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Influence of Water Molar Ratio on Fabrication of Silica Ceramic Membranes via Sol-Gel Dip-Coating Method

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ABSTRACT. Ceramic membranes are an inorganic membrane that received great attention as it overcome the limitation of polymeric membrane. The silica ceramic membrane can be used in gas separation as it able to work at elevated temperature and in chemically aggressive environment. This work is conducted to study the influence of the molar ratio of water (R) on the fabrication of silica ceramic membrane by sol-gel dip-coating method. Commercial support was dipped into the solutions consist of the mixture of tetraethylorthosilicate (TEOS), distilled water and ethanol with the addition of a small amount of acid as a catalyst. The molar ratio of TEOS toethanol was fixed at 1 to 3.8 (TEOS:ethanol, 1:3.8). However, the molar ratio of water are varied at 2, 3, 4 and 4.7. FESEM (Field Emission Scanning Electron Microscope), XRD (X-ray Diffraction) and FTIR (Fourier Transform Infrared Spectroscopy) are utilized to determine the structural and chemical properties of silica ceramics that are fabricated with different R. FESEM images implied that the silica has been deposited on the surface of silica membrane and penetrate into the pore walls. XRD analysis of the fabricated silica ceramic membranes illustrated the existence of silicate hydrate. The fabricated silica ceramic membrane with silica sol molar ratio 1:3:3.8 gave the lowest intensity of XRD peak. FTIR analysis, it was observed that the presence of Si-O-Si functional group is in the range 1060 to 1080 cm^{-1}.

Keywords: *Water molar ratio, Silica sol, Silica ceramic, Sol-gel, Dip-coating;*

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1. INTRODUCTION

The membrane can be defined as a selective barrier between two phases [1], which are selective to one component and while rejecting the other. Type of membranes can be classified into three categories based on their materials, which are organic (polymeric), inorganic and mixed matrix [2]. Polymeric membranes have limitation in some application due to low chemical resistant and cannot work at high temperatures. Hence, inorganic membrane was used as a substitution of the polymeric membrane [3].Inorganic membranes are used in many practical applications such as gas separation, water desalination, treatment of wastewater and ultra-filtration in food industries [4]. Ceramic inorganic membranehad received a great attention as it offers many advantages such as high chemical, thermal, mechanical and physical stability, outstanding separation characteristics, long working life and ecologically friendly [5]. Silica is one of the examples of ceramic membrane that received great attention compared to other type of ceramic membranes, especially for the removal of carbon dioxide at high temperature in chemically aggressive environment [6,7].

The silica ceramic membrane can be fabricated by the sol-gel method, as this technique has been used for a long time in the fabrication of glasses and ceramics [8].The sol-gel technique has been used in the reproduction of new porous nanomaterial that possessed well-defined structures and complex shape [9]. Sol-gel process refers to a process where the transition from liquid (solution or colloidal solution) into a solid (di- or multiphase gel) is happening. The sol-gel technique has received great attention in the research field since the last two decades due to the benefit they are offering, including the production of high purity homogeneous materials, ability to control the reaction of the solution by using chemical techniques, low processing temperature requirement, and it allows the formation of new crystalline phase of the non-crystalline solid. One specific example of a sol-gel method to produce silica ceramic membrane is the polymerization of tetraethylorthosilicate (TEOS) in ethanol and water, which are then resulting in formation of Si-O-Si chemical linkage [10]. Prepared sol can be deposited onto a substrate in order to form a filmby several methods, for example dip coating. Silica sol-gel dip-coating method can be used whenreduction of pore size of fabricated membrane is desired [4].

There are several parameters involved in the sol-gel technique that will affect the structural and textural properties of the synthesized materials. The parameters that will influence the result of this method including the molar ratio of reactant, initial reaction condition (precursor, pH and temperature), aging and drying conditions, and solvent compositions.Hence, this study will focus only on the influence of the molar ratio of water (R) in silica solution on the fabrication of silica ceramic membranes by using sol-gel dip-coating techniques. The fabricated silica were then characterized in order to observe any changes in their physical and chemical characteristics.

2. MATERIALS AND METHODS

***2.1 Silica sol* preparation.** Silica sols was prepared by mixing TEOS, water and ethanol together at 298K with vigorous stirring. The molar ratio of TEOS to ethanol is fixed at 1:3.8, while the molar ratio of water, were varied at 2,3,4 and 4.7, as shows in Table 1. Then, a small amount of acid was added to the solution as a catalyst.

Table 1 Molar ratio of TEOS, ethanol and water used in preparation of silica sol

TEOS	Ethanol	Water
1	3.8	2
1	3.8	3
1	3.8	4
1	3.8	4.7

2.2 Membranes Fabrication. Commercial ceramics (Fig. 1), 10 inch Doulton OBE Ceramic with 32 mm internal diameter, 48 mm outer diameter and length of 200 mm, were used as a substrate or support to the fabricated membranes. The commercial support was dipped into the prepared silica solutions, and thenwere dried at room temperature for 24 hours.After that, the samples were calcined at 773 K with holding time of three hours. The dipping, drying and calcine process was repeated for three times.

2.3 Membranes Characterization .X-ray diffraction (XRD) was used to determine the element of the fabricated membrane.The morphology of the membrane surface and support were determined using the field emission scanning electron microscope (FESEM) (JSM 6700F JEOL). Fourier transform infrared spectroscopy (FTIR) analysis was performed in order to determine all the existed functional groups.

Fig. 1 Raw commercial support

3. RESULTS AND DISCUSSION

Fig. 2 shows the XRD result of the fabricated silica ceramic membranes with different molar ratio of water (R). XRD analysis was performed in order to identify the compound that existed in materials. For this study, XRD is used to confirm the existence of silica if the fabricated ceramic membrane. The XRD pattern shows the existence of silicate hydrate element for fabricated silica ceramic membrane, which mean a trace on water still remain in the fabricated membrane. However, water has no substantial effect on the fabricated silica structure. The fabricated silica ceramic membrane with R=3 gave the lowest intensity which is 413.3 cps. This means that the membrane with this ratio is thicker among other membranes. Moreover, the highest peak, 22° (2θ) was appeared for all fabricated silica ceramic membrane indicates that there was the presence of the expected reaction, which is silica (SiO_2). This proof that silica ceramic had been successfully fabricated from the solution used.

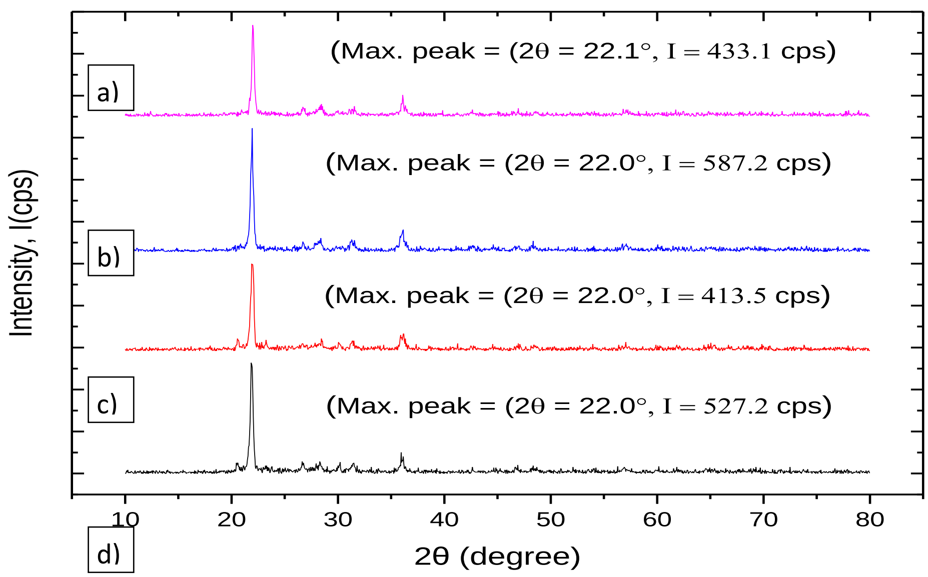

Fig. 2 XRD pattern for fabricated silica ceramic membrane with different silica sol molar ratio (a) R=4.7, (b) R=4, (c) R=3, and (d) R=2

Fig. 3 presented the FESEM images of the fabricated silica ceramic membranes with different R. FESEM analysis was employed in order to study the surface morphology of the fabricated silica ceramic membranes.Fig.4 shows the image of raw commercial support that had been used as a substrate, where silica sol will be deposited onto the support by sol-gel dip-coating method to produce silica membrane. From Fig. 4, the pore of the raw support is visible under the FESEM analysis, where the size of the pore ranges around 0.6 µm. However, in Fig. 3, no pore is observed on the fabricated silica, prove that the pore size had reduced after silica membrane has been fabricated.

Furthermore, before the coating, the image of the raw support in Fig. 4 shows a clear and clean surface without any foreign particle appear the surface. Conversely, after the silica ceramic membrane are fabricated, the FESEM image in Fig. 3shows some agglomeration of fine particle on the surface of the membranes. An agglomeration of particles resulting in particle aggregate formation as the particles adhere to each other. This leads to irregular arrangement on the surface of the fabricated membranes. Fine particle on the membrane surface indicate that the silica have already penetrated onto the surface of the raw support, creating silica ceramic membrane. Besides that, it is observed that, as the R is decreased, the surface of the membrane becoming more consolidated, and less particle aggregate appears on the membrane surface. Fig. 3 (c) illustrates that the surface of membrane fabricated is more consolidated than others, where R=2 is used.

At fixed concentration of TEOS, increasing of R in the solution leads to the increasing of the hydrolysis and rate of condensation. Solution with R=2 make the penetration of silica sol into pore membrane become difficult because the solution become more concentrated as the amount of water used is small, which is insufficient to complete the hydrolysis reaction of the sol. When a very low R is used in silica solution mixture, it will produce a matrix with a more open structure due to incomplete hydrolysis. Hence, there will be fewer cross linking in the sol-gel material. Moreover, a low water content in silica sol will produce a high content of oligomers, either volatile or soluble, and also resulting in reduction of silica generated. The homogeneity and composition of the fabricated silica membrane can be affected by the presence of those oligomers and also monomers that are only partially hydrolyzed.

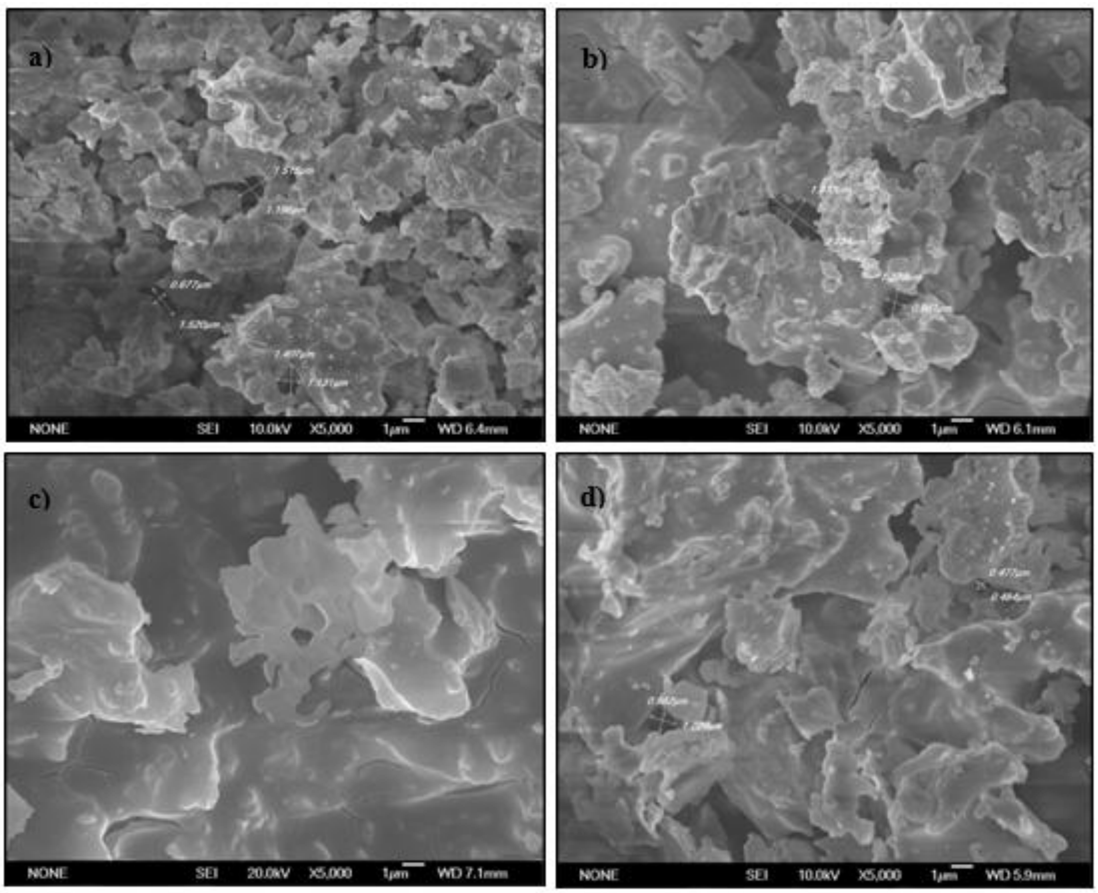

Fig.3 FESEM images of fabricated silica ceramic membrane at different silica sol molar ratio (a) R=4.7, (b) R=4, (c) R=3, and (d) R=2

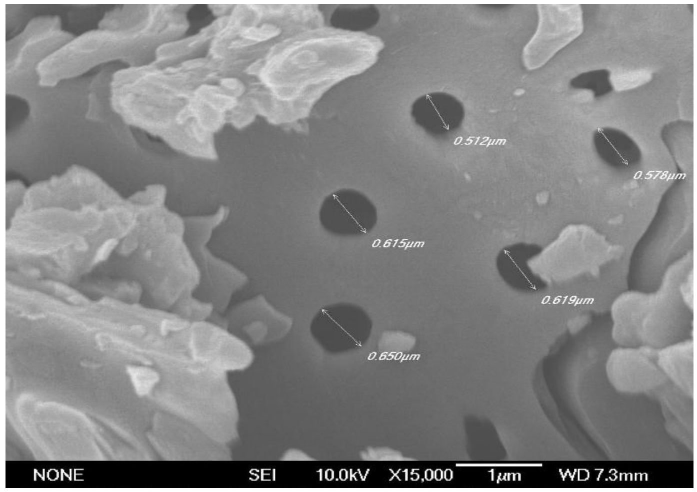

Fig.4 FESEM image of raw support

Fig. 5 represents the FTIR result of the silica ceramics fabricated by various R. FTIR is used to determine the functional group of the fabricated ceramic, and also to analyze the bond existed. From the result illustrated, it displayed that all gels exist at the same absorption band. The band located at around 466 cm^{-1}, 610cm^{-1}, 790cm^{-1} and 1070cm^{-1} are attributed to silica network. The major peak is around 1070 cm^{-1} due to the anti-symmetrical stretching vibration of Si-O-Si, are observed for all fabricated membrane. The strong vibration of silicon–oxygen covalent bonds occur mostly in the 1200-1000 cm^{-1} range exposing the existence of adense silica network. The oxygen atom acts as a bridge that connects the two silicon sites. Secondary bands around 790cm^{-1} related to the symmetric stretching, vibration are observed, while bending mode are detected around 466cm^{-1} [11-13].

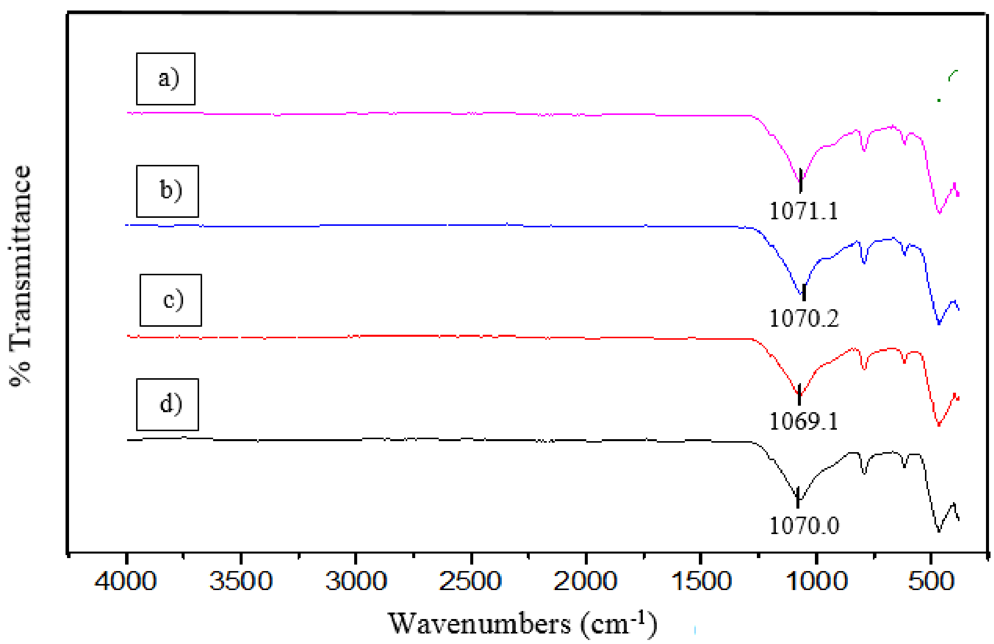

Fig. 5 FTIR resultof fabricated silica ceramic membrane at different silica sol molar ratio(a) R=4.7, (b) R=4, (c) R=3, and (d) R=2

4. SUMMARY

Fabrication of the ceramic membrane via sol-gel dip coating, generate membrane with uniform pore size distribution, while a variation of R affected the physical and chemical characteristics of the fabricated silica ceramic membranes. From the results obtained, it can be concluded that the silica ceramic membrane via sol-gel dip-coating method is successfully fabricated. The fabricated silica ceramic membrane is characterized chemically and physically. Silica solution was prepared by varying the R, then the sol were deposited onto the commercial support as that acts as the substrate. According to FESEM analysis, it is discovered that as the molar ratio of deionized water is decreased, the surface of the membrane becomes more consolidated and pore size was reduced. From the XRD analysis, the result proved the existence silica compound in the fabricated membrane. In addition, FTIR analysis presents the Si-O-Si bond which strengthen the proof of the existence of the silica in the fabricated membrane. Hence, according to analysis result, all fabricated membrane showing the existence of silica.

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