Preparation of Ceramic Support from Waste: Effect of Sintering Temperature

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Abstract— A structure of ceramic support was fabricated from broken ceramic waste. The ceramic wastes which contain high source of alumina and silica were initially crushed into powders. The powders were mixed with binders and formed into cylindrical pellets. The preparation of the support was carried out based on press molding or compaction technique at a pressure of 160MPa. In this study, the effects of sintering temperatures were investigated by evaluating the phase characterization, porosity properties of the supports, average pore size and microstructure of the sintered samples. Sintering temperatures were carried out in a range of 900 to 1200°C with an interval of 100°C for 2.5 hours. The evaluation was carried out through the use of X-ray diffraction (XRD) and scanning electron microscopy (SEM). Based on the analysis, major crystalline phase of the sintered samples was mullite. However, fewer peaks were detected at 1200°C resulting to a decrease in the mullite crystal phase. Results from sintering showed that, with increasing of temperature, open porosity showed a decrease trend in the sintered samples from 45.32% to 14.67% at 1200°C. Also, with increasing sintering temperature, average pore size increases with a maximum average of 17.04 µm at the final sintering temperature of 1200°C. On the other hand, SEM observation on microstructure of the sintered samples showed higher densification at 1200 °C compared to the other temperatures.

Keywords— Ceramic Waste, Support, Sintering Temperature, Mullite

I. INTRODUCTION

Nowadays, with strong environmental demands, ceramic membrane has seen to be leading applications for industrial separation processes such as filtration. This is due to their unique advantages such as excellent productive life, thermal and mechanical resistance, good chemical stability and hardness. These qualities are among the essential properties which have been attracting much attention in established researches as of late [1]. The ceramic membrane mainly consists of three structural layers known as top layer, intermediate layer and support. The support gives a mechanical strength to the top most membrane layer and it is also responsible to provide an integrated structural for the membrane [2]. Most supports are made of inorganic materials such as alumina, titania and zirconia. A ceramic support structure is coated with an intermediate layer to reduce roughness and pore size which then allows the thin top layer to be structured. A typical ceramic support is established largely by controlling the pore size and sintering conditions [3]. These factors are needed to be given attention as it is to determine the performance of a ceramic support. However, the fabrication of the ceramic supports tends to have costs issues because of the expensive raw materials.

As a result, the interest on preparing ceramic membrane supports from utilise recycled waste material has grown. A wide range of materials are said to be the potential usable waste forms include coal fly ash, construction waste and sanitary ware [4]. These utilizations will not only lower the cost of abundant raw materials but also reduced the environment pollution in terms of pollutants. For example, coal fly ash coming from combustion of raw coal in thermal power plants is a source of dust that can resentfully cause air pollution and affects the environment. A research done on the preparation of a porous ceramic membrane support from the coal fly ash has proven to contain enhanced porosity and permeance [5]. The benefit of the utilization also includes the production of high added value products.

A process of manufacturing ceramic is based on sintering which is a heat treatment process. This is because ceramics melt at a high temperature. Sintering process of ceramics will alter and affects several changes and properties of the product. Variables or properties that will be affecting from sintering are strength, hardness, thermal conductivity, average grain number, size and shape, crystal structure, porosity and several other changes.

On the other hand, as a structural material, fabrication of ceramic support is crucial on producing or having mullite crystalline phase on its production. This is because mullite is a promising and the only stable crystalline aluminosilicate phase in the SiO₂-Al₂O₃ binary system. This is due to its minerals which very apt to high temperature including high melting point, chemical stability, low thermal expansion coefficient and corrosion resistance. However, mullite phase of crystallization is difficult to control with the effects of temperature. This is because the phase composition is strongly dependent on the thermal treating temperature to a large extent. The structural application of mullite is depending on the mullite's temperature stability and refractory nature [6]. Previous works observed that alumina mullites are difficult to produce when the alumina properties, quantity and purity are insufficient [7]. Therefore, the mullite phase composition of these supports is expected to have an effect from the sintering temperature.

This study is focusing on two objectives which are to prepare a ceramic support by utilizing waste and determine the effects of sintering temperature on the characterization of support by evaluating the phase composition, porosity, average pore size and microstructure from the effects of sintering temperature.

II. METHODOLOGY

A. Starting Materials

Ceramic waste from a glove manufacturing company in Meru, Klang was used as the starting materials. The raw materials were molds known as formers which were broken and no longer has its function in the manufacturing process. The waste collected was as big as hand-shaped molds. It was then crushed in a former crusher to form powdered size waste. The powder was further sieved using Sieve Shaker (ENDECOTTS Octagon 2000 Digital) to obtain waste with powder particle less than 125µm.

B. Compaction and Sintering

The prepared ceramic waste powder was mixed with PVA-1750 (5wt% solution) which acts as a binder in a laboratory mixer for 1

hour. The mixture then undergoes uniaxial pressed moulding techniques in a 25 tonnes compression machine at a pressure of 160MPa forming cylindrical pellets (30mm in diameter and 0.5-1.0 mm in thickness). The cylindrical pellets were dried in a laboratory oven for 24 hours at 110°C. This is to eliminate the presence of water from the samples.

After sufficient drying, the samples were sintered at various temperatures for 2.5 hours ranging from 900 to 1200°C with an interval of 100°C. The firing was conducted in a programmable electrical furnace (Laboratory Box Furnace VT) with heating rate of 5°C/min from room temperature to 450°C and 3°C/min up to the final sintering temperature. A holding time of 1 hour was carried out at 450°C to remove the added binder. Figure 1 shows the process flow summary of ceramic support fabrication from the starting material until it undergoes the process of sintering.



Figure 1: Process flow of summary of ceramic support fabrication

C. Characterization Techniques

The characterization is emphasis particularly on raw materials and ceramic support properties such as chemical and phase composition, open porosity and average pore size. A detailed microstructural analysis was also performed to observe the sintering effects.

Raw materials were examined by an X-ray fluorescence spectrometer (XRF-1800, Shimadzu Corporation, Japan) in order to obtain their chemical composition. The phase composition of starting materials and sintered samples were characterized using XRD (Rigaku Int. Corp., Japan: Cu K α radiation, working voltage 40kV, working current 40mA, scanning speed of 5°2 θ /min. Porosity, average pore size and microstructure were analyzed by using scanning electron microscopy (SEM; Nova NanoSEM 430, Fei, Holland) and ImageJ.

III. RESULTS AND DISCUSSION

A. Characterization of Starting Materials

Table 1 lists the chemical composition of the starting materials characterized by quantitative X-ray fluorescence spectrum analysis. The ceramic waste mainly comprises of alumina (38.09wt %) and silica (57.17wt %). There is a small amount of titania (0.29wt %),

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magnesium oxide (2.00wt %) and calcium oxide (2.44wt %) in the starting materials. The initial mass of sample after compaction was 12.5g and decreases to 9.1g after firing. The mass loss is mainly due to the removal of structural water and organic substances of binder after firing.

Table 1: Chemical composition of the starting material						
The chemical composition (wt. %)						
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materials	The enermean composition (wa 70)				
	Al ₂ O ₃	SiO ₂	TiO ₂	MgO	CaO
Ceramic	38.09	57.17	0.29	2.00	2.44
waste					

Figure 2 shows the XRD pattern of the ceramic waste before sintering. It was seen that the mullite crystalline phase is the major phase and has already existed in the samples before sintering is done. Other minor crystalline phase present is quartz and corundum. The existence of mullite before sintering belongs to primary mullite which was formed from the first production of ceramic itself [8].



Figure 2: XRD pattern of ceramic waste. C: corundum, M: mullite, Q: quartz

B. XRD Analysis

Figure 3 indicates XRD patterns of the phase composition of samples sintered at different temperatures between 900 to 1200°C with an interval of 100°C. From the XRD analysis, it was found that mullite is the major crystalline phase of the sintered samples. At 900°C, the sintered samples consist of mullite characteristics peaks and weak characteristics of quartz and cristobalite. The XRD analysis observed has started to form an amorphous structure because of the sintering at this temperature. At 1000°C, only a single peak of quartz phase is seen. The decrease in quartz can be related to the phase transition of quartz to cristobalite. As seen in figure 3 at 1000°C, cristobalite was detected and the rest of the phase is mullite. On top of that, the peaks of mullite crystals grew as the temperature reach 1000°C. This indicates that the mullite undergoes a phenomenon known as secondary mullization [9]. At 1100°C, only mullite characteristics peak was found. This is due to the cristobalite contribution to the formation of mullite. However, peak characteristics of mullite started to disappear when sintering temperature exceeds 1100°C. This is probably due to the formation of anorthite and cordierite since the presence of CaO and MgO from the starting materials [10]. At 1200°C, only a few peaks with mullite and quartz phase were detected.

C. Open Porosity

Figure 4 shows the variation of open porosity on the sintering

temperature between 900 to 1200°C. As seen clearly from the figure, the porosity decreases with the increasing of sintering temperature. The porosity is a measure of empty spaces in the samples. At 900°C, the porosity is the highest at 45.32% and decreases upon increasing the temperature. The open porosity of the sintered samples at 1000, 1100 and 1200°C is 39.27, 29.70 and 14.96% respectively. This is associated to the principal objective of sintering which is the elimination of porosity. A complete elimination of porosity can only happen at the final stage of sintering [11]. All pores are connected, grain size started to increase and diffusion paths along grain boundaries becomes shorter. The small pores coalesce becomes larger in size and resulting to the loss of porosity.

D. Average Pore Size

Figure 5 represents the effect of sintering temperature on average pore size of the sintered samples. As can be seen from the figure, the average pore size of the sintered specimens increase as the sintering temperature increases. The growth of pore size with an increase in sintering temperature is due to the formation of well-developed necks [12]. The increases of pore sizes can also attribute to the densification of sintered samples [13]. The average pore diameters are 2.03, 2.67, 6.91 and 17.04 μ m for sintering temperatures at 900, 1000, 1100 and 1200°C.



Figure 3: XRD Pattern of sintered samples at various temperatures for 2.5 hours. C: corundum, M: mullite, Q: quartz, A: cristobalite



Figure 4: Open porosity of ceramic support with different sintering temperatures for 2.5 hours.

Figure 5: Average pore size of ceramic support with different sintering temperatures for 2.5 hours.

E. SEM Observation on Microstructure

Figure 6 shows the cross-sectional micrographs of the sintered samples at different temperatures measured by SEM. Undoubtedly, the temperature shows a significant impact on the structure and morphology of the obtained ceramic sintered samples. As the sintering gradually enhanced, the pore sizes were seen to become more obvious and grew larger. At 900°C, the structure was seen to have the presence of porosity contains higher than the other sintering temperature. On the other hand, at 1200°C, an obvious densification is observed. The increase was anticipated to decrease in open porosity as discussed in Figure 4. The microstructure observation was an effect of the increase in sintering temperature that promotes the presence of vitreous phase correlated to the densification process [14].

Figure 6: Cross-sectional SEM micrographs of sintered samples at different temperature for 2.5 hours: (a) 900°C, (b) 1000°C, (c) 1100°C, (d) 1200°C.

IV. CONCLUSIONS

This study successfully demonstrated the preparation of a ceramic membrane support using ceramic waste of former by uniaxial pressing in a compression machine at a pressure of 160Mpa. By studying the effects of sintering temperature on phase composition, porosity, pore properties and microstructure, the following conclusions can be drawn:

 The major crystalline phase of sintered samples is mullite. Other minor phases are quartz and corundum. The mullite is seen to increase from 900 to 1100°C. However, the phase evolution occurred at temperature 1200°C and mullite started to decrease.

- (2) The support produced shows a decrease in open porosity with increasing sintering temperature. The sample sintered at 1200°C has the lowest open porosity which is 14.96%.
- (3) The average pore sizes increase with an increase in sintering temperature. At 1200°C, the average pore size is the largest at 17.04μm.
- (4) The microstructure of sintered samples observed that the densification of the structure becomes higher in increasing of temperature. Densification results to the decrease in porosity especially at the highest temperature of 1200°C.

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