The Effect of Different Binder Compositions in Fabricating Silica Foam (SiO₂) Via Replication Method

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ABSTRACT

Ceramic foams or reticulated porous ceramic are form highly porous ceramic with closed, fully open and partially interconnected structure of porosity and wide range of application of the catalyst, electrical conductivity, refractory insulation of furnaces, filtration, adsorption and separation. There are various conventional method in fabrication silica foam such as direct forming, steam heating, freeze casting method and polymeric sponge method which is also well-known also as replication method. In this study, silica foam was fabricated using a 55wt. % of silica powder with different binder compositions of polyethylene glycol (PEG) and carboxymethyl cellulose (CMC) via replication method. The present work focuses in the effect of three variable compositions of PEG and CMC at high sintering temperature of 1250ºC in producing silica foams. The polyurethane foam (PU) acts as the template for slurry from silica powder compounds and mixed together with binders and distilled water. All samples are immersed into the SiO_2 slurry and dried at a temperature of 100ºC for 24 hours. The green bodies of foams were further sintered at temperature of 1250ºC. The morphological analyses depicted that the size of the silica foam pores were in the range of 259.15µm to 559.83µm. It was found that the density of silica foam in the range of 0.4953 g/cm³ to 0.7170 g/ cm3. Whereas, the result of porosity percentage obtained from 61.38 % to 71.9 %. Density and porosity analysis shows that the density of foams increased with decreased porosity of the silica foam. Mechanical properties analysis with compressive strength give a result in a range of 0.1310 N/mm² to 0.2329 N/mm² respectively. Thus, the replication method adopted for the SiO_2 foam fabrication was proven a success.

Keywords : *Silica foam, Porous Silica, Replication Method*

Introduction

Ceramic foams are categorised as porous and brittle materials that consist a cellular structure of three dimensional strut. As defined by the characteristics of the pores they are either closed pore, fully open pore, or partially interconnected porosity and the pore is comprised of cells with size ranging starts from 10µm until in a size of 5mm [1,2]. Ceramic foams are known to have applications in the diverse area such as supports for catalytic reaction, filtration, thermal insulation, impact absorbing structures, high specific strength materials and preforms for metal ceramic composites, biomedical implants, high-efficiency combustion burners and industrial process [3,4]. However, the performance of porous ceramics or ceramic foams mainly depends on the properties of the porosity, pore size, morphology and distribution. These properties are dependent on the processing steps involved during the fabrication of the ceramic foams [5].

There are various method that has been used to produce silica foam such as steam heating, direct foaming, freeze casting and polymeric sponge technique or also known as replication method. Previous studies reported that processing of silica foam using steam heating and its characterization showed that the silica content of foaming with interconnecting porosity up to 90% can be successfully processed [3]. Besides, the fabrication for open and closed pore structure are particularly suitable for direct foaming technique with a result porosities obtained in the range 45% to 97% as a previous study [4]. In addition, freeze casting method are also usually used in producing a porous ceramic body with a certain analysis of morphology and scale but a problem often occurs in this process are the low strength of the green bodies. Thus, this situation can cause the process is difficult to handle because the green bodies a very fragile when the frozen suspension is volatilized [6]. Besides, the most conventional method to produce ceramic foam is the polymeric sponge method or the replica technique where this method is involved an impregnated process of a polymeric sponge that is coated with a ceramic slurry then through the drying of the samples and sintering process with a high temperature. The sintering temperature also play a key role in effect of physical and mechanical properties [7]. The previous research stated that 1250ºC is the suitable sintering temperature in fabrication of ceramic foams due to the highest strength and density [8].

The replication method plays a key role in producing a highly open porosity due to the outstanding characteristic of this method that can control the pore size and complex shapes for different applications by using polyurethane as a template [8, 9]. Moreover, the replica technique is indeed a well-established method for producing cellular ceramic or porous ceramic consisting open cells up of to 90% achievable. In addition, this method is simple in which consist of immersion the polyurethane into a slurry and binder, removal of excess slurries followed by the drying and finally sintering process [10]. Therefore, the quality of ceramic slurry that is coated in the polymeric sponge is strongly dependent on the viscosity of the slurry and the density of the sponge [11]. The previous study reported that the slurries can be of variable solid weight percent usually in a range of 50 wt. % until 70 wt. % of solids [12].

In the present work, the SiO_2 foam with the composition of 55 wt. % and three different compositions of binder (PEG and CMC) was fabricated by replication method. Whereas, in this method a cylindrical PU was used as the template for the slurry. Therefore, this paper highlights on the fabrication of silica foams by applying different binder compositions of PEG and CMC and its effect on physical and mechanical characteristics of SiO_2 foams.

Materials and Methods

The main raw materials in this study is a silica (SiO_2) powder obtained from Sigma Aldrich, United Kingdom with a composition of 55 wt. %. The binder (Polyethylene glycol) is obtained from Merck Milipore, Germany and carboxymethyl cellulose was purchased from Multifilla, Malaysia. Binders (PEG and CMC) were dissolved in water. Different binder mixture of 3 wt. %, 5 wt. % and 7 wt. % are mixed together with SiO_2 in distilled water to form slurries. PU foam was selected to acts as a template of the slurry for replication method.

Sample Preparation

The polyurethane as a template was cut in a cylindrical shape with the dimension of 13mm × 26mm. PEG and CMC were first dissolved in a distilled water, as according to the varied pre-determined composition. SiO_2 was then added and stirred using a mechanical stirrer (RW 20 Digital, IKA, German). The polyurethane is then immersed in the prepared SiO_2, PEG and CMC mixtures for about 10 minutes. The impregnated polyurethane was taken out and squeezed out using two parallel plates to remove excess slurry. Drying was performed 24 hours for 100ºC in a drying oven is then followed by sintering process of 1250ºC with 2ºC/min of heating rate in a muffle furnace. Pore sizes and strut are determined by the morphology analyses using scanning electron microscope (SEM). Density and porosity analysis are determined by Archimedes principle using a Mettler Toledo. Mechanical behaviour of SiO_2 foam is determined by compressive test using Universal Testing Machine 10 KN. The fabrication the silica foam method is as summarised in Figure 1.
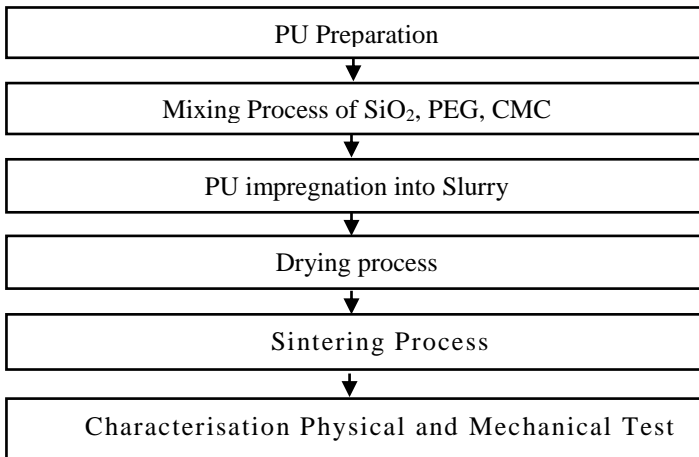
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│              PU Preparation                 │
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│      Mixing Process of SiO₂, PEG, CMC       │
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Figure 1: Summary of SiO_2 foam preparation process using replication method

Sample Characterisation

The morphology analyse of the SiO_2 foam includes the measurement size and pore shapes images obtained by Scanning Electron Microscopy (JSM-6700, JOEL, Japan). Density and porosity were identified adapting Archimedes Principles by ASTM C20-00 using analytical balances (XS6, Mettler Toledo,

Switzerland). The density and porosity were determined by the following equations:

$$\text{Apparent density} = \frac{W_d}{W_w - W_s} \qquad (1)$$

$$\text{Percentage of apparent porosity} = \frac{W_w - W_d}{W_w - W_s} \times 100 \qquad (2)$$

Where;

W_d = Total weight sample in dry condition
W_w = Total weight sample in wet condition
W_s = Total weight samples suspended in submerged condition

Strength of the SiO_2 foam were determined using the Universal Testing Machine 10 KN (AGS-X, Shimadzu, North America) in terms of compressive strength adapting the American Standard Testing Method, ASTM C773-88.

Results and Discussion

Morphology Analysis of Silica Foam

The green porous SiO_2 foam after sintering process for temperature of 1250ºC using three different binder compositions of PEG and CMC as shown in Figure 2.
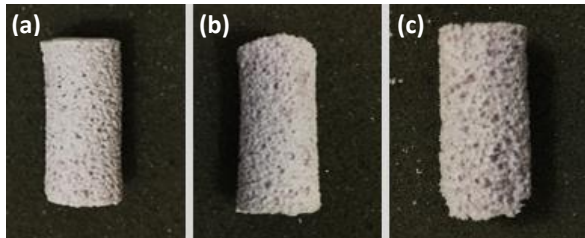
Figure 2 : Images of Silica foam samples using three different binders (PEG and CMC) (a) 3 wt. % (b) 5 wt. % (c) 7 wt. % , sintered at 1250ºC

It was found that type of porosity affects the size of the hole namely open and closed pores. As observed in the representative SEM image in Figure 3.
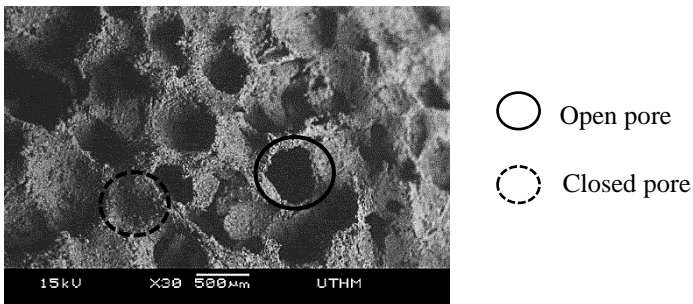

Figure 3: Microstructure SEM images shows the closed pore and open pore

The morpholgy analysis was conducted to find out the size of the porosity and microstructure of strut on the surface of each samples of the silica foam. The average pore sizes of SiO_2 foams are shown in Table 1.

Table 1 : Average size pores of SiO_2 foam

Silica (wt. %)	PEG and CMC (wt. %)	Average Size of Pore (μm)
	3	259.17 ± 29.08
55	5	306.17 ± 22.64
	7	559.83 ± 58.49

Besides, from the result obtained in Table 1, it can be observed that the highest composition binder gives the highest values sizes of the pore. This is due to the very thick slurry that did not fully adsorbed into the PU foam template. Moreover, the inhomogeneous mixture of the slurry causes the incomplete process of the combination powder particles does not combined well with each other so, the process of neck growth does not well-established during the sintering [13]. Moreover, the solid loading and the binders play a key role in controlling the viscosity of the slurry formed. Therefore, in a certain level amount of binder compositions in the aqueous slurry can give a result of increasing viscosity with increasing the solid loading [14].

The representative images in Figure 4 shows the microstructure pore through the SEM image of silica foam with different binder compositions. Based on the percentage composition of the mixture and binder to produce a slurry, the average pore sizes were found to increase with increasing binders (PEG and CMC) compositions. This is because the slurry was increased in viscosity. The increased viscosity would cause incomplete absorbance into the pore structure of polyurethane foam. Therefore, the stabilization of slurries can be determined based on the condition of the slurry. Besides, minimum solid loading would form a thin slurry and it would unable to impregnate polyurethane foam effectively [15]. On the other hand, if the solid loading is too high thick slurry would form caused the non-uniform impregnation polyurethane foam. Moreover, non-uniform impregnation can give a result with inhomogeneous impregnation particularly the inside structure of the template [11]. Therefore, the slurry composition is the most important parameter during the replication method process.
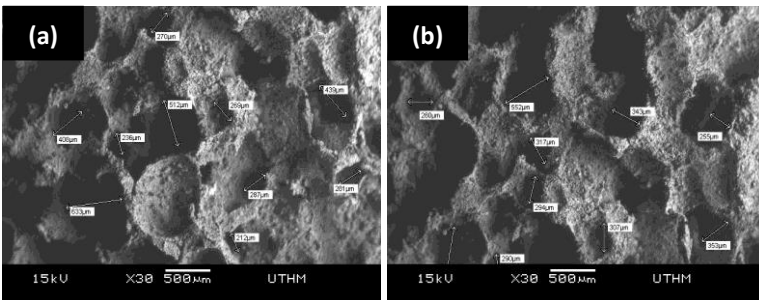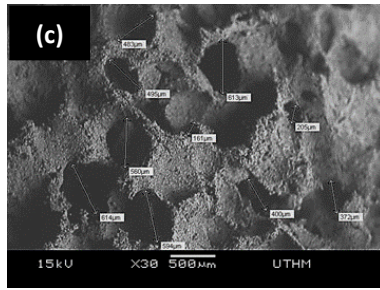
Figure 4: Microstructure pore through SEM image of silica foam with composition silica 55 wt. % and different binders (a) 3 wt. % (b) 5 wt. % and (c) 7 wt. %

Density and Porosity

The purpose of the density and porosity analysis is to determine the properties of the resulting density and total porosity percentage for each sample of SiO_2 foam with different binders PEG and CMC. In theory, the porosity is inversely proportional to the density. It was expected and proven that the resulting porosity correlated with the density. It can be observed that increasing the density yielded the lower percentage of porosity is produced [16]. Figure 5 shows the relations of apparent density and percentage of apparent porosity.
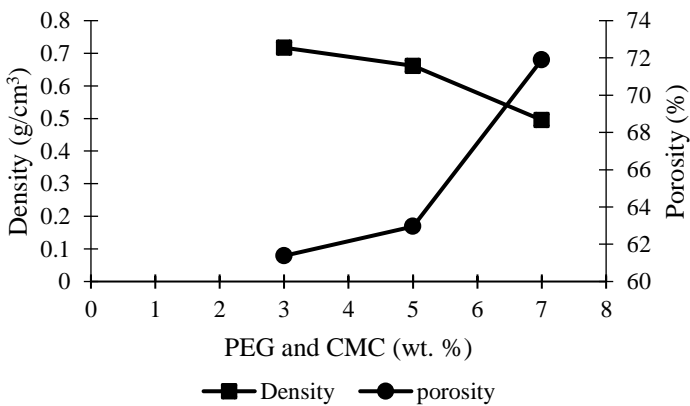

Figure 5: Apparent density and percentage of apparent porosity of SiO2 foam with the varied contents of binders (PEG and CMC)

It was observed that the total density decreased with increasing the composition of binders (PEG and CMC). Thus, the composition of 55 wt. % silica with a high binder composition of 7 wt. % with sintering temperature 1250°C showed the lowest result of density which is 0.4953 g/cm^3. Basically, low viscosity slurry with low compositions binder facilitates the slurry to enter the pore structure of PU as the template. However, when pore density is increased and reduction of pore size do not prevent a perfect and uniform coating around the polymer struts. Meanwhile, the increase of pore density leads to the production of a more relative density and less total porosity of the SiO_2 foam. Thus, with increasing the pore density, the strut in the SiO_2 foam become thinner [16].

In addition, based on observation in Figure 5, it shows that the total porosity obtained from silica foam with a sintering temperature 1250°C at the

lowest binder compositions of PEG and CMC gives a result of porosity 61.38% and the highest total porosity of 71.9% is obtained from the highest compositions of PEG and CMC which is 7 wt. %. This result shows that the compositions of binders (PEG and CMC) indeed influenced the density and total porosity. High viscosity slurry affected the impregnation process of the slurry to meet the structure of polyurethane foam [17].

Mechanical behaviour analysis

The mechanical behaviour for the sample is determined based on the compressive strength. Figure 6 shows the compressive strength against different binders of PEG and CMC.
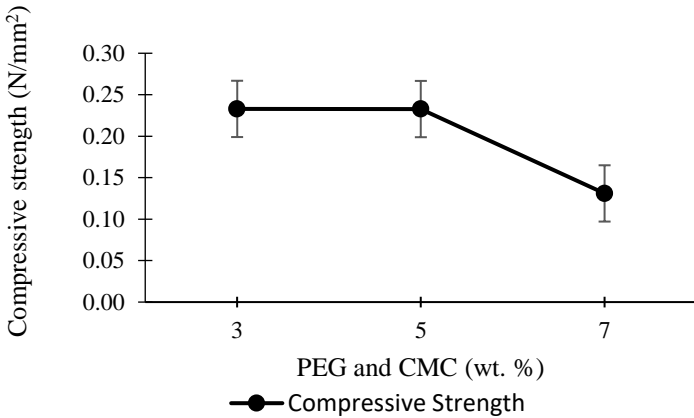

Figure 6: Compressive strength of SiO_2 foam with different binders PEG and CMC

The compressive strength at a composition silica 55 wt. % with 3 wt. % of binders (PEG and CMC) give the highest result of compressive strength which is 0.2339 N/mm^2. On the other hand, the highest weight percentage of binders (PEG and CMC) is 7 wt.% gives the lowest result of the compressive strength which is 0.1310 N/mm^2. Previous studies have shown that the compressive strength for samples sintered at higher temperatures due to the low porosity and pore size is small [18,19]. In the present study, it was found that the binders (PEG and CMC) 3 wt. % gives highest compressive strength with lowest average pore size and porosity of SiO_2 foam at temperature 1250°C. Thus, it can be deduced that the size and arrangement of the pore also affect the compressive strength of the SiO_2 foam. Compressive strength is inversely correlated to the pore size, and pore volume as total porosity. The correlation between the compressive strength and density of silica foam with a different composition of binders (PEG and CMC) as shown in Figure 7.
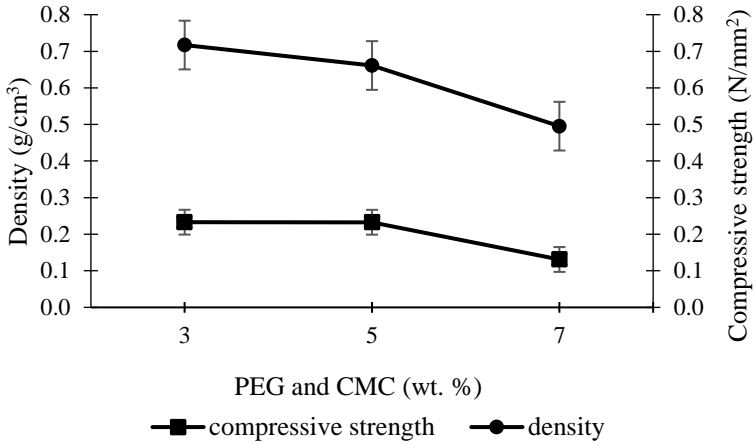
Figure 7: Correlation between density and compressive strength of silica foam

From the observation, it clearly shows that the density decrease with decreasing the compressive strength of the SiO_2 foam. As a mentioned earlier, the higher composition of binders (PEG and CMC) influenced the viscosity of slurry hence affect the impregnation process on the polyurethane foam. Besides, the condition of the slurry should be viscose enough to more deposition on the struts of PU foam. However, the workability of the high viscosity of the slurry can be reduced because of difficulty enter of slurry in the sponge structure. So, that results in non-uniform foam [16,20]. The authors have found that the strength of the strut was depending on the foam density. Moreover, the sample with higher foam density has the thicker struts compared to the other samples [21]. Therefore, high strength values obtained from silica foams in this study is mainly due to the presence a small size of the pore and high strength of struts in the structure network of foam.

Conclusion

Fabrication a silica foam using a different composition of the binders of PEG and CMC with sintering temperature 1250ºC was successfully achieved by adaptation of the replication method. The result found that the composition of binders (PEG and CMC) affects the viscosity of the slurry. The sizes of pores were obtained in a range between of 259.17μm to 559.83. The percent porosity was found to be in the range of 60% to 75%. Thus, it is clear that the density and porosity are interrelating with each other. In addition, compressive strength analysis results of silica foam is decreasing with increasing solids loading. Moreover, it was found that pattern shows that the higher compressive strength is higher the density of the sample is formed.

Thus, from this study the effect of the composition binders (PEG and CMC) influencing the physical and mechanical properties of the SiO_2 foam. Moreover, the finding of composition SiO_2 55wt. % with 3 wt. % binders (PEG and CMC) producing the highest density which is 0.7170 g/cm^3 and 0.2329 N/mm^2 is indeed to be applied in a various industrial application.

Acknowledgement

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