

Preparation of antifouling polypropylene/ZnO composite hollow fiber membrane by dip-coating method for peat water treatment

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ARTICLE INFO

Keywords:

Additive
Fouling
Hydrophilic
Peat water
Polymeric membrane

ABSTRACT

Dip-coating is a facile method for membrane modification. In this work, composite polypropylene-based membrane (PP) with improved anti-organic fouling is prepared by coating polysulfone (PSf)/PEG400/ZnO layer on PP hollow fiber membrane. Separation properties and organic fouling tendency of the composite membrane are investigated during the peat water filtration. Results show that PSf/PEG400/ZnO layer is successfully deposited on PP membrane evidenced by scanning electron microscopy and fourier-transform infrared spectroscopy analysis. ZnO particles increase membrane hydrophilicity shown by decreasing water contact angle from ~ 97.7 to 77.1 , which results in higher pure water permeability (from ~ 16 to $\sim 46 \text{ L.m}^{-2} \cdot \text{h}^{-1} \cdot \text{bar}^{-1}$). Higher hydrophilicity also reduces organic fouling tendency of the membrane during peat water treatment. The flux recovery ratio increases from 63 % to 66 % while relative flux reduction decreases from 53 % to 44 %. The composite PP membrane (M-ZnO-40 %) is able to remove ~ 70 % humic substances from peat water. Results of this study show that the addition PSf/PEG400/ZnO layer can effectively improve membrane permeability, selectivity, and anti-organic fouling of PP hollow fiber membrane.

1. Introduction

Peat water is a potential water source that can be used for supplying clean or drinking water in a remote area especially for those who live in peatland. However, peat water contains a high concentration of dissolved organic matters, thus peat water treatment process is required. Several technologies have been proposed for peat water treatment. Among the technologies, ultrafiltration membrane provides the interesting features of simple operation, less energy consumption, lower footprint, and lower cost [1–5].

In membrane operation, fouling becomes a major challenge [6–9]. Fouling is associated with the accumulation of substances on the membrane surface or within the membrane pores. This fouling not only leads to productivity decline, but also requires of additional energy supply to keep the membrane performance constant [10–12]. To solve this problem, various strategies have been proposed which include pre-treatment optimization, controlling operating conditions, and developing membrane with antifouling properties [13–17].

One of the most important identified foulants found in surface water sources is organic substances. Organic fouling on the membrane surface can be controlled by endowing membrane surface with a more hydrophilic layer. The hydrophilic surface will reduce the fouling tendency of organic substances, which are typically hydrophobic, on the membrane. Membranes with the hydrophilic surface are able to form a hydrogen bond with water molecules, thus produce water layer on the membrane surface. The presence of the water layer is able to prevent or reduce the undesirable adsorption or adhesion of foulants at the membrane surface, thus enhances water permeate flux and mitigate membrane fouling [18–20].

Modification of membrane can be done by blending of the original polymer with polymers having more suitable properties or by addition of polymer layers on the active surface of membrane. There are several methods for modifying the membrane surface such as plasma treatment, in-situ polymerization, dip-coating, etc. Dip-coating is a facile method modifying membrane surface. In previous study [21], it has been demonstrated that the dip-coating method has successfully

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<https://doi.org/10.1016/j.jwpe.2020.101158>

Received 19 September 2019; Received in revised form 17 December 2019; Accepted 17 January 2020

Available online 06 February 2020

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increased the hydrophobicity of a polypropylene (PP) membrane by creating a rougher surface. However, dip-coating method for increasing hydrophilicity of PP membrane in water treatment, especially for peat water filtration application is rarely reported.

Nanoparticles and various hydrophilic additives have been used for hydrophilic modification on PP membrane [22–29]. Among the additives, ZnO has increasingly been used due to its antibacterial and antifouling properties as well as lower cost. The ZnO particles are toxic to several bacteria, both gram-positive and gram-negative [30–34]. Therefore, ZnO can be used for two purposes, i.e. increasing antifouling and antimicrobial properties. In this work, composite polypropylene-based membrane (PP) with lower organic fouling is prepared by coating PSf/PEG400/ZnO layer on PP hollow fiber membrane via the dip-coating method. Separation properties and organic fouling tendency of the composite membrane are studied during the peat water filtration. In addition, membrane morphology and its chemical structure are also analyzed.

2. Materials and method

2.1. Materials

Polypropylene hollow fiber (PP-HF) membrane was supplied by GDP Filter, Indonesia, with average pore size was 0.05 μm and OD/ID diameter was 400/250 μm . Polysulfone (UDEL-P3500 MB7) was provided Solvay Advanced polymer, while the ZnO particle was supplied by a local supplier. Dimethylacetamide (DMAc) obtained from Shanghai Jingsan Jingwei Chemical Co. Ltd (99.9 %) was used as a solvent. Polyethylene glycol (PEG400) and ZnO particles were supplied by a local supplier. Isopropyl alcohol (IPA, Merck, ≥ 99.7 %) was used for membrane characterization. The peat water was obtained from a river at Pekanbaru, Riau, Indonesia. The peat water has a pH of 3–4, absorbance of 1.48 at 254 nm wavelength, and conductivity of 55 $\mu\text{S/cm}$.

2.2. Composite PP membrane preparation

Composite PP membrane was prepared by coating PSf/DMAc solution. Preparation steps are shown in Fig. 1. PSf and PEG400 were dissolved in DMAc. The polymer and solvent were placed in a closed glass and stirred at 200–300 rpm. Specific weight of ZnO was also added into the solution. The composition of PSf solution is summarized in Table 1. The solution was mixed until optically homogeneous. Then, the PSf solution was deposited on the PP membrane by dip-coating method. Dip-coating was carried by a home-made dip-coater as shown in Fig. 1. The PP membrane was dipped into PSf solution for 2 and 10 s and withdrawn from the solution. Afterward, the coated PP fibers were dried at room condition (~ 26 C, overnight) for removing the solvent.

Table 1
Composition of coating solution.

Membrane code	Polymer ** (wt. %)	ZnO (wt. %)
M-ZnO-0 %	100	0
M-ZnO-10 %	90	10
M-ZnO-20 %	80	20
M-ZnO-30 %	70	30
M-ZnO-40 %	60	40

** Solvent : Polymer = 90:10 (w/w); Polymer = PSf + PEG400; PEG400: PSf = 1: 4 (w/w).

2.3. Membrane characterizations

Membrane surface and cross-section were analyzed by scanning electron microscopy (SEM, JSM-6510LV) for observing membrane morphology. Meanwhile, the chemical properties of the composite membrane were analyzed by Fourier transform infrared analysis using The PerkinElmer SpectrumTwo.

Pure water permeability (PWP, $\text{L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) is determined by measuring pure water flux at various trans-membrane pressure ($\Delta P = 0.5\text{--}1.5$ bar or $0.05\text{--}0.15$ MPa). Pure water flux test was performed in a dead-end filtration mode by using demineralized water. The pure water flux, J_w ($\text{L m}^{-2} \text{ h}^{-1}$) is calculated by

$$J_w = V / (A.t) \quad (1)$$

where ($\text{L m}^{-2} \text{ h}^{-1}$), V is the volume of permeate (L), t is permeation time (h), and A is membrane surface area (m^2). PWP is the slope of $J_w - \Delta P$ curve.

Water contact angle (WCA) between a water droplet and the outer surfaces of the membrane was measured using goniometry method. Demineralized water was dropped on the membrane surface by using a micro-syringe. The image of the droplet was captured at 3rd second after dropping. The contact angles were estimated from the water droplet image by using a graphic software program. Contact angles were measured three-times at different locations of a membrane.

Porosity of the membrane was determined by a method explained in Ref. [35]. Dried and wetted membranes (in IPA for 24 h) were weighted and compared. Volume of membrane voids filled by IPA was compared to total volume of polymer + IPA. Porosity was the ratio of IPA volume to total volume.

Filtration performance of the composite membrane was carried out by using peat water. Filtration was operated under a dead-end mode at 1 bar trans-membrane pressure. After the flux was reduced to a half of the initial flux (relatively constant) (J_p), the membrane was back-washed by demineralized water for 1 min at 1 bar pressure. The pure water flux was measured (J_r). Relative flux reduction (RFR) was determined by $\text{RFR} = (1 - J_p/J_w) \times 100$ %. Meanwhile, the flux recovery ratio (FRR) was determined by $\text{FRR} = J_r/J_w \times 100$ %. The fraction of reversible fouling (R_r) was calculated by $R_r = (J_p - J_r)/J_w \times 100$ %.

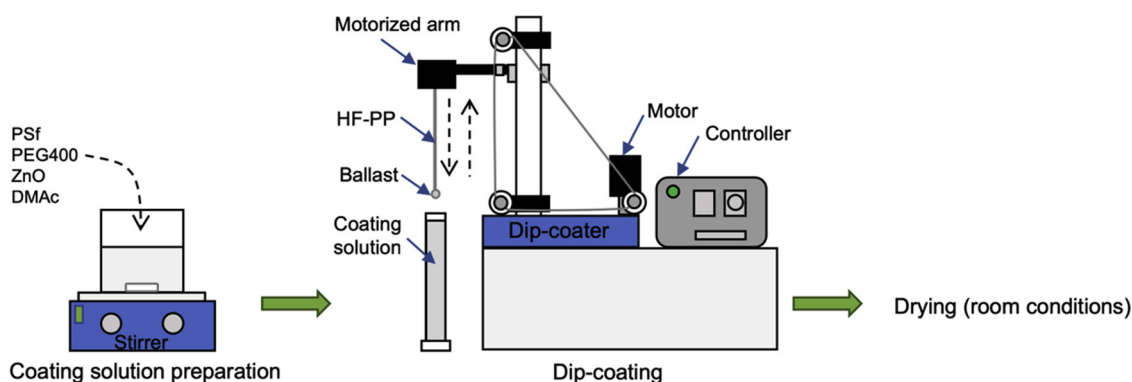


Fig. 1. Composite PP membrane preparation steps.

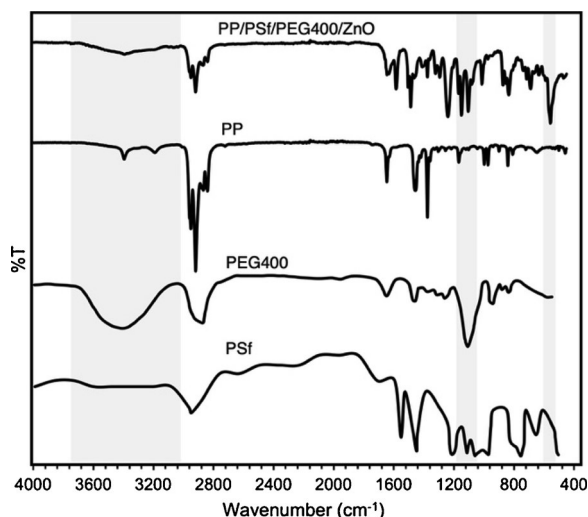


Fig. 2. FTIR spectra of composite PP membranes.

The fraction of irreversible fouling (R_{ir}) was determined by $R_{ir} = (J_w - J_r) / J_w \times 100\%$. Meanwhile, the degree of total flux loss caused by fouling was determined by $R_t = (J_w - J_p) / J_w \times 100\%$. Samples of UF feed and permeate were characterized by UV-vis spectrophotometry. Absorbance of the samples were analyzed at 254 nm wavelength. Then, the absorbances of both samples were compared to determine the ability of the UF in removing humic acid substances [36].

3. Results and discussions

3.1. Chemical structure and morphology of composite membrane

Chemical structure of the membrane was examined by FTIR and the result is shown in Fig. 2. Compared to the bear PP membrane, the composite membrane shows several new peaks. A broad band at ~ 3000 to 3600 cm^{-1} shows functional group of $-\text{OH}$ which indicates the presence of PEG400 in the membrane coating layer. The composite PP membrane also shows peaks at ~ 1200 and $\sim 1100\text{ cm}^{-1}$ which can be attributed to the sulfonic groups. These groups may be associated with the PSf. A sharp peak is also found at $\sim 560\text{ cm}^{-1}$ showing the presence of ZnO. Therefore, IR spectra of the composite membrane show that additional layer containing PSf, PEG400, and ZnO was successfully coated on the PP membrane surface. The results also indicate that the simple dip-coating method may be used as an alternative technique for preparing composite UF membrane.

SEM images of the bear PP and composite PP membranes are shown in Fig. 3. Fig. 3 (b1) shows the surface of PP membrane. The pore size of the PP membrane is about $40 - 50\text{ nm}$. After being coated by PSf, the PP membrane shows a new layer on the membrane surface (Fig. 3(a2) and (a3)). Fig. 3(b2) is the SEM image of M-ZnO-0 % membrane. At a similar magnification (10,000), the M-ZnO-0 % displays the new surface which is attributed to the PSf layer. As shown in Fig. 3(b2), the PSf layer has relatively large pores even though the PSf layer was formed by evaporation method. The formation of relatively large pores may be due to the presence of PEG400. Unlike M-ZnO-0 % membrane, M-ZnO-40 % membrane shows several aggregates on the PSf layer which represents ZnO particles. The presence of ZnO particles in the M-ZnO-40 % leads to the formation of larger pores than those formed in M-ZnO-0 % membrane. The particles decrease the interconnection of the polymer during membrane formation thus it creates voids in the polymeric layer.

3.2. Pure water permeability (PWP) and water contact angle (WCA)

Fig. 4(a) shows the PWP of the composite PP membrane. The PWP

increases with the increase of ZnO concentration. It is well known that PEG400 and ZnO are hydrophilic additives, thus the introduction of these additives results in higher hydrophilicity. It is in accordance with the results of this study where the PWP of the composite membrane is increased by ZnO. In addition, the higher ZnO results in a higher porosity (Fig. 4(a)). The higher porosity may also contribute to the improved permeability. By introducing ZnO nanoparticles, new voids may be formed between ZnO and polymer as the effect of incompatibility. Consequently, the porosity is increased by increasing ZnO content. The increase of membrane hydrophilicity is confirmed by the decrease in WCA (Fig. 4b). As shown in Fig. 4b, the WCA of M-ZnO-40 % membrane is lower than the M-ZnO-0 %. According to those results, the ZnO may have roles in increasing hydrophilicity and inducing a higher porosity.

3.3. Peat water filtration and fouling analysis

To evaluate the separation properties and organic fouling tendency, the composite membrane was used in peat water filtration. Natural peat water obtained from a local river in Riau, Indonesia was used as the feed. The filtration was operated at dead-end filtration mode. Several composite membrane fibers were used to fabricate a mini-module. PP fibers were potted in U-configuration which may simplify membrane operation for those who live in the remote area. The peat water and permeate of composite membrane were analyzed and the absorbances and photographs are shown in Fig. 5(a) and (b), respectively. The composite membrane can reduce the absorbance up to $\sim 68\%$. The decrease in absorbance value is associated with the reduction of humic substances content. In other words, it may imply that the composite membrane is able to remove $\sim 68\%$ humic substances from the peat water. Fig. 5(b) also shows that the composite membranes are able to increase the clarity of the peat water. Even though M-ZnO-40 % membrane displays a drastic increase (almost 3 times) in PWP compared to M-ZnO-0 %, the M-ZnO-40 % membrane exhibits almost a similar humic substances removal (Fig. 5c). This result indicates that combining PEG400 and ZnO particles can produce membrane with better permeability and selectivity.

Peat water contains humic substances which can cause fouling formation on the membrane surface. A rapid decrease of membrane flux was observed since the membrane was operated at dead-end filtration mode. The flux was reduced to almost half of its initial value (Fig. 6(a)). The M-ZnO-40 % membrane exhibited a lower flux reduction than the M-ZnO-0 % membrane (Fig. 6a). This may be due to the presence of ZnO which helps to reduce the fouling tendency of organic substances on the membrane surface, as ZnO increases membrane hydrophilicity [37,38]. M-ZnO-40 % membrane also display higher flux recovery than the M-ZnO-0 %. A flux recovery of $\sim 80\%$ was obtained by a simple back-flushing with the membrane permeate.

For determining fouling mechanism, several models were used: (a) cake filtration, (b) complete blocking, (c) combined cake filtration/complete blocking, and (d) cake filtration followed by complete blocking. The models show the declining water flux (J) as the function of filtration time (t). Models describing those mechanism are as follow [39,40]:

Cake filtration

$$J = J_0 (1 + 2 K_{cf} J_0^2 t)^{-1/2} \quad (2)$$

Complete blocking

$$J = J_0 \exp(-K_{cb} t) \quad (3)$$

Combined cake filtration/complete blocking

$$J = J_0 \exp(-K_{cb}/(K_{cf} J_0^2) ((1 + 2 K_{cf} J_0^2 t)^{1/2} - 1)) / (1 + 2 K_{cf} J_0^2 t)^{1/2} \quad (4)$$

where J_0 is initial water flux and K_{cf} and K_{cb} are fouling parameter for

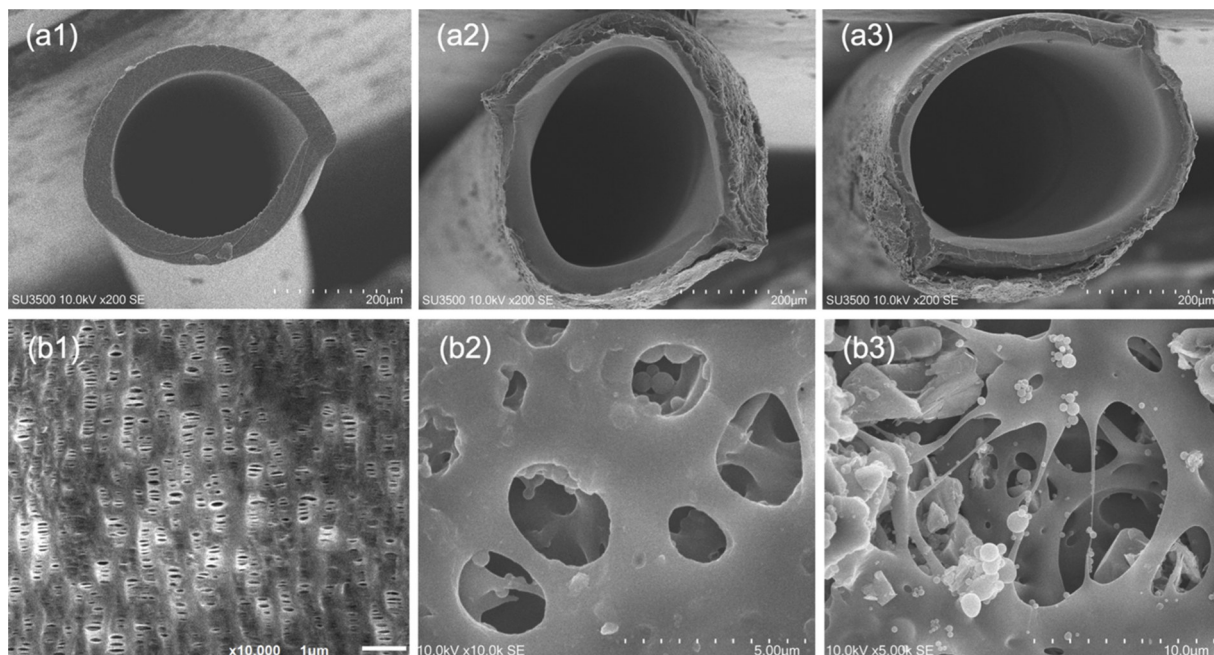


Fig. 3. SEM images of PP and composite PP membranes. (a) Cross-section, and (b) membrane surface. Membranes: (a1), (b1) = PP; (a2), (b2) = M-ZnO-0%; (a3), (b3) = M-ZnO-40%.

cake filtration and complete blocking, respectively.

Fig. 6(b) shows that model cake filtration – complete blocking gives the best result the lowest error of 6.5%. Therefore, it may imply that the fouling mechanism in the peat water filtration was cake filtration followed by complete blocking. Cake filtration occurred in the first 330 s followed by the formation of complete blocking in the next operation time.

Fouling parameters of M-ZnO-0% and M-ZnO-40% membranes are shown in Fig. 6(c). Overall, M-ZnO-40% has lower fouling parameters, i.e. RFR, Rr, Rir, and Rt, than M-ZnO-0% but higher FRR than M-ZnO-0%. This might be due to the weaker attachment of organic substances on the M-ZnO-40% than M-ZnO-0%, therefore, the foulant could be easier to remove from the M-ZnO-40% membrane surface [41]. The weaker attachment may due to the fact that more hydrophilic surface has lower membrane-foulant interaction. As the result, anti-organic fouling of M-ZnO-40% was improved due to the hydrophilic nature of ZnO.

4. Conclusion

In this work, composite PP-based membrane with lower organic fouling tendency was prepared by coating PSf/PEG400/ZnO layer on PP hollow fiber membrane via dip-coating method. Results show that the simple dip-coating method was successfully employed to deposit PSf/PEG400/ZnO layer on PP membrane surface evidenced by the results of FTIR and SEM analysis. ZnO particles could increase membrane hydrophilicity shown by decreasing WCA from ~97.7 to 77.1 which resulted in higher PWP (from ~16 to ~46 L.m⁻².h⁻¹.bar⁻¹). The higher hydrophilicity also reduced organic fouling tendency of the membrane during peat water treatment (FRR: from 63% to 66% and RFR: from 53% to 44%). In addition, the composite PP membrane, M-ZnO-40%, was able to remove about ~70% humic substances from peat water.

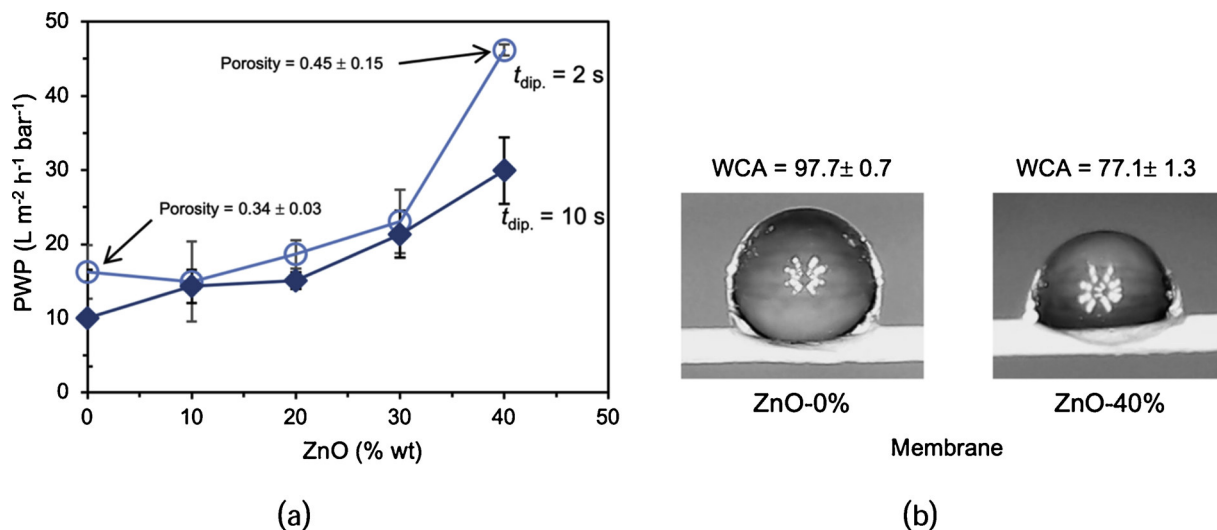


Fig. 4. (a) Pure water permeability (PWP) and (b) WCA.

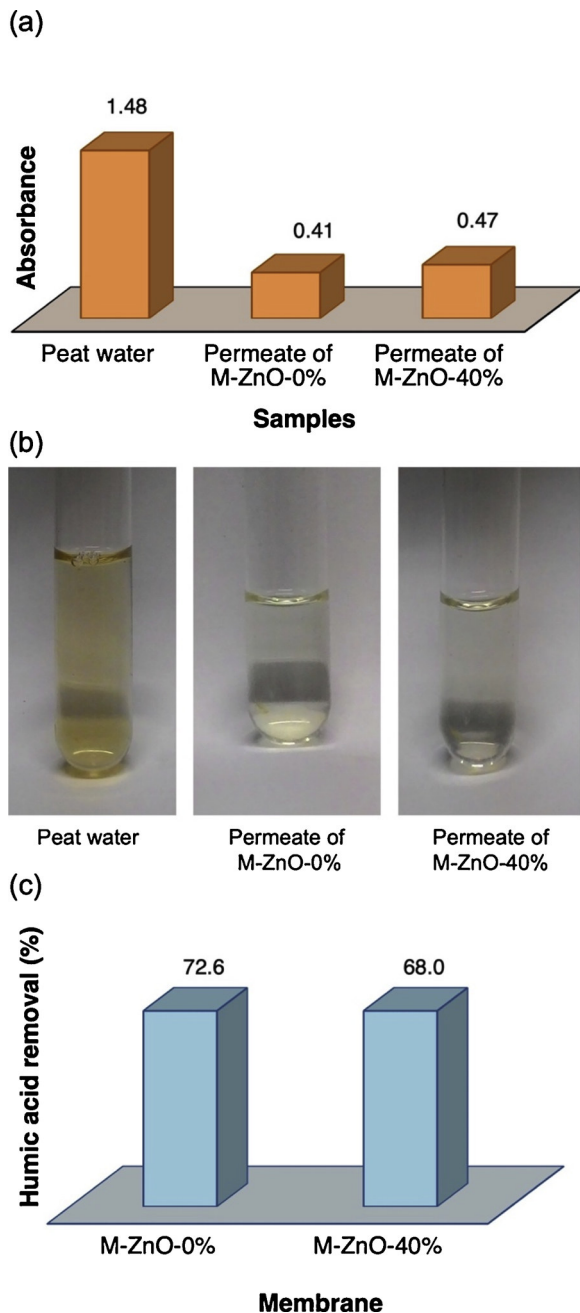


Fig. 5. Performance of composite PP membrane in peat water filtration. (a) absorbance of peat water and UF permeate at 254 nm wavelength, (b) images of peat water and UF permeate, and (c) humic acid removal.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This research is partially funded by Multidisciplinary Research Program provided by Institut Teknologi Bandung and the Indonesian Ministry of Research, Technology and Higher Education under WCU Program managed by Institut Teknologi Bandung.

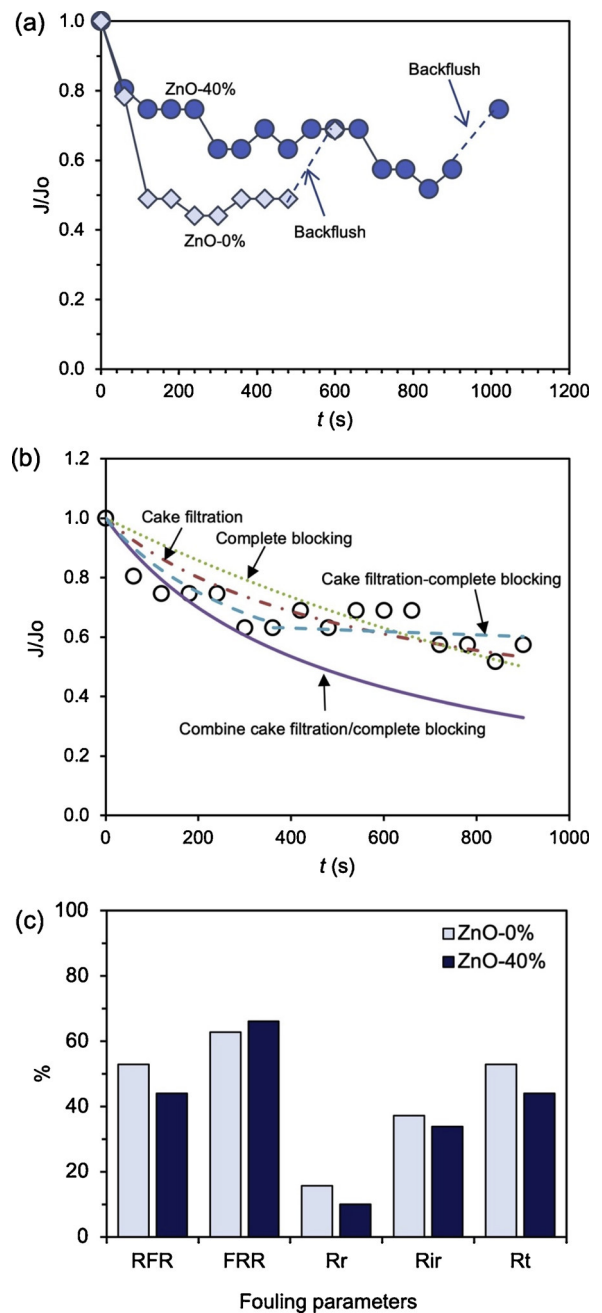


Fig. 6. Fouling on composite PP membrane during peat water filtration. (a) J/Jo vs t for membrane with ZnO-0 % and ZnO-40 %, (b) J/Jo vs t and models, and (c) fouling parameters.

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