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FIRING BEHAVIOUR OF CERAMIC WHITEWARE BODIES INCORPORATED WITH LOCAL FELDSPATHIC SOURCES

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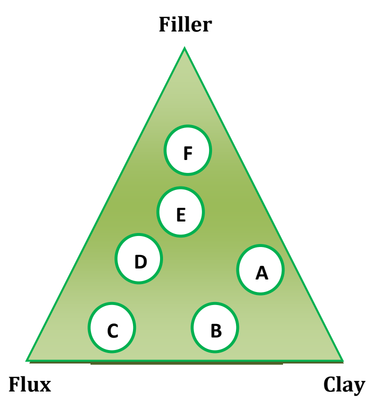

Triaxial body compositions of clay-based ceramic products
A-Wall Tiles B-Vitreous Whitewares C-Floor Tiles D-Electrical Porcelain E-Hard Porcelain F-Semi-vitreous Porcelain

Abstract

Quarry dust is a by-product of granite quarrying activities. Granite fragments, that are normally obtained from blasting granite rocks in a particular quarry, are subjected to a number of machineries of decreasing crushing aperture sizes and each attendant sieving to obtain granite aggregates of the desired size, normally 20 mm, 10 mm, etc. These are to be used in the construction of buildings as coarse aggregates in concrete mixes, as well as in the laying of tarred roads. The finest fraction, nominally below 7 mm or 5 mm in size depending on the practice of the quarrying company, is normally dumped on site. It is considered to be a waste that needs to be disposed of. Apart from attending to the environmental issue, the reuse of such waste material in the development of new products would be an attractive commercial proposition. In this work, granite quarry dust was incorporated into a clay-based ceramic body to replace the use of feldspar (an expensive component) as the fluxing agent in such triaxial clay whiteware compositions. A fluxing agent is the component in a ceramic body composition that melts first and functions as the component that binds together all the other solid particles into a rigid body. Initially, the quarry dust was chemically and mineralogically characterized before mixing with clay at a fixed 45:55 weight ratio. The mixtures were then pressed and fired at various temperatures before testing the properties of the fired products. A comparison was made with a local feldspathic source from Gua Musang to ascertain the feasibility of such replacement to produce high quality ceramic bodies. It was found that mineralogically the granite quarry dust consists of K-feldspar (or orthoclase) as compared to Na-feldspar (or albite) found in the Gua Musang feldspar. This led to melting at a lower temperature of the former but exhibited a much more viscous melt. At the same time, the higher iron content in the granite quarry dust led to a much darker colourisation of the body upon firing. In conclusion, the granite quarry dust has been found to be successful in lowering the maturing temperature of ceramic bodies compared to the Gua Musang feldspar beside conferring stability of the low porosity property over a wider firing temperature.

Keywords: Granite quarry dust, feldspar, ceramic bodies, fluxing agent, clay

1.0 INTRODUCTION

Ceramic whiteware bodies are made up essentially of three components in various proportions, viz. clay, flux and filler [1]. In such triaxial body compositions, clay and the filler, normally silica sand, are easily resourced in Malaysia. On the other hand, the flux, which is normally the mineral feldspar, was for many years imported from the Scandinavian countries by the Malaysian ceramic industries, and tends to be the most expensive of the three components. Over the years, the industries have resorted to the use of feldspars imported from either India or China. However, lately there has been a gradual replacement with a local feldspathic source, i.e. the Gua Musang feldspar (GMF). Nonetheless, efforts are still on-going to source out cheaper and better alternatives. In this work, the use of granite quarry dust (GQD) as a fluxing agent was attempted and the results compared to that of the Gua Musang feldspar. Quarry dust is a by-product of granite quarrying activities which are spurred on by the requirements for granite aggregates which are heavily used in the construction of buildings and roads. The use of the quarry dust as a fluxing agent is driven by two principal factors, namely to attend to the environmental problem created by this quarry dust which is normally dumped on-site as wastes, apart from seeking for an added value to these wastes in the manufacture of ceramic products.

2.0 METHODOLOGY

In this work, the body composition was fixed at 55% ball clay and 45% of either Gua Musang feldspar or granite quarry dust. The third component, the filler which is normally silica sand, is considered to be adequately contributed by the high percentage of free silica (SiO_2) observed in both feldspathic sources. All three raw materials were dry-milled in a Fritsch Pulverisette P5 milling machine to reduce the size using zirconia milling media for 15 minutes duration. The ground powders were then analysed chemically and mineralogically using a Rigaku RIX 3000 x-ray fluorescence spectrometer and a Siemens D500 x-ray diffractometer, respectively. The ball clay was mixed with either the GMF or GQD at the specified weight ratio. The mixture was then uniaxially pressed in a steel die of 23mm diameter at a pressures of 150MPa. The number of pellets formed for each set of conditions were 15. After drying overnight at 100°C, the green pellets were fired at three different temperatures of 1150°C, 1200°C and 1250°C in a Carbolite RHF 1400 furnace with a soaking time of 2 hours. The fired pellets were then analysed for porosity, density, phase analyses by XRD and morphological observations using a Zeiss Supra 35VP field emission scanning electron microscope (FESEM).

3.0 RESULTS AND DISCUSSION

3.1 Chemical Composition

The results of the chemical compositions for the three raw materials are given in Table 1. Firstly, the chemical composition of the ball clay is typical of a kaolinitic clay with kaolinite, quartz and muscovite as the principal minerals as indicated by the high amount of alumina (Al_2O_3) and silica (SiO_2) contents [2]. The high loss upon ignition (LOI) is indicative of a high amount of weight losses due to kaolinite dehydroxylation and possibly the burning out of some organic content [3]. Secondly, upon comparing the two feldspathic sources, the high SiO_2 content in both (69.0 and 70.0) is indicative of a high free silica content and feldspar minerals. The high free silica content thus did not necessitate the incorporation of additional silica filler in the initial body composition of the ceramic bodies. GMF shows a combined alkali contents which are quite high (4.0% Na_2O and 2.7 K_2O), whilst GQD shows a similarly high combined alkali contents (2.2% Na_2O and 5.0% K_2O). It is significant to note that the former is higher in Na_2O (sodium feldspar) whilst the latter is higher in K_2O (potassium feldspar). This difference can be ascertained by the detection of different feldspathic minerals in the two sources by XRD [4]. As the flux is the first component in a ceramic body to form a glassy phase upon firing, it has been reported that K-feldspar will produce a much more viscous glassy phase at a lower firing temperature [3]. In terms of the colouring oxides, which are mainly Fe_2O_3 and TiO_2, they are higher in GQD compared to that in GMF. As a result, the fired GMF bodies with lower colouring oxides exhibit a creamy colour upon firing whilst those containing GQD show a much darker buff appearance [5] (Table 2).

Table 1. Chemical compositions of ball clay, Gua Musang feldspar (GMF) and granite quarry dust (GQD)

Elements	Clay	GMF	GQD
SiO_2	54.2	69.0	70.0
Al_2O_3	29.9	21.0	16.0
Na_2O	trace	4.00	2.20
K_2O	1.50	2.70	5.00
Fe_2O_3	1.10	0.80	1.90
Cr_2O_3	0.04	trace	trace
TiO_2	0.60	0.07	0.28
CaO	0.04	0.24	2.10
MgO	0.22	0.31	1.30
LOI	12.31	2.00	1.50

*LOI is loss on ignition

Table 2. Colours of body upon firing at different temperatures

Body	1150°C	1200°C	1250°C
GMF	Creamy	Creamy	Creamy
GQD	Buff	Dark buff	Darkish

3.2 Mineralogical Composition

The chemical compositions shown in Table 1 shows a higher content of Na_2O in GMF, and hence the main mineral detected by XRD is the mineral Na-feldspar (albite) apart from free silica in the form of the mineral quartz (Figure 1). On the other hand, GQD exhibits the presence of a K-feldspar (orthoclase) apart from free silica, also in the form of the mineral quartz (Figure 2). Hence, these two variants of the feldspathic minerals affect the firing behaviour of the ceramic bodies as detailed in the preceding sub-section on chemical composition.

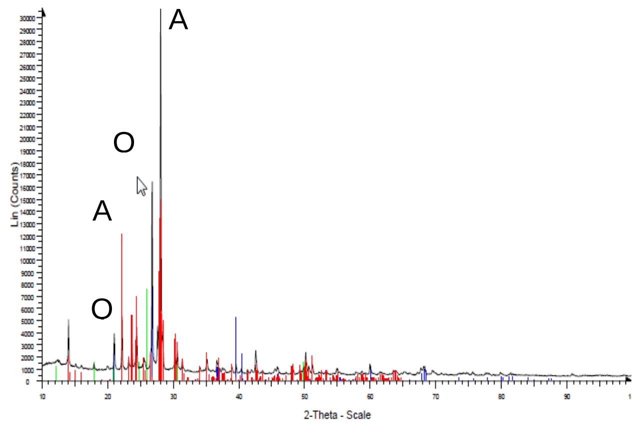

Figure 1 X-ray diffractogram of Gua Musang Feldspar (Legend: A is Albite, Q is Quartz)

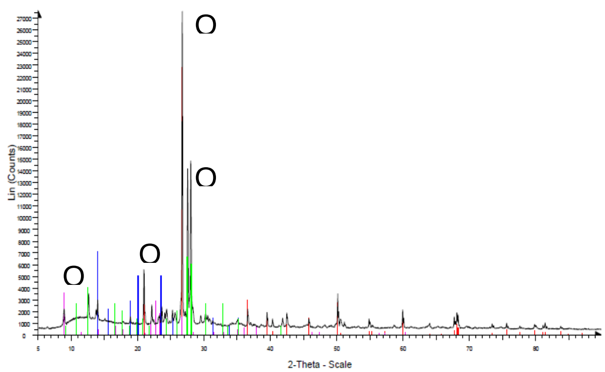

Figure 2 X-ray diffractogram of Granite Quarry Dust (Legend: O is Orthoclase, Q is Quartz)

3.3 Physical Properties

The physical properties reported in this paper include density and porosity. The porosity and density results are presented in Figures 3 and 4. The results show that the GMF body exhibits an increase of bulk density from 1150°C to a maximum at about 1200°C before a sharp fall takes place down to1250°C. The porosity shows a corresponding inverse relationship of a decrease in porosity before it increases back again up to 1250°C. This inverse relationship between porosity and density has been reported in most ceramic systems [6].

On the other hand, the GQD body shows a marked difference to that of the GMF body. At 1150°C, the density shows a gradual decrease down to 1250°C. Correspondingly, the porosity shows a gradual increase as the temperature increases.

This difference in behaviour can be attributed to the glassy phase formed from the two different feldspathic sources. As mentioned earlier, the presence of K_2O will promote the formation of a glassy phase at a lower temperature for the GQD body which have densified at a temperature lower than 1150°C, whilst the GMF body achieves maximum vitrification (or glassy phase formation) at around 1200°C. In summary, it can be concluded that the GQD promotes glassy phase formation or vitrification (hence densification) at a lower temperature than GMF. This can be considered to be advantageous in terms of reducing the firing temperature of the products.

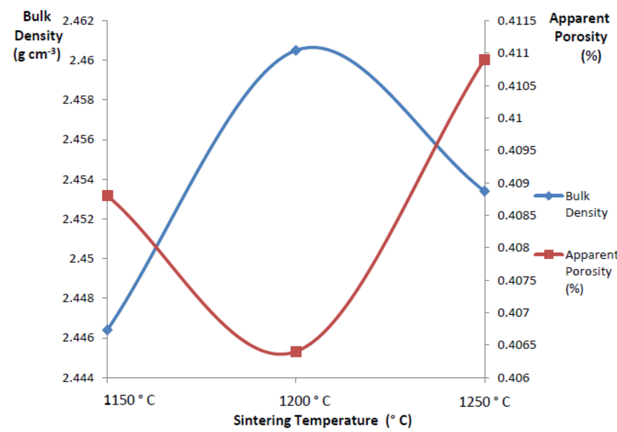

Figure 3 Effect of firing temperature on the porosity and bulk density of a ceramic body added with GMF

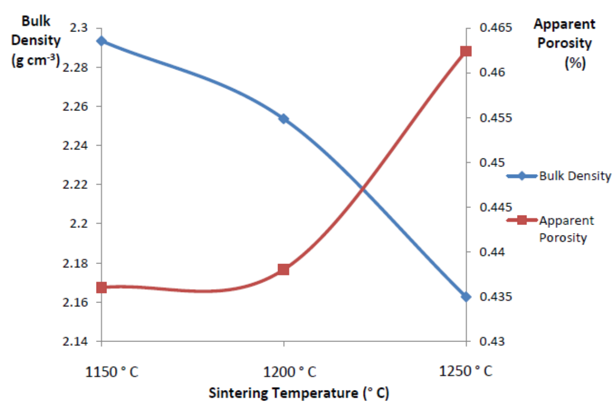

Figure 4 Effect of firing temperature on the porosity and bulk density of a ceramic body added with GQD

3.4 Morphological Observation

Figures 5 shows the micrograph of the fracture surface of a specimen fired at 1150°C. Numerous tiny pores are still observed indicating that vitrification (i.e. densification in the presence of a glassy phase) is still taking place in the body at that temperature. The surface appears fairly smooth and flat indicating that fracture had taken place along a glassy phase and which is of low viscosity, respectively. On the other hand, the GQD body at 1150°C (Figure 6) shows a solid compacted structure with lesser tiny pores but the surface, however, shows a contoured shape indicative that the glassy phase formed (higher in K content) is more viscous compared to the glassy phase (higher in Na content) in the GMF body(Figure 5).

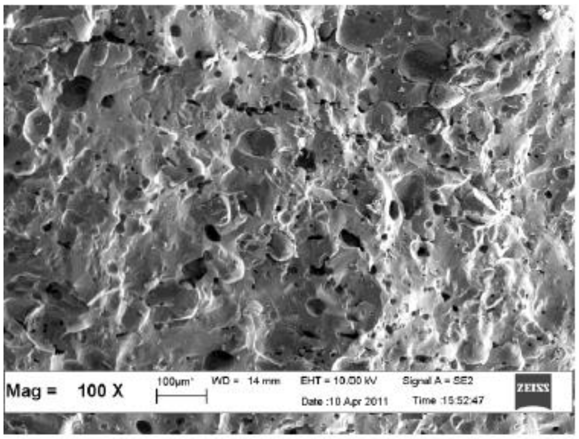

Figure 5 Micrograph of the fracture surface of a clay body with GMF fired to 1150°C

Thus, it can be confirmed that K-feldspar promotes the formation of a glassy phase at a lower temperature but the viscosity of the glassy phase formed is higher than that for Na-feldspar [1, 3]. Apart

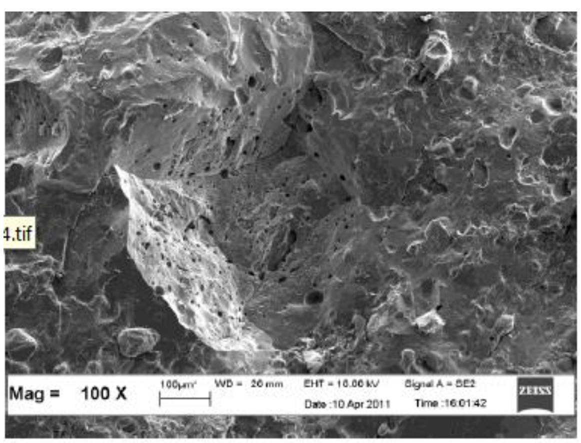

Figure 6 Micrograph of the fracture surface of a clay body with GQD fired to 1150°C

from the lower firing temperature required, the viscosity of the glassy phase is much more stable over a wide range of temperatures and thus contribute to product stability [7, 8]. On the other hand, the GMF body shows a much more fluid glassy phase that shows bloating (expansion of body) at temperatures exceeding 1200°C [9,10,11]. Similarly, the fluxing behavior of other minerals such as illite, muscovite and microcline, which have significant contents of alkali oxides (potassium and sodium), have also been reported [12, 13, 14]. Recently, the use of granite quarry dust in the construction industry, in particular as concrete mixes, and in geopolymer synthesis have also been researched into [15, 16, 17].

4.0 CONCLUSION

This work has successfully shown that both feldspathic sources, viz. the Gua Musang feldspar and the granite quarry dust, can be used as fluxing agents in ceramic body compositions. The results show that chemically and mineralogically these two sources are different in compositions, namely the GMF is richer in Na-alkalis whilst the GQD is richer in K-alkalis. Correspondingly, the GMF is made up mainly of the feldspathic mineral Na-feldspar (albite) whilst GQD is made up of K-feldspar (orthoclase). Consequently, the glassy phase formed upon firing are not the same and the subsequent effects on the densification of the two bodies are invariably different. Apart from that, the higher amount of Fe_2O_3 in GQD contributes to the darker colouring of ceramic bodies made using GQD as the fluxing agent. From the perspective of ceramic processing, the granite quarry dust appears to be of much better potential as a fluxing agent as it contributes to the lowering of the firing temperature (thus contributing to a faster and a more economical processing cycle). Apart from that, the initial material cost of the granite quarry dust (at the moment being considered a waste) is much lower than that of the Gua Musang feldspar, which is only resourced from the state of Kelantan apart from the depletion in reserves.

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