Characterisation of Silica-Alumina Ratio in Ceramic Porcelain Materials

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Abstract. Ceramic porcelain were fabricated with different quantities of alumina-silica (from 0-50 wt% silica) with fixed amount of feldspar (25 wt%) and kaolin (20 wt%). These ceramic porcelain samples were sintered at 900 °C for 4 hours. The effect of different addition ratio of alumina-silica on the structural and physical of the ceramic porcelain materials were assessed by X-ray diffraction, density, apparent porosity, water absorption and firing shrinkage. The results shown that the XRD diffraction pattern exhibited good phase changes in sample A35S15. The water absorption rate were optimum in this composition and the density is 9.76 % gcm-3 and 1.9 % of apparent porosity. Based on the results, the structural and physical properties of ceramic porcelain with 35 wt% alumina and 15 wt% silica (A35S15) is identified as ideal composition for ceramic porcelain industry.

INTRODUCTION

Ceramic porcelain materials are widely used in high temperature applications and wear resistant materials. Ceramic porcelain material are made from natural raw materials such as feldspar, kaolin, silica and alumina. 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]. Previously, the production of porcelain were 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 carryout properly [2]. Therefore, alumina has been chosen as material that is suitable to be partially substituted with quartz. Clay such as kaolin is used as essential material for porcelain meanwhile kaolin possesses different physical and chemical properties depending on geophysical and geological environment. The main constituent of ceramic porcelain materials are clay (as plastic material), fillers (such as alumina and silica) and feldspar ($Na_2OAl_2O_3.6SiO_2$) which acts as flux material [3]. The filler material plays a vital role in ceramic porcelain material to provide additional strength by filling the pores of ceramic body and insulator such as silica which is unreactive at low temperatures and forms highly viscous liquid at very high temperature of sintering.

The motive of the present work is to prepare a mechanically strong ceramic porcelain material by mixing an optimum amount of low cost clay, kaolin and feldspar (up to 50 wt.%) and remaining substituents by silica and alumina to reduce the cost of raw materials. In this study, we increased the silica content and decrease the concentration of alumina in base ceramic porcelain composition. Alumina-based ceramics have high strength, low thermal conductivity as compared to other ceramics [4]. The influence of alumina and silica in ceramic porcelain were observed in order to develop good physical and structural properties ceramic porcelain. The effect of the increasing alumina content and decreasing silica content were observed in every composition. On the other hand, the physical and structural properties of the ceramic porcelain also analysed.

International Conference on Bioengineering and Technology (IConBET2021)
AIP Conf. Proc. 2454, 060060-1–060060-6; https://doi.org/10.1063/5.0079142
Published by AIP Publishing. 978-0-7354-4193-4/$30.00

MATERIALS AND METHODS

Materials Preparation

The base composition for developing a ceramic porcelain material is prepared using 25 wt.% of feldspar, 20 wt.% of kaolin, with varying wt.% of alumina (Al_2O_3) and silica (SiO_2) contents as shown in Table 1. Due to hydroscopic nature of raw materials, the materials are placed in oven at temperature of 105 °C for 24 hours to remove moisture content present. Kaolin powder produced by R & M Chemical Sdn Bhd and feldspar powder from Sibelco Malaysia. Characterization was done by using XRF on feldspar and kaolin while XRD was used to test for all raw materials which are feldspar, kaolin, silica and alumina. Afterwards, the prepared compositions of ceramic porcelain material were mixed together with yttrium stabilized zirconia balls of 10:1 weight ratio in a milling bottle. The compositions were mixed for 30 minutes in ball mill to reduce the particle size. To obtain the homogeneity in particle size, the mixing and milling procedure was repeated up to 8-10 times. Six drops of 5% polyvinyl alcohol (PVA) was used as binder in prepared composition and again mixing was continued for 30 minutes. The prepared compositions were compacted by hand press machine. 0.2g of each composition was hand pressed with 160 MPa.

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

Materials	Feldspar (wt%)	Kaolin (wt%)	Alumina (wt%)	Silica (wt%)
Molecular formula	$Na_2OAl_2O_3 \cdot 6SiO_2$	$Al_2H_4O_9Si_2$	Al_2O_3	SiO_2
Code				
A50S0	25	20	50	0
A40S10	25	20	40	10
A35S15	25	20	35	15
A30S20	25	20	30	20
A25S25	25	20	25	25

Sintering Process

The prepared samples with different compositions were placed in furnace to perform sintering process. The samples were sintered at 900 °C for 4 hours. When 900°C is reached, the atmospheric water present on the ceramic body was completely evaporated. Sintering is an effective process where porosity (open space), is removed from compacted powder particles to form a solid mass. The present materials move to the contact points between particles and fills in the open space.

Characterisation

The density test were carried out using liquid density meter. The further results were utilized in calculation apparent porosity and bulk density using MS ISO 10545-3:1995. The raw materials of ceramic porcelain which are kaolin, feldspar, alumina and silica were analysed through X-Ray Diffraction method and X-Ray Fluorescence method. The ceramic porcelain samples prepared were undergone XRD test using Bruker 2D Phaser.

Water Absorption Method

Water absorption test was carried out for the compositions using standards MS ISO 10545-3:1995 that shown below. Determination of apparent porosity and bulk density provides the means to classify ceramic porcelain with their method of manufacture according to the standards.

Equation used to calculate Water Absorption:

$$E\ (b, v) = \frac{m2(b,v)}{m1} - \frac{m1}{m1}\ x100 \quad (1)$$

Where, m1 : mass of dry pellet
m2 : mass of wet pellet

Equation used to calculate Apparent Porosity:

$$p = \frac{m2v}{V} - \frac{m1}{V}\ x\ 100 \quad (2)$$

Where, m1 : mass of dry pellet
m2 : mass of wet pellet
V : volume

Equation used to calculate Bulk Density:

$$B = \frac{m1}{V} \quad (3)$$

Where, m1 : mass of dry pellet
V : volume

RESULTS AND DISCUSSION

The composition of kaolin was analysed using X-Ray fluorescence instrument and the obtained results shows that kaolin containing 63.8% SiO_2 and 33.7% Al_2O_3. Based on the proximity of silica and alumina compositions obtained, there are still contains of impurities such as Fe_2O_3, K_2O, MgO, Na_2O, and TiO_2. Meanwhile, feldspar containing SiO_2, K_2O and Al_2O_3 with 70%, 17.5% and 11.2% approximately. There are also few impurities detected alongside the oxides which are CaO, Fe_2O_3 and Na_2O.

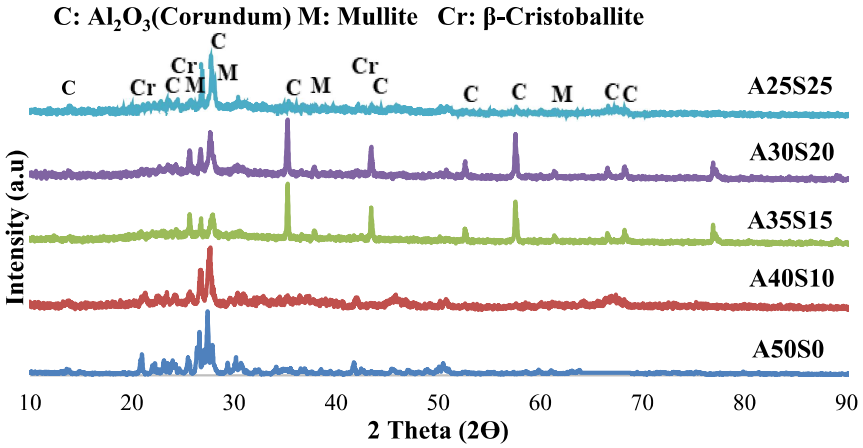

FIGURE 1. XRD patterns for samples with different amount of alumina and silica

The XRD pattern of composition with increasing concentration of silica and decreasing concentration of alumina are plotted in Fig. 1. The diffraction patterns revealed the presence of corundum (COD 9008094), cristobalite (COD 1010954) and mullite (COD 705575) phase. The XRD results show that the A35S15 and A30S20 sample which contains 35 wt% and 30 wt% alumina appears in high crystalline phase compare to other samples when refres to the

intensity of the peak. As the concentration of silica increases and concentration of alumina decreases, it was observed the crystallization of sample is increased as the intensity of peak increased. However, with highest silica in the composition A25S25, the intensity start to decreased. This is might due to the higher silica cause the low ability of the samples to have fully sintered and the melting of silica is higher. The major peak intensity is corundum and valued at (2θ) of 27.68°.

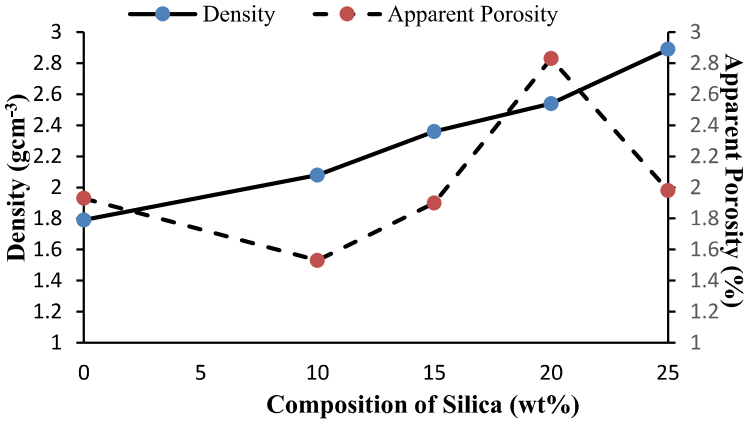

FIGURE 2. Graph between density and apparent porosity of ceramic porcelain with increasing concentration of silica

Figure 2 shows the value of bulk density of ceramic porcelain with decreasing concentration of alumina and increasing concentration of silica. As the concentration of silica increases from 0 % to 25 %, the bulk density of the ceramic porcelain increases from 1.79 to 2.89 g/cm^3. However, the apparent porosity shows insignificant results with become lower at 10 wt% silica and then increased with increasing silica but then decreased at the highest silica content. We can relate that the lower value at high silica to the XRD of this samples which shows that samples having low crystalline value compare to samples 15 and 20 wt% silica. Low crystallization means that samples are not fully crystalline and thus appears to have high porosity. This shows that the difference in apparent porosity value will be resulting in different surface microstructures. The insignificant results of apparent porosity shows that the apparent porosity is not affected by the composition of alumina-silica ratio. Based on the graph, the maximum apparent porosity is 2.83 % at A30S20 and the minimum porosity is 1.53 % in sample A40S10. Porosity increased with decreased of average pore diameter and vice versa but it must be taken to note that porosity also depend on the impurities present in the raw materials of ceramic porcelain. The application of high sintering temperature is the main factor that helps the formation of crystallization phase in the ceramic bodies and the sintering process also enhanced the inner structure by closing the pores present, thus reduced the porosity.

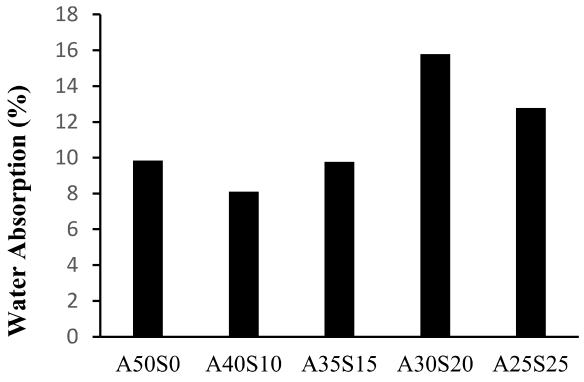

FIGURE 3. The percentage of water absorption of all samples as a function of silica addition and alumina reduction

Figure 3 shows the rate of water absorption and composition of increasing silica concentration. The graph shows that samples A30S20 gave the highest percentage of water absorption with 15.79% meanwhile A40S10 gave the lowest percentage of water absorption, 8.11%. This water absorption are relate to the apparent porosity by giving the same insignificant trend. The water absorption and apparent porosity are related to each other as can be seen from the equation 3.1 and 3.2. The insignificant trend for both properties are also due to the crystallization of the samples from the different ratio between alumina and silica used. Higher alumina will have formation of liquid phase, meanwhile higher silica used, the sintering temperature need to be higher as silica have higher melting temperature.

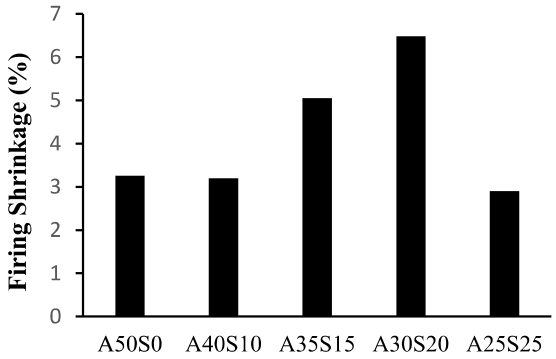

FIGURE 4. The percentage of firing shrinkage of all samples as a function of silica addition and alumina reduction

Figure 4 shows the percentage of firing shrinkage decreases from composition with alumina 50 wt%, silica 0 wt% (A30S0) to composition with alumina 40 wt%, silica 10 wt% (A40A10) from 3.26% to 3.20%. Increasing of silica then shows that the percentage of firing shrinkage increases from composition with alumina 40 wt%, silica 10 wt% to composition alumina 30 wt%, silica 20 wt% from 3.20% to 6.48%. However, the percentage of firing shrinkage dropped to 2.90% in the composition with alumina 25 wt%, silica 25 wt% (A25S25).

Firing shrinkage of ceramic will influence and impact accuracy of the composition of ceramic porcelain and the sintering temperature. Because of ceramic products that contain different composition has varying pore in size and shape, firing shrinkage percentage of ceramic porcelain will differ. Based on the composition used, it can be seen that A35S15 sample which contained 35 wt% alumina and 15 wt% silica is the most preferable compared to other samples. The optimum amount of alumina and silica 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 silica which is 15 wt%, the concentration in the sample 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 [5].

Looking up at the XRD results for K35S15 and K30S20 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. K50S0, sample with 0 wt% of silica is a based structure, and we can see that the intensity of peak are lower than other samples.

CONCLUSION

The effect of different ratio of alumina-silica concentration on ceramic porcelain structural and physical properties was discussed. Ceramic porcelain has been prepared using alumina-silica as mullite ceramics as well as its influence on the porcelain properties after sintering, and their structural and elemental analysis have been studied. In this study, ceramic porcelain was prepared by using different composition of alumina and silica and an optimum amount of kaolin and feldspar. The composition with 35 wt% alumina and 15% wt % silica sintered at 900 °C found to be the most preferable ceramic porcelain with good physical and structural properties. In the above composition with increasing amount of silica and decreasing amount of alumina, the density increases due to low amount of liquid phase produced

by alumina which can fill up the open pores. 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 and β-cristobalite found in the diffraction patterns of XRD have led toward the significant changes in the characteristics of the samples, including bulk density, water absorption and firing shrinkage.

For further study, the research can be conducted by increasing the sintering temperature by 1200 °C – 1300 °C to enhance the crystallization of raw materials because ceramic porcelain sintered at high temperature will results in production of good physical and structural properties. In additional, the usage of additive such as Fe_2O_3 also can be used to reduce the sintering temperature of ceramic porcelain.

ACKNOWLEDGMENTS

This work was supported by a grant from Universiti Malaysia Kelantan (R/SGJP/A1300/00881A/001/2019/00584).

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