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Influence of Different Compositions of Fly Ash as Fluxing Agent in Porcelain

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ABSTRACT. Fly ash is alumina-silica waste products from the combustion of palm fiber and shells in the boiler which are collected at the multi-cyclone collectors where almost 3 million tonnes per annum being produced in palm oil industry in Malaysia. The aim of this paper is to investigate the replacement of feldspar by fly ash as potential fluxing agent in triaxial porcelain. Clay, feldspar, quartz and fly ash were mixed for 12 hours, pressed into pellets and sintered at 1250 ºC. The samples were measured according to the physical and mechanical properties. Microstructure study was done through SEM analysis. The optimum composition of fly ash was observed at 5 wt.% where the maximum compressive strength was achieved at 105.04 MPa and shows the decreasing results in volume shrinkage. SEM study shows intense interlocking between the primary and secondary mullite needles in glassy matrix which contribute in improving the strength of the porcelain at this composition. Therefore the substitution of fly ash is suitable as a fluxing agent in porcelain that improved its physical and mechanical properties.

Keywords: *Fly ash, Porcelain, Fluxing agent, Physical, Mechanical;*

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1. INTRODUCTION

Investigations of using fly ash in ceramics are widely reported over the decade ago. Fly ash is fine residue that has been produced from the combustion process of palm solid waste in the boiler where it can be an alternative source of alumina-silicate materials and has a good potential as raw materials of porcelain [1]. Porcelain is well-known as a type of ceramics which high in complexities in each aspect which start from its raw materials, forming and sintering, microstructure and phase formation [2]. Porcelain is comprised of three different raw materials which are clay, feldspar and quartz [2,3]. Clay are the main raw materials which provides plasticity and helps in giving strength at green state during forming process [2]. Feldspar as a fluxing agent in which it forming a viscous liquid at lower temperature where lead to a better vitrification and resulting to higher in densification. Quartz act as a filler where it has higher melting point which could reduce the shrinkage during firing [2,3].

Much work has been done by using fly ash as part of the raw materials in porcelain compositions. The replacements of quartz with fly ash are widely reported compared to feldspar due to higher of SiO_2 content which fulfill the essential criteria as filler in porcelain. Previous studies by Dana et al. [3], quartz has been progressively replaced by fly ash in triaxial porcelain compositions. They observed that the strength of the porcelain was increased with the increasing of fly ash due to higher presence of the mullite content. The maximum strength (70.5 MPa) has been achieved at 15 wt.% of fly ash content that sintered at 1300 ºC. Other

observation, the earlier formation of mullite and glassy phase in fly ash porcelain at 1200 ºC compared to normal porcelain at 1250 ºC where it was improved the microstructure and the strength of the porcelain at lower temperature [4]. Meanwhile, Kumar et al. [5] found that the strength of the tiles was decreased with the additions of fly ash greater than 25 wt.% due to higher content of glassy phases in fly ash porcelain. On the basis of literature, it was found that feldspar and quartz has been only partially and fully replaced with fly ash simultaneously in porcelain compositions by Mukhopadhyay et al. [6]. They obtained the optimum compositions were at 30 wt% of fly ash with the lowest apparent porosity and the highest strength achieved at 72.3 MPa. Therefore, it was found that the replacements of fly ash on feldspar are still not widely investigated in triaxial porcelain compositions.

This study involves triaxial porcelain compositions with substitution of fly ash progressively on feldspar. Thus, the aim of this study is to optimize the compositions of fly ash as fluxing agent with experienced the best physical and mechanical properties. Various compositions of fly ash were substitute on feldspar and the physical and mechanical properties and microstructure behavior were analyzed.

2. MATERIALS AND METHODS

Fly ash used in this study was collected from Bukit Lawiang Palm Oil Mill in Johor, Malaysia. Standard porcelain compositions used are 50 wt.% of Clay, 25 wt.% of quartz and 25 wt.% feldspar. Six different mixtures were prepared with various compositions of fly ash as shown in Table 1.

Table 1 Compositions of the mixture (wt%)

Sample No.	Clay	Quartz	Feldspar	Fly ash
A1	50	25	25	0
A2	50	25	20	5
A3	50	25	15	10
A4	50	25	10	15
A5	50	25	5	20
A6	50	25	0	25

Fly ash was dried in an oven at 110 ºC for 24 hours to remove the moisture content and then grounded for 12 hours using ball mill in order to reduce the particle size. After that, it was calcined at 800 ºC for 3 hours to remove the volatiles residue and excess unburned carbon. The porcelain powder and calcined fly ash powder were mixed together for 12 hours in order to obtain the homogenized mixture with additional of 2 wt.% PVA powder as a binder. Then, it was pressed into pellets under 3 tons pressure by using hydraulic press machine (Carver 3851-0). All the pellets were isostatically pressed at 100 MPa using cold isostatic pressing (CP360) for more compaction. Finally the samples were sintered using electrical protherm furnace at 1250 ºC for 2 hours. The physical and mechanical properties are measured. The bulk density testing was conducted based on Archimedes' principle follow the standard of ASTM C373. The compressive strength was determined using universal testing machine (Testometric) as ASTM C773-88 and Vickers microhardness was determined by using Shimadzu (HMV-2 series). Characterization on the concentration of the crystalline phase was determined by using x-ray diffraction (Bruker D8 Advance) with Cu Kα radiation and analyzed by using X'Pert High Score Plus software (PANalytical). The sintered samples were polished and etched in 4% hydrofluoric acid for 3 min and the microstructure was observed using Scanning Electron Microscope (Hitachi-SU1310).

3. RESULTS AND DISCUSSION

Fig. 1(a) and Fig. 1(b) show the XRD results of the raw materials of porcelain and fly ash. Based on the XRD results, clay contains of kaolinite and quartz whereas feldspar contains of albite, quartz, microcline and muscovite. Fig. 1(b) indicates the presence of quartz, albite, mullite and cristobalite

in fly ash. It was observed that feldspar and fly ash have almost similar mineralogical compound where both of the materials was higher in quartz (SiO_2) and albite ($NaAlSi_3O_8$) content. The presence of quartz and albite as the main mineralogical compositions in fly ash is 56.6% and 21.2% whereas mullite and cristobalite is 19.2% and 3%. Albite are one of the feldspar mineral which is generally known as the pure sodium Na-feldspar [7]. The interesting fact was the presence of sodium-aluminium silicate (albite) in fly ash are suitable as a fluxing components where above 1100 ºC, the albite will be dissolved in order to helps in the formation of glassy phase in the porcelain [8].

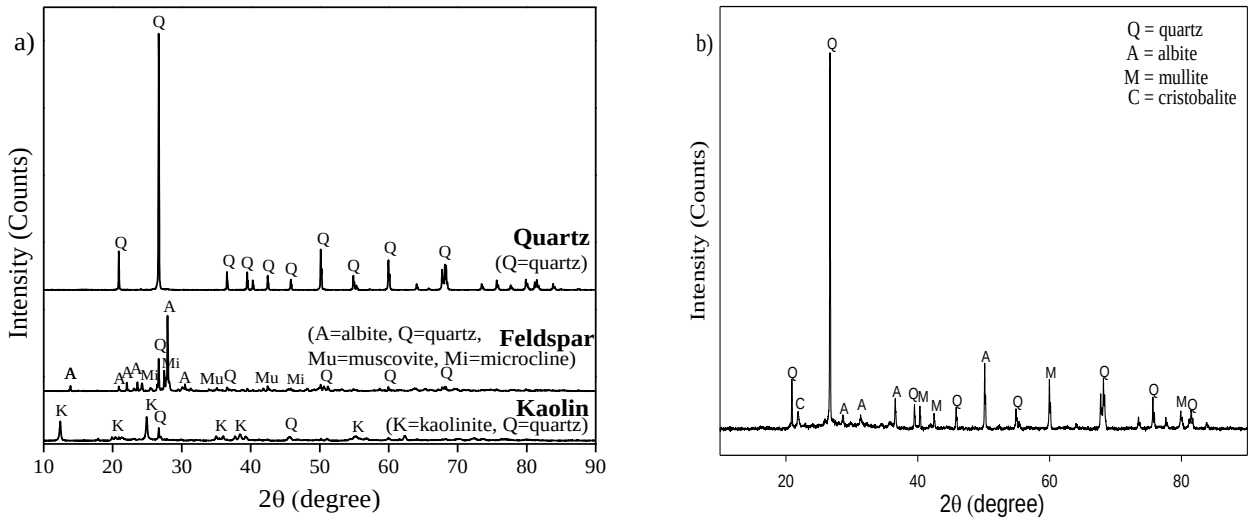


Fig. 1 (a) XRD patterns of the raw materials of porcelain; (b) XRD pattern of fly ash

The apparent porosity and volume shrinkage of the samples is presented in Fig. 2(a). It may be observed that the shrinkage was decreased evenly with increasing in fly ash compositions due to the presence of fly ash. Fly ash was higher in SiO_2 content which leads to reduce the shrinkage in porcelain. Generally, volume shrinkage describes the reverse trend to the porosity. However, the porosity is much related to the bulk density of the samples. The apparent porosity was only increased at the substitution of 5 wt.% of fly ash and declined gradually afterwards. Dana and Das [4] reported that this behavior is cause by the higher formation of glassy phase due to the presence of excess amount of fluxing oxides from fly ash in porcelain compositions which is less viscosity and could fill up the open pores. According to the Yürüyen and Toplan [9], incorporation of 5 wt.% of fly ash in porcelain compositions increase the shrinkage compared to normal porcelain due to the formation of liquid phases from the melting process of fly ash.

The variation of bulk density and mass loss are shown in Fig. 2(b). The mass loss was increased with increasing in fly ash compositions. This result indicates that the additional of fly ash into the normal porcelain has a great potential of becoming lightweight porcelain. The bulk density plotted decreases with the increasing in fly ash compositions. The maximum bulk density, 2.40 g/cm^3 was obtained at 0 wt.% of fly ash and gradually decreased to the lowest bulk density, 2.12 g/cm^3 at 25 wt.% fly ash. These results are closely related to the apparent porosity where both of the results shows decreasing value in density and porosity with increasing in fly ash compositions. Mukhopadhyay et al. [6] reported that this behavior is due to the increasing in number or size of closed porosity caused by bloating formation due to the dissociation of oxygen (O_2) from the transformation of Fe_2O_3 to Fe_3O_4 and other possibilities is attributed to anisotropic grain growth in fly ash porcelain samples at a sintering temperature of 1250 ºC [10].

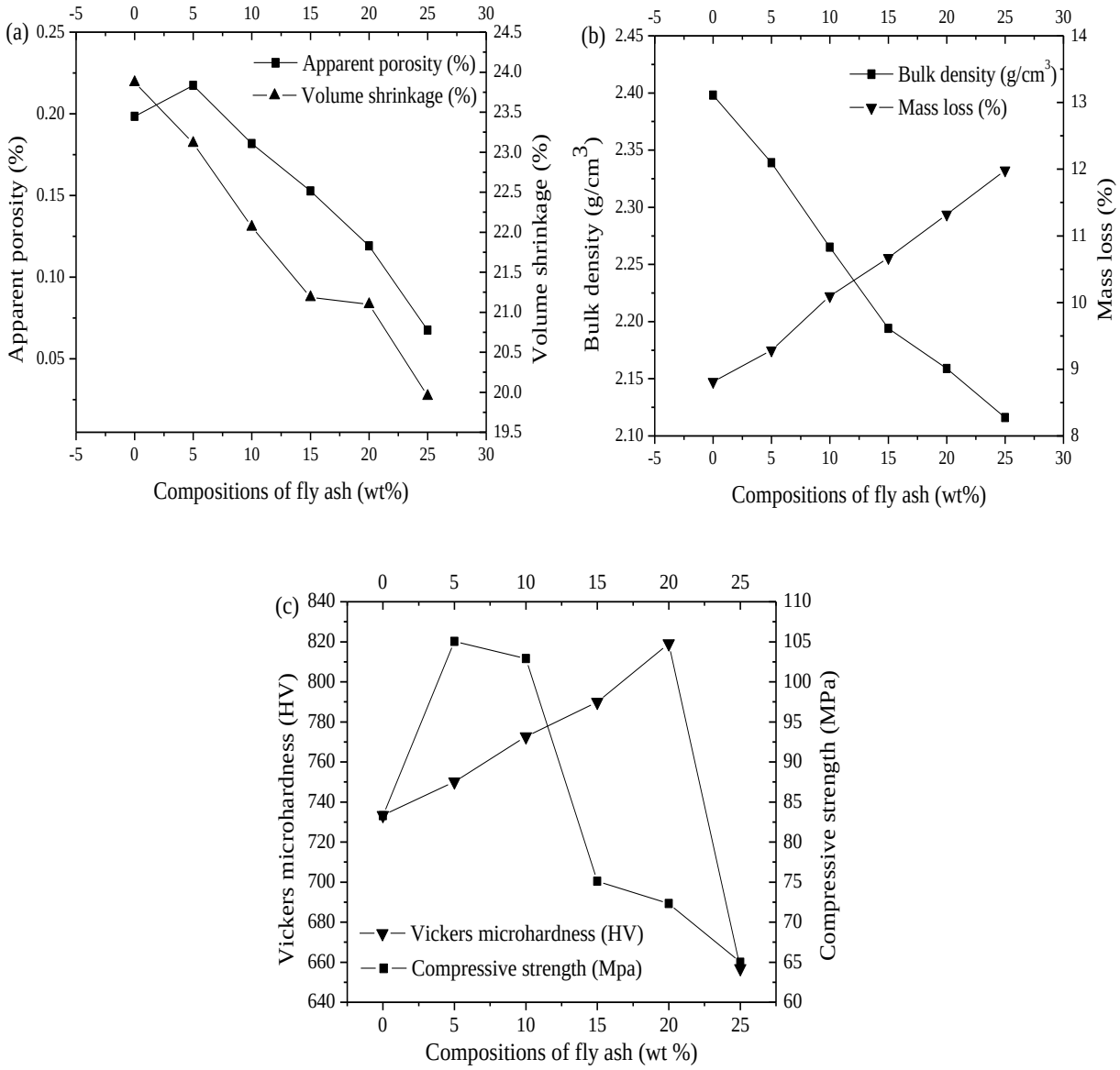
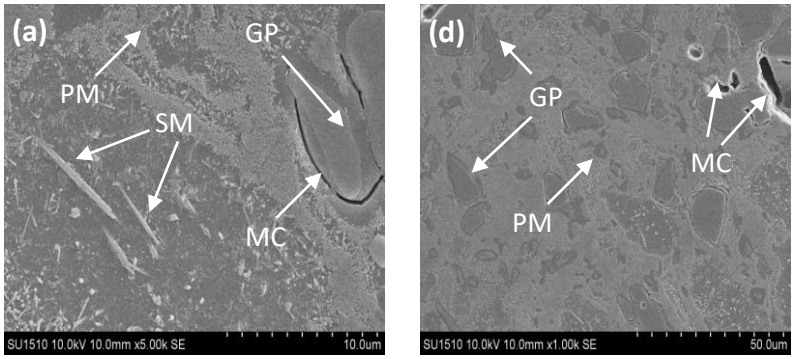


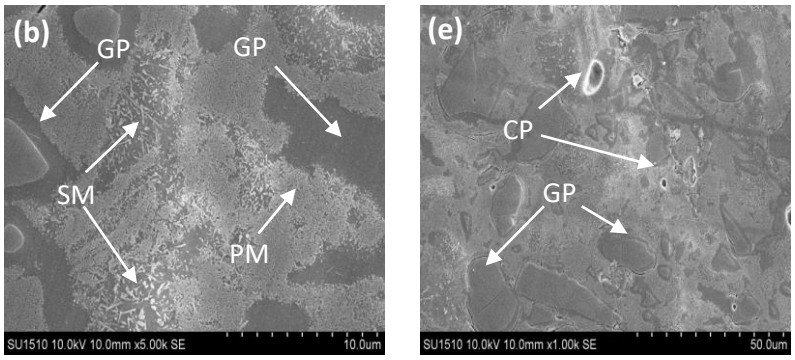
Fig. 2 (a) Variation of the apparent porosity and volume shrinkage; (b) Variation of bulk density and mass loss; (c) Variation of the Vickers microhardness and compressive strength; according to the fly ash compositions at a sintering temperature of 1250 ºC.

The compressive strength and the Vickers microhardness of the porcelain samples were shown in Fig. 2(c). At the substitution of 5 wt.% of fly ash, the highest compressive strength was achieved with value of 105.04 MPa. The compressive strength was dropped dramatically at the composition of more than 10 wt.% due to increasing in closed porosity cause by the bloating pores in fly ash porcelain samples [6]. This behavior is also due to the increasing in glassy phase rather than crystallization of mullite which may be caused by the presence of alkali (K_2O + Na_2O) content in fly ash [5]. Meanwhile, there are difference trend observed in Vickers microhardness result shows a steady increase and obtained the maximum value, 819.1 HV at 20 wt% fly ash. As been reported by Kituoni et al. [10], different trend in both of the mechanical measurement is due to the appearance of different particle size of the materials between the raw materials of porcelain and fly ash. Zhang et al. [11] explained that the relationship between hardness and strength in ceramic could be influenced by the indentation of 'sink-in' morphology and different for coarse-grained since it only involves

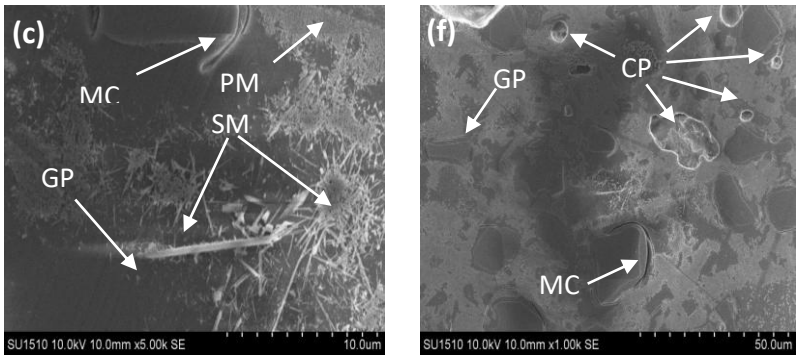
the surface of the samples. However, at the compositions of 5 wt.% of fly ash, the hardness could be consider higher since it shows an increasing value of 750.10 MPa compared to normal porcelain which is 733.40 MPa.



0 wt.% fly ash (normal porcelain)



5 wt.% fly ash



15 wt.% fly ash

Fig. 3 SEM photomicrograph of the sample in different magnification (a-b) 5kx (d-f) 1kx (GP = glassy phase, PM = primary mullite, SM = secondary mullite, MC = microcrack, CP = closed pore)

The SEM photomicrographs of the samples were presented in Fig. 3 where it shows the presence of quartz and mullite in porcelain. According to Fig. 3(a), there are uneven surface structures in normal porcelain. It was observed that the extensive cracking occurred around the quartz grain at normal porcelain

in Fig. 3(a) and at 15 wt.% fly ash in Fig. 3(c) due to the quartz conversion stress during cooling process [6]. However, at 5 wt.% of fly ash in Fig. 3(b) shows strong reinforcement and uniform distribution between fine primary mullite and secondary mullite needles in glassy matrix which probably lead to achieved higher compressive strength as reported by Dana et al. [3]. Fig. 3(f) shows higher formation of glassy phase which lead to increasing number of closed porosity where attributed to a reduction in strength of the fly ash porcelain.

4. SUMMARY

Replacement of fly ash on feldspar in the compositions of porcelain has a great effect to production of porcelain. The optimum compositions of fly ash were obtained at 5 wt.% according to the results of apparent porosity and compressive strength that shows the highest value respectively. The bulk density is obtained decreases as the increase in fly ash composition and contradict pattern with mass loss curve. The highest value of compressive strength and higher Vickers microhardness was achieved at composition of 5 wt.% fly ash with the value of 105.04 MPa and 750.10 HV respectively. Incorporation of porcelain with 5 wt.% of fly ash obtained intense interlocking of fine mullite needles in the glassy phase which contribute in achieving higher strength of porcelain.

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