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Article

Combination of Coagulation, Adsorption, and Ultrafiltration Processes for Organic Matter Removal from Peat Water

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Abstract: The high content of natural organic matter (NOM) is one of the challenging characteristics of peat water. It is also highly contaminated and contributes to some water-borne diseases. Before being used for potable purposes, peat water must undergo a series of treatments, particularly for NOM removal. This study investigated the effect of coagulation using aluminum sulfate coagulant and adsorption using powdered activated carbon (PAC) as a pretreatment of ultrafiltration (UF) for removal of NOM from actual peat water. After preparation and characterization of polysulfone (Psf)-based membrane, the system's performance was evaluated using actual peat water, particularly on NOM removal and the UF performances. The coagulation and adsorption tests were done under variable dosings. Results show that pretreatment through coagulation-adsorption successfully removed most of the NOM. As such, the UF fouling propensity of the pretreated peat water was substantially lowered. The optimum aluminum sulfate dosing of 175 mg/L as the first pretreatment stage removed up to 75–78% NOM. Further treatment using the PAC-based adsorption process further increased 92–96% NOM removals at an optimum PAC dosing of 120 mg/L. The final UF-PSf treatment reached NOM removals of 95% with high filtration fluxes of up to 92.4 L/(m²·h). The combination of three treatment stages showed enhanced UF performance thanks to partial pre-removal of NOM that otherwise might cause severe membrane fouling.

Keywords: coagulation-adsorption; membrane; organic matter; peat water; ultrafiltration; polysulfone

1. Introduction

The supply of high-quality freshwater is a crucial problem in rural areas. In many cases, water resources are of inferior quality (i.e., peat water), making it inconsumable without implementing advanced treatments. Peat water is one of the water sources that are still untapped. It is characteristically acidic (pH 5.9) and high in natural organic matter (NOM), identified using three standard parameters of the non-specific indicator: dissolved organic carbon (DOC, 36.40 mg/L), UV absorbance 254 nm (0.955 1/cm), and organic substances (113.76 mg KMnO₄/L). NOM in peat water may exert odors, aromatization, biological instability, and corrosion of water distribution networks [1]. Conventional water treatments for removal NOM have been widely applied by standalone processes such

as coagulation–flocculation and sedimentation [2], activated carbon adsorption [3], and filtration [4]. However, they do not provide optimal treatment for removing NOM.

Previous studies have reported types of water and wastewater treatments that contain high NOM using standalone coagulation, with about 60–70% removal of hydrophobic fraction of NOM and 30–40% of hydrophilic fractions [5–8]. Another work also reported performance of adsorption for NOM removals of up to 98% that were obtained by powdered activated carbon (PAC) with an optimum dosage 500 mg/L [9], which can remove organic materials with a molecular weight (MW) ranging from 0.5–1 to 1–3 kDa. Nevertheless, it could not remove NOM with an MW of <0.5 kDa [10,11]. In addition, the adsorbent may be saturated due to the complete occupation of the adsorption site, while reactivation of the adsorbent results in a complex operation, which may lead performance to decrease [12].

Membrane technology is an advanced treatment process for treating NOM in water, such as wetland or peat water [13–17]. Several studies were reported successful treatment of wetland saline water by pervaporation using silica-based membranes [18–21], wetland saline water by pure silica membrane and organosilica-based membranes [22–28]. Another study showed ultrafiltration (UF) membrane for removal fraction of NOM from peat water [29]. The UF technology is more applicable and better for reducing NOM in water compared with pervaporation. The pervaporation setup is more complex than UF. However, despite NOM's effective removal by the membrane, it is also able to decline membrane performance through membrane fouling [30,31]. Fouling is a major factor that may decrease membrane flux during the separation process, especially the ultrafiltration.

Membrane fouling in peat water treatment is mainly caused by NOM through both the hydrophilic and the hydrophobic fractions [32]. The most common methods to reduce membrane fouling are by altering the physical and chemical properties of the membrane materials by adding additives in the fabrication stage [33–36] or by applying pretreatment of the feed in the operational stages [32,37]. In this study, both coagulation and adsorption were investigated for the first time as a pretreatment of UF for the treatment of real peat water.

This paper reports a preliminary study on NOM removal from peat water by using both aluminum sulfate-based coagulation and PAC-based adsorption as a pretreatment of UF. The polysulfone (Psf) UF membrane was first prepared and characterized. Before being used for the pretreated peat water filtration. The NOM composition in the peat water samples was then characterized. Finally, actual peat water was treated using a series of treatments, namely aluminum sulfate-based coagulation, PAC-based adsorption, and filtration using the developed Psf-UF membrane.

2. Materials and Methods

2.1. Peat Water Characterization

The peat water sample was taken from Banjar Regency, South Kalimantan, Indonesia. Preliminary characterization of peat water included measuring pH using a pH meter (Hanna Hi2211), KMnO_4 —oxidizable organic substances using the permanganate titrimetric method, and aromatic organic matter absorbance of UV_{254} , and DOC by a total organic carbon analyzer (Shimadzu TOC-L). The permanganate titration method was conducted according to Standard (SNI 06-6989.22-2004). The UV_{254} parameter was measured by a UV visible (UV-1600 Spectrophotometer). On the other hand, DOC was analyzed by high-temperature catalytic oxidation with non-dispersive infrared (NIDR) detection. As a pretreatment, the samples were filtered using Whatman 0.45 μm before being tested by TOC analyzer. Meanwhile, specific UV absorbance (SUVA_{254} , $\text{L}/\text{mg}\cdot\text{m}$) was used to represent TOC normalized aromatic moieties (UV_{254}). Meanwhile, specific UV absorbance (SUVA_{254} , $\text{L}/\text{mg}\cdot\text{m}$) was used to represent TOC normalized aromatic moieties (UV_{254}) by dividing of the UV_{254} with the DOC value.

2.2. Membrane Preparation and Characterization

The dope solution for Psf UF membrane preparation was made using 18 wt.% of Psf (Merck) as the polymer, 64 wt.% dimethylacetamide (DMAc, Merck) as the solvent, and polyethylene glycol (PEG, Merck) PEG 600 as the additive (18 wt.%). According to earlier reports, the membranes were prepared using the phase inversion method [35]. The polymer, solvent, and additive were mixed and stirred until homogeneous. Then, the solution was left idle overnight to release the entrapped bubbles. Subsequently, the dope solution was cast on a glass plate at a wet casting thickness of 165 μm using a casting applicator. The phase inversion was then continued by immersing the cast film into a coagulation bath containing nonsolvent solution comprising of DMAc 35 wt.% and KCl 0.5 wt.% in water.

The hydraulic resistance of the prepared membrane was characterized by measuring the clean water permeability, and a scanning electron microscopy (SEM) was used to determine the membrane Psf morphology and membrane thickness. The pore size of the membranes was determined using image-J software from the surface of the SEM image [38]. The permeability test was conducted by flowing the distilled water on a dead-end system filtration device. The permeate volume was then measured every 5 min intervals for 60 min operation time under different pressures of 1, 1.5, 2, 2.5, and 3 bar.

2.3. Coagulation, Adsorption, and Ultrafiltration

The coagulation tests were done by varying doses of aluminum sulfate (one of the most common coagulants) in a range of 125–250 mg/L using the Jar-test method at adjusting pH 6 (regulated by drop-wise adding 0.1 M NaOH (Merck)) with a working volume of 1.2 L. During the jar test, the coagulant mixture in peat water was stirred at 100 rpm for 1 min, followed by slow stirring at 40 rpm for 20 min and sedimentation for 20 min, according to a protocol reported earlier [39]. The range of the coagulant dosage was defined based on a previous study [31].

The PAC adsorption tests were done using the Jar-test under varying 20–200 mg/L doses. It was carried out for the pretreated peat water through coagulation/flocculation. The feed and PAC (particle size of 100 mesh; surface area of 800 m^2/g , Merck) were mixed with a rotary shaker at 180 rpm for 3 h.

After coagulation and adsorption, the treated supernatant underwent a UF—200 mL of the supernatant was filtered using the developed UF Psf membrane by using a standard dead-end filtration cell (Figure 1) according to a protocol detailed elsewhere [30]. The filtrations were done at variable pressures of 1, 1.5, 2, 2.5, 3 bar for 60 min at room temperature of feed (25 $^{\circ}\text{C}$), stirred at 50 rpm. A gas compressor generated the pressure, and the permeate volume was collected every 5 min. All of the coagulation, adsorption, and filtration experiments were done in triplicate.

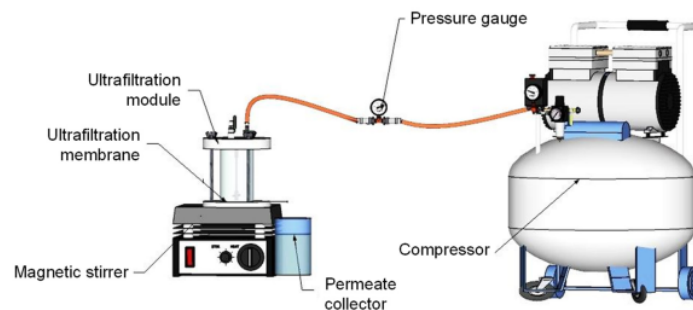


Figure 1. Illustration of the ultrafiltration dead-end experimental setup.

3. Results

3.1. Peat Water Characteristics

The characterizations of peat water were carried out for four periods to monitor the changes of NOM content of the peat water samples, as summarized in Table 1. It shows that the peat water had a neutral pH, similar to previous reports where the pH value on surface water ranged from 5.0–8.1 [31]. The high NOM content was indicated by the DOC values [39,40], the absorbance value of UV_{254} , which are high compared with the results obtained by Kang and Choo [41] and Jeong et al. [42] of $UV_{254} < 0.1 \text{ cm}^{-1}$ for surface water. However, compared with the results obtained elsewhere Herwati, Mahmud, and Abdi [30], Mahmud, Abdi, and Mu'min [31], Saputra [37], Aisyahwalsiah [39] showed the UV_{254} absorbance of the peat water samples was relatively low. Similar to the UV_{254} absorbance value, the $SUVA_{254}$ values of the peat water sample deviated from others (Zularisam et al. [43]). Based on their reports, the $SUVA_{254}$ characteristic of peat water contained a high hydrophobic fraction. The $SUVA_{254}$ values of the peat water sample in this study ranged at 2–3 L/mg.m suggesting the mixture of hydrophobic and hydrophilic substances NOM characteristics, with a large range of MWs. Similar results were found in previous research on surface water that reported $SUVA_{254}$ of $< 2 \text{ L/mg.m}$ (low hydrophobic character) [41,42].

Table 1. Characteristics of the peat water sample.

No	Parameter	Units	Week				Average	STDEV
			I	II	III	IV		
1	pH		6.3	6.3	6.3	6.3	6.3	0
2	DOC (dissolved organic carbon)	mg/L	36.40	-	-	36.40	36.4	-
3	UV_{254} absorbance	1/cm	0.968	1.005	0.977	0.955	0.976	0.02
4	$KMnO_4$ organic substances	mg $KMnO_4/L$	120.08	126.4	120.08	113.76	120.08	5.16
5	$SUVA_{254}$	L/mg.m	2.659	2.761	2.684	2.624	2.682	0.006

3.2. Characterisation of UF-PSf Membranes

The surface and cross-section SEM images of the prepared membrane are shown in Figure 2. The UF PSf membrane had a tight pore arrangement and sponge-like cross-section morphology without macrovoids (large cavities), which was similar to the membrane structure reported earlier [35]. Based on image-J surface SEM image processing (Figure 3), the surface pore size of the membrane was $0.061 \mu\text{m}$, falling under UF range of $0.001\text{--}0.1 \mu\text{m}$ [44].

The polysulfone membrane used in this study had a pore size of less than $0.1 \mu\text{m}$ (Figure 3). However, from the SEM image it is not possible to determine exactly what the pore size is. However, the pore size distribution can determine by utilized Image-J software following the previous research used digital SEM image data. The result of processing SEM images by Image-J can be seen in Figure 3B,C. The results show the average pore diameter of the membrane is $0.061 \mu\text{m}$. Based on literature, the polysulfone membrane used in this work can be categorized as well as ultrafiltration membrane.

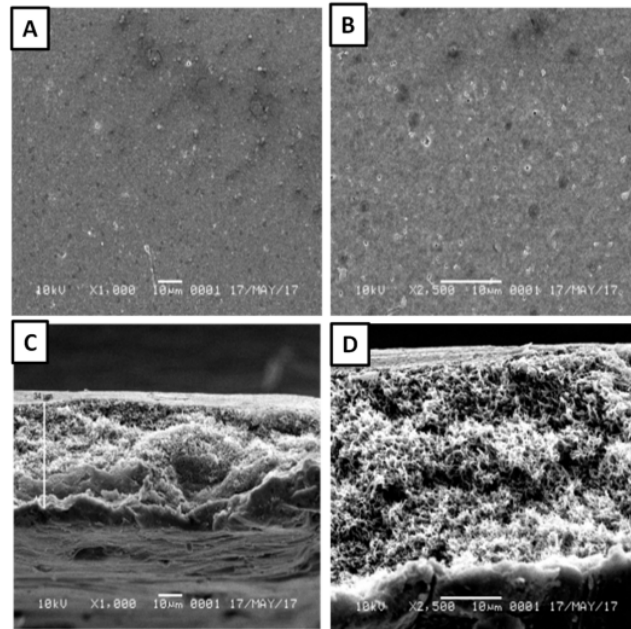


Figure 2. Ultrafiltration polysulfone membrane surface microstructure with (A) magnification of 1000× and (B) magnification 2500×, and cross-section microstructure with (C) magnification of 1000× and (D) magnification 2500×.

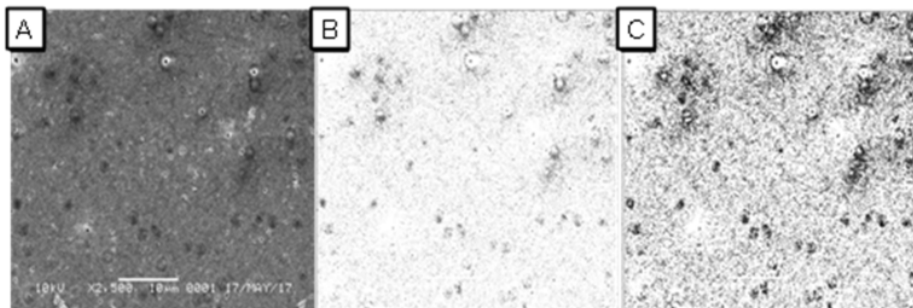


Figure 3. Membrane surface SEM image (2500×) processed with Image-J for pore size determination (A) before editing, (B) after threshold and (C) outline image result.

3.3. Coagulation-Flocculation

Figure 4 shows that for aluminum sulfate dosings of 125–250 mg/L, the removal efficiency of NOM increased from 125 mg/L to 175 mg/L and then a slight decrease until 250 mg/L. Beyond that value, the NOM removal efficiency decreased. The loading restabilization can explain the pattern on the NOM removals as a function of the dosing rate on the addition of $\text{Al}_2(\text{SO}_4)_3$ coagulant [37].

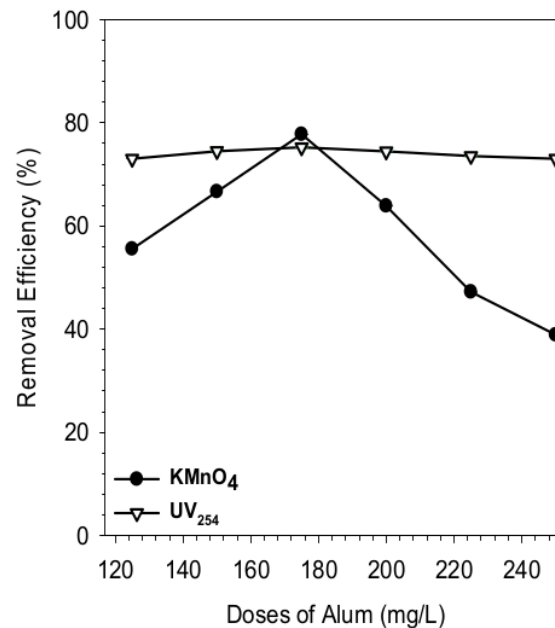


Figure 4. NOM removal rate represented by oxidation with KMnO_4 and UV_{254} absorbances as a function of doses of alum in the coagulation–flocculation test.

The oxidation of organic substances by KMnO_4 peaked at the optimum dose of 175 mg/L. Meanwhile, a slight decrease in UV_{254} removals was observed. In the process of coagulation–flocculation, the dominant fraction of removed NOM is the one that hydrophobic or with large MWs as detailed elsewhere [31,45]. In addition, according to Suslova et al. [46] that KMnO_4 can oxidize various types of organic components irrespective of the MWs.

The NOM removal in the coagulation–flocculation process was achieved optimum at a dose of 175 mg/L corresponding to organic substances KMnO_4 and UV_{254} absorbance of 77.78% and 75.24%, respectively. The removal of NOM obtained in this study was higher than the previous studies [47,48]. After adding the coagulant, the coagulation rate decreased, as well as the pH value from 6 to 3.65. It was caused by the reaction of aluminum sulfate with water that produces H^+ ions. The acidification of water lowered the coagulation/flocculation efficiencies.

3.4. Coagulation-Adsorption

The PAC adsorption was carried out after the coagulation/flocculation of the raw peat water sample. Figure 5 shows the rate of NOM removal in the PAC adsorption process. The NOM removal rate was higher than the standalone coagulation–flocculation or adsorption processes reported earlier [32,49]. Increased NOM removal at low pH during the PAC adsorption can be attributed due to the low pH of the solution due to the preceding coagulation/flocculation stage [39]. The pH has a significant effect on activated carbon adsorption and the removal efficiency is higher in acidic than in neutral and alkaline conditions. The presence of H^+ ions in solution leads to competition between H^+ ions and NOM bonding [50].

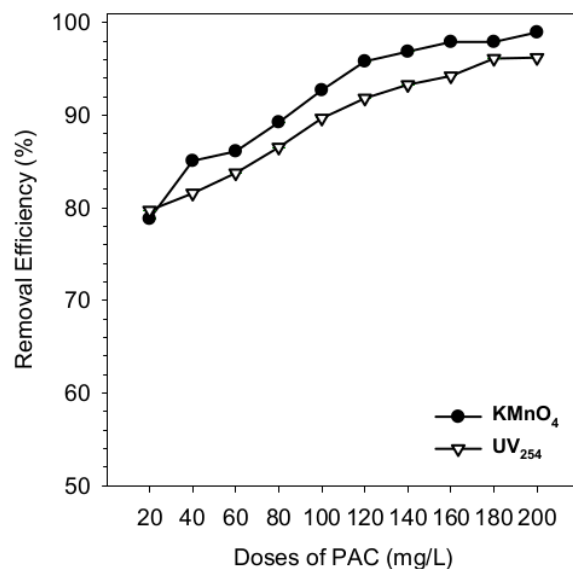


Figure 5. The NOM removal rate represented by organic substances of KMnO₄ and UV₂₅₄ absorbances as function of PAC dosages.

The removal rate of KmnO₄—oxidizable organic substances in the coagulation–adsorption process is higher than the UV₂₅₄ (representing aromatic moieties). The results were the opposite of the coagulation/flocculation process, which could be attributed to the NOM content with large MW removed in the coagulation–flocculation process. As such, only the low MW NOM remained in the PAC adsorption. In previous research, the adsorption process could remove NOM hydrophilic fractions with small MWs [9–11]. The removal rate of KmnO₄ organic substances was higher than the UV₂₅₄ absorbance. These results indicated that KmnO₄ organic substances easily oxidized NOM due to their small MWs as stated elsewhere [51].

The best PAC dose was 120 mg/L of PAC, judging from the highest removal of KmnO₄ and UV₂₅₄ parameters of >90%. In addition, the efficiency of NOM removal slightly increased up to 120 mg/L. Due to the NOM removal of 120 to 200 mg/L being relatively similar, the 120 mg/L of PAC was chosen because it does not need too much PAC. The removal efficiencies of KMnO₄ organic substances and UV₂₅₄ under the optimum dosing were 95.83 and 91.83%, respectively, as shown in Figure 5. The results obtained in this study were in line with others, e.g., Lee et al. [32] and Aisyahwalsiah [39] using PAC as the adsorbant. Nonetheless, higher NOM removals were obtained in this study. After adding PAC, the pH increased as reported by others [49] due to the soluble ash, which is rinsed out of the media during use, and the effluent pH will eventually approach neutral.

3.5. Coagulation-Adsorption-Membrane Experiments

The permeability of pure water (aquadest), pretreatment feed, and non-pretreated feed are shown in Figure 6. Based on the results, pure water permeability was obtained of 38–180 L/h.m² by the prepared UF polysulfone membrane. This result exhibits the higher water flux of pure water permeability by low transmembrane pressure compared to commercial polysulfone membrane (Merck) of 150–350 L/h.m² (6–20 bar), which was reported by Adams et al. [52].

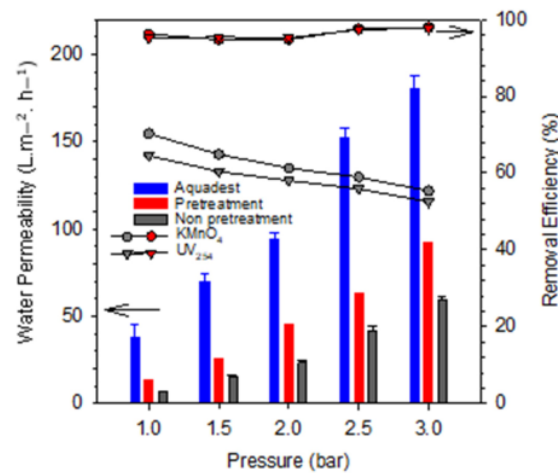


Figure 6. The performance of ultrafiltration of pretreated peat water at different pressures. Aquadest denotes distilled water and represents the permeability of clean water.

Figure 6 shows that the NOM removal efficiency decreased by increasing transmembrane pressure. The results obtained are by previous research, in which the magnitude of NOM rejection is inversely proportional to the applied pressure [53,54]. The deformation of the membrane most probably causes it due to high pressure, which causes membrane compaction that constricts the pore size and the thicker foulant layer that became the secondary filter on top of the PSf membrane.

The NOM removal efficiency reflected from the KMnO₄ organic substances was higher than the UV₂₅₄ absorbance obtained in the adsorption process with PAC. The rejection of NOM by membrane was determined by the adequate pore size [53] and an additional dynamic layer formed on the membrane surface.

In addition to the NOM removal rate, water flux value was also an indicator of the optimum pressure. Figure 6 shows that the water flux value was directly proportional to the pressure. The smallest water flux value was obtained at 1 bar of 13.3 L/h.m² and the highest water flux was at 3 bar of 92.5 L/h.m². The permeability of each pressure to percent removal of NOM for KMnO₄ organic substances and UV₂₅₄ parameters were determined to determine the optimum pressure. The highest water flux was obtained with the removal rate of KMnO₄ and UV₂₅₄ of 94.79 and 94.66%, respectively. The UV₂₅₄ rejection of the polysulfone membrane in this work is extremely high over commercial PSf membrane that was only able to remove about 41% of NOM at 6 bar [52].

The water permeability of treated peat water was smaller than the clean water permeability (Figure 6) due to membrane fouling. However, it was higher by almost two-fold than peat water permeability without pretreatment, which was also similar reported in earlier studies [31,41,54]. It was shown that the pretreatment contributed substantially to reducing the membrane fouling [53].

The permeability decrease in the pretreated peat water filtration can be attributed to the fouling by the residual NOM that escaped from the pretreatment. However, previous works by Kang and Choo [41] and Zhang, et al. [55] ascribed the small water permeability to the use of PAC. The bonding of NOM with PAC particles caused the PAC-NOM particles to become an additional foulant that blocks the membrane pores or forms a cake layer on the membrane surface. In this study, the PAC was separated. Hence the foulant was originated from residual NOM in the feed.

Overall findings suggested that the application of coagulation–adsorption pretreatment of UF is promising to reduce the fouling potential on the feed as indicated by in-

creasing the water permeability value and the removal rate of NOM represented by DOC, KMnO_4 organic substance, and absorbance UV_{254} .

In addition, the results obtained were also reinforced with SEM UF-PSF membrane image after treatment. The SEM images in Figure 7 also show the thickness of the UF-PSF membrane (determined from the cross-section SEM image) after being compressed at 3 bars was $85.4\ \mu\text{m}$. The pore structure of the membrane after passing the feed water was approximately the same as the pristine membrane. It could be seen on the surface of the membrane there is only a thin layer which is thought to be a cake layer. The additional cake layer helped enhance the rejection of NOM and the final quality of the permeate. It is worth noting that a significant difference in thickness was seen from data in Figures 2C and 7B. The high variability was originated from the cutting process.

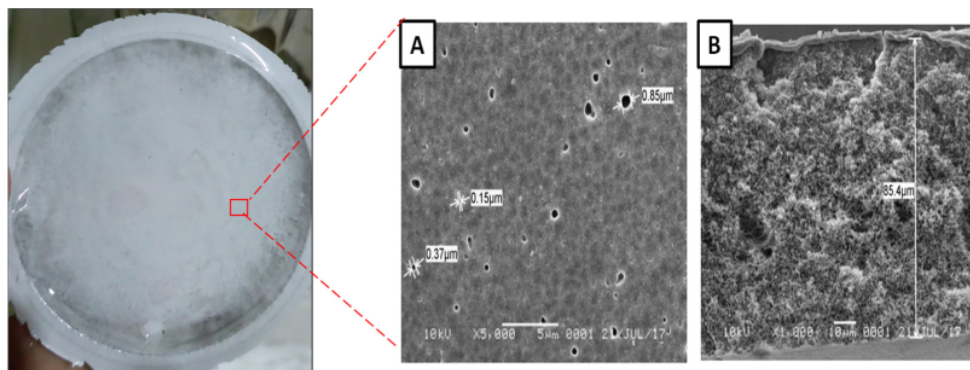


Figure 7. SEM image of used ultrafiltration polysulfone membrane after ultrafiltration process: (A) surface section and (B) cross-sectional.

4. Conclusions

This study demonstrated the advantages of combining the coagulation–adsorption process and membrane filtration to treat fouling-prone actual peat water. The coagulation-adsorption showed a positive effect as a pretreatment for the ultrafiltration. The pretreated feed showed a lower membrane fouling propensity. The optimum coagulation/flocculation and adsorption condition was at $\text{Al}_2(\text{SO}_4)_3$ dosing of $175\ \text{mg/L}$ and PAC dosing of $120\ \text{mg/L}$, respectively. Higher filtration pressure enhanced the peat water permeability. The optimum pressure on the hybrid process was 3 bar with a permeability value of $92.5\ \text{L/m}^2\cdot\text{h}$ and an organic removal rate of 95%. The findings highlight the importance of the hybrid system for treating challenging feeds that otherwise proven difficult when applying a standalone system. Moreover, long-term studies are still required to accurately gauge the performance of the hybrid system for treatment of peat water.

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