Membrane coupled with physic chemical treatment in water reuse

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Abstract

In this study, short-term experiments were conducted with different configurations of membrane hybrid systems to treat biological treated sewage effluent containing refractory organic pollutants: (i) submerged hollow fiber microfiltration (SMF) alone; (ii) spiral flocculator (SF)-SMF without settling; (iii) SF-PAC-SMF without settling and (iv) SMF with magnetic ion exchange resin MIEX® pretreatment. The results indicated that the pre-flocculation of SF could improve the mitigation of membrane fouling significantly even when the system was operated at a high membrane filtration rate. The transmembrane pressure (TMP) of SF-PAC-SMF only increased marginally (0.8 kPa over 8 hours). SF-SMF without the addition of powdered activated carbon (PAC) also took a relatively long duration for the TMP to increase. The TMP only increased by 2.5 kPa over 8 hours. The SF-PAC-MF system resulted in a high dissolved organic carbon (DOC) removal of more than 96%. When used as pre-treatment to submerged membranes, the fluidized bed MIEX® contactor could remove a significant amount of organic matter in wastewater. This pre-treatment helped to reduce the membrane fouling and kept the TMP low during the membrane operation.

Keywords: ion exchange, MIEX®, refractory organic matter removal, spiral flocculator, submerged membrane adsorption hybrid system

1. INTRODUCTION

Fouling is a major problem when operating a membrane system for water and wastewater treatment. It is caused by deposition and accumulation of mainly effluent organic matter (EfOM) on the surface and inside pores of the membrane resulting in reduced flux and higher energy requirements. Pre-treatment of the feed water can significantly enhance the performance of membrane systems and includes coagulation/flocculation, adsorption by activated carbon and ion exchange. Although pre-treatment leads to an increase in additional cost, it can be recovered over the life of the plants in terms of extending the life of the membrane and improving the performance of the system.

Coagulation-microfiltration hybrid system

Mircofiltration (MF) with a large pore size can remove a very small amount of dissolved organic compounds especially trace organic compounds; taste and odor-causing materials and synthetic organic chemicals. In order to increase the rejection by the membrane, the dissolved organics need to be aggregated into particulates. This can be done by adding coagulants and/or flocculants such as alum, aluminium sulfate and ferric chloride. Consequently, coagulation has been employed widely as a pre-treatment to MF/UF.

A combined system of coagulation and MF was studied by Vickers et al.(1995), Al-Malack & Anderson (1996), and Mo & Huanga (2003) in treating secondary wastewater for reuse. Their results showed that pre-treatment with coagulation helped reduce suspended solids, phosphorus and arsenic; optimised backwash efficiency and prevented fouling (Mo & Huanga 2003; Shonet al.2004). By increasing the particle sizes, coagulation reduces the potential of internal pore blockage of the membrane while increasing cake porosity (Junget al.2006).

In addition, coagulation pre-treatment was also found to improve the removal of hydrophobic compounds, resulting in the increase of permeate flux (Oh & Lee 2005; Chen et al.2007). Correspondingly, the flux was reported to recover completely after the backwash of each filtration cycle compared with a gradual 20% flux decline when the membrane was operated alone (Chenet al.2007). However, the performance of coagulation/membrane combined system is significantly dependent on the nature of the organic matter, coagulant dosage and floc characteristics leading to a process which is difficult to control (Schafer 2001).

Adsorption-microfiltration hybrid system

Combining activated carbon (AC) adsorption with membrane filtration MF is another promising pretreatment to improve DOC removal efficiency. In the hybrid system, the small molecular species not usually rejected by the membrane alone are adsorbed onto the AC particles which subsequently can be separated easily by the MF process. As a result, organic compounds are removed. There are two types of AC available, which are powdered activated carbon (PAC) and granular activated carbon (GAC). Of the two, PAC is prefered due to its large surface area per unit volume and its affinity for a wide range of dissolved organics (Guo et al.2005).

In the combined system, PAC can be dosed continuously to the membrane reactor or dosed at the beginning of the membrane filtration cycle. Previous studies showed that

PAC pre-adsorption contributed to higher DOC and disinfection by-products (DBPs) removal, flux enhancement and fouling reduction, thus prolonging the continuous filtration time (Lebeau et al.1998). One type of the hybrid system known as submerged membrane adsorption hybrid system (SMAHS) integrates the entire treatment activity including adsorption/biodegradation, liquid-solid separation and sludge accumulation into one single unit, which can offset the disadvantage of the large equipment size and space requirement (Mozia et al.2005; Guo et al.2006). The system can be operated consistently for a long period with a very low energy requirement (0.2 kWh/m^3) and without any major sludge problems (Seo et al. 1996). The studies conducted by Kimet al. (2008) showed a relatively high removal efficiency of 85% of total organic carbon (TOC) with 40 g/L of PAC for 40 days. Using a lower dose of 5 g/L(reactor volume) of PAC added at the beginning of the experiment, Guoet al.(2006) obtained approximately 84% of TOC removal for a 15-day operation.

While PAC can provide better physical removal of organic pollutants, the addition of high PAC concentration may lead to the accumulation of carbon on the membrane surface, resulting in membrane fouling (Fabris et al.2007). Furthermore, previous researchers have suggested that the removal efficiency and membrane fouling are dependent on several parameters including the pre-adsorption, PAC dose, aeration rate, filtration flux and backwash duration (Guo et al.2005).

Ion-exchange

MIEX® contactor, an ion-exchange, has been used as pretreatment for a submerged membrane hybrid system and has produced a higher effluent quality and a longer operation time (Zhanget al.2006). Short-term experiments also showed that the submerged membrane process with MIEX® pre-treatment can achieve superior organic removal. The MIEX® process has mainly been used as a batch process, which requires large area for accommodating both contact tank and settling tank in the treatment process.

The objective of the study was to investigate a spiral flocculator (SF) as pre-flocculation process prior to a submerged hollow fibre microfiltration (SMF) system for membrane fouling reduction. Short-term experiments were also conducted with flocculation (SF) as pretreatment to PAC-SMF system. A further set of experiments was carried out to investigate the performance of a fluidized bed MIEX® contactor (a continuous flow system) as a pre-treatment to a submerged membrane system in removing refractory organic pollutants.

2. EXPERIMENTAL

2.1. Wastewater

The synthetic wastewater used in this study contained refractory organic compounds such as humic acid, tannic acid, lignin, polysaccharide and other high molecular weight carbohydrates. This wastewater used represents the biologically treated sewage effluent. The characteristics of the synthetic wastewater are shown in Table 1. The TOC concentration of the synthetic wastewater was 10.8–12.6 mg/L. The variation of the TOC value of synthetic wastewater was due to the fact that the synthetic wastewater use prepared using tap water at different occasions. The tap water TOC varied from experiment to experiment. The pH was 7.6–7.7. This wastewater composition was first recommended by Seo et al.(1996).

2.2. Flocculation and membrane reactor

A spiral flocculator (SF) used consists of two parts: (i) rapid mixing device made by winding a 3 mm inner diameter polyvinyl chloride (PVC) tube around a column of 11 cm in diameter; and (ii) slow mixing device made by winding a 6 mm inner diameter PVC tube around an 11 cm diameter column. The wastewater was pumped through the bottom of the rapid mixing device and was then delivered to the slow mixing device. A dosing pump was used to add 60 mg/L of FeCl3prior to the rapid mixing device. The rapid mixing time and slow mixing time were 1 minute and 15 minutes respectively. The flow rate of SF was determined by the filtration flux of the SMF, which was maintained at 60 $L/m^2.h$. The velocity gradient (G, the rate of change of velocity of propagation with distance normal to the direction of flow, which is related to the flow rate and tube diameter) of SF (6 mm diameters tube) was calculated using the formula $G=4.46(v/d)^{1.18}$ (Ngo et al.2000). Based on the filtration flux and the tube diameter, the G value was found to be 30 s^{-1}.

After that, pre-flocculated wastewater was delivered through the top of slow mixing device directly to the membrane tank by gravity. The schematic diagram of the SF-SMF system is shown in Figure 1. A hollow fibre membrane module was placed in the membrane tank and its characteristics are summarized inTable 2. The total volume of the membrane reactor was maintained at 6 L (HRT = 2 hr). An aeration rate of 14.4 $m^3/h\ m^2$ (membrane area) was provided using a soaker hose air diffuser. A pressure gauge was used to measure the transmembrane pressure (TMP). The experiments were conducted at room temperature of 25^{o} C.

2.3. Adsorption

In the SF-PAC-SMF experiments, a predetermined low dose of PAC (12 g in total) was added into the membrane tank. The PAC (80% min finer than 75mm) used was wood based carbon with a surface area of 882 m^2/g and a mean pore diameter of 30.61Å. The adsorption capacity of PAC was found to be 5.67 mg (TOC)/g (PAC) (with the wastewater used in this study).

2.3. Ion-Exchange

The effect of the pretreatment of fluidised bed MIEX® contactor on organic removal efficiency of the membrane system was studied with a MIEX® concentration of 100 mL. The removal of the organic matter was studied in terms of dissolved organic carbon (DOC). DOC was measured using a Multi N/C 2000 analyser. The schematic diagram of the submerged hollow fibre membrane system with fluidised bed MIEX® contactor as pre-treatment is shown in Figure 2.

3. RESULTS AND DISCUSSION

The short-term experiments were conducted to study the reduction in fouling as a result of flocculation as pretreatment to SMF and PAC-SMF hybrid system. In all cases, the membrane was operated at a very high filtration rate at 60 L/m^2h. During the operation, as soon as TMP reached to 50 kPa, a 2 minute-backwash with membrane filtrate was applied at a flow rate of 150 L/m^2h, (Guo et al. 2004). Figure 3presents the TMP development of the following three SMF systems: (i) SMF alone; (ii) SF-SMF and (iii) SF-PAC-SMF.

The TMP development of SMF alone was higher than that of pre-flocculation treatment. Within the operation period, the membrane was backwashed 7 times which indicated an irreversible fouling phenomenon had occurred. The TMP development in both pre-flocculation systems increased marginally during the 8 hours of high rate filtration (e.g. 2.5 kPa and 0.8 kPa TMP developed for SF-SMF and SF-PAC-SMF respectively). Thus, the flocculation is very helpful in reducing the membrane fouling, resulting in stable membrane productivity. Genkin et al. (2006) reported that the critical flux of a hollow fiber MF system increased from 17 to 46 L/m^{2}h with flocculant addition. Decarolis et al. (2001) also elucidated that the continuous in-line addition of ferric chloride prior to membrane filtration significantly reduced the rate of flux value decline for experimental flux values ranging from 54 to 88 L/m^{2}h. Furthermore, pre-flocculation coupled with SMF exhibited higher organic removal than SMF, as in addition to particle aggregation, ferric chloride could enhance organic foulant removal. The removal efficiency increased nearly 30% from 43.7±8% to 70.8±1.5%. In conjunction with PAC adsorption, the removal of organic pollutants increased up to 96.1±0.8%, which indicated that the SF-PAC-SMF could successfully remove the major refractory organics present in the wastewater. Hence, when flocculation was combined with adsorption as pretreatment to SMF, a significant reduction of the membrane fouling was observed.

Figure 4shows the total DOC removal efficiency of the submerged membrane hybrid system with fluidized MIEX® contactor as pre-treatment. The total DOC removal efficiency by the MIEX® reactor-membrane hybrid system can be as high as 75–85% with a pre-treatment of 100 ml (80.3 cm depth during fluidization) of MIEX® resin in the fluidized column. Submerged membranes alone could only remove an average of 35% of DOC from the wastewater. A detailed experiment conducted by Zhang et al.(2008)at different conditions of MIEX pre-treatment also showed the advantage of using MIEX fluidised bed as a pre-treatment. Although MIEX as a pre-treatment was efficient in removing DOC, the use of membranes as post-treatment is essential to remove the suspended solids and the carry-over resins.

Figure 5shows the TMP development during 8 hours of operation of the submerged membrane hybrid system with fluidized bed MIEX® as pre-treatment. With the MIEX® pre-treatment, the submerged membrane system resulted in a marginal increase of TMP. The TMP development was less than 10 kPa during the 8 hours of operation. Zhang et al. (2008) observed that a higher amount of MIEX® and a longer contact time resulted in a slightly higher TMP development of up to 19 kPa (in 8 hours of operation). This was attributed to the overflow of the small MIEX® particles from the fluidised bed reactor into the membrane tank and their accumulation in the submerged membrane tank. The escape of MIEX® resin into the membrane tank was visually observed during the 8 hour filtration operation.

4. CONCLUSIONS

The results indicated that the pre-flocculation of SF could reduce membrane fouling significantly even when system was operated at a high flow rate. The TMP of the SF-SMF system and the SF-PAC-SMF system developed marginally. The SF-PAC-MF system resulted in a high DOC removal of more than 96%. Therefore, the SF- PAC-SMF hybrid system can be a prospective solution for membrane fouling problem. The long-term experiments should be carried out to investigate how in-line flocculation as pretreatment can prevent the PAC-SMF system from fouling without affecting the biodegradation in the reactor. Moreover, when used as pre-treatment to submerged membranes, the fluidized bed MIEX® contactor could remove a significant amount of organic matter in the wastewater. This pre-treatment helped to reduce the membrane fouling and keep the TMP low during the membrane operation.

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Table 1. Constituents of the synthetic wastewater

Compounds	Weight (mg/L)
Beef Extract	1.8
Peptone	2.7
Humic acid	4.2
Tannic acid	4.2
(Sodium) lignin sulfonate	2.4
Sodium lauryle sulphate	0.94
Acacia gum powder	4.7
Arabic acid (polysaccharide)	5
$(NH_4)_2SO_4$	7.1
NH_2PO_4	7.0
KH_2HCO_3	19.8
$MgSO_4 \cdot 3H_2O$	0.71

Table 2. Characteristics of the hollow fiber membrane module used

Item	Characteristics
Material	Hydrophilic polyethylene
Nominal pore size	0.1 μm
Outer diameter	0.41 mm
Inner diameter	0.27 mm
No. of fiber	320 (16 * 20)
Length of fiber	12 cm
Surface area	0.05 m^2
Membrane packing density	9858 m^2/m^3
Membrane manufacturer	Mitsubishi-Rayon, Tokyo, Japan

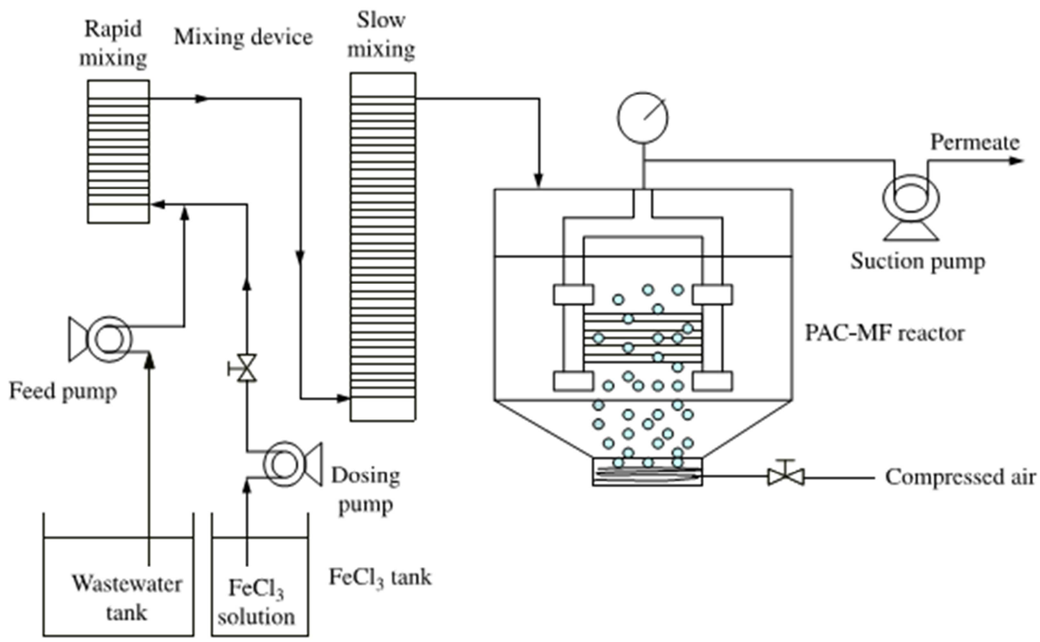

Figure 1. Experimental set-up of SMAHS

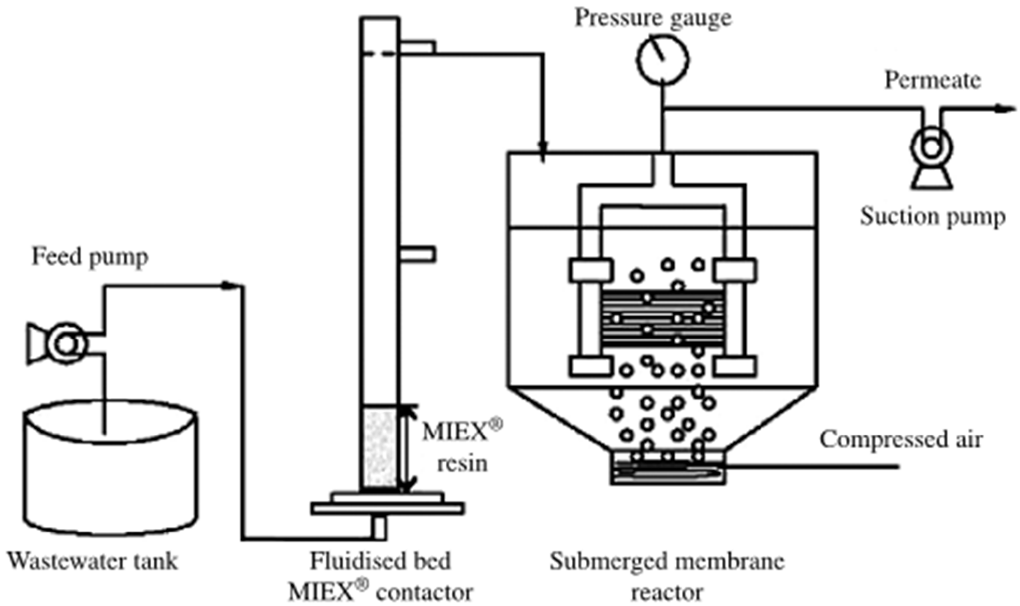

Figure 2. Schematic diagram of submerged membrane system with fluidized bed MIEX® contactor as pre-treatment.

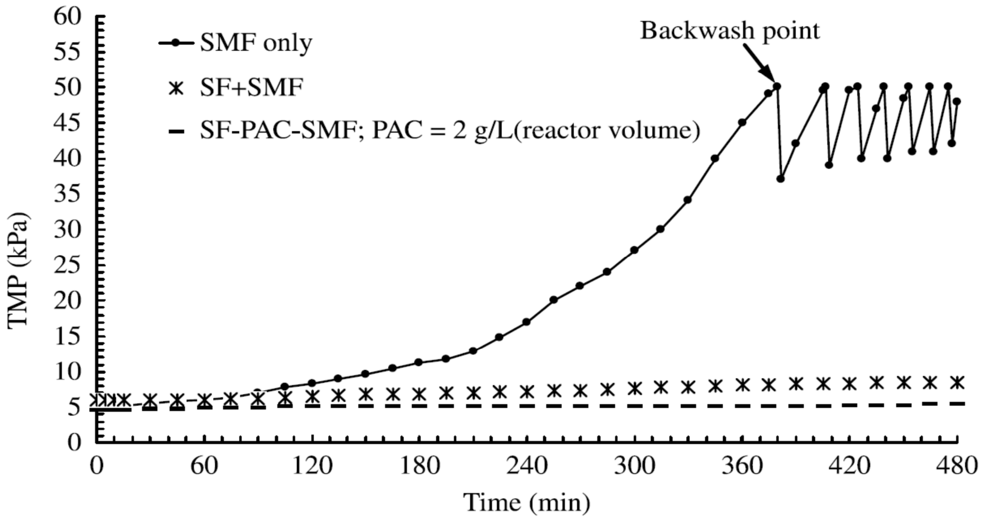

Figure 3. TMP profile of SMF alone, SF-SMF and SF-PAC-SMF systems (filtration Flux = 60 L/m^2h; PAC dose = 2 g/L; $FeCl_3$dose = 60 mg/L; flocculation velocity gradient =30 s^{-1}; aeration rate = 12 L/min)

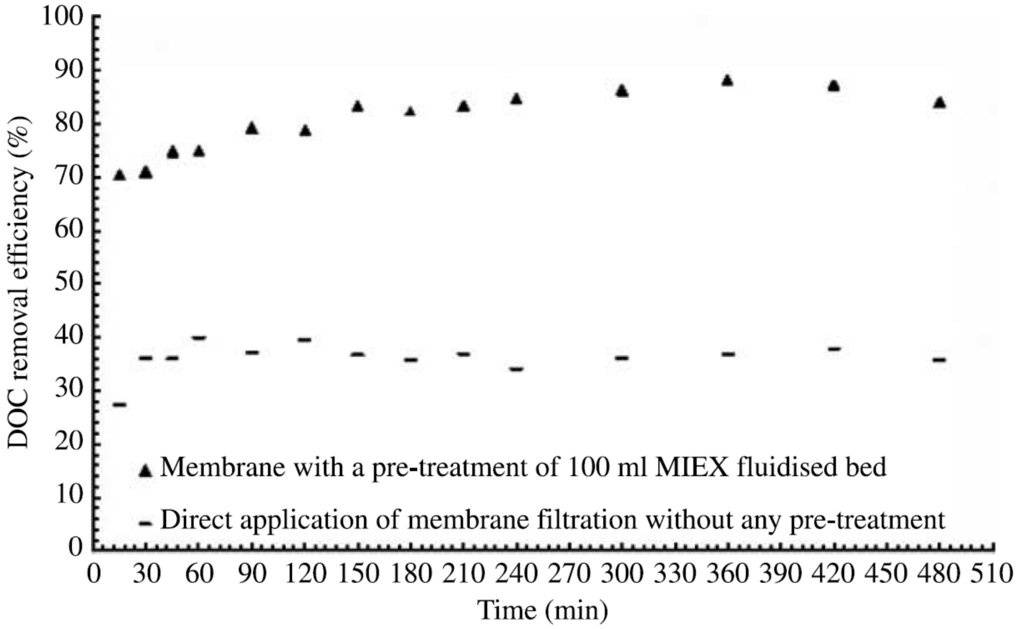

Figure 4. Total DOC removal efficiency of submerged membrane hybrid system with fluidized bed MIEX® contactor as pretreatment (fluidized velocity = 8.6 m/h; DOC of the wastewater = 10 mg/L; permeate flux = 96 L/m^2/h, backwash rate = 240 L/m^2/h, membrane backwash duration = 1 min, membrane backwash frequency = 1h).

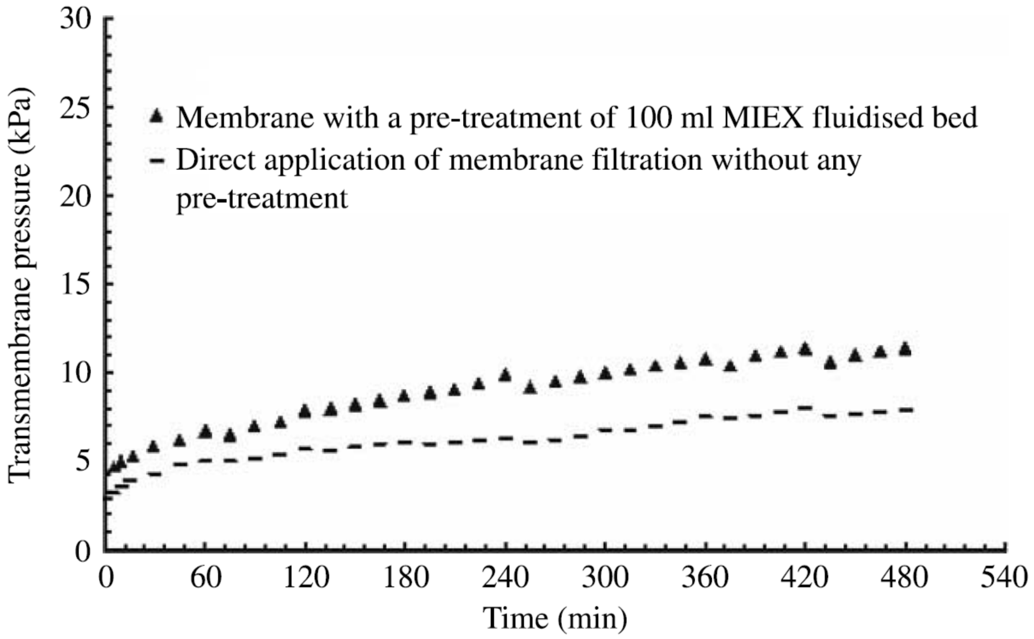

Figure 5. TMP profile of submerged membrane hybrid system with fluidized bed MIEX® contactor as pre-treatment (DOC of the wastewater = 10 mg/L; permeate flux = 96 $L/m^2/h$, backwash rate = 240 $L/m^2/h$, membrane backwash duration = 1 min, membrane backwash frequency = 1h).