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Development of Ultrafiltration Membrane from Polyethylene Terephthalate (Pet) Bottle Waste

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Abstract: Polyethylene Terephthalate (PET) bottle is used as beverage packaging, which is very convenient as one time use packaging. However, the huge amount of PET bottle waste has been becoming a serious problem for the environment. The utilization of PET bottle waste is very important to reduce the environmental problem. In this work, PET bottle waste was used a raw material to develop an ultrafiltration (UF) membrane. The membrane was prepared by using a phase inversion technique. The effect of the type of solvent, additive, and non-solvent on the microstructure and ultrafiltration performance of the membrane was studied. Different type of solvent, phenol, m-cresol, and DMSO were used to dissolve PET bottle as the source of membrane polymer. Two different additives, Polyethylene Glycol (PEG) and Polyvinyl Pyrrolidone (PVP) were used. Membrane 3 with the composition of PET, phenol as solvent, and PEG as additive was prepared successfully. The variation of aqueous alcohol solutions as non-solvent resulted in different microstructures of the membranes as shown by the scanning electron microscopy (SEM). The permeation experiment result using pure water as the feed showed that membrane 3 using aqueous butanol as non-solvent (membrane 3-ButOH) exhibited the highest permeate flux compared to that of membrane 3 using aqueous propanol (membrane 3-PrOH) or ethanol as non-solvent (membrane 3-EtOH). The ultrafiltration experiment was carried out using a feed solution of water containing polyethylene glycol (PEG) 20,000. The membrane 3-EtOH showed the lowest permeate flux of 3.24 kg/m^{2}h, but the highest rejection of PEG 20,000 of 65.87%. The membrane 3-PrOH had a permeate flux of 11.57 kg/m^{2}h and a rejection of 64.73%. Whereas the membrane 3-ButOH showed the highest permeate flux of 27.78 kg/m^{2}h, but the lowest rejection 16.93%. This result was obtained due to the different membrane microstructures which were strongly affected by the type of non-solvent.

Keywords: Membrane, Ultrafiltration, Polyethylene Terephthalate (PET), PET bottle

1. Introduction

Membrane is a selective barrier that allows the passages of certain component while retains other. Various types of membranes such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis membranes have been widely used in food, beverage, pharmaceutical and chemical industries for various separation and purification processes. Pervaporation membranes and gas separation membranes have also become alternatives of conventional separation techniques due to their high selectivity and low energy (Kusumocahyo, 2005; Ohta, 2008). Membranes which are commercially available in the market are mostly made from polymer materials such as cellulose acetate, polysulfone, polyamide, etc (Baker, 1991). Recently, membranes made from ceramic materials have been receiving a high attention due their chemical and thermal stability (Kusumocahyo, 2016; Huang, 2014; Xu, 2016), however the price of ceramic membranes is much more expensive compared to the polymer membranes. There are many studies on the use of other polymer materials with the aim to improve the membrane performance (Liu, 2017; Pulido, 2018) or to reduce the cost of the membranes (Ahmad, 2018). The use of cheap membrane material is still important in the developing countries like Indonesia, since most polymer materials to produce the membranes are not produced in Indonesia and have to be imported.

There is an alternative to obtain a membrane material from our daily waste, namely used plastic bottles. Most plastic bottles used for the packaging of water, juices, tea, coffee, soft drink, etc are made from polyethylene terephthalate (PET). Millions PET bottles are disposed every day causing a serious

environmental issue. According to the Forbes Magazine in 2017, only less than 10% of bottle plastics are recycled (Nace, 2017). It means over 90% of plastic are disposed without further treatment or just combusted causing another environmental problem. The reuse of PET bottles as a polymer material to produce membranes is a conversion of waste into a high tech and more valuable product, yet at the same time it contributes in the efforts to solve the environmental problem.

The aim of this study is to utilize PET bottle waste as the polymer material to develop ultrafiltration membranes. The ultrafiltration membranes were prepared using a phase-inversion technique. The influence of type of solvent, type of additive, and type of non-solvent on the microstructure and the ultrafiltration performance of the membranes were studied. Ultrafiltration experiments were carried out using pure water and water containing polyethylene glycol (PEG) to study the permeate flux and the rejection of the membranes.

2. Experimental

2.1. Materials

Used PET clear bottles were collected from various sources. Phenol, dimethyl sulfoxide (DMSO) and m-cresol were used as the solvent to prepare the polymer solution. All solvents were purchased from Merck. Polyethylene glycol (PEG) and polyvinyl pyrrolidone (PVP) were purchased from local suppliers, and were used as the additive for the polymer solution. Ethanol, n-propanol, and n-butanol were used as the non-solvent of the coagulation bath, and were also obtained from local suppliers. Purified water was used as the non-solvent of the coagulation bath and as the feed solution for the ultrafiltration experiment.

2.2. PET bottle pre-treatment

Transparent and colorless used PET bottles were collected, then the bottle caps and the labels attached on the bottles were removed. The bottles were washed using water, and after drying they were cut into small pieces of plastic shards with an approximate size of 3 mm x 3 mm.

2.3. Membrane preparation

The ultrafiltration membrane was prepared by using the phase inversion technique. First, the solvent was heated to 35℃ for 30 minutes, then the PET shards and the additive were added into the solvent. The weight ratio of the solvent, PET and the additive was 70:25:5. The solution was stirred and heated at 100℃ for 3 hours. Then the solution was casted onto a glass plate using a home-made casting knife, and it was immersed into a coagulation bath at room temperature. The coagulation bath contained water-alcohol mixture as the non-solvent with a weight ratio of water and alcohol of 1:12. Three different alcohol, namely ethanol, n-propanol, and n-butanol, were used.

2.4. Characterization of the membranes

The appearance of the membrane was observed using a microscope, whereas the microstructure of the membrane was observed using a scanning electron microscope (SEM). The thickness of the membrane was measured using a micrometer.

2.5. Ultrafiltration experiment

The ultrafiltration experiment was carried out using an ultrafiltration membrane apparatus equipped with a pump (Masterflex L/S Economy Drive Pump, Cole Parmer, USA) and a membrane cell with an effective membrane area of 51.8 cm^2. The experiment was conducted at room temperature and a pressure of 1 bar. Figure 1 shows the schematic diagram of the ultrafiltration experiment. Pure water and water containing polyethylene glycol (PEG) 20,000 with a concentration of 500 ppm were used as

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the feed solution to study the permeate flux and the rejection of the membranes. The permeate sample was collected after a steady state condition was achieved, and the permeate flux was calculated from the flow rate of the permeate using the following formula:

$$F = \frac{m}{A\,\Delta t}$$

Where F is the permeate flux (kg/m^{2}h), m is the weight of the permeate (kg), A is the effective membrane area (m^2), and Δt is the time interval (h).

The concentration of PEG 20,000 in the permeate was measured using a UV-vis spectrophotometer (T60 UV, PG Instruments Ltd., England) with a wavelength of 150 nm, and the rejection R was calculated using the following formula:

$$R = \frac{C_F - C_P}{C_F} \times 100\%$$

Where R is the rejection (%), C_F is the concentration of PEG in the feed solution (ppm), and C_P is the concentration of PEG in the permeate (ppm).

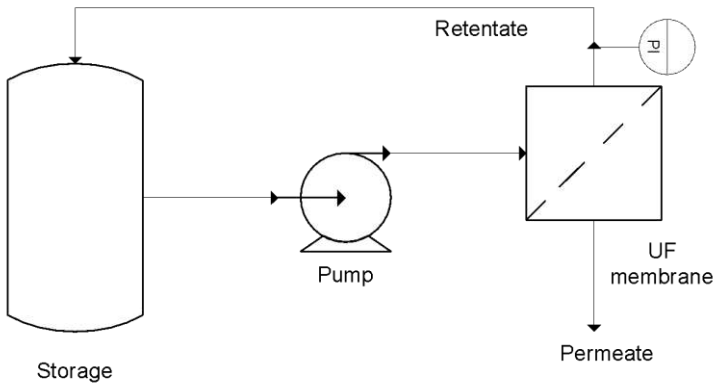

Figure 1. Schematic diagram of the ultrafiltration experiment

3. Results and Discussion

3.1. Effect of Additive

To study the effect of additive, three different membrane solutions using phenol as the solvent were prepared. The first membrane (Membrane 1) was prepared without additive, the second membrane (Membrane 2) was prepared using polyvinyl pyrrolidone (PVP) as additive, whereas polyethylene glycol (PEG) was used to prepare the third membrane (Membrane 3). These three membrane solutions were clear, transparent and homogeneous, however after the solution was casted and immersed in the coagulation bath containing water-ethanol solution, the appearances of the resulted membranes observed using a microscope were different as can be seen in Figure 2.

Membrane 1 which contained no additive had a white color and a smooth surface but the membrane was stiff and not flexible enough to be used for the ultrafiltration experiment. Membrane 2 which contained PVP as additive had more flexibility than Membrane 1, but it had defects on its surface as can be seen in Figure 2. Thus, this membrane was leak and could not be used for the ultrafiltration experiment. The defects occurred because PVP had a strong interaction with phenol (Molyneux, 1961). When the casted solution was immersed in the coagulation bath containing non-solvent, PVP was possibly diffused from the casted membrane together with phenol into the coagulation bath, creating a defect of the membrane. A study by Arahman et al (Arahman, 2017), reported that PVP can act as a pore former and can increase the pore size of a membrane, however in our study most likely PVP is too strong to be used as pore former for PET membrane, and resulted in a defect membrane. On the other hand, Membrane 3 which used PEG as the additive had a no-defect

smooth surface, and showed a good flexibility. Thus, this membrane could be used for the ultrafiltration experiment. Other study reported that PEG acts as a plasticizer to produce a flexible membrane (Hee, 2014). Thus, in this study PEG was used to prepare PET membranes with the variation of solvent and non-solvent.

Figure 2. Surface appearance of Membrane 1, Membrane 2, and Membrane 3 as observed using microscope

3.2. Effect of Solvent

Three different solvents, namely phenol, DMSO and m-cresol, were used to study the effect of solvent on the performance of the PET membrane. PEG was used as the additive, while water-ethanol mixture was used as the non-solvent. The use of phenol as the solvent resulted in a clear and homogeneous polymer solution, as well as a membrane with good flexibility. However, when DMSO was used as the solvent, the PET bottle shards and PEG were not completely soluble in DMS, even the polymer solution was stirred and heated for more than 3 hours at 100℃. When m-cresol as used as the solvent, the PET shards was completely soluble in m-cresol, however the addition of PEG into the solution resulted in an agglomeration of the solution. Therefore, for the next experiment, phenol was used as the solvent.

3.3. Effect of Non-Solvent

Coagulation bath containing non-solvent is one of the important things in the phase inversion technique. Here, three different types of non-solvent were used to prepare the membranes. They were water-ethanol mixture, water-propanol mixture, and water-butanol mixture. All membranes used phenol as the solvent and PEG as the additive. The resulted membranes were indicated as Membrane 3-EtOH, Membrane 3-PrOH and Membrane 3-ButOH.

Figure 3 shows the surface appearance of Membrane 3-EtOH, Membrane 3-PrOH and Membrane 3-ButOH. These three membranes showed the same appearance as observed using microscope. They were white in color, mechanically strong, and had a good flexibility. As shown in the picture below, there were no defects on their surface, thus ultrafiltration experiment can be carried out using these three membranes. The thickness of the membranes ranged between 110 to 146 μm.

Figure 3. Surface appearance of Membrane 3-EtOH, Membrane 3-PrOH and Membrane 3-ButOH as observed using microscope

3.4. Results of Ultrafiltration Experiment

Membrane 3-EtOH, Membrane 3-PrOH, and Membrane 3-ButOH were then tested through an ultrafiltration experiment using a feed solution consisting of pure water and PEG with an average molecular weight of 20,000 Da to measure the permeate flux and the selectivity. The concentration of PEG in the feed solution was 500 ppm, and the concentration of PEG in the permeate was measured using a UV-vis spectrophotometer with the aid of a standard calibration curve. The result of the ultrafiltration experiment is shown in Table 1 below. As can be seen in Table 1, Membrane 3-EtOH exhibited the lowest permeate flux, but the highest rejection of PEG 20,000 with a rejection of 65.78%. Membrane 3-PrOH exhibited a higher permeate flux but a lower rejection of 64.73%. The highest permeate flux was achieved using Membrane 3-ButOH, however the rejection is the lowest. The use of ethanol as the non-solvent resulted in the lowest permeate flux due to the smoothest microstructure which led to the formation of the smallest pores. On the other hand, the use of propanol and butanol, which are more non-polar compared to ethanol, led to the formation of bigger pores, thus higher permeate fluxes.

Table 1. Permeate flux and rejection of the membranes

Membrane	Permeate flux (kg/m^{2}h)	Rejection
Membrane 3-EtOH	3.24	65.78 %
Membrane 3-PrOH	11.57	64.73 %
Membrane 3-ButOH	27.78	16.93 %

4. Conclusion

Ultrafiltration membranes from used polyethylene terephthalate (PET) bottles were successfully developed in this work. The use of phenol as the solvent and PEG as the additive resulted in a smooth and flexible membranes which could be used for ultrafiltration experiments. The microstructures of the membranes were strongly affected by the type of non-solvent for the coagulation bath. The use of water-ethanol mixture as the non-solvent resulted in the smoothest microstructure as observed using SEM, and as a result Membrane 3-EtOH exhibited the lowest permeate flux of 3.24 kg/m^{2}h but the highest rejection of PEG 20,000 with a value of 65.87%. On the other hand, the use of water-propanol and water-butanol as the non-solvent resulted in coarser microstructures, thus a larger pore size. The highest permeate flux with a value of 27.78 kg/m^{2}h was achieved using water-butanol as the non-solvent, but the selectivity was the lowest with a rejection of PEG 20,000 of 16.93%.

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