Effect of Kaolin-Alumina Ratio on Physical and Structural Properties of Ceramic Porcelain

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Abstract. Ceramic porcelain was prepared by sintering process at temperature of 850 °C, using minerals such as kaolin ($Al_2H_4O_9Si_2$) and feldspar ($Na_2OAl_2O_3{\cdot}6SiO_2$) as main raw materials with addition of alumina (Al_2O_3) and silica (SiO_2). This study was focus on the effect of kaolin addition and alumina reduction on the physical and structural properties of ceramic porcelain. Fixed value of feldspar and silica which are 20 wt% and 10 wt% respectively was mixed with x wt% of kaolin and 70-x wt% of alumina (with x= 30, 35, 40, 45 and 50). The composition was then mixed and sintered at 850 °C for 4 hours. Characterization were done to observe the properties of ceramic porcelain samples through X-Ray diffraction (XRD), X-Ray fluorescence (XRF), density, water absorption and firing shrinkage. XRD results show that the amount of corundum as main phase has increased with the increasing amount of kaolin and decreasing amount of alumina. XRF results show the percentage of elements presence in the raw materials. Density of the samples show decreasing values from 2.85 g/cm^3 to 2.49 g/cm^3 with the increasing amount of kaolin-alumina ratio. Both water absorption and firing shrinkage results show increasing values with increasing amount of kaolin and decreasing amount of alumina. Result obtained shows strong evidence that proved the addition of kaolin and reduction of alumina give significant effect on the physical and structural properties of ceramic porcelain.

INTRODUCTION

Porcelain is a ceramic material that is made by heating the ceramic raw materials such as kaolin, feldspar, alumina and silica by using high sintering temperature. The application of high sintering temperature is the main factor that helps in the formation of crystallization phase in the ceramic bodies. Porcelain materials have good properties for applications in many industries and they usually made up from the mixtures of kaolin, flux feldspar and quartz [1]. In recent years, the study on the properties of ceramic porcelain has been widely conducted in order to develop high density ceramic porcelain while having excellent mechanical strength, high electric insulation and good thermal properties [2]. The issue of low strength ceramic porcelain which led to the formation of crack after being exposed to environment has gained interest from researchers to further their studies in this field.

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Previously, the production of porcelain was made by mixing the raw material with quartz. However, researchers have found out that the presence of quartz in porcelain insulator will be the source of micro crack when the production was not carry out properly [3]. Therefore, alumina has been chosen as material that is suitable to be partially substituted with quartz. Alumina carries good electrical and mechanical properties and at once can avoid any crystallographic changes during heating. Alumina also will increase the mullite content which acted as agent for increasing the strength. Moreover, alumina can increase the electrical resistivity and reduces the loss tangent to act as good insulator [4]. On the other hand, the amount of kaolin in the composition also plays role in the formation of the ceramic plasticity. Kaolin which acts as body former will provide the plasticity towards the ceramic thus can increase the density.

In this study, the influence of kaolin and alumina in ceramic porcelain will be further observed in order to develop good physical and structural properties ceramic porcelain. The effect of the increasing kaolin content and decreasing alumina content will be observed in every composition. On the other hand, the physical and structural properties of the ceramic porcelain also will be analysed.

MATERIALS AND METHODS

Raw Materials

Kaolin powder produced by R & M Chemical Sdn Bhd and feldspar powder from Sibelco Malaysia were dried in oven at 105 ± 5 °C for 24 hours. Characterization was done by using XRF (Bruker S2 Ranger) on feldspar and kaolin while XRD (Bruker D2 Phaser) was used to test for all raw materials which are feldspar, kaolin, silica and alumina. The raw materials were sieved using mesh sieve and weighed according to their composition which are 10 wt% of silica, 20 wt% of feldspar, 70-x wt% of alumina and x wt% of kaolin (with x = 30, 35, 40, 45, 50)] as shown in Table 1. Silica and alumina were obtained from Progressive Scientific Sdn Bhd.

Table 1. Composition of raw materials used and the formulation for each composition

Materials	Kaolin (wt%)	Alumina (wt%)	Feldspar (wt%)	Silica (wt%)
Molecular formula	$Al_2H_4O_9Si_2$	Al_2O_3	$Na_2OAl_2O_3{\cdot}6SiO_2$	SiO_2
Code				
$K_{30}A_{40}$	30	40	20	10
$K_{35}A_{35}$	35	35	20	10
$K_{40}A_{30}$	40	30	20	10
$K_{45}A_{25}$	45	25	20	10
$K_{50}A_{20}$	50	20	20	10

Ceramic Fabrication

Raw materials were homogeneously mixed using milling machine by alumina ball of 10:1 weight ratio for 30 minutes to reduce the particle size. The mixing process was repeated for 8-10 times to achieve homogeneity in particles size. Four drops of 5% polyvinyl alcohol (PVA) were used as a binder. Prepared raw materials were compacted with a constant holding time of 2 minutes by using hydraulic press machine with pressure load of 160 MPa for making testing pellets. The size of pellet die was 10 mm diameter. The samples were sintered at temperature of 850 °C for 4 hours.

Structural Properties

Samples were analysed with XRD by using Bruker D2 Phaser machine. The XRD pattern was scanned at angle of 10° to 90°.

Physical Properties

Bulk Density

The density of the samples was calculated by using water displacement technique using Archimedes principle. The sample was tested through water immersion technique by using density testing machine. The result also was calculated again by using Equation (1).

$$\text{Density, } \rho = Ma/(Ma-Ml)\ \rho o \quad (1)$$

With, Ma = sample mass in air, Ml = sample mass in liquid and ρo = density of liquid (1 g/cm3)

Water Absorption

The water absorption of the sample was measured as a function of the sample's weight difference in air and in liquid. The sample was weighted before and after being immersed in liquid water. The water absorption test was computed by using Equation (2).

$$\text{Water absorption} = (Wl-Wa)/Wl \times 100 \quad (2)$$

With Wa = weight in air and Wl = weight in liquid

Firing Shrinkage

The diameter changes of the samples were taken and the results were used to determine the firing shrinkage after sintering at 850 °C. The firing shrinkage was determined by using Equation (3).

$$\text{Firing shrinkage} = (Lb-La)/Lb \times 100 \quad (3)$$

With, Lb = diameter before sintering and La = diameter after sintering

RESULTS AND DISCUSSION

Figure 1 shows the diffraction patterns for the effect of kaolin-alumina ratio analysed by using XRD. The diffraction patterns revealed the different phases present due to the increasing amount of kaolin and decreasing amount of alumina in each sample. The diffraction patterns revealed the presence of corundum (COD 9008094), cristobalite (COD 1010954), mullite (COD 705575) and albite (COD 9000681) phase.

The XRD results show that the K30A40 sample which contains 30 wt% kaolin and 40 wt% alumina appears in amorphous phase. The diffraction pattern shows large amorphous hump and no crystalline peak for the samples. This means that the mixture of raw materials in the sample do not starts to crystallize at 850 °C. This is also occurs due to the insufficient amount of kaolin and high amount of alumina does not provide enough concentration for crystallization to occur during sintering of the sample. However, the addition of 5 wt% of kaolin in every sample has created the formation of crystalline phase. The addition of kaolin has contributed to the formation of crystalline phase such as corundum and mullite. The corundum phase proved that the dissolution of Al_2O_3 has been occurred results from the raw material such as alumina and kaolin.

The XRD patterns starts to shows crystalline phase from the addition of kaolin starting from 30 wt% to 50 wt% in all compositions. The diffraction pattern also shows the formation of mullite which presence only in small peaks due to the low sintering temperature used. As for the corundum, it was produced from the crystallization of Al_2O_3 after undergo sintering process. The XRD patterns for K35A35 shows corundum as the major peaks intensity among other peaks at angle $2\Theta = 25°, 35°, 43°$ and 57°. This occurs as the content of alumina (35 wt%) in the sample is sufficient to contribute in the formation of corundum phase. At the angle of 35°, the peak for K35A35 is slightly broad compared to other samples. The broader peak indicates that the amount of alumina used is higher for the formation of corundum. The peaks continued become narrow with the decreased of alumina for the other composition.

The formation of mullite phase is not quite significant due to insufficient sintering temperature used for kaolin to crystallize into mullite phase. The amount of mullite phase only increases with increasing of sintering temperature.

The insufficient sintering temperature of the sample makes the kaolin failed to transform completely into mullite phase. On the other hand, albite phase also shows some small and sharp peaks centered at 2Θ = 27°. The presence of some albite peaks indicates the crystallization of feldspar is not done completely during sintering process. This occurs may be due to the insufficient of sintering temperature used which is 850 °C. As for the formation of cristoballite phase from silica, it depends on the sintering temperature and increases with increasing in temperature. For this case, the low temperature used for sintering is insufficient to form high cristobalite phase resulting in formation of small peak at 2Θ = 21°.

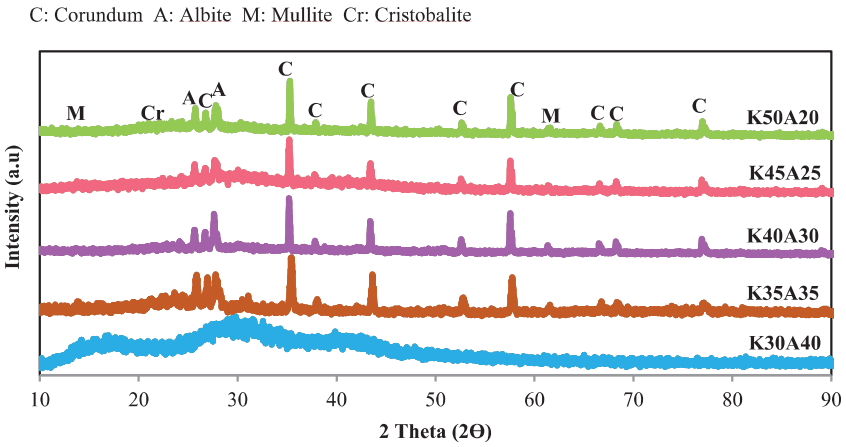

FIGURE 1. XRD patterns for samples with different amount of kaolin and alumina

The effect of kaolin-alumina ratio on the bulk density is shown in Fig. 2. The result shows that the addition of kaolin in the sample gives an impact towards density where the density value experienced slight reduction with increasing amount of kaolin and decreasing amount of alumina. The density also influence by the insufficient sintering temperature which is 850 °C. K30A40 sample with 30 wt% of kaolin produced the highest density which is 2.85 g/cm^3. As for K35A35 sample which contains 35 wt% of kaolin, the density reduced slightly towards 2.72 g/cm^3. The density of the sample continued to decrease for K40A30 sample which is 2.70 g/cm^3 and rapidly reduced with increasing amount of kaolin for K45A25 sample and K50A20 sample which are 2.51 g/cm^3 and 2.49 g/cm^3 respectively.

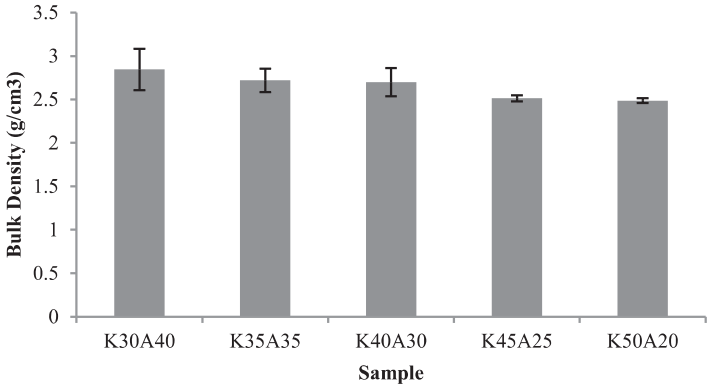

FIGURE 2. Bulk density of all samples as a function of kaolin addition and alumina reduction

Figure 3 shows the percentage of water absorption of sample with different amount of kaolin and alumina. The K30A40 sample showed the lowest percentage of water absorption which is 35.18%. The percentage of water

absorption slightly increases for the other samples as a function of the addition of kaolin and reduction of alumina started with 36.66%, 36.93%, 39.87% and 40.62%.

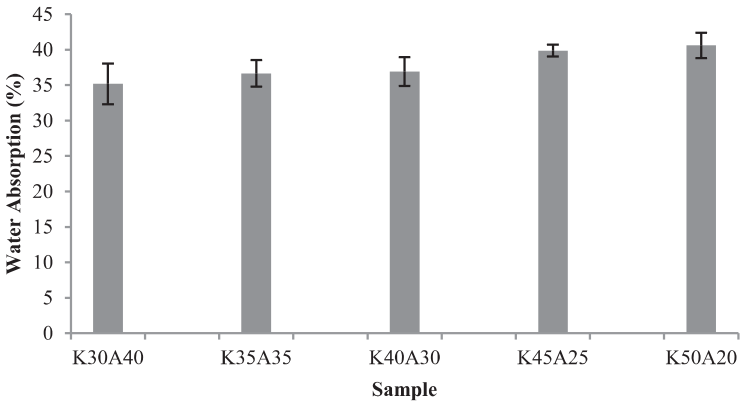

FIGURE 3. The percentage of water absorption of all samples as a function of kaolin addition and alumina reduction

The addition of kaolin in the sample has increased the percentage of water absorption. The water absorption increased according to open porosity in the sample structure. The alumina content in the sample help to form liquid phase during sintering process and fill up the open pores. The decreasing amount of alumina in each sample has contributed to the increased in percentage of water absorption.

The percentage of water absorption also was affected by the insufficient sintering temperature of the sample which is 850 °C. According to Mehta et al [2], water absorption decrease with the increase of sintering temperature as the close pores and open porosity being reduced while increasing the amount of glassy phase in the sample. However, the insufficient amount of feldspar that acts as flux in the composition also can be the reason towards the increase in the percentage of water absorption. Feldspar acts as fluxing agent which reduces the melting point of the mixture. The low amount of feldspar does not promote the densification of the ceramic green body thus increasing the percentage of water absorption.

Figure 4 shows the percentage of firing shrinkage values of samples prepared with influence of different kaolin and alumina content sintered at temperature of 850 °C. The percentage of firing shrinkage increases with the addition of kaolin and reduction of alumina content. The K30A40 sample shows the lowest percentage of firing shrinkage which is 1.99%. As the content of kaolin increase, the percentage of firing shrinkage rises from the K35A35 sample until the K50A20 samples which are 2.37%, 2.41%, 2.79% and 2.98% respectively.

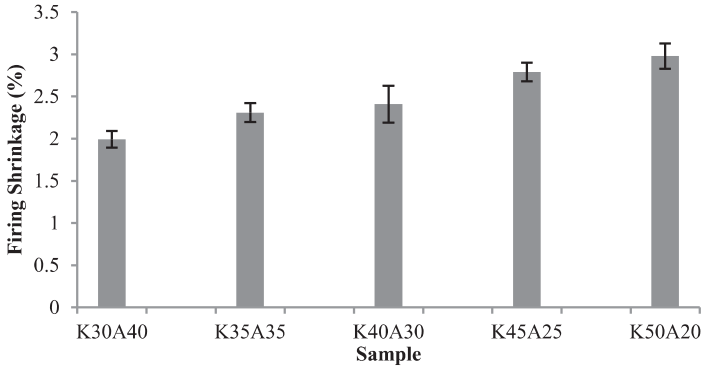

FIGURE 4. The percentage of firing shrinkage of all samples as a function of kaolin addition and alumina reduction

The amount of kaolin affected the firing shrinkage of the samples. The drying shrinkage refers to the degree of plasticity that provides by the kaolin itself. Firing shrinkage indicates how the mixture fused together and increasing

the sample shrinkage with increasing the kaolin content [5]. On the other hand, reducing the amount of alumina also contributed to the increases of the firing shrinkage of samples. This is because low amount of alumina will form low mullite phase which will decreased the effect of impurity phases by forming solid solution during sintering process process.

Based on the composition used, it can be seen that K35A35 sample which contained 35 wt% kaolin and 35 wt% alumina is the most preferable compared to other samples. The equal amount of kaolin and alumina in the sample produced good physical and structural properties of ceramic porcelain. The high amount of alumina also helps in formation of liquid phase which will fill up the open pores in the ceramic body thus enhancing the physical properties.

Same goes to the amount of kaolin which is 35 wt%, the concentration of kaolin in the sample as body former has provide a good structural properties for the sample. On the other hand, the 10 wt% of silica used also contributed in maintaining the strength of ceramic porcelain. Previous researcher mentioned through their work that the increasing amount of silica up to 15% has decreased the strength of ceramic porcelain due to non-bonding additional free silica content that results in the formation of closed pores [2].

Looking up at the XRD results for K35A35 sample, the high amount of corundum phase in the sample is due to high concentration of alumina used. The high amount of corundum helps in increasing the strength of ceramic by filling up the open pores. This is because corundum will form liquid phases which can fill up the open pores in the ceramic structure.

The relationship between bulk density and firing shrinkage is shown in Fig. 5. The bulk density of the samples decreased with the addition of kaolin and reduction of alumina content. On the other hand, the percentage of firing shrinkage shows an increase when the amount of kaolin being increase and alumina decrease. As the density being decrease with the addition of kaolin, the firing shrinkage increase as a function of the degree of plasticity provides by kaolin. Kaolin acts to provide plasticity in the sample which causes the samples to experience more shrinkage during sintering process. Kaolin will act as binder upon introduced with high temperature. However, the reduction of alumina content has reduced the formation of liquid state which can fill up the open pores thus has resulted in the decrease of sample bulk density.

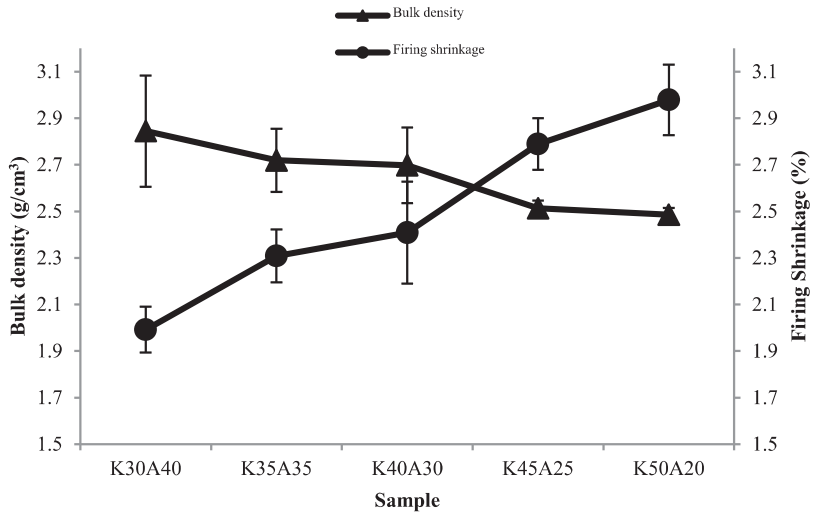

FIGURE 5. Graph between bulk density and firing shrinkage of all samples as a function of kaolin addition and alumina reduction

CONCLUSION

In this study, ceramic porcelain was prepared by using different composition of kaolin and alumina and an optimum amount of silica and feldspar. The composition with 35 wt% kaolin and 35 wt % alumina sintered at 850 °C for 2 hours found to be the most preferable ceramic porcelain with good physical and structural properties. In the above composition with increasing amount of kaolin and decreasing amount of alumina, the bulk density decreases due to low amount of liquid phase produced by alumina which can fill up the open pores. Increasing amount of kaolin also makes the ceramic particles tend to fused together and undergo high percentage of firing shrinkage due to plastic behaviour of kaolin. Upon reduction of the amount of alumina, water absorption of the ceramic increased due to insufficient amount of alumina needed to reduce the open pores. The formation of different phases such as corundum, mullite, cristobalite and albite found in the diffraction patterns of XRD have led toward the significant changes in the characteristics of the samples, including reducing of bulk density, increasing of water absorption and firing shrinkage.

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