Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2013.826333

52 (2014) 670–677
January

Taylor & Francis
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Comparison of the performance of ceramic microfiltration and ultrafiltration membranes in the reclamation and reuse of secondary wastewater

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Received 5 May 2013; Accepted 12 May 2013

ABSTRACT

Advanced treatment of secondary wastewater generally has been achieved using polymeric microfiltration and ultrafiltration membranes. Newly developed ceramic membranes offer distinctive advantages over the currently employed membranes and were recently introduced for the purpose. This paper presents results of a pilot study designed to investigate the application of ceramic microfiltration (MF) and ultrafiltration (UF) membranes in the recovery of water from secondary wastewater. Synthetic wastewater similar to the quality of secondary treated wastewater was fed to ceramic MF and UF system in a cross-flow mode. The filtration experiments revealed that the flux recovery through tubular ceramic MF membrane was more sensitive to the variation in TMP compared with the tubular ceramic UF membrane over the range of TMP studied. The resistance in series model was used for the evaluation of the resistance to the permeate flux. The results revealed that for ceramic UF membrane, the contribution to the total resistance of fouling was higher than the inherent of the clean membrane resistance. However, both the clean membrane resistance and the fouling resistance contribute equally in the case of MF membrane. Various wastewater indices were measured to evaluate the effectiveness of the filtration treatment. The ceramic UF membrane consistently met water quality in the permeate in terms of colour, turbidity, chemical oxygen demand and absorbance, suggesting that the permeate water could be made to be reused or recycled for suitable purposes. However, MF membrane appeared to be incompetent with respect to the removal of colour. The unified membrane fouling index (UMFI) was used to measure the fouling potential of both the membranes. The result showed that for UF membrane, the value of UMFI is one order of magnitude higher than MF membrane. The overall results suggest that there were significant differences in the performance of both the ceramic UF and MF membranes that are likely to impact on the operation and maintenance of the membrane system.

Keywords: Ceramic membrane; Microfiltration; Ultrafiltration; Secondary wastewater; Removal efficiency

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Presented at the Fifth Annual International Conference on "Challenges in Environmental Science & Engineering—CESE 2012" Melbourne, Australia, 9–13 September 2012

1. Introduction

It has been widely recognised that the application of low-pressure membranes, such as microfiltration (MF) and ultrafiltration (UF) processes to treat secondary wastewater provides enhanced water quality compared with the conventional treatment processes [1,2]. This has resulted in the increased uptake of MF and UF in meeting the stringent environmental discharge standards and producing recycled water for higher value uses. However, these membranes have been perceived to incur higher capital and operating costs as well as operational challenges as a result of fouling problems particularly the polymeric membranes compared with the traditional wastewater treatment methods. The nature and complexity of fouling problems are generally depend to the type of feedwater [3] and the increase in operating costs are associated with the higher fluid pumping, more membrane cleaning and replacements. For example, membrane fouling encountered in reclamation of municipal wastewater is largely due to the extensive variability of wastewater quality and the complex and unstable nature of the organic materials present in the wastewater, which poses a serious operational problem for efficiently operating these systems [4].

However, with the recent development of ceramic membranes, it appears that these operational challenges could be effectively addressed improving both the technical and economic viability of the application of membranes for the treatment of wastewater. Ceramic membranes offer numerous advantages as they have superior physical integrity, chemical resistance and thermal stability compared with polymeric membranes [5]. Also, ceramic membrane exhibits good stability to organic media, resistant to bacterial action, ability to process highly viscous fluids and finally backwashing with possibility of regeneration after fouling [6]. These unique properties of ceramic membranes coupled with long and reliable lifetime and decreasing membrane cost have significant advantages over polymeric membranes. Recent research demonstrated that ceramic membranes treating wastewater can produce a high permeate flux, high water recoveries and with less frequent chemical cleaning [6,7].

Even though few researches have shown that the ceramic membrane is a potential candidate for the treatment of sewage effluent, it has not been used widely in the wastewater treatment areas as the performance of these membranes has not been rigorously investigated and understood as compared to the polymeric membranes. Our earlier studies investigated the effect of membrane materials by comparing the performance of polymeric and ceramic UF membranes and have shown that the ceramic UF membrane consistently met water quality in the permeate [4].

This study is a continuation of our previous study, which evaluates and compares the performance of ceramic membranes with different pore sizes (MF and UF) with respect to fouling and removal efficiency. The objective of this manuscript is to describe pilot-scale experiments, where ceramic MF and UF membranes were tested with the same water sources to determine their filtration performance under similar operating conditions. This study has focused on (1) using commercially available ceramic MF and UF membranes as representative samples (2) evaluating and comparing their filtration performance (3) interpreting the results. The effectiveness of filtration treatments were assessed based on the removal of various pollution indices of the wastewater such as chemical oxygen demand (COD), colour, turbidity and absorbance at 254 nm. Further, the fouling mechanism in the present case was evaluated by fitting the experimental data to a resistance in series model [6,8]. Also, the unified membrane fouling index (UMFI) was used to quantitatively assess the fouling potential of the membranes [9].

2. Materials and methods

2.1. Characteristics of synthetic secondary wastewater

A synthetic secondary wastewater was prepared from a sterile concentrated solution with the composition shown in previous study [10] and used throughout the experiments. The synthetic secondary wastewater contains organic compounds such as humin, tannin, lignin, protein and high-molecular carbohydrates (Table 1). The physical and chemical characteristics of the synthetic wastewater are shown in Table 2.

2.2. Membranes and pilot-scale filtration system

The experiments were performed with tubular ceramic MF and UF membranes. Both the ceramic membranes used in this work were supplied by Tami industries. The characteristics of these membranes are shown in Table 3. The same experimental set-up and filtration protocol as described in our previous paper has been used for this study [4]. Experiments were carried out using a pilot-scale cross-flow mode filtration apparatus supplied by Liquids Technology, Australia. A diagram of the membrane pilot system is shown in Fig. 1. A synthetic wastewater sample was prepared and diluted with freshwater in a 1,000-L container and mixed thoroughly and continuously to create uniform concentration of secondary wastewater. The container was connected with the buffer

Table 1
Composition of the synthetic secondary wastewater

Substances	Composition (mg/l)
Beef extract	1.8
Humic acid	4.25
Peptone	2.7
Sodium lignin sulfonate	2.4
Tannic acid	4.18
Sodium lauryle sulfate	0.9
Gum powder	4.7
K_2HPO_4	7
$(NH_4)_2SO_4$	7.1
$MgSO_4$	0.71
NH_4HCO_3	19.8

Table 2
Physicochemical characterisation of synthetic wastewater

Parameters	Value
Chemical oxygen demand (mg/l)	39
Electrical conductivity (μS/cm)	340
Turbidity (NTU)	22.8
Colour (filtered through filter paper 0.45 μm), ADMI	81
Absorbance (254 nm)	0.591
pH	7.73

feed-tank and feed from the buffer tank was pumped through a 100-μm prefilter unit to remove any large particles present in the feed. The temperature of the feed could be raised or lowered using water fed heat exchanger/chiller. The feed was then passed through either a ceramic MF or UF membrane stage with the feed pressure controlled using a diaphragm valve.

Table 3
Characteristics of membranes

Specification	Ceramic MF membrane	Ceramic UF membrane
Manufacturer	Tami industries	Tami industries
Material	Zirconium/ Titanium dioxide	Zirconium/ Titanium dioxide
Molecular weight cut off/pore size	1.4 μm	1 kDa
Membrane	Hydrophilic	Hydrophilic
membrane area	0.5 m^2	0.35 m^2
Operating pressure	<10 bar	<10 bar
Operating pH	0–14	0–14
Operating temperature	<350℃ but change in temperature must lower than 10℃/min	<350℃ but change in temperature must lower than 10℃/min

The cross-flow velocity of 0.2 m/s was used during the experiments. The pilot-scale unit was equipped with three pressure gauges at feed inlet, retentate outlet and permeate outlet. Operation modes with different transmembrane pressures (TMPs) were created by adjusting the control valves appropriately. The hydraulic flow metres installed on the system were used to measure the flow rates of permeate and retentate.

2.3. Membrane characterisation

Both the ceramic membranes were operated with clean tap water prior to wastewater filtration experiments, in order to evaluate the dependence of the

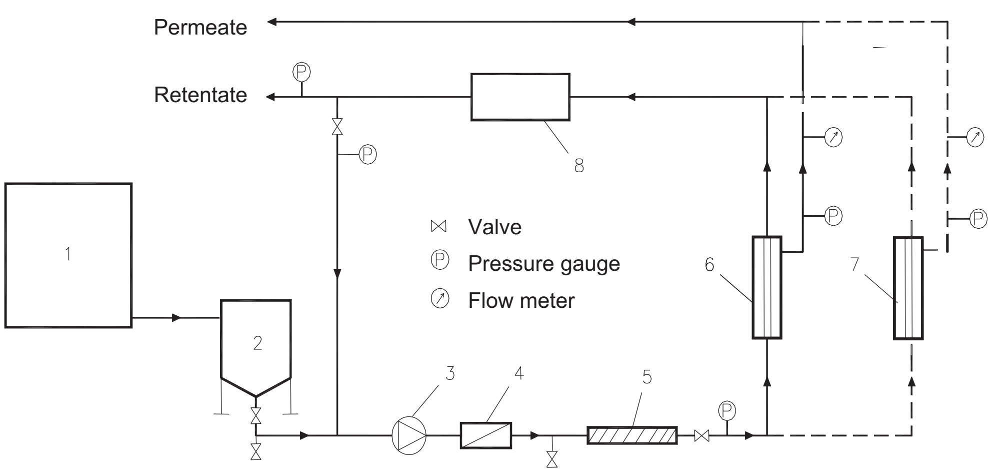

Fig. 1. Schematic diagram of experimental installation. (1) 1,000-L feed tank; (2) 50-L buffer tank; (3) pump; (4) pre-filtration unit; (5) chiller; (6) ceramic MF membrane; (7) ceramic UF membrane; and (8) heater.

water flux on the TMP. The applied pressures during this process were adjusted according to the membrane filtration ranges: thus, the maximum TMP was 0.9 bar for the MF membrane and 3.3 bar for the UF membrane. The water flux increases with increasing TMP, obtaining a linear relationship with high correlation coefficients. The slope of this straight line is the pure water hydraulic permeability (L_p) which is better characterise a membrane in filtration processes. The increase in the water filtration rate over the pressure range was higher in the MF than the UF membrane with L_p values of 378 and 17 l/h m^2 bar, respectively, at 25˚C. Although both the membranes are hydrophilic, the UF membrane with a smaller pore size has larger membrane resistance affects the permeability compared with larger pore size MF membrane.

2.4. Analytical method

Permeate samples were collected at predetermined intervals and stored at 4˚C until analysis. All the analytical procedures were followed according to the standard methods [11]. All experiments were conducted in duplicate. The COD in the samples was determined by spectroquant Nova 60. Absorbance was measured as the absorbance values at 254 nm by spectroquant Photo 300. Absorbance at 254 nm provides an indication of aromatic organic compounds present in the sample. Colour was determined using Spectrophotometer DR/4000 V. Other parameters including: turbidity pH and electrical conductivity were analysed with 2100P, pH 315 i/SET and LF 330/SET metres, respectively.

3. Results and discussion

3.1. Comparison of ceramic MF and UF membranes

A comparison between tubular ceramic MF and UF membranes treating the synthetic secondary wastewater is shown in Fig. 2. The result is consistent with the expectation that MF membrane exhibits higher flux rates compared with the UF membrane due to the larger pore sizes. There is about 50% permeate flux decline in MF membrane and more than 60% in UF membrane across all the TMPs studied. For both MF and UF, organic matter is the primary reason for irreversible fouling, since the dissolved and colloidal organic matter is much smaller than the pore sizes of MF or UF membrane that enable them to adsorb onto or block the pores of the membranes. On the other hand reversible fouling occurs as external fouling or cake formation, which is mainly caused by filtration-induced macro-solute or

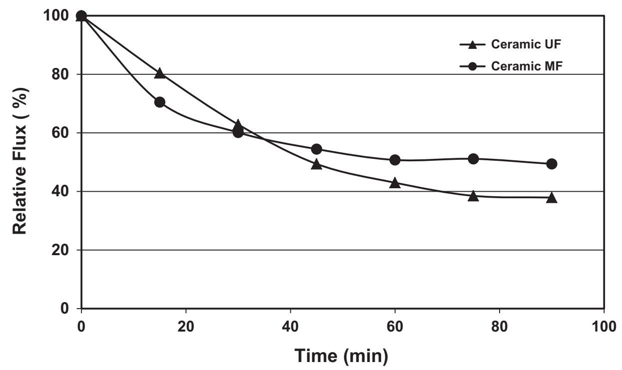

Fig. 2. Flux decay patterns of both ceramic UF (TMP = 2.8 bar and T = 25˚C) and MF membranes (TMP = 0.7 bar and T = 25˚C) as a function of time.

particle deposition. During the initial stages of the filtration, the undesired particulate pollutants are rejected by the size of the membrane pores. After this stage, the particles start accumulating near the membrane surface to form a cake layer that assists in pollutant removal. The result shows that for both the membranes, there is a large initial variations in the permeate flux with the variations diminishing significantly as cake layer is formed during the filtration process which is consistent with other studies [12]. Both the pore blockage and cake layer decrease the permeate flux rate, which are key factors governing the application of membrane system. In these trails, the steepest permeate flux decline was observed in MF membrane upto 45 min, and a final stage, where the permeate flux started to stabilise and reached a pseudo steady state. On the other hand, UF membrane showed a similar trend as MF during the initial period, but with continuous decline upto 60 min and later reached a pseudo steady state. This result shows that for both the membranes, pore blocking and adsorption of the substance onto the membrane surface is intense during the initial stage following concentration polarisation, and cake formation dominates in the later stages. The results of this study are consistent with other studies which show that the membrane with higher permeability consistently fouls faster at the initial filtration than membranes with lower permeability [13]. However, the overall flux decline for UF membrane is very significant compared with MF membrane. This fouling mechanism was confirmed by fitting the resistance in series model that is discussed in the following section (Section 3.3).

According to Zheng et al. the most pronounced fouling was caused by large colloids and dissolved organic substances whose size fraction would be in the range of 0.45–1.2 μm and less than 0.45 μm, respectively, and contributes more than 50% of the total fouling [13]. Both colloids and dissolved substances that are smaller than the pore size of the MF membrane cause internal pore blocking, while the larger particles cause cake layer formation. Whereas for the UF membranes the dissolved substances whose size fraction less than the pore size of the UF membranes cause internal fouling and the colloids and particles cause cake layer formation. In our study, it can also be concluded that a MF membrane with a pore size of 1.4 μm will not completely remove foulants as most of the organics pass through the membrane without causing significant membrane fouling compared with UF membrane with a pore size of 1 kDa. These results are qualitatively supported by the removal efficiencies determination discussed below (Section 3.5).

3.2. Effects of TMP on the permeate flux

The effects of TMP on the steady-state permeate flux in the experiments carried out with MF and UF membranes are shown in Fig. 3. It was observed that for the MF membrane, the steady-state flux increased gradually from 0.5 to 0.7 bar and more rapidly from 0.7 to 0.9 bar. The overall flux was increasing linearly with increasing pressure in the range of TMP used which is consistent with other studies [14,15]. On the other hand, the linear variation of the permeate flux with TMP was not observed, showing that the fouling phenomena is more severe in UF compared with MF membrane. This is consistent with other studies, where they have found that increase in TMP from 1.5 bar to 2.5 bar showed no increase in steady-state permeate flux, when ceramic membrane was used for the treatment of sewage effluents. This is because high TMP could lead to a more rapid flux decline and

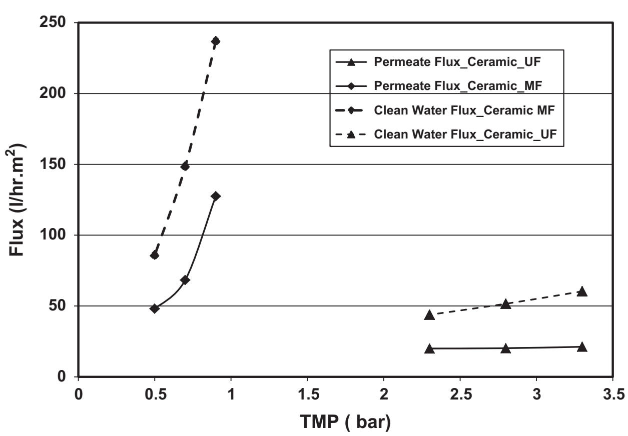

Fig. 3. Effect of TMP on steady-state permeate flux ($T = 25$°C).

lower steady-state flux due to more compact cake formation and greater in-pore plugging [12]. Further the steady-state permeate flux shows closer agreement with the non-fouled membrane (clean water flux) performance in the case of MF compared with UF membrane.

In addition, regression analysis conducted on the plots in Fig. 3 led to a value of 200 l/h m^2 bar for the slope of the straight line for MF membrane compared with the value of the hydraulic permeability for clean water obtained for MF membrane (378 l/h m^2 bar). For the UF membrane, the regression analysis provided a slope of 1.2 l/h m^{2}bar. This is a very low value compared with the value of the hydraulic permeability for pure water (17 l/h m^2 bar). This also confirms the higher fouling in UF membrane compared with MF membrane.

3.3. Filtration resistance

The determination of filtration resistances from flux data has provided additional insight on the fouling mechanism. According to constant pressure theory, the permeate flux J is expressed by the resistance in-series model, where ΔP is the TMP, μ the permeate viscosity and R_t the total hydraulic resistance.

$$J = \frac{\Delta P}{\mu R_{\mathrm{t}}} \tag{1}$$

$$R_{\mathrm{t}} = R_{\mathrm{m}} + R_{\mathrm{f}} \tag{2}$$

$$R_{\mathrm{f}} = R_{\mathrm{if}} + R_{\mathrm{ef}} \tag{3}$$

where R_{m} is the hydraulic resistance of clean membrane and R_{f} the total (overall) fouling resistance. The external fouling resistance R_{ef} (reversible resistance) includes concentration polarisation and deposition of solids on the membrane surface. The internal fouling resistance R_{if} (irreversible resistance) is due internal fouling such as pore blocking. Table 4 summarises the resistances in every experiment conducted using MF and UF membranes.

It can be seen that in general terms, the contribution to the total resistance of the fouling resistance (combined external and internal) is same as the inherent resistance of the clean membrane in the case of the MF membrane and higher in the case of the tubular UF membrane. Thus, for the UF membrane, R_{m} contributed to 36% of R_{t} and R_{f} provided the remaining 62%. In particular irreversible resistance (R_{if}) is significantly higher than reversible resistance (R_{ef}). In the case of the MF membrane, both R_{m} and

Table 4
Values of filtration resistances obtained in the MF and UF experiments performed

TMP, bar	$R_m \times 10^{-13}$ (m^{-1})	$R_t \times 10^{-13}$ (m^{-1})	$R_f \times 10^{-13}$ (m^{-1})	$R_{ef} \times 10^{-13}$ (m^{-1})	$R_{if} \times 10^{-13}$ (m^{-1})
Ceramic MF membrane					
0.9	0.137	0.262	0.125	0.017	0.108
0.7	0.170	0.375	0.206	0.128	0.078
0.5	0.210	0.385	0.175	0.052	0.123
Ceramic UF membrane					
3.3	1.968	6.162	4.194	1.166	3.028
2.8	1.955	5.964	4.010	0.282	3.727
2.3	1.890	4.315	2.425	0.232	2.193

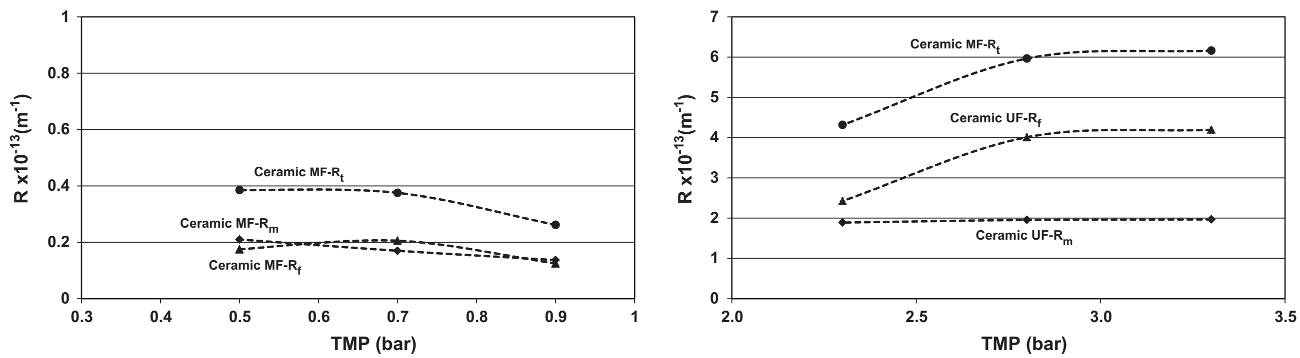

Fig. 4. Resistance values as function of TMP for both ceramic UF and MF membranes.

R_f contributes equally. Further, both the R_{if} and R_{ef} contributes equally for the MF membrane and is much lower compared with the UF membrane. Overall, the membrane resistance and fouling resistance of the UF membrane is greater than the MF membrane.

With respect to the influence of TMP, the R_m presented almost the same value across all the TMP for both the membranes (Fig. 4). The small changes in the values of R_t and R_f shows a there is a slight influence of TMP on resistance that agrees well with the linear increase in permeate flux at higher TMP in case of the MF membrane. On the other hand, for the UF membrane, an increase in TMP leads to an increase in R_t and R_f and not much increase in permeate flux at high TMP. As a consequence, a TMP increase results first in greater internal fouling due to pore blocking and adsorption of dissolved components and then an increment of the external cake fouling due to the colloids and particles present in the wastewater.

3.4. *Unified membrane fouling index (UMFI)*

UMFI is a measure of the total fouling capacity of the feedwater. The UMFI was established based on Hermia's filtration model, assuming that cake filtration was the principal fouling mechanism but included a potential contribution from cake layer formation and pore blocking [16]. This is explained as the slope of the curve of the reciprocal of the normalised flux (J_0/J) vs. accumulated specific permeate volume (v), due to the following linear relationship,

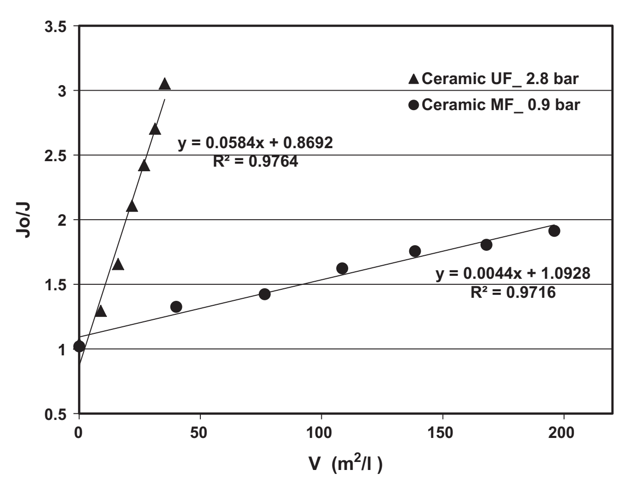

Fig. 5. J/J_0 as a function of specific permeate throughput.

$$\frac{J_0}{J} = \left(\frac{\alpha C_W}{R_m}\right)v + 1 \quad (4)$$

where J_0 is the permeate flux at time $t = 0$, J is the permeate flux through the membrane for the wastewater being tested, α is the specific resistance of the cake layer, C_w is the foulant concentration in the feed, v is the accumulated specific permeate volume (permeate volume per unit membrane area). As shown in Fig. 5, UMFI for total fouling was calculated by unforced linear regression of the experimental data using Eq. (4). A larger UMFI value indicates a faster decrease in the normalised specific flux and represents the greater the membrane fouling potential.

The UMFI value for the ceramic MF and UF membranes are 0.0044 and 0.0584 m^2/l, respectively. This result shows that for UF membrane, the value of UMFI is one order of magnitude higher than MF membrane. This result is again consistent with the above results indicating ceramic UF has high-fouling rate compared with ceramic MF membrane.

3.5. Removal efficiency

The effectiveness of the filtration processes in the removal of organic matter present in wastewater was evaluated by removal efficiencies. As previously explained, the pollution indices selected in the present study were COD, absorbance at 254 nm, turbidity and colour. The removal efficiency in the case of colour was defined by the following (f_{Colour}):

$$f_{Colour} = \frac{Colour_F - Colour_P}{Colour_F} \times 100 \quad (5)$$

where $Colour_F$ and $Colour_P$ represent the colour on the feed and permeate streams, respectively. The removal efficiencies were determined using Eq. (5), for all the experiments performed and shown in Fig. 6.

In the case of MF membrane, there was a very high removal of turbidity (>95%) and high removal of COD (>80%) and moderate removal of absorbance (>75%) under all TMP studied and very low removal of colour (0–50%) as the TMP was increased. This membrane has limitation that it cannot remove contaminants that are smaller than the membrane pore size and this limitation can be overcome by a combination with proper treatment processes. On the other hand, with UF membrane there was very high removal of turbidly (>95%), COD (>95%) and colour (>92%) and high removal of absorbance (>85%). This result again confirms that the high rejections of pollution parameters with UF membrane cause lower permeate flux and higher fouling resistance compared with MF membrane.

4. Conclusion

This study provides the comparison between ceramic UF and MF in the recovery of water from secondary wastewater and arrives at the following conclusions.

(1) Permeate flux increases with TMP for tubular ceramic MF membrane and increases but at a lower rate for tubular ceramic UF membrane over the range of TMP studied.
(2) Membrane fouling, in particular in-pore adsorption/deposition of particles, had a critical influence on the dynamic behaviour of flux reduction and change of rejection characteristics

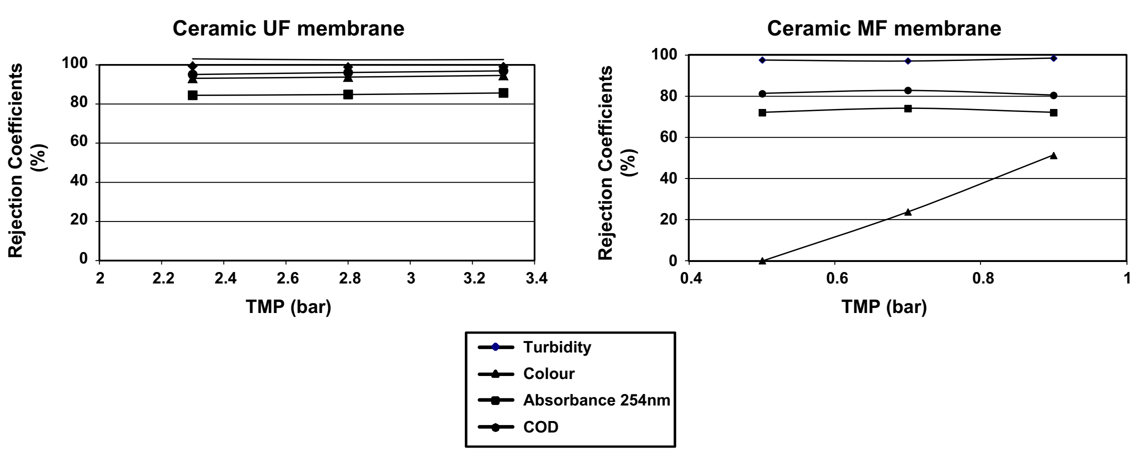

Fig. 6. Influence of TMP on the removal efficiencies ($T = 25°C$).

of the membrane system during the filtration processes.

(3) The results revealed that for ceramic UF membrane, the contribution to the total resistance of fouling was higher than the inherent of the clean membrane resistance. In particular irreversible resistance was higher than reversible resistance. This indicated that pore blocking and adsorption in the membrane predominated over cake layer and concentration polarisation. However, both the clean membrane resistance and the fouling resistance contribute equally in the case of MF membrane.

(4) The ceramic UF membrane consistently met water quality in the permeate in terms of colour, turbidity, COD and absorbance, suggesting that the permeate water could be made to be reused or recycled for suitable purposes. However, MF membrane appeared to be incompetent with respect to the removal of colour.

Although the tubular ceramic UF membrane obtained high removal efficiencies for the selected pollutant parameters, this membrane is suitable only with significant pre-treatment due to the high fouling resistance and low permeates flux. On the other hand, MF membrane seems to be suitable, when it is provided with additional treatment required to remove the colour. These results suggest that different design criteria and operation and maintenance protocol have to be used in order to achieve similar level of performance outcomes for MF and UF membrane types. However, further studies on real secondary wastewater are needed from a practical point of view.

Acknowledgements

The authors wish to acknowledge the student Duy Anh Nguyen of the School of Engineering, Deakin University for his support to operate the membrane unit.

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