Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2023.29787

303 (2023) 1–10
August

Integration of direct microfiltration and reverse osmosis process for resource recovery from municipal wastewater

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Received 7 April 2023; Accepted 16 July 2023

ABSTRACT

For the sustainability of water resources, the recovery of water, organic matter (OM), energy, and nutrients from municipal wastewater become very attractive resources. As direct application of water, nutrient, and energy recovery from municipal wastewater cannot be feasible, the wastewater needed to be concentrated. In this study, the molecular weight distribution of OM content was determined in wastewater samples, up-concentration potential of direct microfiltration (DMF) of municipal wastewater and water recovery were investigated. In OM fractionation studies, around 52% of the chemical oxygen demand (COD) in wastewater was particulate or colloidal (>10 kDa) and 48% was soluble (<300 Da). In DMF tests, the COD concentration was concentrated up to 1,573 mg/L after sequential DMF experiments. Additionally, the theoretic total energy requirement of the DMF process was found around 0.3 kWh/m^3 and it would be potentially energy positive. In crossflow experiments, the reverse osmosis (RO) process was performed using DMF effluent. When microfiltration and RO membranes were chemically cleaned, flux recovery rates of 100% and 99% were achieved, respectively. However, the foulants could not be completely removed during the cleaning according to scanning electron microscopy, atomic force microscopy, and attenuated total reflection-Fourier-transform infrared spectroscopy results of the virgin, fouled, and cleaned membranes. This study reveals that the DMF+RO process is a promising technology for the recovery of OM and water from municipal wastewater.

Keywords: Direct membrane filtration; Reverse osmosis; Resource recovery; Water recovery; Membrane fouling

1. Introduction

Sustainable wastewater treatment technologies have become crucial in the transformation of the global economy from linear to circular. Nevertheless, conventional municipal wastewater treatment plants (WWTPs) consume a large amount of energy to remove mainly organic matter (OM) and nutrients [1]. The transition toward energy-neutral and more sustainable municipal wastewater treatment processes needs a new perspective since globally generated 330 km^3 annual municipal wastewater theoretically contains resource potential to supply millions of households' energy needs and to irrigate and fertilize millions of hectares of the agricultural area [2].

Energy recovery from OM instead of aerobic oxidation requires the upgrade of the existing plants. Anaerobic methane (CH_4) production, which is the most common application for the conversion of OM in wastewater into energy, has limited direct application for energy generation from domestic wastewater due to its low OM content [3]. Different treatment processes are being studied for up-concentrating municipal wastewater, such as high-rate activated sludge

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system for OM recovery [4], chemically enhanced primary treatment [5], dynamic membrane [6], and direct membrane filtration process [7–9].

Direct microfiltration (DMF) is a promising process thanks to extreme compactness, a small footprint, and reduced energy consumption [10]. The most important advantage of the DMF process is that it is purely a physical process with a small amount of chemical requirement [11]. In this way, the DMF process can be readily implemented by dimensioning it in the real-scale application. In addition to OM and nutrients, municipal wastewater contains many other organic and inorganic pollutants including heavy metals and micropollutants [12]. So far, reverse osmosis (RO) coupled with various processes for municipal wastewater reclamation and reuse has been attracting interest owing to its great effectiveness in removing dissolved inorganics, organics, and microbial matters [13–15].

Membrane fouling is the key limitation for both the DMF and RO processes, though they have significant potential for recovery of OM, nutrients, and water from wastewater. Several membrane fouling mitigation strategies were investigated including coagulation/flocculation, aeration, flushing for DMF and feed pretreatment, membrane surface modification, and chemical cleaning for RO process [16–21]. Despite the implementation of membrane-based pretreatment to ensure the production of superior feed for RO, it is noteworthy that dissolved matter, comprising both organic and inorganic constituents, may still permeate through the aforementioned pretreatment process. Thus, these constituents adhere, accumulate, or precipitate onto the surface of the RO membrane, resulting in the formation of membrane fouling [22–24]. In order to address the issue of membrane fouling, it is imperative to investigate the fouling mechanisms and assess the impact of physical and/or chemical cleaning on the membrane flux recovery [25,26]. The existing body of literature provides information on the conventional techniques utilized to mitigate fouling and the effects of these mitigation strategies on the membrane surface in membrane bioreactors (MBRs), which have gained widespread adoption due to their maturity and prevalence in numerous installations [27]. However, investigations are still required for the DMF process in order to analyze the fouling mechanism and the impact of the cleaning procedure. Furthermore, the existing literature regarding DMF operation indicates that most applications perform in a submerged configuration, with only a limited number of studies where crossflow configuration is performed [28].

To the best of our knowledge, there is a lack of information on detailed membrane surface characterization and fouling mechanisms of each membrane for the integrated DMF+RO process for wastewater reclamation and resource recovery. It should be emphasized that membrane fouling, particularly in pressure-driven membrane filtration, limits the use of DMF in wastewater treatment and resource recovery. Furthermore, understanding membrane fouling mechanisms and their impact on membrane surface properties are critical for sustainable long-term DMF process operation. This paper presents the results of a study evaluating the integrated DMF+RO process performed on a laboratory scale for increasing energy and water recovery from municipal wastewater. Regarding the fouling characteristics of membranes, the surface of the virgin and physically/chemically cleaned membranes were characterized using spectroscopic and microscopic techniques. As a preliminary test, for estimating process performances, OM content was determined by using fractionation of wastewater samples in terms of chemical oxygen demand (COD) and total organic carbon (TOC) in dead-end filtration mode. After fractionation, DMF was performed using a microfiltration (MF) (0.1 μm) membrane for increasing the energy potential of wastewater and for producing permeate suitable to feed the RO process. In addition, RO treatment has a high potential for nutrient recovery by enriching them in the concentrate as well as producing high-quality water for reuse. This study provides information for future research work related to resource and water recovery using membrane processes.

2. Materials and methods

2.1. Wastewater analysis and membranes

Raw wastewater was collected from the effluent of the pre-settling tank in Kayseri municipal WWTP, in which a conventional activated sludge process is applied. A characteristic of wastewater is given in Table 1. All the analyses were performed according to the "Standard Methods for Water and Wastewater" published by the American Public Health Association [29].

The most common and commercially available flat sheet MF, ultrafiltration (UF), and nanofiltration (NF) membranes and MF and RO membranes were used in the fractionation and up-concentration experiments, respectively. The properties of these membranes are listed in Table 2.

2.2. Experimental set-up

2.2.1. COD fractionation of wastewater samples

Determination of COD and TOC fractions provides detailed information about the OM content of wastewater and the results can be used in the design of treatment processes. The OM fractionation experiments were conducted using the Sterlitech HP4750 dead-end stirred cell (Sterlitech, USA) with a 14.6 cm^2 filtration area and the instrument

Table 1
Characteristics of a sample of wastewater treatment plant primary settling tank effluent

Parameters	Values
pH	7.1 ± 0
Conductivity (μS/cm)	1,420 ± 22
Turbidity (NTU)	82.0 ± 5.2
Chemical oxygen demand (mg/L)	564 ± 9
Total organic carbon (mg/L)	212 ± 5
Total nitrogen (mg/L)	35.8 ± 1.6
PO_4^{3-} (mg/L)	10.3 ± 0.4
Cl^- (mg/L)	169.2 ± 0.9
SO_4^{2-} (mg/L)	21.7 ± 0.4

Table 2
Specifications of membranes used in this study

Process	Membrane	Membrane material	Pore size/Molecular weight cut-off	From
Fractionation	MF	Cellulose	10 μm	Macherey-Nagel, Germany
	MF	Nitrocellulose	0.45 μm	Millipore, Germany
	UF	PES[a]	10 kDa	GE, USA
	NF	PA[b]	150–300 Da	GE, USA
DMF	MF	PES[a]	0.1 μm	Sterlitech, USA
RO	BW30-XFR	PA[b]	99% NaCl/100 Da	Dupont Filmtech, Netherlands

[a]Polyethersulfone;
[b]Polyamide.

setup. The stirred filtration cell was endowed with a magnetic stirrer, mixing at 300 rpm to simulate crossflow filtration mode and thereby minimize concentration polarization. For fractionation, wastewater samples were sequentially filtrated by using MF and UF membranes. In the sequential filtration experiments using MF membranes with 10 and 0.45 μm pore sizes and UF membranes with molecular weights of 10 kDa and NF membrane with molecular weights of 150–300 Da were used, respectively.

2.2.2. Membrane filtration tests

A crossflow membrane filtration system (Sterlitech, Sepa CF, USA) was used to perform the DMF and RO process with 5 L of the feed sample. The effective area of the membrane module was 150 cm^2. During the filtration experiments, the temperature was 25 ± 2°C. DMF and RO filtration tests were performed according to the operational conditions given in Table 3.

For each stage of DMF and RO tests, flux was calculated by using Eq. (1).

$$J = \frac{V}{A \times t} \quad (1)$$

where J is the water flux (L/m^2·h, LMH), V is the permeate volume (L), A (m^2) is the effective membrane area, and t (h) is the filtration duration. Filtration experiments were conducted in triplicate and average flux values were provided.

In the crossflow membrane filtration tests, the water fluxes were measured under steady-state conditions after physical and chemical cleanings of the membranes. The fouling behavior of the membranes was investigated using pure water in terms of flux recovery ratio (FRR) and relative flux reduction (RFR) according to Eqs. (2) and (3), respectively.

$$\text{FRR} = \frac{J_2}{J_1} \times 100 \quad (2)$$

$$\text{RFR} = \frac{J_1 - J_2}{J_1} \times 100 \quad (3)$$

where J is the flux value of wastewater at steady state (LMH), J_1 is the pure water flux value before wastewater filtration at steady state (LMH), and J_2 is the pure water flux value after physical or chemical cleaning at steady state (LMH).

Table 3
Operational parameters for DMF and RO process

Operational parameters	DMF	OF
Crossflow velocity (m/s)	0.5	0.8
Pressure (bar)	0.5	15
Recovery rate (%)	80	60

2.2.3. Membrane cleaning

To investigate water flux recovery for the MF and RO membranes physical and chemical cleaning was applied. After wastewater filtration, the cleaning of the fouled membrane for 15 min was directly carried out in the membrane module for cleaning the fouled membrane using flushing at 1.2 m/s cross flow velocity. The cleaning process involved using DI water for physical cleaning and an alkaline solution (0.75% NaOCl) for chemical cleaning.

2.2.4. Membrane characterization

The surface of membranes was characterized after each filtration and cleaning process to evaluate the fouling phenomena. Before the characterization tests, the samples were dried at 50°C in a laboratory-scale oven.

The morphology of the virgin and fouled membrane surface was investigated by scanning electron microscopy (SEM) on a Zeiss GeminiSEM 500 Field Emission Microscope operated at 3 kV. Before the measurements, membranes were coated with gold. The SEM images for the surface of the membranes were taken at 20 and 30 K magnification for MF and RO membranes, respectively.

Atomic force microscopy (AFM) was used to determine the effect of fouling on membrane topography. Membranes were compared in terms of surface roughness (average surface roughness (R_a), root mean square roughness (R_q), and the maximum height of the profile (R_{max}). AFM measurement was conducted by MultiMode 8-HR and Veeco operated in tapping mode (Model: RTESP-300). The analyses were performed using 10 × 10 μm and 5 × 5 μm image sizes for MF and RO membranes, respectively. AFM measurements were performed at two different locations.

The surface roughness values which are R_a, R_q, and R_{max} were given average values with these two locations.

ATR-FTIR was employed for the identification of functional groups of the virgin and fouled membranes of the deposited foulants using an FTIR spectrometer (Thermo Nicolet Avatar 370). The analysis was conducted in the wavelength range of 4,000–400 cm^{-1}.

3. Result and discussion

3.1. COD fractionation of wastewater samples

In order to estimate the effectiveness of the OM recovery process, OM content was evaluated using fractionation of wastewater samples in terms of COD and TOC in dead-end filtration mode. The molecular weight distribution of OM content was determined in wastewater samples using different MF, UF, and NF membranes. As shown in Fig. 1, around 52% of the COD in the raw municipal wastewater consists of particulate or colloidal (>10 kDa) and 48% was soluble (<300 Da). Besides, 49% of TOC of the municipal wastewater consists of particulate or colloidal (>10 kDa) and 51% was soluble (<300 Da). Hence, more than half of the OM in the wastewater sample can be recovered with MF membranes. The outer layer of rejected particles may act as a dynamic membrane, screening out the more highly fouling species of smaller size [30]. It should be noted that the wastewater was sampled from the effluent of the primary settling tank. Therefore, some of the OM can also be recovered in the sludge of the primary settling tank. Assuming that approximately 25% of the COD is removed in the primary settling tank and 52% of the remaining OM is recovered by the proposed process, the recovery potential can be calculated as approximately 64%. Similarly, Noyan et al. [31], reported that particulate and soluble COD account for between 70%–71% and 29%–30%, respectively in municipal wastewater. Additionally, the COD/TOC ratio was 2.73, and values for this ratio in the literature ranged from 2.15 to 3.00 [32,33].

3.2. DMF+RO experiments

3.2.1. Sample characterization

In COD fractionation tests, the efficiency of COD removal through MF membranes with pore sizes of 10 and 0.45 μm was reached at about 30%. Since COD is one of the most essential characteristics for improving the energy recovery potential of municipal wastewater, a more selective MF membrane with a pore size of 0.1 μm was used in DMF testing to increase removal efficiency and produce more concentrated wastewater. The initial concentration of COD in the feed wastewater was measured at 564 mg/L. Following the DMF experiment, the concentration of COD was observed to increase to 1,573 mg/L, indicating a concentration factor of approximately 3. Assuming around 25% of the COD was removed in the primary settling tank and the CH_4 production potential of 1 g COD is around theoretically 0.35 L, the proposed process can promote energy-neutral or positive wastewater treatment [34]. According to the calculations, around 0.39 kg/m^3 COD can be recovered using primary sedimentation and DMF process. Theoretically, 0.135 m^3-methane/m^3-wastewater or around 1.35 kWh/m^3-wastewater energy recovery potential can be calculated. If we assume that the efficiency of electricity generation from methane is around 45%, approximately 0.6 kWh/m^3 of electricity can be generated. Assuming that 0.2 m^3-air/(m^2-membrane·h) aeration requirement to scour cake from the membrane surface in the DMF process, around 30 LMH flux in the membrane, 5 m water depth, and 0.8 blower efficiency, the aeration energy requirement to scour cake layer would be around 0.12 kWh/m^3. Physical treatment and other pumping energies would be lower than 0.2 kWh/m^3 [30]. Theoretically, the total energy requirement of the process would be around 0.3 kWh/m^3. Hence, the process would be potentially energy-positive, especially DMF treatment is met the effluent quality requirements for agricultural irrigation. Additionally, the degree of commercialization of the DMF processes should be considered. Consequently, preliminary assessments must be performed at the pilot scale to establish the feasibility of DMF usage before its implementation on a larger scale [35].

COD, pH, conductivity, PO_4^{3-}, Cl^-, and SO_4^{2-} results of the collected samples from the DMF and RO process are shown in Table 4. While the COD concentration increased in the DMF concentrate sample, there was no significant increase in SO_4^{2-} concentration. In anaerobic methane production processes, some part of the OM is used for SO_4^{2-} reduction [31]. In the proposed process, the membrane does not reject sulfate and its concentration per OM is also low compared to the original wastewater content, and the consumption of the OM for sulfate reduction is reduced in the DMF process.

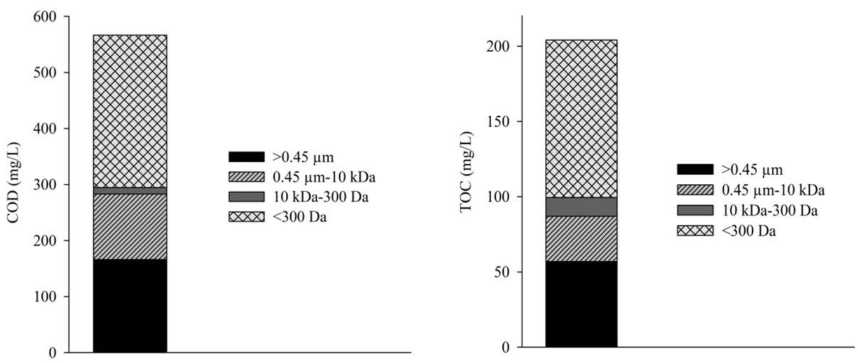

Fig. 1. Molecular weight fractions of total organic carbon and chemical oxygen demand of municipal wastewater.

If higher quality water is required for various reuse alternatives, for example, industrial reuse, the DMF process can be fed to the RO process. In this way, higher-quality water is generated, and the nutrients can be concentrated for further recovery. For example, PO_4^{3-} concentration was increased to 16.8 mg/L in the concentrate of the RO process, which makes it potentially feasible to recover PO_4^{3-} [36]. The RO process is a powerful alternative for obtaining high-quality product water and sustainability of water. However, the permeate of the DMF process has the potential to be reused for agricultural irrigation.

3.2.2. Membrane characterization

3.2.2.1. SEM images

SEM analysis provides a better understanding of the characterization and development of fouling on the membrane surface after wastewater filtration. The cake layer development is a crucial part of membrane fouling, and its characteristics are projected as the main factor affecting membrane fouling mitigation [37]. The surface images of the MF membranes that were used in DMF tests, are shown in Fig. 2. The thick and dense cake layer was formed on the membrane

Table 4
Sample characteristics after performing DMF and RO experiments

Parameters	Raw wastewater	DMF		RO	
		Concentrate	Permeate	Concentrate	Permeate
pH	7.1 ± 0	7.3 ± 0.1	7.3 ± 0.1	8.0 ± 0.3	9.1 ± 0.4
Conductivity (µS/cm)	1,420 ± 22	1,352 ± 105	993 ± 52	2,520 ± 226	72.8 ± 8.1
Chemical oxygen demand (mg/L)	564 ± 9	1,573 ± 9.4	104.7 ± 5.2	308.0 ± 4.7	<5
PO_4^{3-} (mg/L)	10.3 ± 0.4	12.2 ± 1.7	9.4 ± 0.2	16.8 ± 1.0	ND
Cl^- (mg/L)	169.2 ± 0.9	99.4 ± 1.3	141.9 ± 0.9	322.5 ± 1.4	6.1 ± 0.5
SO_4^{2-} (mg/L)	21.7 ± 0.4	24.5 ± 0.6	12.3 ± 1.5	30.9 ± 0.2	ND

*ND, not detected: indicates less than detectable value.

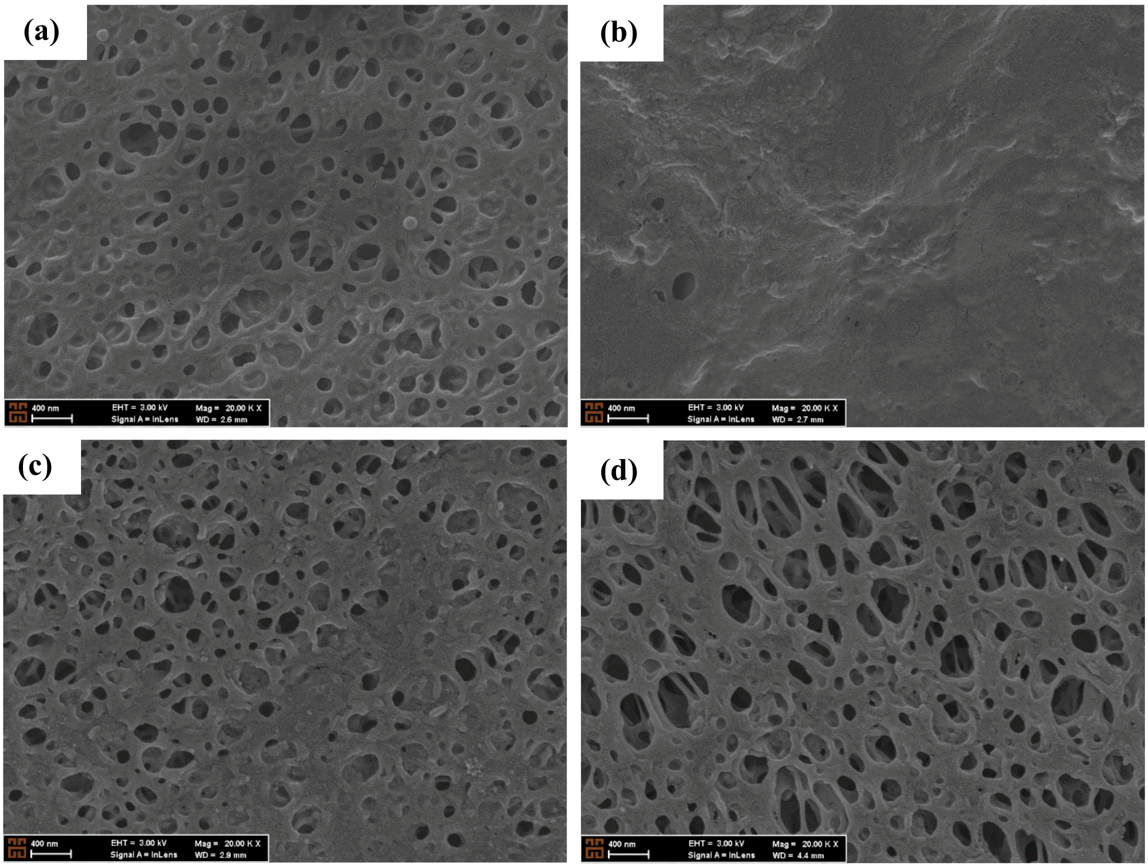

Fig. 2. Scanning electron microscopy images of the virgin (a), fouled after DMF (b), physically cleaned (c), and chemically cleaned (d) MF membrane.

surface (Fig. 2b). After physical cleaning, although the cake layer was eliminated, membrane pores were still blocked by pollutants (Fig. 2c). The SEM images suggest that the size of pores in chemically cleaned membrane membranes is larger (Fig. 2d). Polyethersulfone (PES) membranes are considered highly chlorine-tolerated and stable in hypochlorite solutions [38]. However, Arkhangelsky et al. [38] showed that escalated hydrophilicity of chemically cleaned membranes can also be represented by the formation of bigger pores due to degradation of the membrane.

The surface images of the BW30-XFR membranes used in RO tests are shown in Fig. 3. The cake layer formed on the membrane surface during filtration (Fig. 3b). The removal of most of the deposited materials from the membrane surface is clearly observed after two-step physical and chemical (0.75% NaOCl) cleaning in Fig. 3c and d. Similarly, in the literature, it was shown that basic cleaning reagents provide effective cleaning instead of acids for the recovery of fouled RO membranes [39].

3.2.2.2. Atomic force microscopy

The three-dimensional surface morphology of the virgin, fouled, physically cleaned, and chemically cleaned membranes were analyzed for both DMF and RO processes using tapping mode AFM and are shown in Fig. 4. The roughness of the virgin MF and RO membrane is not different from typical MF and RO membranes [40,41]. The effect of fouling on the membrane surface was clearly observed in AFM images. The porous structure could not be specified on the surface of the fouled membrane because of the cake layer. However, as physical and chemical cleaning was carried out, the roughness of the MF and RO membrane's surface might be observed.

The roughness parameters, R_q, R_a, and R_{max} are summarized in Table 5 for both MF and RO membranes. The virgin membrane had the lowest roughness for both MF and RO. Similarly, chemically cleaned MF and RO had the highest roughness because of the deterioration of the membrane surface [38]. After physical cleaning, the roughness of the MF membrane was more similar to the virgin MF membrane. While R_a, R_q, and R_{max} were 35.2, 37.8, and 553.1 nm for virgin MF membrane, they were 34.8, 44.0, and 339.4 for physically cleaned MF, respectively. However, physical cleaning did not have the same effect on the RO membrane in terms of roughness.

3.2.2.3. Attenuated total reflection-Fourier-transform infrared spectroscopy

The ATR spectra that include the vibration bands specifics of the membranes' substrate and the accumulated

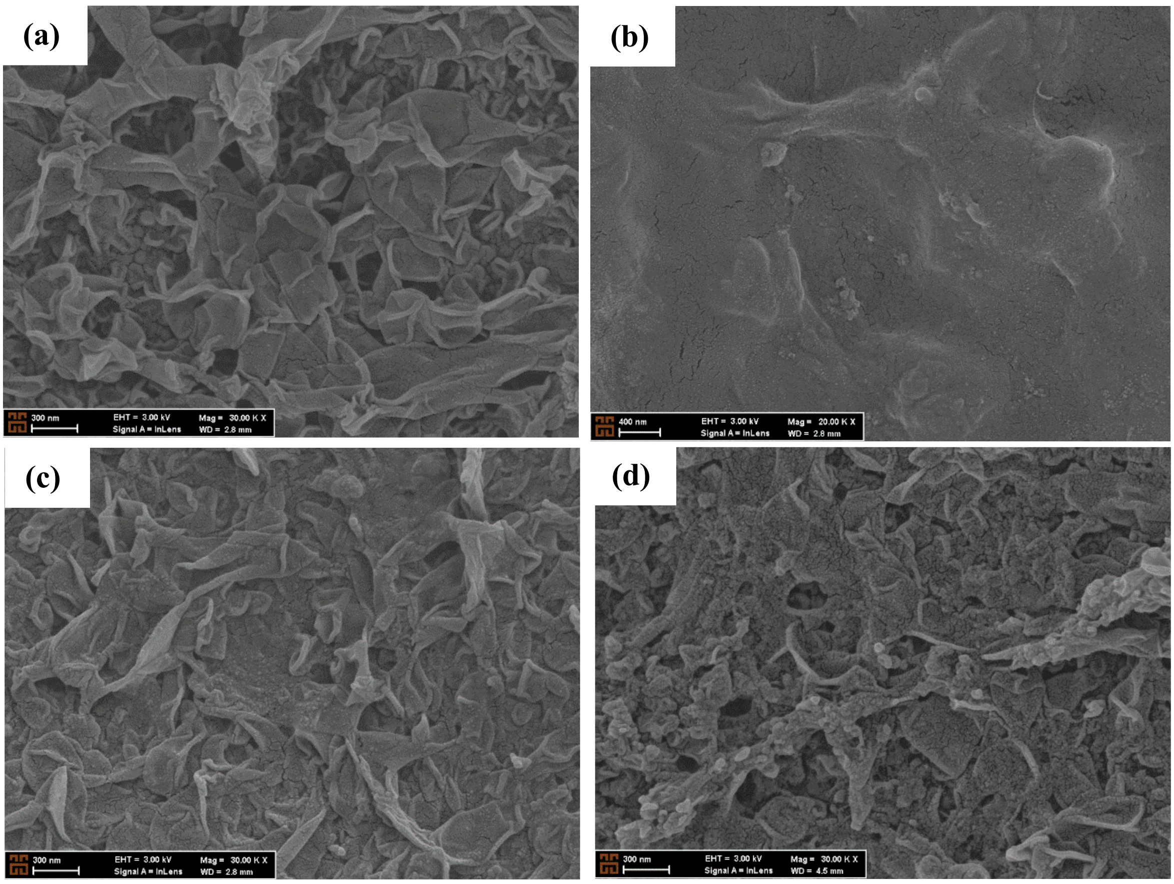

Fig. 3. Scanning electron microscopy images of the virgin (a), fouled after filtration (b), physically cleaned (c), and chemically cleaned (d) RO membrane.

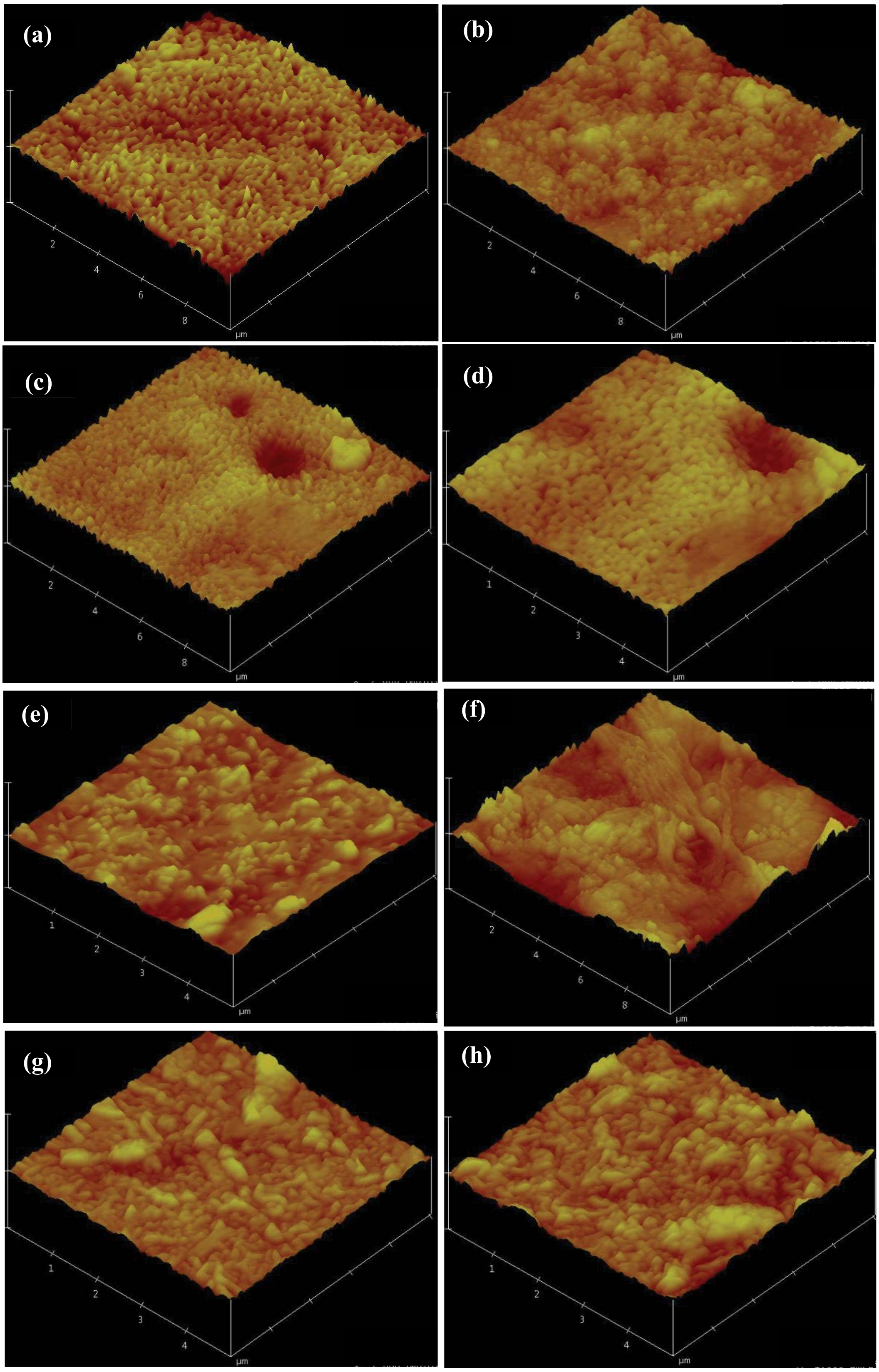

Fig. 4. AFM images of the virgin (a), fouled after filtration (b), physically cleaned (c), chemically cleaned (d) MF membrane, and the virgin (e), fouled after filtration (f), physically cleaned (g), chemically cleaned and (h) RO membrane.

Table 5
Roughness properties of MF and RO membranes

Process	Membrane	R_a (nm)	R_q (nm)	R_{max} (nm)
DMF	Virgin MF	35.2 ± 3.3	37.8 ± 6.7	533.1 ± 19.0
	Fouled MF	56.1 ± 6.5	70.2 ± 9.6	553.1 ± 111.8
	Physically cleaned MF	34.8 ± 7.6	44.0 ± 9.6	339.4 ± 83.9
	Chemically cleaned MF	72.8 ± 6.8	98.4 ± 14.4	907.6 ± 252.2
RO	Virgin RO	39.0 ± 0.4	48.2 ± 1.8	361.9 ± 32.6
	Fouled RO	65.5 ± 15.9	79.6 ± 16.2	476.7 ± 69.7
	Physically cleaned RO	66.5 ± 23.8	85.7 ± 29.9	776.2 ± 250.4
	Chemically cleaned RO	77.0 ± 20.1	97.2 ± 24.2	726.1 ± 129.7

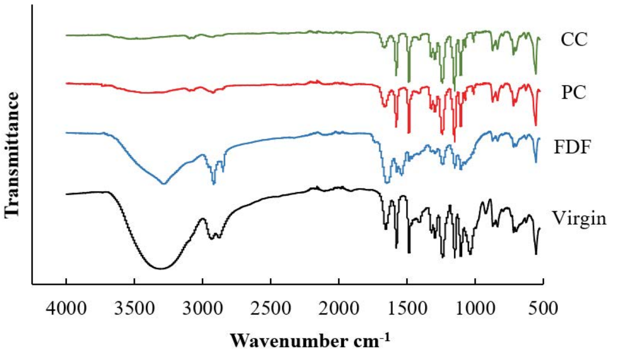

Fig. 5. ATR-FTIR spectra of the virgin, fouled after filtration (FDF), physically cleaned (PC), and chemically cleaned (CC) MF membrane.

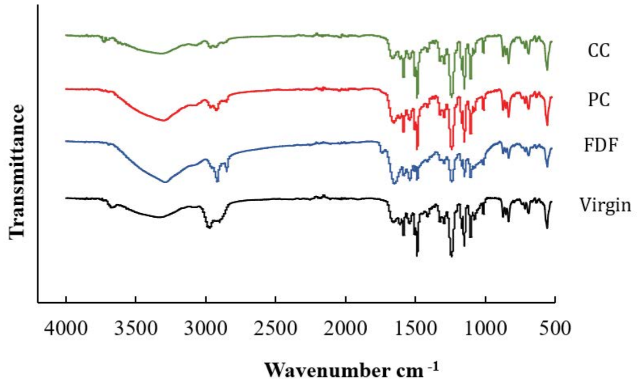

Fig. 6. ATR-FTIR spectra of the virgin, fouled during filtration (FDF), physically cleaned (PC), and chemically cleaned (CC) RO membrane.

foulant were conducted taking into consideration the weakening of the substrate-specific bands. The intensity of these bands was directly related to the wavenumber of IR radiation and the thickness of the accumulated foulant layer. The FTIR spectra in the range of 500–4,000 cm^{-1} of the MF membranes are shown in Fig. 5. ATR-FTIR spectra specify the presence of multiple lines resulting from the vibrations of the sulfonic groups in the range of wavenumbers between 500 and 1,800 cm^{-1}, as well as relatively weak lines in the range of wavenumbers between 2,400 and 3,800 cm^{-1} for MF membrane [42]. They are specific peaks for the PES membrane. The FTIR spectrum in the latter range of wavelength also had weak bands associated with the presence of residual foulant even in the chemically cleaned membrane. C–S vibration reveals due to the intensity of the peak at 1,485 cm^{-1}, which was lower in the chemically cleaned MF membrane than in the virgin MF membrane. This intensity indicated a weakening of the C–S vibration [39].

The FTIR spectra in the range of 500–4,000 cm^{-1} of the BW30-XRF membranes are shown in Fig. 6. In the 500–4,000 cm^{-1} region, peaks were associated with the PSF sublayer and the polyamide (PA) layer [43]. The strong peaks in the region between 700 and 1,750 cm^{-1} were associated with carbonyl groups and amide bands where the PA thin-film membranes presented their characteristic peaks [44]. The second specific peak with the presence of carbonyl groups

Table 6
Membrane characteristics during DMF and RO experiments

Membrane	J_1 (LMH)	J (LMH)	J_2 (LMH)		FRR (%)		RFR (%)	
			PCM	CCM	PCM	CCM	PCM	CCM
MF (2 bar TMP)	77.5	28.7	76.3	79.8	98.5	>100	1.5	–
RO (15 bar TMP)	48.6	36.3	45.2	42.6	93	99	7	12

J_1: pure water flux values before wastewater filtration;
J_2; pure water flux values after wastewater filtration for physically and chemically cleaned membrane;
J: wastewater filtration flux values.
FRR, flux recovery ratio; RFR, relative flux reduction.
PCM, physically cleaned membrane; CCM, chemically cleaned membrane.

and amide bands was indicated between 2,700 and 3,700 cm^{-1} [45]. There was a broad absorbance peak at 3,350 cm^{-1}, corresponding to the overlap of the amine groups (–N–H) and hydroxyl groups (–O–H) stretching vibrations. In order to demonstrate the effectiveness of membrane cleaning, virgin, and chemically cleaned membranes had quite similar spectra except for regions between 2,800 and 3,000 cm^{-1}. The decrease in the peak strength was consistent with the reduced hydrogen bond. All these results suggested that chlorine had been attached to the PA chains [46].

3.2.2.4. Flux recovery

In membrane processes, the reduction of productivity is directly related to fouling development on the membrane surface. Calculated pure water fluxes, wastewater filtration fluxes, FRR, and RFR are given in Table 6. In DMF, physical cleaning provided 98.5% flux recovery. Furthermore, pure water flux was higher than the initial flux after chemical cleaning. The increase in pure water flux is caused by the degradation of the PES membrane due to chemical exposure [47]. In RO, it was achieved to 93% and 99% of flux recovery by physical cleaning and chemical cleaning, respectively.

4. Conclusion

In this study, resource recovery potential in wastewater collected from the effluent of the primary settling tank of a municipal WWTP was evaluated using an integrated DMF+RO process. DMF is a promising technology for concentrating OM from municipal wastewater for improving the energy generation potential by subsequent anaerobic processes. Regarding the RO process, higher-quality of water was generated, and the nutrients were concentrated for further recovery processes. This study provides a promising approach for resource recovery from municipal wastewater. However, SEM, AFM, and ATR-FTIR findings indicated that chemical cleaning led to the deterioration of the membranes, especially for MF membranes.

In future works, periodic flushing should be evaluated for high performance in MF membrane fouling control with also considering the energy consumption. Additionally, more studies are needed to determine and optimize the cleaning procedures for the membrane and to evaluate the nutrient recovery potential of RO concentrate, all of which would help advance efforts toward a circular economy.

References

[1] S. Longo, B.M. d'Antoni, M. Bongards, A. Chaparro, A. Cronrath, F. Fatone, J.M. Lema, M. Mauricio-Iglesias, A. Soares, A. Hospido, Monitoring and diagnosis of energy consumption in wastewater treatment plants. A state of the art and proposals for improvement, Appl. Energy, 179 (2016) 1251–1268.

[2] J. Mateo-Sagasta, L. Raschid-Sally, A. Thebo, Global Wastewater and Sludge Production, Treatment and Use, P. Drechsel, M. Qadir, D. Wichelns, Eds., Wastewater, Springer, Dordrecht, 2015, pp. 15–38. Available at: https://doi.org/10.1007/978-94-017-9545-6_2

[3] Z. Jin, H. Gong, K. Wang, Application of hybrid coagulation microfiltration with air backflushing to direct sewage concentration for organic matter recovery, J. Hazard. Mater., 283 (2015) 824–831.

[4] D. Jiménez-Arias, S.-M. Sierra, F.J. García-Machado, A.L. García-García, A.A. Borges, J.C. Luis, Exploring the agricultural reutilisation of desalination reject brine from reverse osmosis technology, Desalination, 529 (2022) 115644, doi: 10.1016/j.desal.2022.115644.

[5] H. Zhuang, J. Guan, S.Y. Leu, Y. Wang, H. Wang, Carbon footprint analysis of chemical enhanced primary treatment and sludge incineration for sewage treatment in Hong Kong, J Clean Prod. 272 (2020) 122630, doi: 10.1016/j.jclepro.2020.122630.

[6] J. Xiong, S. Yu, Y. Hu, Y. Yang, X.C. Wang, Applying a dynamic membrane filtration (DMF)process for domestic wastewater preconcentration: organics recovery and bioenergy production potential analysis, Sci. Total Environ., 680 (2019) 35–43.

[7] S.K. Lateef, B.Z. Soh, K. Kimura, Direct membrane filtration of municipal wastewater with chemically enhanced backwash for recovery of organic matter, Bioresour. Technol., 150 (2013) 149–155.

[8] Z. Jin, H. Gong, H. Temmink, H. Nie, J. Wu, J. Zuo, K. Wang, Efficient sewage pre-concentration with combined coagulation microfiltration for organic matter recovery, Chem. Eng. J., 292 (2016) 130–138.

[9] T.A. Nascimento, F.R. Mejía, F. Fdz-Polanco, M. Peña Miranda, Improvement of municipal wastewater pretreatment by direct membrane filtration, Environ. Technol. (United Kingdom), 3820 (2017) 2562–2572.

[10] T.A. Nascimento, M.P. Miranda, Continuous municipal wastewater up-concentration by direct membrane filtration, considering the effect of intermittent gas scouring and threshold flux determination, J. Water Process Eng., 39 (2021) 101733, doi: 10.1016/j.jwpe.2020.101733.

[11] A.M. Ravazzini, A.F. van Nieuwenhuijzen, J.H.M.J. van der Graaf, Direct ultrafiltration of municipal wastewater: Comparison between filtration of raw sewage and primary clarifier effluent, Desalination, 178 (2005) 51–62.

[12] P.M. Melia, A.B. Cundy, S.P. Sohi, P.S. Hooda, R. Busquets, Trends in the recovery of phosphorus in bioavailable forms from wastewater, Chemosphere, 186 (2017) 381–395.

[13] F. Tang, H.-Y. Hu, L.-J. Sun, Y.-X. Sun, N. Shi, J.C. Crittenden, Fouling characteristics of reverse osmosis membranes at different positions of a full-scale plant for municipal wastewater reclamation, Water Res., 90 (2016) 329–336.

[14] Y. Li, L.N. Sim, J.S. Ho, T.H. Chong, B. Wu, Y. Liu, Integration of an anaerobic fluidized-bed membrane bioreactor (MBR) with zeolite adsorption and reverse osmosis (RO) for municipal wastewater reclamation: comparison with an anoxic-aerobic MBR coupled with RO, Chemosphere, 245 (2020) 125569, doi: 10.1016/j.chemosphere.2019.125569.

[15] Y.H. Wu, Z. Chen, X. Li, Y.H. Wang, B. Liu, G.Q. Chen, L.W. Luo, H.B. Wang, X. Tong, Y. Bai, Y.Q. Xu, N. Ikuno, C.F. Li, H.Y. Zhang, H.Y. Hu, Effect of ultraviolet disinfection on the fouling of reverse osmosis membranes for municipal wastewater reclamation, Water Res., 195 (2021) 116995, doi: 10.1016/j.watres.2021.116995.

[16] Z. Jin, F. Meng, H. Gong, C. Wang, K. Wang, Improved low-carbon-consuming fouling control in long-term membrane-based sewage pre-concentration: the role of enhanced coagulation process and air backflushing in sustainable sewage treatment, J. Membr. Sci., 529 (2017) 252–262.

[17] Y.x. Zhao, P. Li, R.h. Li, X.y. Li, Direct filtration for the treatment of the coagulated domestic sewage using flat-sheet ceramic membranes, Chemosphere, 223 (2019) 383–390.

[18] S. Hube, J. Wang, L.N. Sim, T.H. Chong, B. Wu, Direct membrane filtration of municipal wastewater: linking periodical physical cleaning with fouling mechanisms, Sep. Purif. Technol., 259 (2021) 118125, doi: 10.1016/j.seppur.2020.118125.

[19] T. Yu, L. Meng, Q.B. Zhao, Y. Shi, H.Y. Hu, Y. Lu, Effects of chemical cleaning on RO membrane inorganic, organic and microbial foulant removal in a full-scale plant for municipal wastewater reclamation, Water Res., 113 (2017) 1–10.

[20] Y.H. Cai, N. Galili, Y. Gelman, M. Herzberg, J. Gilron, Evaluating the impact of pretreatment processes on fouling of reverse osmosis membrane by secondary wastewater, J. Membr. Sci., 623 (2021) 119054, doi: 10.1016/j.memsci.2021.119054.
[21] J. Wu, Y. Zhang, J. Wang, X. Zheng, Y. Chen, Municipal wastewater reclamation and reuse using membrane-based technologies: a review, Desal. Water Treat., 224 (2021) 65–82.
[22] P. Xu, C. Bellona, J.E. Drewes, Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: membrane autopsy results from pilot-scale investigations, J. Membr. Sci., 353 (2010) 111–121.
[23] X. Tong, Y.H. Wu, Y.H. Wang, Y. Bai, X.H. Zhao, L.W. Luo, Y. Mao, N. Ikuno, H.Y. Hu, Simulating and predicting the flux change of reverse osmosis membranes over time during wastewater reclamation caused by organic fouling, Environ. Int., 140 (2020) 105744, doi: 10.1016/j.envint.2020.105744.
[24] H. Özgün, H. Sakar, M. Ağtaş, İ. Koyuncu, Investigation of pretreatment techniques to improve membrane performance in real textile wastewater treatment, Int. J. Environ. Sci. Technol., 20 (2023) 1539–1550.
[25] S. Jiang, Y. Li, B.P. Ladewig, A review of reverse osmosis membrane fouling and control strategies, Sci. Total Environ., 595 (2017) 567–583.
[26] A. Matin, T. Laoui, W. Falath, M. Farooque, Fouling control in reverse osmosis for water desalination & reuse: current practices & emerging environment-friendly technologies, Sci. Total Environ., 765 (2021) 142721, doi: 10.1016/j.scitotenv.2020.142721.
[27] T. Xiao, Z. Zhu, L. Li, J. Shi, Z. Li, X. Zuo, Membrane fouling and cleaning strategies in microfiltration/ultrafiltration and dynamic membrane, Sep. Purif. Technol., 318 (2023) 123977, doi: 10.1016/j.seppur.2023.123977.
[28] T.A. Nascimento, F. Fdz-Polanco, M. Peña, Membrane-based technologies for the up-concentration of municipal wastewater: a review of pretreatment intensification, Sep. Purif. Rev., 491 (2020) 1–19.
[29] APHA, Standard Methods for the Examination of Water and Wastewater Federation, Water Environmental American Public Health Association, Washington, DC, USA, 2017.
[30] V.T. Kuberkar, R.H. Davis, Modeling of fouling reduction by secondary membranes, J. Membr. Sci., 1681 (2000) 243–258.
[31] K. Noyan, B. Allı, D. Okutman Taş, S. Sözen, D. Orhon, Relationship between COD particle size distribution, COD fractionation and biodegradation characteristics in domestic sewage, J. Chem. Technol. Biotechnol., 928 (2017) 2142–2149.
[32] T. Siwiec, L. Reczek, M.M. Michel, B. Gut, P. Hawer-Strojek, J. Czajkowska, K. Jóźwiakowski, M. Gajewska, P. Bugajski, Correlations between organic pollution indicators in municipal wastewater, Arch. Environ. Prot., 44 (2018) 50–57.
[33] D. Dubber, N.F. Gray, Replacement of chemical oxygen demand (COD) with total organic carbon (TOC) for monitoring wastewater treatment performance to minimize disposal of toxic analytical waste, J. Environ. Sci. Health. Part A Toxic/Hazard. Subst. Environ. Eng., 45 (2010) 1595–1600.
[34] J. Tauber, V. Parravicini, K. Svardal, J. Krampe, Quantifying methane emissions from anaerobic digesters, Water Sci. Technol., 809 (2019) 1654–1661.
[35] S. Hube, M. Eskafi, K.F. Hrafnkelsdóttir, B. Bjarnadóttir, M.Á. Bjarnadóttir, S. Axelsdóttir, B. Wu, Direct membrane filtration for wastewater treatment and resource recovery: a review, Sci. Total Environ., 710 (2020) 136375, doi: 10.1016/j.scitotenv.2019.136375.
[36] M. Kumar, M. Badruzzaman, S. Adham, J. Oppenheimer, Beneficial phosphate recovery from reverse osmosis (RO) concentrate of an integrated membrane system using polymeric ligand exchanger (PLE), Water Res., 41 (2007) 2211–2219.
[37] W. Wang, Q. Yue, K. Guo, F. Bu, X. Shen, B. Gao, Application of Al species in coagulation/ultrafiltration process: influence of cake layer on membrane fouling, J. Membr. Sci., 572 (2019) 161–170.
[38] E. Arkhangelsky, D. Kuzmenko, V. Gitis, Impact of chemical cleaning on properties and functioning of polyethersulfone membranes, J. Membr. Sci., 305 (2007) 176–184.
[39] S.S. Madaeni, S. Samieirad, Chemical cleaning of reverse osmosis membrane fouled by wastewater, Desalination, 257 (2010) 80–86.
[40] L.Y. Ng, A. Ahmad, A.W. Mohammad, Alteration of polyethersulphone membranes through UV-induced modification using various materials: a brief review, Arabian J. Chem., 10 (2017) S1821–S1834.
[41] M.Y. Ashfaq, M.A. Al-Ghouti, N. Zouari, Functionalization of reverse osmosis membrane with graphene oxide to reduce both membrane scaling and biofouling, Carbon, 166 (2020) 139500, doi: 10.1016/j.carbon.2020.05.017.
[42] H. Guo, X. Tang, G. Ganschow, G.V. Korshin, Differential ATR-FTIR spectroscopy of membrane fouling: contributions of the substrate/fouling films and correlations with transmembrane pressure, Water Res., 161 (2019) 27–34.
[43] S. Saki, N. Uzal, Surface coating of polyamide reverse osmosis membranes with zwitterionic 3-(3,4-dihydroxyphenyl)-L-alanine (L-DOPA) for forward osmosis, Water Environ. J., 34 (2020) 400–412.
[44] I.M. El-Azizi, R.G.J. Edyvean, Performance evaluation and fouling characterisation of two commercial SWRO membranes, Desal. Water Treat., 51 (2009) 34–41.
[45] O. Akin, F. Temelli, Probing the hydrophobicity of commercial reverse osmosis membranes produced by interfacial polymerization using contact angle, XPS, FTIR, FE-SEM and AFM, Desalination, 278 (2011) 387–396.
[46] G.D. Kang, C.J. Gao, W.D. Chen, X.M. Jie, Y.M. Cao, Q. Yuan, Study on hypochlorite degradation of aromatic polyamide reverse osmosis membrane, J. Membr. Sci., 300 (2007) 165–171.
[47] B. Malczewska, A. Żak, Structural changes and operational deterioration of the UF polyethersulfone (PES) membrane due to chemical cleaning, Sci. Rep.-UK, 9 (2019) 422, doi: 10.1038/s41598-018-36697-2.