENT FILLER LOADING

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DECLARATION

I declare that this thesis entitled “The Effect of Heating Rate and Sintering Temperature on Glass Ceramic Composite at Different Filler Loading” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the award of Master of Science in Manufacturing Engineering.

Signature :……………………
Supervisor Name :…………………….
Date :…………………….

DEDICATION

This thesis is special dedicated to:

My supportive husband, Mohd Rasyuyadi bin Salleh

Our cores of heart, Nur Raisya binti Mohd Rasyuyadi and Muhammad Rasydan bin Mohd Rasyuyadi.

My life inspiration parents, Hj. Salleh bin Maidin and Hjh. Fatimah binti Muda

My understanding mother in-law, Hjh. Aishah binti Daud

My intelligent supervisors, Dr.Zurina Shamsudin and Profesor Madya Dr. Jariah Mohamad Juoi

& to all of my siblings and close friends

ABSTRACT

The present work aims to develop fundamental information for the fabrication of glass ceramic composite (GCC) from recycled soda lime silicate (SLS) glass integrated with high loading of filler which is spent bleach earth (SBE). The aim of this study is to investigate the effects of different heating rate and sintering temperature on physical properties, microstructure and mechanical properties of GCC at different SBE loading. The recycled SLS glass and SBE were prepared by sieving to particle size < 45 µm and removing the oil in SBE by sonification process, respectively. The particle size distribution for both powders was determined using Laser Particle Size Analyzer (LPA). The main elements of recycled SLS glass were silicon dioxide (SiO_2), and calcium oxide (CaO), while the main elements of SBE were silicon dioxide (SiO_2) and aluminium oxide (Al_2O_3) determined via X-ray Fluorescence (XRF) analysis. Glass transition temperature (T_g) of recycled SLS glass was observed around 514 ºC. The compositions were selected based on high SBE loading which were SLS to SBE; 60:40, 55:45 and 45:55 wt. %. The type of sintering involved for 60:40 and 55: 45 wt. % of SLS to SBE was viscous flow sintering (VFS). Meanwhile for 45:55 wt. % of SLS to SBE, the sintering type involved was liquid phase sintering (LPS). The GCC was then formed using uniaxial dry pressing method with pressure at 2.5 tonnes for square mold and at 8 tonnes for rectangular mold. In the first part, the GCC sample was sintered at 700 ºC with heating rate of 2, 4, 6, and 8 ºC/min. Observation on crystalline phases showed that main crystalline phases appeared were quartz (SiO_2) and wollastonite ($CaSiO_3$). Increasing the heating rate did not give remarkable results of physical properties on green GCC. Microstructure observation revealed the presence of pores at minimal quantity. In this parameter, 2 ºC/min was chosen as an optimum heating rate. In the second part, the GCC was sintered at different sintering temperature of 750 ºC and 850 ºC. The results showed distinctive differences in physical properties, phases, and microstructure. This indicates that the composite was strongly influence by the sintering temperature. Increasing the sintering temperature showed high percentage of shrinkage, low percentage of porosity, and water absorption. The main phases appeared were quartz ($Si0_2$), wollastonite ($CaSi0_3$), cristobalite ($Si0_2$), and carneigeite ($NaAlSiO_4$). Crystalline phases showed high intensity and additional of new phases when the sintering temperature increased. Observation on microstructure disclosed more uniform and densified microstructure. An optimised green GCC of 45 wt. % SBE was successfully fabricated at sintering temperature of 850 ºC and heating rate of 2 ºC/min. The mechanical properties of hardness was 0.12 GPa and flexural strength of 9.50 MPa. A good prospect of addition some binder should be further explored on the fabrication of glass ceramic composite with high loading of SBE.

ABSTRAK

Kajian bertujuan untuk menyediakan maklumat asas dalam pembuatan komposit seramik kaca (GCC) daripada kaca soda kapur silikat (SLS) dikitar semula dengan komposisi tertinggi dari tanah peluntur terpakai (SBE). Tujuan kajian ini adalah untuk mengkaji kesan kadar pemanasan dan suhu sinter yang berbeza terhadap sifat fizikal, mikrostruktur dan sifat mekanik GCC pada komposisi SBE yang berbeza. Kaca SLS yang dikitar semula ditapis untuk mendapatkan saiz serbuk <45 mikron manakala SBE telah melalui proses sonifikasi untuk membuang kandungan minyak. Taburan saiz zarah ditentukan menggunakan Penganalisa Zarah Laser (LPA). Elemen utama kaca SLS dikitar semula adalah silicon oksida (SiO_2), dan calcium oksida (CaO) manakala unsur utama SBE adalah SiO_2 dan Aluminium oksida (Al_2O_3) melalui analisis XRF. Suhu peralihan kaca (T_g) kaca SLS dikitar semula adalah 514 ºC. Komposisi dipilih berdasarkan kepada kandungan SBE yang tinggi iaitu SLS kepada SBE; 60:40, 55:45, 45:55 wt. %. Jenis pensinteran yang terlibat untuk 60:40 dan 55: 45 wt. % kaca SLS terhadap SBE adalah pembakaran aliran likat (VFS). Sementara 45:55 wt. % SLS terhadap SBE, jenis pembakaran yang terlibat adalah cecair fasa pensinteran (LPS). GCC kemudiannya dibentuk menggunakan kaedah mampatan kering ekapaksi pada tekanan 2.5 tan untuk acuan segi empat sama dan 8 tan untuk acuan segi empat tepat. Bahagian pertama, sampel GCC disinter pada 700 ºC dengan kadar pemanasan 2, 4, 6 dan 8 °C/min. Pemerhatian pada fasa hablur menunjukkan bahawa fasa hablur utama adalah quarza (SiO_2) dan wollastonite ($CaSiO_3$). Peningkatan kadar pensinteran tidak memberi kesan yang terbaik pada ciri-ciri fizikal GCC. Pemerhatian terhadap mikrostruktur menunjukkan kuantiti liang berkurang. Walau bagaimanapun, permukaan kasar diperolehi dan kuantiti lubang berbentuk terhasil secara bertaburan. Dalam parameter ini, 2 ºC/min dipilih sebagai kadar pemanasan yang optimum. Pada bahagian kedua, GCC disinterr pada suhu pensinteran berbeza iaitu 750 ºC dan 850 °C. Perubahan ketara pada sifat fizikal, fasa dan mikrostruktur diperhatikan. Ini menunjukkan komposit ini amat dipengaruhi oleh suhu pensinteran. Peningkatan suhu pensinteran menunjukkan peratusan yang tinggi terhadap pengecutan. Peratusan yang rendah pada keliangan dan penyerapan air. Fasa utama adalah kuarza ($Si0_2$), wollastonite ($CaSi0_3$), cristobalite ($Si0_2$) dan carneigeite ($NaAlSiO_4$). Kristaliniti menunjukkan fasa intensiti yang tinggi dan penambahan fasa baru apabila suhu pensinteran bertambah. Pemerhatian pada mikrostruktur mendedahkan keseragaman dan kemampatan. GCC hijau yang optimum adalah 45 wt. % SBE, berjaya direka pada suhu pembakaran 850 ºC dan kadar pensinteran 2 ºC/min. Sifat mekanik, ujian kekerasan micro adalah 0.12 GPa dan kekuatan lenturan adalah 9.50 MPa. Prospek yang lebih baik perlu dikaji terhadap penambahan beberapa bahan pengikat terhadap penghasilan komposit seramik kaca yang mengandungi kandungan SBE yang tinggi.

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LISTS OF ABBREVIATIONS

Al_2O_3	Alumina
ASTM	American Society for Testing and Material
$CaCO_3$	Calcium Carbonate
CaO	Calcium Oxide
DTA	Differential Thermal Analysis
EDX	Energy Dispersion X-ray
EU	European Union
GCC	Glass ceramic composite
GPa	Giga Pascal
LOI	Loss of ignition
LPA	Laser Particle Size Analyzer
LPS	Liquid Phase Sintering
MPa	Mega Pascal
SBE	Spent Bleach Earth
SEM	Scanning Electron Microscopy
SFE	Spent Filtered Earth
SiO_2	Silica
SLS	Soda Lime Silicate
SSS	Solid State Sintering
T_c	Crystallization temperature
T_g	Glass transition temperature
T_m	Melting temperature
T_n	Nucleation temperature
T_{ng}	Nucleation and glass transition temperature
T_s	Softening temperature

VFS	Viscous Flow Sintering
XRF	X-ray Fluorescence

LIST OF PUBLICATIONS

(i) Peer reviewed journals

1. Shamsudin, Z., **Salleh, N.,** Juoi, J.M., Mustafa, Z., Zulkifli, M.R., 2016. The Effect of Spent Bleach Earth Loading on the Sintered Properties of Green Glass Ceramic Composite. Key Engineering Materials, 694, pp.179-183.

2. Shamsudin, Z., **Salleh, N.,** Mustafa, Z., Bakar, M.A.A., Hasan, R., 2016. Influence of Size Particles of SLS Glass on Properties of Sintered SBE Reinforced Glass Waste Composite. Proceedings of Mechanical Engineering Research Day, pp. 1-2.

(ii) Conference

1. 24th Scientific conference of the Microscopy Society Malaysia (SCMSM) 2015 (Oral), Melaka, 2nd – 4th December 2015.
    - Title: The Effect of Spent Bleach Earth Loading on the Sintered Properties of Green Glass Ceramic Composite.
2. Mechanical Engineering Research Day (MERD' 16) 2016 (Poster), Melaka, 30th – 31st March 2016.
    - Title: Influence of Size Particles of SLS Glass on Properties of Sintered SBE Reinforced Glass Waste Composite.

CHAPTER 1

INTRODUCTION

1.1 Background of study

Technological development, economic prosperity, cultural evolution, and urbanization have affected the Earth greatly in all aspects and this includes the increasing waste production. As urban population is increasing and consumption pattern is changing, solid waste management has been applied. In Asia, immediate attention towards waste management is required in all countries especially for developing countries including Malaysia. In the context of developing countries, the urgency is needed as health and environmental implication is always associated with solid waste management (Marshall and Farahbakhsh, 2013).

Recycling is considered as a very safe method and not expensive. In Kuala Lumpur, the government focuses on increasing the recycling rate, managing the costs of solid waste disposal, and deploying the use of technology in managing solid waste (Saeed et al., 2009). Due to this situation, studies have been done to protect and preserve the earth by implementing more reduce, reuse, and recycle (3R) alternatives. The 3R initiative is not only run in Malaysia. This is a common practice in the other developed and developing countries too. The usage of recycled soda lime silicate (SLS) glass is also an initiative that has been proposed by researchers who study the fabrication of glass ceramics or so- called glass ceramic composite with various applications (Ponsot and Bernardo, 2013). This has been recommended due to the unique behaviour of glass that can be recycled many times without reducing its strength. It is also adaptable in different techniques and shapes of fabrication (Juoi et al., 2013). Besides reducing pollution, it also offers less energy in the processing of secondary materials (Chinnam et al., 2013).

The establishment of recycled SLS glass in the fabrication of glass ceramic composite (GCC) has inspired other researchers to integrate other natural waste materials into the recycled SLS glass. A combination of various types of natural waste was found improving the properties of glass ceramic composite (GCC). Spent bleach earth (SBE) is a natural waste from palm oil refineries. It is a clay type and has been used as an absorbance in palm oil refineries to absorb all the impurities including colours. It contains a major composition of SiO_2 and Al_2O_3, it is expected to have a possibility in improving the properties of GCC when integrated with recycled SLS glass. Boey et al. (2011a) stated that SBE has a potential to be used as a clay substitute in brick or tile manufacturing processes.

Sintering process is a controlled heat treatment used to fabricate GCC. This process is very common in which it can produce strong and dense-glass ceramics from waste at very low sintering temperature (Bernardo et al., 2007; Fernandes et al., 2012). Properties and densification of GCC can be adjusted by controlling the sintering profile (Pascual et al., 2002). Different parameters such as method, composition, temperature, and heating rate give advantages in producing glass ceramics (Juoi et al., 2013) as it influences the types of sintering occur during sintering process.

This sintering profile of composition, heating rate and sintering temperature need to control to achieve a good properties of GCC with low porosity, low water absorption, and high density. The formation of phases and development on the microstructure is expected to show correlation between the selection of parameters with the physical and mechanical properties of GCC.

1.2 Problem Statement

In Malaysia, recycled soda lime silicate glass is easily accessible. Besides that, it has a good bending strength and chemical stability (Bernardo et al., 2007). Also, it improves water absorption of ceramic tiles (Pontikes et al., 2007). In general process, recycling soda lime silicate glass was seen as a solution of landfilled space (Saeed et al., 2009).

Spent bleach earth (SBE) is well known as one of vegetable industrial waste has dramatically increased. It is used as an absorbance in the vegetable refineries process including from palm oil industries. In Malaysia, over 150,000 tonnes of SBE is estimated to be produced annually with more than 17 million tonnes of palm oil production (Boey et al., 2011a). Generally, it is predicted that the production of SBE waste around the world is 600,000 tonnes (Suhartini et al., 2011). The current practice to dispose the high volume of SBE is through landfill. However, this disposal method is problematic due to the oil on surface area of the SBE particles which exposed to oxygen from the environment. Thus, producing rapid oxidation and adequate heat to ignite the oil. Hence, the disposal activity need to be done immediately to avoid rapid oxidation. This disposal method has been banned by European Union (EU) landfill directive (Beshara and Cheeseman, 2014). A consequence of this disposal method also was very costly and need to be paid by refineries (Loh et al., 2013). Hence, alternative is extensively needed to control consequence mentioned. Recycling method can be used extensive in maintaining good levels in disposal of waste from refineries especially SBE.

SBE was utilized by regenerating the SBE with acid or alkaline to recover it. This regenerated SBE will be used again as an absorbance to absorb other unwanted matter such as fluoride (Malakootian et al., 2011), basic dye (Mana et al., 2006) and herbicide paraquat

(Tsai and Lai, 2006). Further studies on properties of SBE has led to a discovery of new technology on bio-organic fertilizer and briquette made from SBE. In bio-organic fertilizer, the adequate amounts of valuable mineral elements from co-composed of SBE with some agricultural and palm oil milling by-products has a positive impact on soil physical. Meanwhile, the SBE was used as briquette due to the catalytic partial oxidation's (CPO) hydrocarbon component and high calorific value in it (Suhartini et al., 2011; Loh et al., 2013).

Besides that, the potential of SBE to be used as a clay substitute in the production of brick and tile manufacturing also has been highlighted by many researchers (Boey et al., 2011a; Beshara and Cheeseman et al., 2014). Recent research on the formation of pore forming brick by Eliche-Quesada and Corpas-Iglesias (2014) reported that only 10 wt. % of SBE was added into clay brick formulation that showed an optimum result of low bulk density, mechanical strength and thermal conductivity with higher total porosity and water absorption with respect to the pure clay brick. It was sintered at sintering temperature of 950 °C and heating rate of 3 °C/min. Increase the SBE loading higher than 10 wt. % result in high mechanical strength which was 30–25 MPa. However, higher water absorption value was obtained.

In theory, sintering profile at various heating rate, sintering temperature and holding time can affect the end properties of sintered GCC (Guo et al., 2010). However, the holding time favour less significant effect to the properties as compared to heating rate and sintering temperature (Bernardo et al., 2007). Therefore, by differentiate the heating rate and sintering temperature, it will be expected to produce a good properties of GCC with maximum used of weight percentage SBE between 40 to 55 wt. % in one GCC batch. Moreover, the effect of these parameters on physical properties, microstructure and phases