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Utilization of Natural Zeolite for Development of Ceramic/Polymer Composite Membrane for Ultrafiltration

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Abstract

Natural zeolites are aluminosilicate materials with tetrahedral structure. Natural zeolites have several advantages for many purposes such as adsorbent, catalyst support and ceramic membrane. This research explored the utilization of natural zeolite for the ceramic/polymer composite membrane. The results of the research show that membrane has the density of 1.64 gr/cm³ and porosity of 37.39% at the concentration of PVA of 7%. The performance test shows that composite membrane can be used for methylene blue separation and can be categorized as ultrafiltration. The coefficient of rejection that can be achieved is 93.53%. The membrane still has still a weakness. The flux of the membrane is very low.

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1. Introduction

Natural zeolites are abundance resource in the world. There are thousands of kinds of natural zeolites. Every mining has specific characteristics. Commonly, natural zeolites have special structures that are framework silicates consisting of interlocking tetrahedrons of SiO₂ and Al₂O₃ [1]. Because of these special structures, natural zeolites have pores, namely macropores and micropores with dimensions of 3 to 10 Angstroms.

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Natural zeolites are widely used for many purposes. Natural zeolite can be used as an adsorbent for organic and inorganic materials. As adsorbent, the natural zeolite can adsorb until 80% of heavy metal from wastewater [1][2]. Natural zeolite is also effective to adsorb organic materials of Rhodamine B from aqueous solution [3].

Natural zeolite can also be used as a catalyst support. The utilization process of natural zeolite as catalyst support is by impregnating the catalyst to the pores of the natural zeolite. A series of KOH/zeolite catalyst was prepared by impregnation of zeolite from Pacitan with potassium hydroxide in a various concentration as a catalyst for the transesterification of palm oil [4]. Nickel supported natural zeolite was successfully synthesized as a bi-functional catalyst for conversion of citronella oil crude to menthols.

Another utilization of the natural zeolite is for the production of the ceramic membrane. Hristov et al. [5] produced the ceramic membrane from natural zeolite by semi-dry pressing. Ha et al. [6] also produced ceramic membrane from diatomite for microfiltration. The main process production of the ceramic membrane is sintering process at the high temperature of 900 – 1000°C. To improve the grade of the ceramic membrane from microfiltration to become ultrafiltration or nanofiltration is usually done by coating process with the polymers.

This research aimed to produce and characterize the ceramic/polymer composite membrane from natural zeolite and polyvinyl alcohol (PVA). The characteristic of the composite membrane such as bulk density and porosity were determined. The performance test of ceramic/polymer was done for textile wastewater purification.

2. Materials and Methods

2.1. Materials

Natural zeolite was purchased from the local market at Purwokerto, Indonesia. TiO_2 was purchased from Merck. PVA polymer was technical grade purchased from local market. Rice starch and methylene blue (from Mbangun-Trisno wenter) were purchased from the local market at Purwokerto, Indonesia.

2.2. Ceramic Membrane Support Preparation

Natural zeolite was crushed to the size of $< 45 \mu\text{m}$. The natural zeolite was mixed with TiO_2 of 5% and rice starch of 7%. The mixture was then placed at O-ring casting with the diameter of 3.2 cm and the thickness of 1 cm and pressed at 5 tons for 30 minutes. The mixture was then sintered at the temperature of 900°C for 6 hours.

2.3. Composite Membrane Development

The composite membrane was developed by dip coating process. PVA polymer and citric acid (crosslinker) were dissolved in the water and then homogenized and heated by ultrasound bath at 60°C for 30 minutes. Ceramic membrane support was dip coated at the polymer solution for 24 hours. The hybrid membrane was drying at 60°C for 6 hours then at 120°C for 2 hours.

3. Results and Discussion

3.1. Characterization of Composite Membranes

Fig. 1 shows the bulk density of the composite membranes. The bulk density of the membranes will increase with the PVA concentration. Without PVA coating, the bulk density of the ceramic membrane only 1.45 gr/cm^3 , and with PVA coating the bulk density will increase until 1.64 gr/cm^3 at the concentration of PVA is 7%.

Fig. 2 shows the porosity of the composite membranes. The porosity of the membranes will decrease with the PVA concentration. Without PVA coating, the porosity of the ceramic membranes reach 53.27%, and with PVA coating, the porosity will decrease until 37.39% at the concentration of PVA is 7%. It can be explained, that the polymer will fill the pores of the ceramic membrane. It causes increasing bulk density and decreasing porosity.

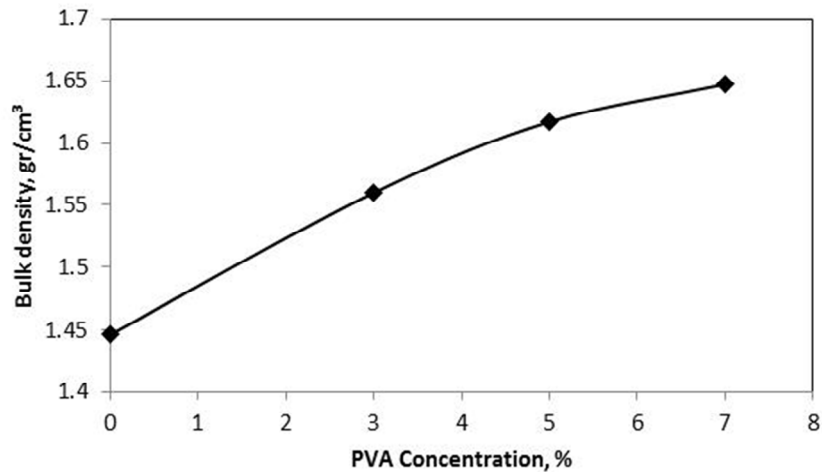


Fig. 1. Bulk density of composite membranes at various PVA concentrations

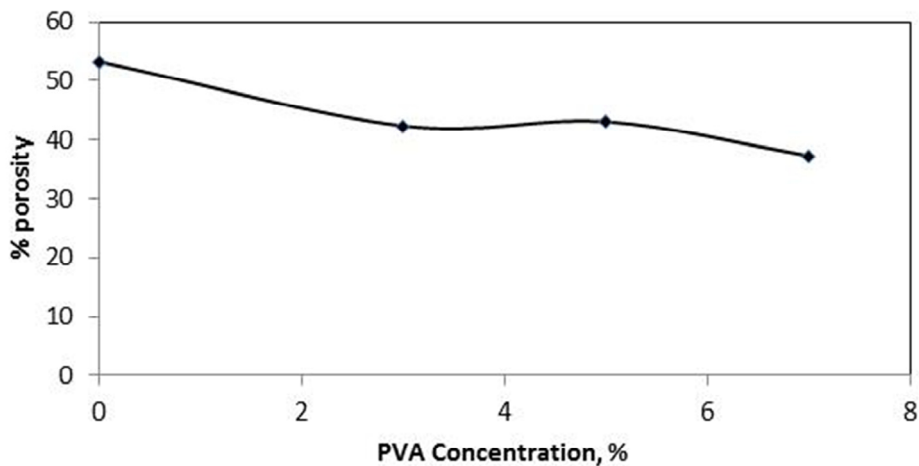


Fig. 2. The Porosity of composite membranes at various PVA concentrations

Fig. 3 shows the morphology of the membranes. It can be seen that before the coating process, the ceramic membrane is very porous (porosity of 53.27%). The average diameter of the pores is 1 μm . After the coating process, the pores are covered and filled with the polymer. The grade of membranes will increase from microfiltration to ultrafiltration.

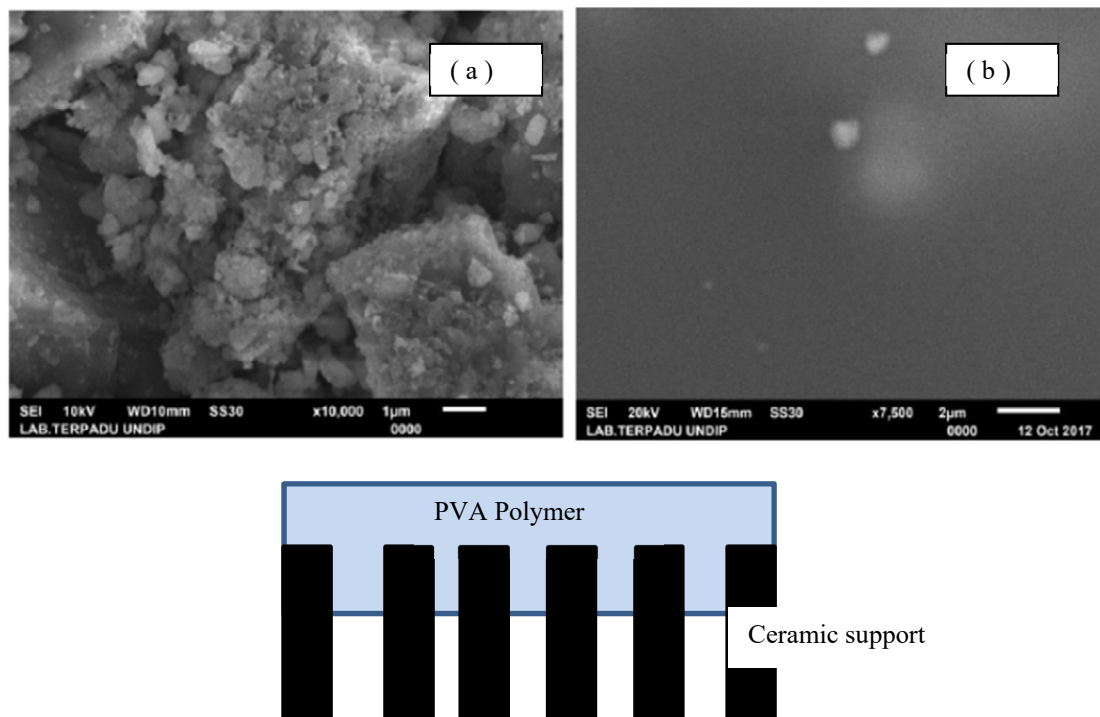


Fig. 3. The morphology of the composite membranes: (a) before coating; (b) after coating; (c) composite membrane

3.2. Application of Composite Membrane for Textile Wastewater

The performance test of the composite membrane was done for textile wastewater purification. The synthetic textile wastewater is done by dilute of 1 gram of methylene blue in 1 liter of demineralized water. Without coating with PVA polymer, the ceramic membrane can't separate the color (Rejection of 0%). After the coating process, the composite membranes can separate the color. Table 1 shows the rejection of the membranes. The higher rejection is reached at the PVA concentration of 7%.

The composite membranes still have a weakness. At the ultrafiltration process with the pressure of 5 bar, the flux of permeate is very low. The flux of the membrane ranges from 0.033 – 0.0059 ml/minute.cm² (Fig. 4) at the 7% PVA concentration. As a comparison, Majewska-Nowak and Kawiecka-Skowron [7] utilized the commercial ceramic membranes from Tami industries for anionic dye separation with various cut-off values (15 and 150 kDa). The ultrafiltration (UF) processes were done at a trans-membrane pressure of 0.2 MPa with the use of the cross-flow system. The rejection coefficient of direct green, helion blue and direct black increased from 78%, 80%, and 94% (at a linear velocity of 1.5 m/s) to 86%, 94%, and 98% (at linear velocity of 6.9 m/s), respectively.

Table 1. Rejection of textile wastewater during ultrafiltration process

No	PVA Concentration (%)	Rejection
1	0	0
2	3	9.12
3	5	67.68
4	7	93.53

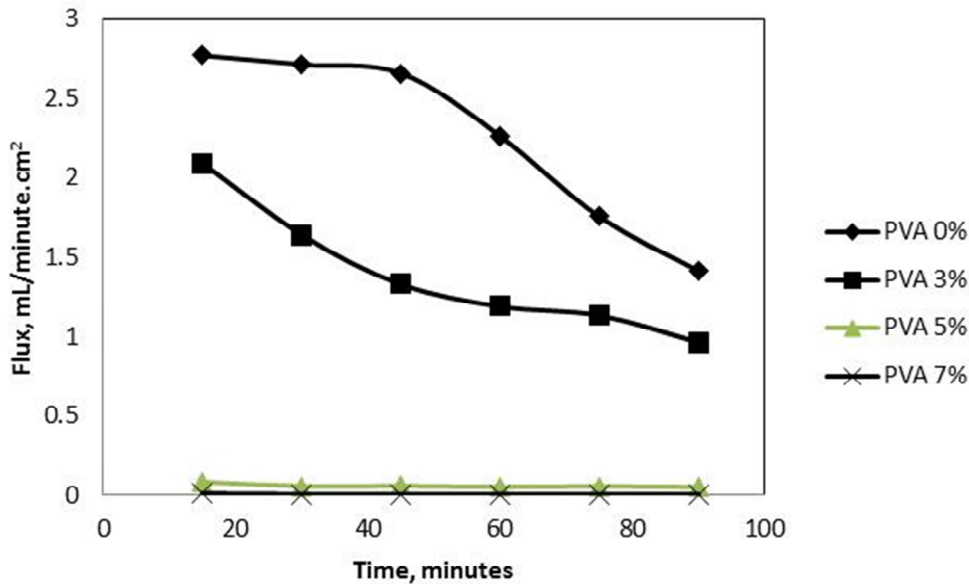


Fig.4 The flux of composite membranes at various PVA concentrations

4. Conclusion

The natural zeolites can be used for ceramic/polymer composite membranes. The membrane has the density of 1.64 gr/cm^3 and porosity of 37.39% at the concentration of PVA of 7%. The performance test shows that the composite membrane is categorized as ultrafiltration. The coefficient rejection that can be achieved is 93.53%.

Acknowledgements

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