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Improved permeate flux and rejection of ultrafiltration membranes prepared from polyethylene terephthalate (PET) bottle waste

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Abstract

The vast amount of not-recycled polyethylene terephthalate (PET) bottle waste is a serious threat to the environment. In order to utilize the waste, PET ultrafiltration membranes were prepared using PET bottle waste as the raw material by using the phase inversion technique. Low molecular weight polyethylene glycol (PEG 400) was used as the additive for the membranes. PET resin was also used as the membrane material to compare the properties of the membrane from PET bottle waste and those from the PET resin. The membrane prepared from PET bottle waste and that prepared from PET resin showed similar membrane characteristics such as IR spectra, morphology, hydrophilicity and porosity, indicating that instead of using PET resin, PET bottle waste can be utilized as a source of the polymer material to fabricate low-cost membranes. The morphology, hydrophilicity and porosity of the membranes were strongly affected by the PEG 400 concentration. The analysis of the membrane morphology using Scanning Electron Microscopy showed that the membranes had an asymmetric structure that consisted of a macroporous cross section and a smooth active layer. Increasing the PEG 400 concentration resulted in a smaller pore size, however the hydrophilicity and the porosity of the membranes increased. As a result, the membranes showed an increase in both permeate flux and rejection with increasing concentration of PEG 400 as observed from the results of the ultrafiltration experiments. Using Bovine Serum Albumin as a solute model in the feed, high values of rejection of up to 94% were achieved. Thus, ultrafiltration membranes with improved permeate flux and rejection could be prepared from PET bottle waste by the addition of PEG 400 as the additive.

Keywords: Ultrafiltration membrane, Polyethylene terephthalate (PET), Plastic bottle, Recycle, Water treatment

Introduction

Ultrafiltration membranes are widely used for separation processes of aqueous solutions in various industries such as food, dairy, beverage, pharmaceutical, textile, electronics, and chemical industries. They are mostly applied to water treatment such as the production of pure water to remove microorganisms, bacteria, virus, colloidal substances, and suspended micro particles from the water [1, 2]. Another application of ultrafiltration membrane is for the concentration of protein or enzyme [3, 4]. Ultrafiltration membranes have usually a porous asymmetric structure with a macroporous cross section and a smooth active layer that is able to reject high molecular weight solutes such as protein, virus, bacteria, etc., whereas water or low molecular weight solutes can permeate through the membrane. The separation using the ultrafiltration membrane is a pressure-driven separation process, which can be simply operated using a pump without the use of heat. Therefore, the use of the ultrafiltration membrane for separation processes has many advantages due to the lower energy consumption and the high selectivity.

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Commercial ultrafiltration membranes available in the market are usually made from cellulose acetate, polysulfone, polyethersulfone or polyvinylidene fluoride. Many studies to develop ultrafiltration membranes using other polymer materials such as polyetherimide, polyvinyl chloride, chitosan, and other materials have been reported [5–7]. Studies on the modification of the membranes to improve the permeate flux and the rejection have also been reported [1, 8, 9]. Recently, our previous study on the use of polyethylene terephthalate (PET) bottles to prepare PET ultrafiltration membranes has been reported [10]. PET packaging is widely used by the food and beverage industries because of its excellent mechanical strength, good chemical resistance, good transparency, and excellent gas-barrier resistance. PET films are also suitable for many other applications due to their excellent mechanical properties [11, 12], and good chemical resistance against acids and low concentration of alkalies [13]. The outstanding mechanical and chemical properties of PET create the opportunity to fabricate ultrafiltration membranes from PET. The source of the polymer material can even be found in used PET bottles or other used PET packaging that are usually considered as waste. A study on the development of PET ultrafiltration membranes made from PET waste was reported by Rajesh and Murthy [14] who reported that the membranes exhibited a molecular weight cut-off (MWCO) of 500 kDa or higher. Since ultrafiltration membranes having lower MWCO values are desired for the industrial application, our previous study was conducted to develop lower-MWCO ultrafiltration membranes from PET bottle waste [10]. In our previous study, it was observed that the membranes had a MWCO value of 66 kDa, and the permeate fluxes could be increased by decreasing the polarity of the non-solvent, by increasing the molecular weight of the additive, or by increasing the additive concentration. However, it was observed that the permeate flux enhancement was followed by a decline of the rejection rate, because of the enlargement of the membrane pore size [10]. The same phenomenon has been also reported in other studies [5, 15]. Ultrafiltration membranes with high permeate fluxes are desired since the ultrafiltration membranes have been known to have a drawback, namely the fouling problem, that is the permeate flux decline with the operating time because of the concentration polarization on the surface of the membranes. In order to eliminate fouling, many studies have been done to develop membranes with improved permeate fluxes [1, 7–9]. However, the increase in the permeate flux is usually followed with the decrease in the rejection of the membranes. Thus, it is crucial to develop ultrafiltration membranes with improved permeate flux without any decrease in the rejection.

The objective of this work is to develop PET ultrafiltration membranes which exhibit improved permeate fluxes with high rejection values. The membranes were developed using PET bottle waste as the polymer material using polyethylene glycol (PEG) with molecular weight of 400 Da as the additive. The aim of the utilization of PET bottle waste is also to make a contribution to plastic recycling in order to reduce plastic waste. Since PET bottles are originally produced from PET resin, PET resin was also used in this work as the polymer material to prepare the membranes with the aim to compare the characteristics of the membrane developed from PET bottles and those from PET resin. The effect of the PEG 400 concentration on the microstructure, the hydrophilicity and the porosity of the membranes was studied by using Scanning Electron Microscopy (SEM), water contact angle measurement, and gravimetric method, respectively. The membranes were characterized using Fourier Transform Infrared (FTIR) spectroscopy to study the chemical properties. Furthermore, the membranes were characterized for their ultrafiltration performances through ultrafiltration experiments using pure water and a feed solution containing Bovine Serum Albumin (BSA) molecules as a feed model.

Materials and methods

Materials

Plastic bottles made from PET were used. The PET bottles were previously used as packaging for mineral water and was obtained from the local supermarket in Indonesia. PET resin was also used as the polymer material, and was manufactured and supplied by PT Indorama Ventures Indonesia. Phenol (≥ 99%) was used as the solvent, and was supplied by Merck, Germany. Polyethylene glycol with a molecular weight of 400 Da (PEG 400) was used the additive, and was supplied by Merck, Germany. BSA (molecular weight: 66 kDa) was also supplied by Merck, Germany. Technical grade ethanol (96%), monosodium phosphate (NaH_2PO_4, ≥ 99%), disodium phosphate (Na_2HPO_4, ≥ 99%), potassium chloride (KCl, ≥ 99%), and sodium chloride (NaCl, ≥ 99%) were all supplied by Merck, Germany. All chemicals were used as received. Distilled deionized water was used.

Preparation of PET membranes

After removing the labels and bottle caps, the bottles were thoroughly washed. The clean and dry bottles were then cut to obtain small PET shards. PET resin was also used as the membrane material with the aim to compare the characteristics of the membranes with those prepared from the used PET bottles. To prepare the casting solution, phenol was heated at 40 °C to liquify it as phenol is a solid at room temperature. Then, the PET bottle shards or the PET resins were added into the phenol under continuous stirring and heating at 100 °C using a

hot plate (Barnstead Thermolyne) equipped with a magnetic stirrer. Meanwhile, a solution of PEG 400 in phenol was prepared separately by dissolving PEG 400 in phenol at the same condition as above. Both polymer solutions were then mixed at 100 °C for 1 h under continuous stirring to obtain a homogeneous polymer solution. The composition of the PET, the PEG 400 and the solvent in the casting solutions can be seen in Table 1. The membranes were then prepared from the casting solutions by the phase-inversion technique. The polymer solution was cast onto a glass plate and then submerged in a non-solvent bath containing solution of water-ethanol (1:12 v/v) at room temperature. As a result, a white solid flat membrane was obtained. After rinsing several times using distilled deionized water, the membranes were stored in plastic containers containing distilled deionized water for further use.

Characterization of PET membranes

These flat-sheet membranes were characterized for their average thicknesses using a micrometer (Tricle, China) from the measurement of five different locations of the membrane. Analysis using FTIR spectroscopy was conducted to study the chemical structure of the membranes using FTIR spectrometer (Shimadzu IR Prestige-21, Japan). Analysis using SEM (Quanta 650) was conducted to study the microstructures of the membranes. Gravimetric method was used to determine the membrane porosity using the following equation [16, 17]:

$$\varepsilon = \frac{(w_1 - w_2)/d_w}{((w_1 - w_2)/d_w) + w_2/d_p} \quad (1)$$

where w_1 and w_2 are the weight of the wet membrane and that of the dry membrane, respectively, whereas d_w and d_p are the density of the water and that of the polymer, respectively. The wet membrane was obtained by immersing the membrane in distilled deionized water at room temperature for 24 h, while the dry membrane was obtained by drying the membrane in an oven at 110 °C for 3 h. Five membrane samples were used to obtain the average value of the porosity. The membranes were characterized for their hydrophilicity by measuring the water contact angle using a water contact angle meter (Face CA-D, Kyowa Kaimengaku, Japan). The measurement was conducted using distilled deionized water at room temperature, and repeated six times to obtain the average value of the contact angle.

Measurement of permeate flux and rejection through ultrafiltration experiment

Ultrafiltration experiments were performed to measure the pure water permeate flux using distilled deionized water as the feed that was pumped through a membrane cell. The membrane cell had an effective area of 51.8 cm^2. The experiment was conducted in a cross-flow mode at a trans-membrane pressure of 100 kPa at room temperature. The permeate flux J was determined from the weight of the collected permeate m_p divided by the membrane area A and the time interval Δt using the equation below:

$$J = \frac{m_p}{A\,\Delta t} \quad (2)$$

The membranes were then characterized for their ability to reject macromolecules through ultrafiltration experiments using an aqueous phosphate buffered-saline solution containing 1000 ppm BSA. BSA is a common model of solute macromolecule to study the ultrafiltration performance of the membranes [18, 19]. The BSA molecule has a molecular weight of 66 kDa that is within the MWCO range of ultrafiltration membranes. In this experiment, a feed solution with BSA concentration of 1000 ppm was used since this is the typical solute concentration for ultrafiltration experiment representing the common solute concentration of the real feed solutions in ultrafiltration processes [18]. The method to prepare the phosphate buffered-saline solution can be found elsewhere [10]. To determine the rejection R, the following equation was used:

$$R = \left(1 - \frac{C_P}{C_F}\right) \times 100\% \quad (3)$$

where C_F and C_P are the concentration of BSA in the feed solution and that in the permeate, respectively. A

Table 1 Composition of the membrane casting solutions

Membrane casting solution	Weight ratio of PET:phenol: PEG 400	PEG 400 concentration (wt%)
Membrane PET bottle	4:20:0	0
Membrane PET resin	4:20:0	0
Membrane PET-PEG-1.25	4: 20:1.25	4.95
Membrane PET-PEG-1.5	4:20:1.5	5.88
Membrane PET-PEG-2	4:20:2	7.69
Membrane PET-PEG-3	4:20:3	11.11

UV-Vis spectrophotometer (PG instrument T-60, UK) was used to measure the BSA concentration at a wavelength of 280 nm.

The membranes were also characterized for their antifouling performances by determining the flux recovery ratio (*FRR*) and the flux decline ratio (R_t) using the following equations [18]:

$$FRR = \frac{J_2}{J_1} \times 100\% \quad (4)$$

$$R_t = \left(1 - \frac{J_P}{J_1}\right) \times 100\% \quad (5)$$

where J_1 is the initial pure water permeate flux, and J_P is the permeate flux that was measured using the aqueous BSA solution as the feed. After measuring J_P, the feed solution was changed with pure water again, and the pure water permeate flux J_2 was measured after flushing the membrane for 30 min with water.

Results and discussion

Comparison of membranes prepared from used PET bottles and PET resin

It has been known that the morphology of a membrane prepared by using the phase-inversion technique is strongly affected by the polymer, the solvent, the non-solvent and the additive. In this study, the polymer used to prepare the membranes was PET. Two different sources of the polymer material were used, namely used PET bottles and PET resin which is the raw material to produce the PET bottles. Since used PET bottles are considered as waste, it is important to compare the characteristics of the membrane developed from the PET bottles and that prepared from the PET resin. The FTIR spectra of the PET membrane developed from the PET bottles are shown in Fig. 1a, whereas those from the PET resin are shown in Fig. 1b. Both membranes were prepared without additive. As can be seen, both membranes showed similar IR spectra, indicating that there is no difference in the chemical structure of the membrane from the used PET bottles and that from the PET resin. The FTIR spectra of both membranes are similar to the spectra of PET film that have been analyzed by other studies [20, 21]. Figure 2 shows the chemical structure of PET. Based on the known chemical structure of PET as shown in Fig. 2, the interpretation of the FTIR spectra of both PET membranes is listed in Table 2.

Furthermore, the microstructure of the membrane was analyzed by using SEM. Figure 3a and b show the SEM images of the cross section and surface of the membrane from used PET bottles and those from PET resin, respectively. The magnification of the SEM images in Fig. 3a and b were 1000 and 5000 times, respectively. No additive was used to prepare both membranes. Both membranes had an asymmetric structure that consists of a macroporous cross section and a smooth surface as the active layer of the membrane. As seen in Fig. 3a and b, the whole cross section of the membrane from PET bottle consisted of globular clusters and the pores were not interconnected, whereas a small part of the cross section of the membrane from PET resin had interconnected pores. The formation of the pore interconnectivity in the membrane from PET resin was possibly caused by the presence of water in the PET resin. It is known that PET resin can absorb moisture with a water content of about 50 ppm [22]. During the immersion of the cast membrane in the water-ethanol, the phase separation between PET and the non-solvent (water-ethanol) occurred. The presence of small amount of water in the PET resin affected the phase separation process since water acted as the non-solvent, and as a result a small part of the pores were connected [23]. However, in general there was almost no significant difference between the morphology of the membrane from used PET bottles and that from PET resin. This finding is in accordance with the FTIR analysis of both membranes that showed no difference of the FTIR spectra as described previously.

The membranes had an average thickness of 149 ± 13 μm as measured using a micrometer. Furthermore, the porosity and the water contact angle of both membranes are listed in Table 3. There was no significant difference between the porosity of both membranes. Both membranes also showed almost the same hydrophilicity as measured using the water contact angle method. Again, these results are in agreement with the results of the FTIR analysis and the SEM analysis as described previously.

Since both membranes prepared from used PET bottle and PET resin exhibited the same properties such as the chemical structure, the microstructure, the porosity and the hydrophilicity as described above, it can be concluded that instead of using PET resin, used PET bottles that are usually considered as waste can be utilized as the source of the polymer material to prepare the PET ultrafiltration membranes. The utilization of used PET bottles is advantageous since it will not only reduce the cost of the membrane material, but also will contribute in the efforts of plastic recycling process for a sustainable environment.

Moreover, PET has been known to have excellent mechanical properties [24]. Gupta et al. reported that the tensile strength and the Young modulus of PET membranes are 48.9 MPa and 2.25 GPa, respectively [25, 26]. Similar values of the mechanical properties of PET membranes were also reported by other researchers [27–29]. The good mechanical properties of the PET membranes are advantageous for the application of the membranes.

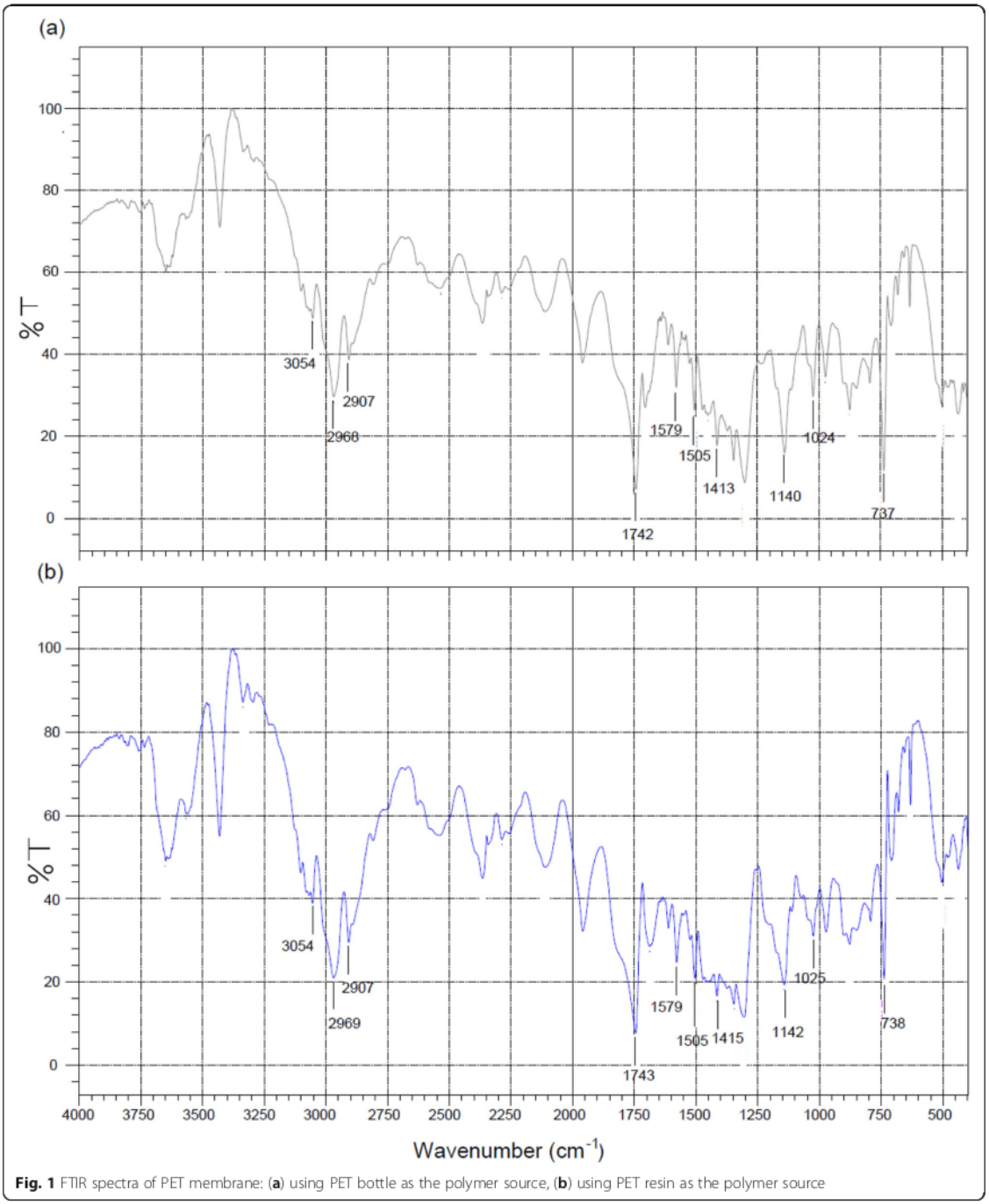

Fig. 1 FTIR spectra of PET membrane: (**a**) using PET bottle as the polymer source, (**b**) using PET resin as the polymer source

Effect of PEG 400 concentration on the microstructure, hydrophilicity and porosity

The use of additives for the preparation of membrane by using the phase inversion technique has been known to be effective to achieve the desired membrane characteristics such as microstructure, hydrophilicity, porosity, and flexibility [19]. In this study, PEG with a molecular weight of 400 Da (PEG 400) was used as the additive.

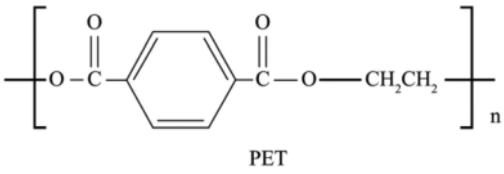

Fig. 2 Chemical structure of PET

PEG has been known as a pore forming agent for various polymers [19, 30, 31], however studies conducted by many researchers reported that low molecular weight of PEG such as PEG 200 or PEG 400 can be used as a pore-reducer rather than a pore-former [5, 32]. In our previous study, it was observed that the use of high molecular weight of PEG such as PEG 4000 resulted in the membranes with too large pore size that decreased the rejection rate of the membranes since PEG 4000 acted as a pore-former [10]. In the present work, a low molecular weight of PEG, namely PEG 400, was chosen as the additive for the PET membranes with the aim to suppress the formation of large pores. Figure 4a and b show the SEM images of the cross section and surface of the membranes with various concentrations of PEG 400. The magnification of the SEM images in Fig. 4a and b were 1000 and 5000 times, respectively. All of the membranes were prepared using PET bottles as the polymer material. All membranes showed an asymmetric structure that consisted of a macroporous cross section and a smooth surface as the active layer. Interestingly, the morphology of the membranes changed as the formation of the pores was influenced by the PEG 400 concentration. It can be observed from the SEM images that the increment of the PEG 400 concentration resulted in a smaller pore size of the membrane cross section. It has been known that low-molecular weight PEG such as PEG 400 acts as a pore-reducer for various polymer membranes [5, 19]. The formation of pores occurred when the casted polymer solution consisting of PET, phenol (the solvent) and PEG 400, was immersed in the water-ethanol as the non-solvent. Due to the solvent and non-solvent exchange, precipitation took place, and PEG 400 acted as a pore-reducer for the membrane. The growth of the pore formation was hindered when the membrane contained high concentration of PEG 400. As a result, the pore size of the of the membrane cross section decreased with increasing concentration of PEG 400. Moreover, it can be seen in Fig. 4b that the pores were partly interconnected when the PEG 400 concentration was 4.95 wt%. The pore interconnectivity disappeared when the PEG 400 concentration was increased to 5.88 wt%. As mentioned previously, during the immersion of the membrane in the water-ethanol coagulation bath, the phase separation between PET and the non-solvent (water-ethanol) occurred. During the phase separation process, the PEG 400 interacted with water-ethanol due to the hydrogen bonding. The increase in the PEG 400 concentration increased the hydrogen bonding that improved the phase separation process between PET and the non-solvent. As a result, the pore interconnectivity at the PEG 400 concentration of 5.88 wt% disappeared.

Furthermore, Fig. 5a shows the effect of the PEG 400 concentration on the membrane porosity. It can clearly be seen that the porosity of the PET membrane increased by the addition of PEG 400 as the additive. The porosity of the PET membrane without additive was 69.7 ± 0.5%, and the porosity increased sharply to 79.4 ± 0.3% through the addition of 4.95 wt% of PEG 400. A further increase in the porosity with increasing PEG 400 concentration was observed, and then the value of the porosity became stable at high concentrations of PEG 400. High values of porosity of 82.4 ± 0.4% and 82.2 ± 0.2% were achieved by adding 7.69 wt% and 11.11 wt% of PEG 400, respectively. This phenomenon occurred since PEG 400 acted as a pore-former that increased the membrane porosity as described above.

Figure 5b shows the effect PEG 400 concentration on the water contact angle of the membranes. It can clearly be seen that the water contact angle decreased significantly with increasing concentration of PEG 400. This indicated that the hydrophilicity of the membranes increased. The hydrophilic characteristic of PEG was effective to increase the hydrophilicity of the membranes. Other studies have reported a similar phenomenon for polysulfone and polyethersulfone membranes that showed an increase in the hydrophilicity by the addition of polyethylene glycol as the additive [19]. The increase in the porosity and the hydrophilicity is desirable as the ultrafiltration membranes are mostly applied for water treatment.

Table 2 Interpretation of FTIR spectra of membranes from PET bottle and PET resin

No	Wavenumber (cm^{-1})	Functional group	References
1	3054	C-H bond of the phenyl ring	[20]
2	2968–2969	C-H bond of the ethyl group	[20]
3	2907	C-O bond of the ester group	[21]
4	1742–1743	C-O bond of the ester group	[21]
5	1578–1579	C-H bond of the phenyl ring	[20]
6	1504–1505	C-C bond of the phenyl ring	[21]
7	1413–1415	C-C bond of the phenyl ring	[20]
8	1140–1142	C-O bond of the ester group	[20]
9	1024–1025	C-O bond of the ester group	[21]
10	737–738	C-H bond of the ethyl group	[20]

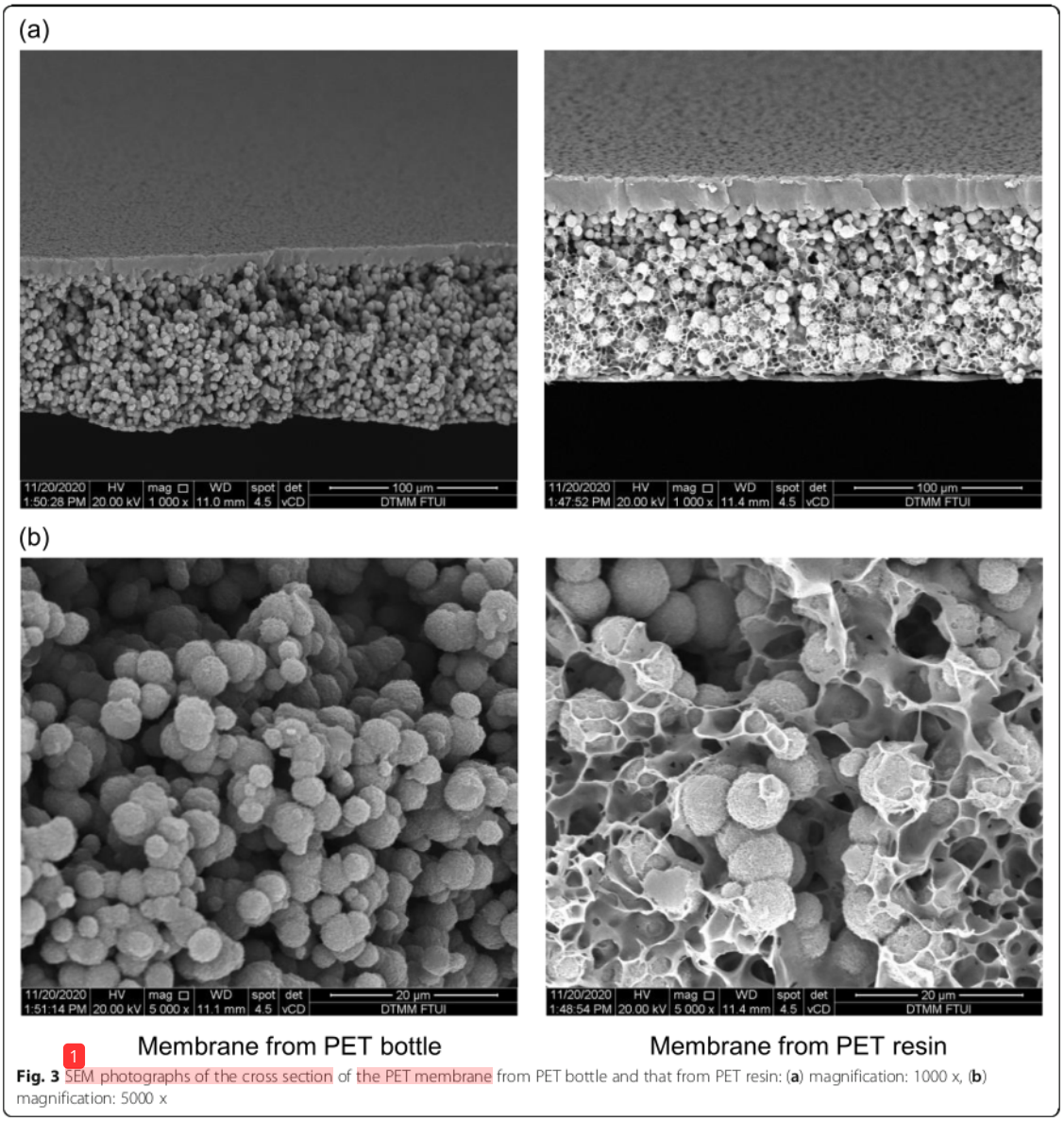

Fig. 3 SEM photographs of the cross section of the PET membrane from PET bottle and that from PET resin: (**a**) magnification: 1000 x, (**b**) magnification: 5000 x

Table 3 Comparison of porosity and water contact angle of the membrane from PET bottle and those from PET resin without additive

Membrane	Polymer source	Porosity (%)	Water contact angle (°)
Membrane - PET bottle	Used PET bottle	69.7 ± 0.5	65.5 ± 1.4
Membrane - PET resin	PET resin	71.0 ± 1.2	65.8 ± 1.1

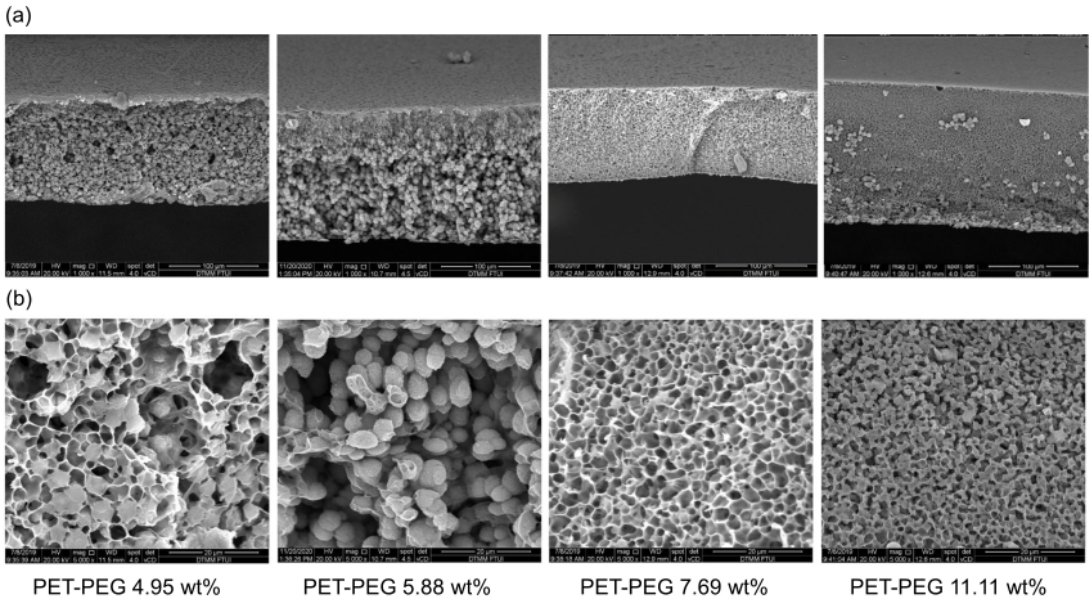

Fig. 4 SEM photographs of the cross section and surface of the PET membranes prepared from PET bottle with different PEG 400 concentrations: (**a**) magnification: 1000 x, (**b**) magnification: 5000 x

Results of ultrafiltration experiment using PET membranes

Comparison of ultrafiltration performance of the membrane from PET bottle and that from PET resin

The membranes prepared from used PET bottle and those from PET resin were then tested through ultrafiltration experiments to measure the permeate flux of pure water. The membranes prepared from PET bottle and those from PET resin with the addition of PEG 400 showed a good flexibility, since PEG acted as a plasticizer for the membranes [30, 31]. However, the membrane developed from used PET bottle and that from PET resin without PEG 400 were so stiff that they could not be fitted in the membrane cell for the ultrafiltration experiment. Figure 6 shows the permeate flux of pure water for the membrane developed from PET bottle in comparison with that from PET resin using PEG 400 as the additive. Both membranes were prepared using the same PEG 400 concentration of 11.1 wt%. It can be seen that both membranes exhibited almost the same values and profiles of water permeate fluxes as a function of the permeation time. In the beginning, the water permeate fluxes decreased with time, then they became stable after around 120 min. The decline of the permeate flux with time was caused by the physical compaction of the newly prepared membranes. The phenomenon of the physical compaction of polymer membranes has been also found in many other membranes [33–36]. As described previously, both membranes prepared from PET bottle and PET resin showed similar membrane properties such as porosity and hydrophilicity. The similar membrane properties of both membranes resulted in similar permeate fluxes during the ultrafiltration experiment. The slight difference between the morphology of the membrane from PET bottle and that from PET resin as seen in Fig. 3 did not have a significant impact on the permeate flux. This result revealed that instead of using PET resin as the source of the polymer, the PET ultrafiltration membranes could be prepared using PET bottle waste. Since PET bottle waste needs to be recycled, the conversion of used PET bottles into PET ultrafiltration membranes has great potential to contribute towards environmental conservation.

Ultrafiltration performances of PET membranes with different PEG 400 concentrations

To study the influence of additive concentration on the ultrafiltration performance, the membranes that were prepared from PET bottles with the addition of various concentrations of PEG 400 were tested through ultrafiltration experiments. Figure 7 shows the permeate fluxes of pure water for the membranes prepared from PET bottle with various PEG 400 concentrations. The membranes showed a decline of the permeate flux in the beginning of the permeation time, and then the permeate flux became stable after around 2 h, because of the physical compaction as described above. Interestingly, the membranes showed an increment of the permeate

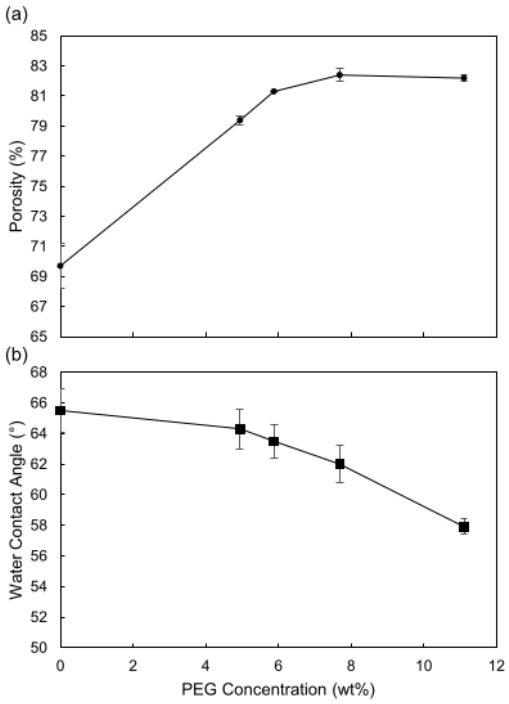

Fig. 5 a Porosity of the PET membranes as a function of PEG 400 concentration, (**b**) Water contact angle of the PET membranes as a function of PEG 400 concentration

flux when the PEG 400 concentration was increased. It can be seen that the membrane with a low PEG 400 concentration of 4.95 wt% exhibited the lowest water permeate flux. The permeate flux increased sharply as the PEG 400 concentration was increased to 5.88 and 7.69 wt%. A further increase in the PEG concentration of 11.11 wt% resulted in the highest permeate flux, however in the steady state condition the permeate flux values

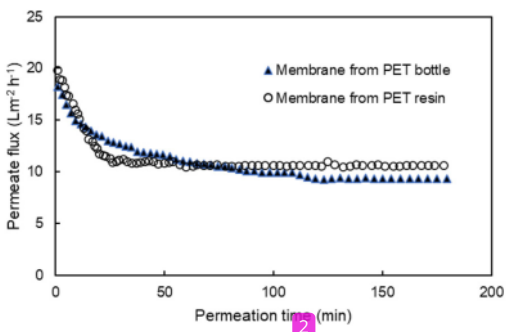

Fig. 6 Comparison of the permeate flux of the membrane prepared from PET bottle and that from PET resin with PEG 400 as the additive

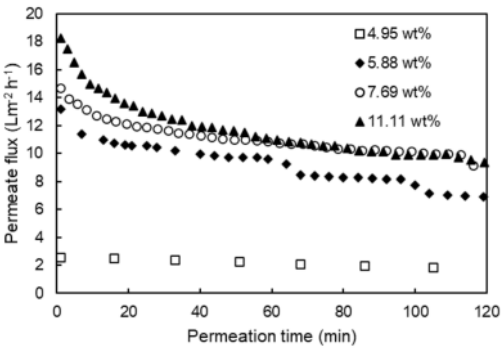

Fig. 7 Permeate fluxes of the PET membranes with various PEG 400 concentrations

became almost the same with the permeate flux of the membrane with PEG 400 concentration of 7.69 wt%. This result is in agreement with the results of the membrane characterization as explained previously. The increment of the permeate flux with increasing concentration of PEG 400 was caused by the increase in the hydrophilicity and the porosity of the membranes. As described previously, the PET membrane without PEG 400 showed a low hydrophilicity. When PEG 400 was introduced into the PET, the membrane became more hydrophilic, and water was attracted stronger onto the membrane, resulting in a higher water permeate flux. At the same time, the increment of the PEG 400 concentration also increased the membrane porosity, resulting in an increment of the permeate flux.

Furthermore, the membranes were tested through ultrafiltration experiments using an aqueous feed solution containing 1000 ppm BSA. The permeate samples were collected after attaining a steady state condition, and the rejection was determined from the BSA concentration in the permeate and that in the feed using the Eq. (3). Table 4 depicts the results of the experiments, showing the BSA rejection of the PET membranes prepared with different PEG 400 concentrations. The membrane with a PEG 400 concentration of 4.95 wt% showed a low rejection value of 61%, and the rejection increased by increasing the PEG 400 concentration. A high value

Table 4 Rejection of BSA molecules for PET membranes with different concentrations of PEG 400

Membrane	PEG 400 concentration (wt%)	Rejection (%)
Membrane PET-PEG 1.25	4.95	61
Membrane PET-PEG 1.5	5.88	73
Membrane PET-PEG 2	7.69	91
Membrane PET-PEG 3	11.11	94

of rejection rate of 94% was achieved by the membrane having PEG 400 concentration of 11.11 wt%. This result is very interesting since the increment of the permeate flux did not decrease the rejection as usually observed in the development of ultrafiltration membranes as reported in many studies [5, 14]. Here, the PET membranes exhibited an increase in both permeate flux and rejection when more PEG 400 was added into the membranes. A similar phenomenon has also been observed by Eren et al. [35], who reported an improvement of both permeate flux and BSA rejection of polysulfone membrane containing hydrophilic modifiers. In this work, the increment of the rejection rate of the PET membranes with increasing PEG 400 concentration was caused by the decrease in the pore size of the membranes as revealed by the results of the SEM analysis, whereas the increment of the permeate flux was caused by the increase in the hydrophilicity and the porosity of the membranes. A high hydrophilicity resulted in a strong sorption of water to the membranes, whereas a high porosity increased the diffusivity of water through the membrane. As an additional information, a PET membrane from used PET bottle with PEG 400 concentration higher than 11.11 wt%, namely 14.29 wt%, was also prepared, however defects of the membrane were observed so that the membrane could not be used for the ultrafiltration experiment. Thus, it was concluded that the PEG concentration of 11.11 wt% was the optimum concentration to obtain a good membrane with a good ultrafiltration performance.

Table 5 depicts the flux recovery ratio FRR and the flux decline ratio R_t of the PET membranes with various concentrations of PEG 400. The *FRR* values increased with increasing PEG 400 concentration, indicating an improved antifouling property of the membrane by increasing the PEG 400 concentration. The improvement of the antifouling property was also confirmed by the decline of the R_t values when more PEG 400 was added into the membranes.

Moreover, in future applications it is possible to recycle the used PET membranes through a mechanical recycling method to produce PET flakes that can be used as a material to make various products such as fibers, sheets, and bottles. This mechanical recycling method is widely used in recycling industries to recycle PET waste including PET films [37]. The treatment of the PET films waste includes washing, crushing, drying, and then shredding into small pieces to produce PET flakes.

Table 5 Effect of PEG 400 concentration on the flux recovery ratio and the flux decline ratio

Membrane	PEG 400 concentration (wt%)	Flux recovery ratio (%)	Flux decline ratio (%)
Membrane PET-PEG 1.25	4.95	51	80
Membrane PET-PEG 1.5	5.88	55	78
Membrane PET-PEG 2	7.69	60	71
Membrane PET-PEG 3	11.11	64	68

Conclusions

The ultrafiltration membrane prepared from used PET bottles showed the same chemical property, morphology, porosity and hydrophilicity with that prepared from PET resin, indicating that instead of using PET resin, used PET bottles that are considered as waste can be utilized as a polymer source to prepare the PET membranes. The use of additive PEG 400 for the PET membranes increased the porosity and the hydrophilicity of the membranes, but decreased the membrane pore size as observed by the SEM analysis. As a result, both permeate flux and rejection of the PET membranes were improved by increasing the PEG 400 concentration. A high value of BSA rejection of 94% was attained using the membrane prepared from PET bottles with the PEG 400 concentration of 11.11 wt%. The result of this study revealed that the low-cost ultrafiltration membranes with improved permeate flux and rejection could be prepared from used PET bottles as the polymer material with the addition of PEG 400 as the additive. The PET membranes have great potential to be applied for water treatment processes and to contribute in the environment conservation through the recycling of PET bottle waste.

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Authors' contributions

Samuel P. Kusumocahyo conducted conceptualization, methodology, supervision, writing, review and editing. Syarifa K. Ambani conducted investigation and visualization. Shelly Marceline conducted investigation, data curation, formal analysis and validation. All authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are available from the corresponding author, upon reasonable request.

Declarations

Competing interests

The authors declare they have no competing interests.

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