



Chemical cleaning of flat sheet ultrafiltration membranes fouled by effluent organic matter



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ABSTRACT

This paper examines the efficiency of chemical cleaning of flat sheet UF membranes severely fouled by rendering plant secondary effluent. The rendering plant secondary effluent was treated in a laboratory cross-flow batch ultrafiltration (UF) set-up with three membranes (MW, CQ, and GM). Effluent organic matter (EfOM) caused a severe fouling, which led to an increase in hydraulic membrane resistance (HMR), 6.5–11.3 times the HMR of pristine membranes (R_m). In the first part, the fouled membranes were flushed with water and cleaned by chemical cleaning in two phases. The first phase included flushing and soaking with a commercial alkali cleaning agent, as in the second phase a commercial acidic cleaning agent was used. After each phase, the HMR was determinate and one membrane was removed and characterized (FTIR and SEM). The chemical cleanliness was determined from the FTIR spectra; while the hydraulic cleaning efficiency (HCE) was determined from the drop in HMR. In the second part, the cleaning was performed with individual cleaning agents. The alkali agent was tested at 25 °C and 35 °C as well as after the UF at different pressures. The alkali cleaning agent had the greatest impact on the cleaning of membranes fouled by EfOM.

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1. Introduction

The reclamation of industrial and municipal wastewater represents a substantial source of water that minimizes the environmental impact [1]. Membrane technologies can achieve the water quality needed for reclamation in various industries [2–6]. Wastewater reclamation with membrane technologies often includes multiple phases. Ultrafiltration (UF) and microfiltration (MF) are often used as the first phase. However, UF is preferred [7], as it is better at preventing the fouling formation on nanofiltration (NF) or reverse osmosis (RO) membranes in the second phase [8]. Ultrafiltration is applied during or after the biological treatment [7,9], with or without additional pretreatment [9,10]. Nonetheless, the UF of secondary effluents is accompanied by severe fouling caused by effluent organic matter (EfOM) [11].

The EfOM consists of three main fractions: (i) soluble microbial products (SMPs) secreted by microorganisms during biological treatment, (ii) natural organic matter (NOM) from drinking water sources, and (iii) trace levels of synthetic organic compounds [12,13]. Depending on the study, the main cause of fouling during UF was either hydrophilic colloidal compounds [14] or hydropho-

bic SMPs [13]. However, a recent study, Zheng et al., 2014 showed that biopolymers from the SMPs fraction, as an isolated fraction, have greater impact on the fouling phenomenon than the other constituents [11]. Other than the impact of specific fractions, a synergistic effect is present. Even in small concentration, extracellular polymeric substances (EPSs), which are represent a fraction of SMP, increase the fouling potential of other fractions [15]. Furthermore, the concentration of EfOM has a great influence on the fouling mechanism. Esfahani et al., 2015 argued that at higher concentrations of humic acids and proteins form larger aggregates causing external pore-blocking and cake-formation, a more reversible fouling; while at low concentration they form smaller aggregates that can penetrate in the pores causing internal pore-blocking, a fouling less prone to removal [16]. An effective fouling control would facilitate the application of UF for the treatment of secondary effluents.

There are several approaches to membrane cleaning. The adequate choice depends on the foulants, compatibility of the cleaning agent and membrane, and its cost-effectiveness. The EfOM causes hydraulically irreversible fouling [11,17–19], especially for flat sheet membranes where back-flushing is often not allowed due to structural vulnerability; hence, chemical agents should be added to promote fouling removal. Chemical cleaning removes the fouling by soaking and flushing the membrane with a solution that can contain acids, alkalis, oxidants, surfactants, chelants, and

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enzymes [20]. If a commercial chemical agent is used, its composition is often unknown to public domain [21]. The choice of chemical agents depends on the properties (composition) of the fouling layer among other parameters. Alkalis remove proteins and polysaccharides (the main foulants in the secondary effluent), while acids remove inorganic material [20,22]. So, a combination of chemical cleaning with alkali and acid agents should effectively remove the fouling caused by EfOM.

Even though UF of secondary effluents can cause severe fouling, other than studies on pretreatment methods [9,23] and fouling formation [12,14,15,18], only few recent studies on chemical cleaning of flat sheet membranes fouled by secondary effluents [24–27] were conducted. Those studies applied chemically enhanced back-flush with two chemical agents (NaOH [26] and NaOCl [24,25,27]) on one type of UF membrane, i.e. PTFE, achieving removal efficiency up to 80%.

This paper examines the chemical cleaning-in-place of fouling formed on three flat sheet membranes (MW, CQ, and GM) during UF of rendering plant secondary effluent. Cleaning-in-place was conducted by flushing and soaking the fouled membranes with a solution of an alkali and an acidic cleaning agent. Other than flux recovery, physical-chemical cleanliness of membranes after each cleaning phase was examined by FTIR and SEM. Moreover, to determine more favorable operation conditions, the impact of different working pressure during UF and temperature of the alkali agent were tested.

2. Material and methods

2.1. Membranes

Three commercial flat sheet UF membranes (MW, CQ, and GM from GE Osmonics, USA) were used. The main membrane characteristics are given in Table 1. The membranes are made of different polymers, with a wide range of molecular weight cut-off (MWCO) (8000–50,000 Da).

2.2. Secondary effluent

The rendering plant wastewater from Agroproteinka, Sessvetki Kraljevec, Croatia was subjected to biological treatment in a sequential batch reactor (SBR) with a hydraulic retention time of 7 h. The samples of secondary effluent (50 L) was taken and kept in plastic containers at low temperature (<10 °C) and used within 2 weeks. The samples of secondary effluent were characterized for dissolved organic carbon (DOC, sample 1: 29.67 mg L⁻¹, sample 2: 50.45 mg L⁻¹) and inorganic carbon (IC, sample 1: 39.32 mg L⁻¹, sample 2: 4.18 mg L⁻¹), along with the turbidity (sample 1: 4.1 NTU, sample 2: 24.38 NTU), conductivity (sample 1: 1111 µS cm⁻¹, sample 2: 1010 µS cm⁻¹), chemical oxidation demand (COD, sample 1: 116 mg L⁻¹, sample 2: 75.25 mg L⁻¹) and pH (sample 1: 6.89, sample 2: 6.29).

2.3. Cleaning agents

Two commercial chemical cleaning agents were used, PermaClean 77 (PC 77) and PermaClean 99 (PC 99) from NALCO, Netherlands. The PC 77 is an acidic formulation of acid cleaners and sequestrants developed for the removal of inorganic fouling such as iron oxide and calcium carbonate. The PC 99 is an alkali formulation (tetrapotassium pyrophosphate 5–10%, potassium hydroxide 5–10%) of dispersants and inorganic builders with amphoteric surfactants developed for the removal of organic debris and biofouling.

2.4. Filtration set-up

Ultrafiltration was conducted in a batch cross-flow laboratory set-up with a detailed description in Dolar et al., 2011 [28]. Six UF cells, with equal membrane surface area (11 cm²), were connected in parallel. Prior to the experiment, the membranes were washed with demineralized water (10 L) and pre-compress for 6 h at the corresponding working pressures (Table 1). The UF of the secondary effluent was conducted for 8.5 h for sample 1, while during the testing for the influence of pressure (2, 4 and 6 bar), temperature (25 and 35 °C) and type of cleaning agent (PC 99 and PC 77) on MW, UF was conducted until 0.5 L of permeate was collected.

2.5. Chemical cleaning conditions

In the first part the membrane cleaning was performed with demineralized water, 1% solution of PC 99, and 1% solution of PC 77. Initially, the membranes were flushed with 25 L of demineralized water for 30 min. During the first phase of chemical cleaning, a 5 L of 1% solution of PC 99 at 33.7 ± 2.5 °C were recycled at working pressure for 30 min, followed by 30 min of soaking and an additional flushing with 10 L of demineralized water to remove the cleaning agent. The final phase repeated the previous procedure, but with the solution of PC 77. One membrane was removed after each cleaning phase to characterize the membrane surface.

In the second part the cleaning of MW membrane was performed with the same protocol, but with a single cleaning agent and by varying one parameter: working pressure (2, 4 and 6 bar) or temperature of cleaning agent (25.7 ± 0.8 °C and 34.4 ± 1.7 °C).

2.6. Membrane surface analysis

To characterize the fouling layer and chemical cleanliness of the membrane; the surface of pristine, fouled, and cleaned membranes were analyzed by FTIR spectrometer (Bruker Vertex 70, Germany) equipped with a Platinum ATR single reflection diamond (n = 2.4) crystal-based module in the mid IR range (400–4000 cm⁻¹). The FTIR spectra were recorded at 4 cm⁻¹ resolution with 32 scans. Prior to analysis, the membranes were dried for 24 h in a desiccator to remove any water residue.

Table 1

The MWCO, pure water permeability (PWP), working pressure (p_w), and contact angle (θ_w) of membranes.

Membrane	Polymer	pH range	MWCO (Da)	PWP (L m ⁻² h ⁻¹ bar ⁻¹)	p_w (bar)	θ_w (°)
MW	Polyacrylonitrile	2–9	50,000	80.34	2	28.3 ± 0.6
CQ	Cellulose acetate	2–9	20,000	46.06	4	53.6 ± 3.9
GM	Polyamide (TFC ^a)	2–10	8000	24.59	5	60.0 ± 3.1

^a TFC-thin film composite.

A visual examination of the fouled and cleaned surface was conducted using SEM (Tescan Vega III Easyprobe, Czech Republic) operated at 10 kV. Samples were dried and coated with gold and palladium.

Contact angles of pristine membranes were measured on a DataPhysics OCA 20 Instrument goniometer (Germany). Sessile drops (2 mL) of MilliQ water at 23 °C were used for contact angle measurements. The contact angle of the membranes was measured 5 times at different locations. The measurement error is expressed by the standard deviations (SD).

2.7. Membrane hydraulic resistance, fouling type, and cleaning efficiency

Membrane hydraulic resistance (MHR) was calculated with the following equation:

$$R = \frac{\Delta p}{\eta \cdot J}, \quad (1)$$

where J is the permeate flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$), Δp is the transmembrane pressure (Pa), η is the absolute viscosity (Pa s), and R is the MHR (m^{-1}); which was determinate according to the flux at constant working pressure. The total MHR (R_t) can be divided, as resistance-in-series, into the inherent membrane resistance (R_m) and resistance caused by fouling (R_f):

$$R_t = R_m + R_f. \quad (2)$$

The R_f can be divided into reversible (R_{rev}) and irreversible resistance (R_{irrev}):

$$R_f = R_{rev} + R_{irrev}, \quad (3)$$

while the R_{rev} can be divided according to the applied cleaning agent:

$$R_{rev} = R_w + R_{alk} + R_{aci}, \quad (4)$$

where R_w is the resistance reversed by flushing with demineralized water, R_{alk} with an alkali agent, and R_{aci} with an acidic agent. The R_m was determined after the membrane pre-compression with demineralized water; the R_t was measured after 8.5 h of secondary effluent UF. The R_w , R_{alk} , and R_{aci} were calculated from the drop in the resistance after each cleaning phase. Unlike R_{alk} and R_{aci} , caused by the fouling removed by the respective cleaning agent, R_w includes the resistance caused by osmotic pressure.

The hydraulic cleaning efficiency (HCE) was calculated with the R_f and the measured resistance after each cleaning phase (R_{fc}):

$$\text{HCE (\%)} = \frac{R_f - R_{fc}}{R_f} \cdot 100\%. \quad (5)$$

3. Results and discussion

3.1. Secondary effluent

The rendering plant wastewater had a high content of dissolved organic carbon ($\sim 230 \text{ mg L}^{-1}$) and TKN ($\sim 340 \text{ mg L}^{-1}$), along with high COD ($\sim 5000 \text{ mg L}^{-1}$), turbidity ($\sim 700 \text{ NTU}$) and conductivity ($\sim 3000 \mu\text{S cm}^{-1}$). These characteristics arise from the high content of biological contaminants such as blood, fat, skin, feathers, bones etc. When treated in SBR, the wastewater characteristics substantially improved, but with a high content of EfOM that remains (values described in Materials and methods section).

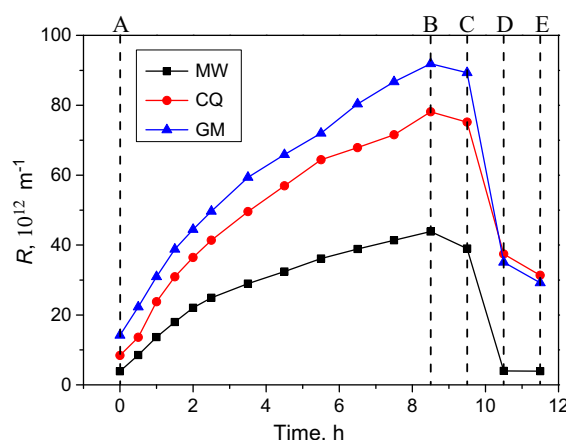


Fig. 1. Change in MHR in different phases of the process: A-beginning of secondary effluent UF, B-end of secondary effluent UF, C-MHR after cleaning with demineralized water, D-MHR after cleaning with PC 99, and E- MHR after cleaning with PC 77.

3.2. Membrane hydraulic resistance

Fig. 1 shows the change in MHR during the UF with three types of membranes of secondary effluent and after each cleaning phase. The initial MHR (R_m) (Fig. 1A) correlates with the MWCO (Table 1); thus, MW has the lowest and GM the highest R_m (Fig. 2). The MHR of the examined membranes drastically increased during the treatment of the secondary effluent (Fig. 1A and B). The increase in resistance is attributed to the fouling potential of EfOM, and might be amplified by the EPSs produced during the biological treatment [15]. Extracellular polymeric substances, even with a relatively low content compared to other EfOM fractions, ease the adhesion of other foulants, such as colloids, amplifying their fouling potential [15]. The initial growth in MHR can be attributed to the adsorption of foulants on the membrane surface and pore blocking followed by a rapid cake formation caused by the high initial flux. The following increase in MHR is attributed to cake formation. The tighter membranes (GM and CQ) accumulated a larger quantitative increase in resistance (Fig. 1B); as a result of higher retention and cake compression due to higher working pressure (Table 1). The looser membrane (MW) gained a greater relative increase in resistance (RIR) (Table 2). The correlation between RIR and MWCO was attributed to a greater relative reduction in pore size and faster blocking of larger pores by internal pore blocking; foulants can infiltrate in the larger [29]. The greater relative reduction in pore sizes for looser membrane results in a similar retention, critical flux and gel formation. This is confirmed by the similar final flux and retentions (COD and DOC) given in Table 2.

3.3. Hydraulic cleaning efficiency

The cleaning efficiency is calculated with Eq. (5) and presented in Fig. 2. Demineralized water mildly reduced the MHR, as expected, i.e. 12.2% for MW, 4.2% for CQ, and 3.4% for GM. This reduction (R_w) is attributed to the gel-layer destabilization caused by the dissolution of salts from the fouling layer. The chemical cleaning with PC 99 (alkali agent) reduced most of the fouling resistance (R_{alk}) (Fig. 2), i.e. 87.5% for MW, 54.2% for CQ, and 71.2% for GM. MW achieved a cumulative HCE of 99.7% after the washing with PC 99, as the MHR dropped almost to the R_m value. The superior performance of PC 99 is expected, as the fouling layer is made of EfOM and the alkali agent destabilizes the fouling layer via hydrolysis of proteins and polysaccharides, expansion of humic acid, neutralization of acidic organics, etc. [20]. In the final phase,

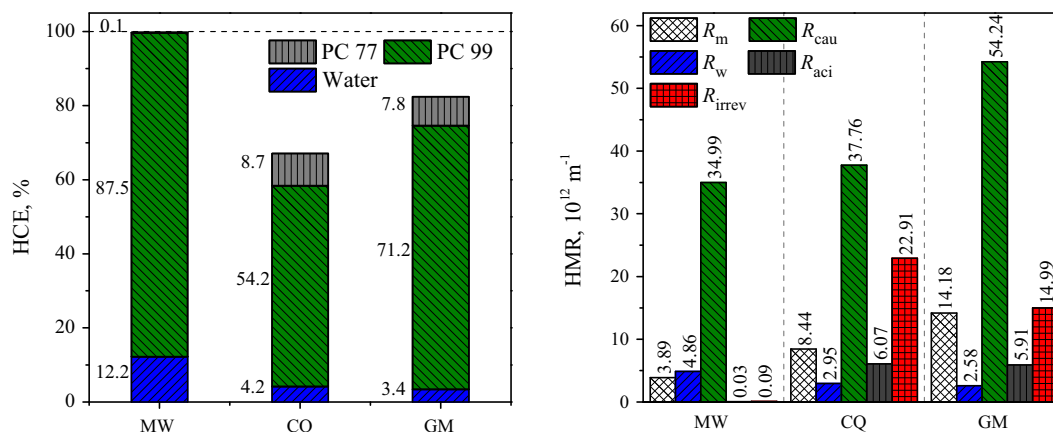


Fig. 2. Values of specific HCE of demineralized water and chemical cleaning agents (PC 99 and PC 77); and specific MHR.

Table 2

The RIR (R_i/R_m), total resistance (R_t), sieving coefficient for DOC (S_{DOC}) and COD (S_{COD}), and final flux (J_f) of the tested membrane.

	R_i/R_m , –	R_t , 10^{12} m^{-1}	S_{DOC} , –	S_{COD} , –	J_f , $\text{L m}^{-2} \text{ h}^{-1}$
MW	11.3	43.86	0.508	0.932	16.42
CQ	9.3	78.13	0.450	0.944	18.43
GM	6.5	91.9	0.454	0.937	18.75

PC 77 (acidic agent) mildly lowered the resistance (R_{aci}) (Fig. 2), i.e. 0.1% for MW, 8.7% for CQ, and 7.8% for GM, indicating the absence of inorganic foulants and its ineffectiveness in the destabilization of the organic fouling layer. After the cleaning procedure, a notable difference in HCE can be seen from the irreversible fouling (R_{irrev}); while the R_{irrev} of MW is negligible, the R_{irrev} of other membranes is a comparable or twice the value of R_m (Fig. 2).

There are few studies on chemical cleaning of flat sheet membrane fouled by EfOM, but with a lower content of foulants compared to the secondary effluent used in this work [24–27]. One study expatiated the HCE of chemically enhanced backflush with NaOCl solutions ($100\text{--}600 \text{ mg L}^{-1}$) for a membrane fouled by SMP and a membrane fouled with dissolved organic matter (DOM) [24]. They achieved approximately 70% HCE for SMP and approximately 60% for DOM with a 600 mg L^{-1} solution after 2 h of cleaning, but if looking at the results after 1 h (as in our work) they achieved approximately 55% HCE for SMP and 60% for DOM. Another work examined the chemically enhanced cleaning combined with physical cleaning methods for flat sheet membrane fouled during the MF of secondary effluent [27]. An excellent HCE of 80% was achieved after applying a 600 mg L^{-1} solution of NaOCl on the permeate side and microbubbles on the retentate side after 2 h, but with 60% efficiency after 1 h. Those two studies had a lower efficiency, especially compared to the results for MW, but it's important to note that in those studies they applied the NaOCl solutions on the permeate side and let it diffuse, as in this study, the chemical agents and shear force were applied directly on the fouling.

The second batch of SE was treated with MW, the membrane that had the best performance in the previous experiments. The fouling was more reversible, i.e. the HCE of water was between 45.74% and 65.97%, as shown in Fig. 3. The reversibility increased with the decrease of working pressure. That can be attributed to cake layer compression and increased permeate flux which leads to a more rapid fouling formation as the permeate flux is farther from the critical flux. On the other hand, the decrease in pressure decreased the HCE of PC 99 as more fouling was previously removed with water. As shown in Fig. 3, temperature plays an

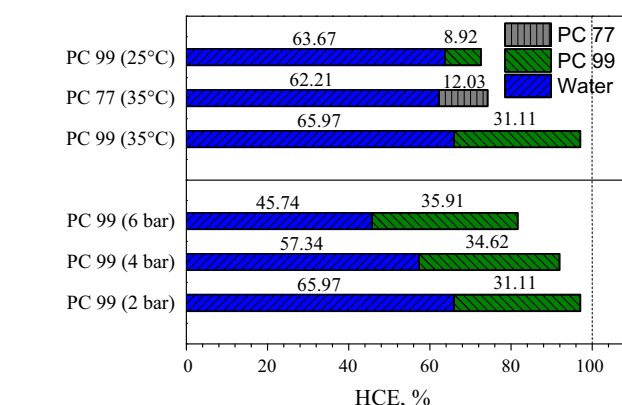


Fig. 3. Hydraulic cleaning efficiency with different cleaning agents and at different pressures and temperatures.

important role in the cleaning efficiency, i.e. increased temperature increases the efficiency. Moreover, Fig. 3 shows that PC 99 performs 2.5 times better cleaning efficiency than PC 77, confirming the superiority of PC 99 for fouling removal.

3.4. Membrane chemical cleanliness

To characterize the fouling and determine the chemical cleanliness, the membrane surface was analyzed with ATR-FTIR. The FTIR spectra of fouled membranes are presented in Fig. 4. The similarity in the spectra indicates that the fouling layers on all membranes have the same composition. The absence of characteristic peaks of the membrane polymer indicates the presence of a substantial fouling layer on the surface, blocking the detection of characteristic polymer bonds of the pristine membranes (Fig. 4). The spectra of the fouling layer has broad absorption peaks at 3266 , 2968 , 2924 , and 2854 cm^{-1} attributed to stretching vibrations of O–H, N–H, and C–H bonds. The carboxylic group is represented by the peak at 1722 cm^{-1} , amide I group (CO stretching vibration) at 1634 cm^{-1} , amide II group (NH plane bond and CN stretching vibration) at 1549 cm^{-1} , and the $\text{CH}_2\text{C=O}$ group from amino sugars at 1377 cm^{-1} . According to those peaks the chemical composition of the fouling layer can be determine [11,12,14,30,31]. The peaks at 1634 , 1549 , and 1040 cm^{-1} are characteristic for hydrophilic colloids from EfOM composed of proteins, polysaccharides, and amino sugars. The signal at 1634 cm^{-1} suggests a β -sheet secondary protein structure [31]. The peak at 1722 cm^{-1} is

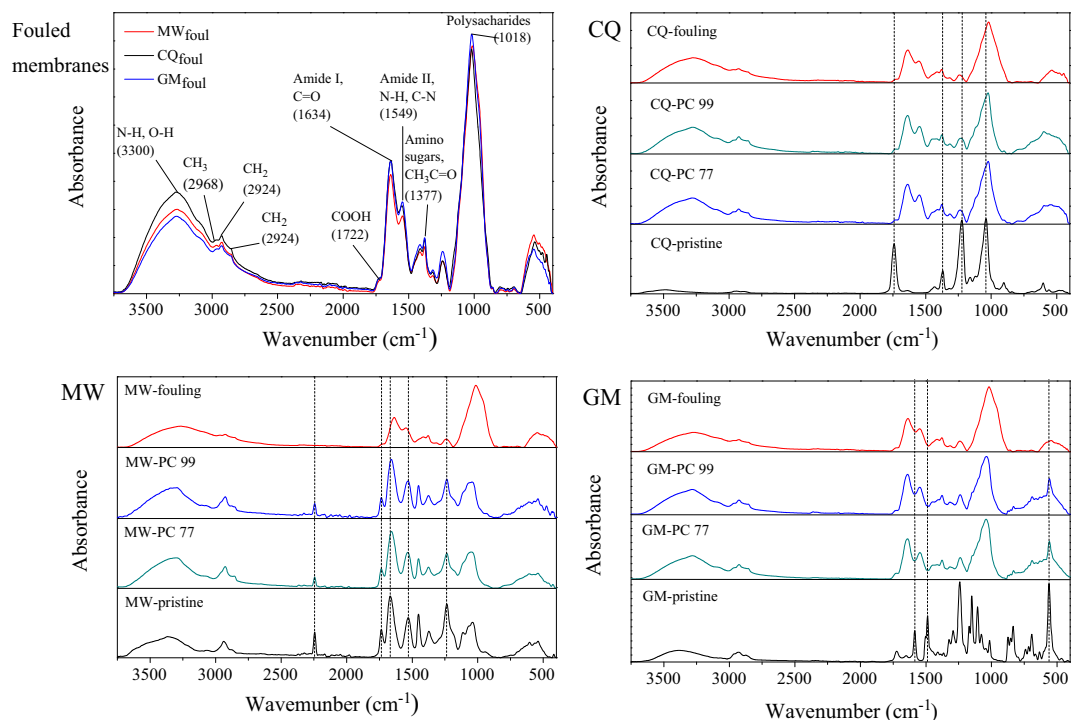


Fig. 4. FTIR spectra of fouled membranes; pristine, fouled, and cleaned MW membranes; pristine, fouled, and cleaned CQ membranes; and pristine, fouled, and cleaned GM membranes.

characteristic for hydrophilic and hydrophobic NOM, and the peak at 1377 cm^{-1} for nitric acid in hydrophilic and neutral NOM.

The FTIR spectra of pristine, fouled, and cleaned (PC 99 and PC 77) membrane are presented on Fig. 4 (MW, CQ, and GM). The spectra of the pristine MW (polyacrylonitrile) shows the character-

istic peaks of nitrile (2243 cm^{-1}), ester (1735 cm^{-1}), amide I (1655 cm^{-1}), amide II (1529 cm^{-1}), and C–N group (1235 cm^{-1}). These peaks are not visible, or have different intensities, on the fouled MW spectra. After the chemical cleaning with PC 99, the spectrum has the same peak position with almost the same

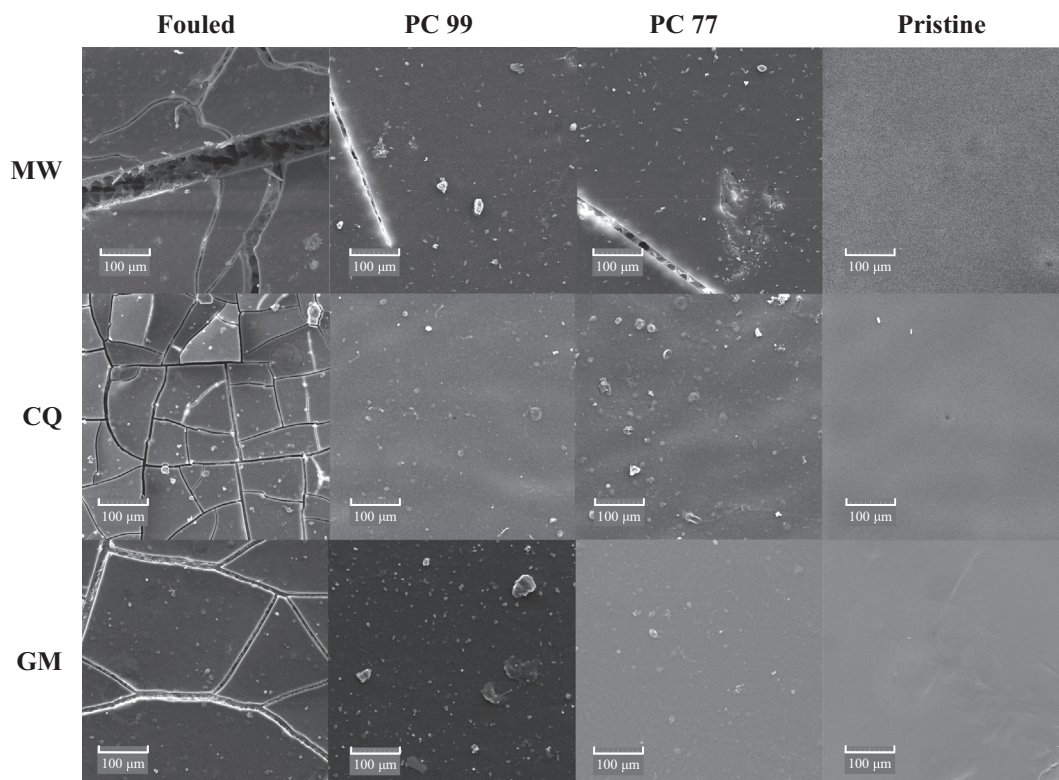


Fig. 5. SEM images (magnification $500\times$) of MW, CQ, and GM membrane (from left to right: fouled, after PC 99, after PC 77, and pristine).

intensity as the spectra of the pristine membrane, demonstrating an excellent fouling removal, which is in accordance with the HCE (Fig. 2). The pristine CQ membrane (cellulose acetate) has characteristic peaks of ester (1740 cm^{-1}), C—H (1369 cm^{-1}), and C—O (1224 and 1038 cm^{-1}) groups. Those peaks appear in the fouling spectra, but with different intensities; as mentioned before, the spectra of the fouled membranes are not influenced by the membrane polymer. The spectra of the cleaned membrane (with PC 99 and 77) didn't change suggesting an incomplete fouling removal, i.e. the presence of irreversible fouling. Similarly to the CQ membrane, the GM membrane (polyamide) has the characteristic peaks of the ester (1585 cm^{-1}) and N—H group (1487 and 560 cm^{-1}) that didn't appear on the spectra of the fouled and cleaned membrane, indicating an incomplete fouling removal.

The FTIR spectra of MW membrane fouled at different pressure with the second batch of SE are presented in the [supplementary material](#) (Figs. S1–S3). Those spectra show a similar composition of the fouling layer and a homogeneously fouled surface, as Fig. S2 shows the spectra measured at different places on the same membrane. Fig. S3 shows the FTIR spectra of the fouling layer and of the solids that remained after evaporation of SE. The spectra of the fouling layer (MW SE 2) compared to the spectra of dried solids from SE 2 shows higher adsorption of amide I and II (proteins, amino sugars) and a lower adsorption at 1040 cm^{-1} (polysaccharides) indicating that the fouling layer contains less polysaccharides compared to the polysaccharides content in the SE 2.

Fig. 5 presents the surface morphology of fouled and cleaned membranes. All fouled membranes have a substantial fouling layer, which cracked as it dried. The images of cleaned membrane show that even with a high HCE there was still some residual fouling visible as attached particles.

3.5. Comparison of membrane performance

Two membrane characteristics can explain the high cleaning efficiency of MW membrane, as opposed to less efficient cleaning of CQ and GM membranes. MW membrane has a higher MWCO that retains mostly macromolecules (polysaccharides and proteins) and colloids, as the alkali agent can destabilize this foulants and facilitate their removal. GM and CQ membranes with a lower MWCO retain smaller organic molecules that along with a higher working pressure result in a more stable and denser fouling layer, which is less prone to destabilization. The other main difference is the surface hydrophilicity. The membrane contact angle of water, as a measure of hydrophilicity, is given in Table 1. MW membrane has a lower contact angle than CQ and GM membranes, indicating the higher wettability of the surface. This surface propriety can have a high impact on fouling removal.

4. Conclusion

In this study three UF membranes were used to treat the rendering plant secondary effluent. During the UF severe fouling appeared due to high content of EfOM and permeate flux above the critical value. The highest increase in resistance was noted on GM ($91.9 \cdot 10^{12}\text{ m}^{-1}$), the tightest membrane. Even though the membranes had different initial flux and increase in resistance, the final fluxes reached similar values ($17.69 \pm 1.17\text{ L m}^{-2}\text{ h}^{-1}$).

The HCE differed among membranes. In first phase, MW had the highest HCE (99.8%), which can be attributed to its hydrophilicity, i.e. lower contact angle. Meanwhile, the fouling on CQ and GM membranes was partially removed, 67.1% and 82.4% respectively. The better cleaning agent for this type of fouling was the alkali cleaning agent, PC 99. Temperature has a positive influence on the HCE, while pressure lowers the HCE.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.seppur.2017.07.041>.

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