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The evaluation of fouling effects in membrane process dealing with the biologically pre-treated textile effluents

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ABSTRACT

Wastewater reuse is necessary in the textile industry due to its consumption of large amounts of freshwater. However, the main problem with the membranes is the decline of permeate flux due to the accumulation of organic and inorganic molecules on the membrane surface when the raw wastewater is used. This study focused on the fouling effect of aerobically and anaerobically pre-treated textile industry effluents using ultrafiltration (UC010) and nanofiltration (NP010 and NP030) membrane processes. Ultrafiltration (UF) and nanofiltration (NF) membranes were applied sequestered (UF or NF) and combined (UF + NF) to treat effluents from a full-scale aerobic sequencing batch reactor (SBR), which is present in the factory, and a laboratory-scale anaerobic treatment plant, namely the static granular bed reactor (SGBR). Membrane experiments at 10 bar operating pressure were carried out on the laboratory scale to obtain better results. Initial fluxes with NP010 seriously declined after 24 h during the long-term experiments (96 h). According to the membrane flux decline and the fouling rates, aerobically pre-treated textile wastewater was better than anaerobically pre-treated textile wastewater, suggesting that dissolved organic matter formed in the anaerobic treatment processes.

Keywords: Reuse; Membrane; Fouling; Aerobic treatment; Anaerobic treatment; Textile wastewater

1. Introduction

The textile industry, which consumes large volumes of fresh water, generates wastewater that contains high colour, suspended solids, pH, temperature and COD values. Conventional treatment methods, which have been used for textile wastewater treatment, are chemical coagulation, electrochemical oxidation, filtration and biological treatment. However, many of these methods may not be used individually because they have disadvantages such as the presence of large amounts of sludge, which is generated by chemical coagulation processes, and the high cost of electrochemical oxidation processes. Textile industry wastewater is usually treated by using activated sludge treatment plants so that the water meets discharge standards [1]. However, the efficiency of colour removal from activated sludge is very low, and effluents cannot be reused [2].

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Advanced treatment methods such as membrane processes are very promising. Nowadays, a large number of studies have drawn attention to the treatment of industrial wastewater by using membrane systems, and technical and economical improvements have been made [2–9].

Ultrafiltration (UF) is an effective membrane process for the removal of particles and macromolecules, while nanofiltration (NF) allows for the separation of low-molecular weight organic compounds and divalent ions. Effective colour removal has been achieved using NF and reverse osmosis (RO) [3–5], while decolourisation of the wastewater has not been achieved using UF. Van der Bruggen et al. [6] suggested using NF membranes instead of UF membranes because of membrane fouling during filtration of textile wastewater.

The use of a combined process employing membranes has been suggested recently to overcome the disadvantages of conventional treatment plants and promote the reuse of wastewater. Bes-Pia et al. [7] showed that the combination of physicochemical treatment and NF provided high-quality treated and reusable water. The reuse of biologically pre-treated textile wastewater has been studied using membrane processes by Badani et al. [8] and Fersi et al. [5].

Because textile industry wastewater is usually treated aerobically in Turkey, further experiments are necessary to evaluate the performance of different membrane processes for biologically pre-treated textile wastewater. Anaerobic treatments of textile effluents are a good alternative to aerobic treatments because of their ability to remove colour and COD as well as methane production [10–15]. Although not yet widely used (food industry wastewater, municipal wastewater, leachate, poultry slaughterhouse wastewater) [16–20], the static granular bed reactor (SGBR) is an attractive anaerobic treatment option because it removes COD with high efficiency. SGBR works like an anaerobic biofilter since it involves no mixing systems and has stable granule media. Because SGBR is one of the newly developed anaerobic reactors, this is the first study to investigate the effectiveness of the system during the treatment of textile effluents. In this study, effluents from both a full-scale aerobic treatment plant named the sequencing batch reactor (SBR) and an anaerobic laboratory-scale SGBR were compared for their effects to treatment with different UF and NF membranes.

To perform this study, the NF membranes used for the treatment effluents (i.e., the full-scale aerobic treatment plant and the laboratory-scale anaerobic reactor) were evaluated for total flux decline, fouling and concentration polarisation after long-term experiments, which was carried out for 96 h.

2. Materials and methods

2.1. Wastewater origin

In this study, effluents from an aerobic treatment plant and a laboratory-scale anaerobic treatment plant were used. Wastewater was produced from the process in a factory that was used to created dyes for 95% cotton and 5% synthetic fibres. The domestic wastewater content of the textile wastewater flow was 1%. Sodium chloride and urea were used as fixing materials during the dyeing process.

Domestic and processed wastewater originating from the factory was treated using a nitrifying aerobic SBR. The SBR system had six parallel reactors with a cycle of 14 h.

Colour content of the wastewater was not removed, whereas a 72% COD removal rate was obtained from the SBR system installed in the factory. The characteristics of the wastewater generated from the factory and the effluent of the aerobic treatment process are given in Table 1.

Besides aerobic treatment in the factory, the raw textile wastewater was also treated by using a laboratory-scale anaerobic reactor, namely the SGBR [21], which was developed in 2000 and has been used in several laboratory and pilot plant studies [16–20] (Fig. 1). The laboratory-scale SGBR was 15 cm in diameter and 50 cm in length and had an effective volume of 3 l. Granulated anaerobic sludge was used as the seed and filling

Table 1
Characteristics of the raw (untreated), aerobically and anaerobically pre-treated wastewater

Wastewater	pH	COD, mg/l	SS, mg/l	NH_3-N, mg/l	TP, mg/l	Sulfate, mg/l
Raw (untreated) wastewater	12±1	1660±300	130±50	10±2	1.55±1	150±50
Aerobically pre-treated wastewater	7.1±0.3	450±200	70±10	47.6±5	0.41±0.1	370±20
Anaerobically pre-treated wastewater	7.2±0.5	440±10	130±50	13.2±3	0.5±0.3	150±100

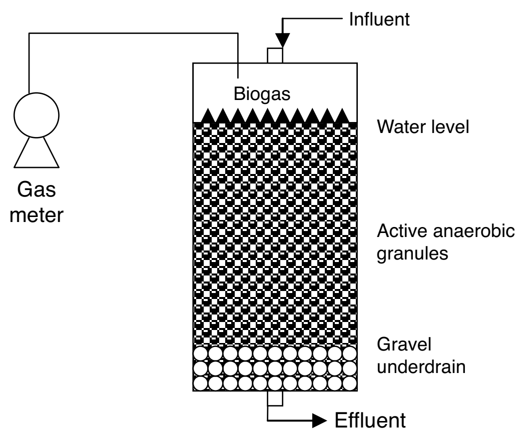

Fig. 1. Schematic diagram of the SGBR process [15].

material in the anaerobic reactor. The acclimation period was set at five weeks with a synthetic solution (2000 mg O_2 l^{-1}) containing milk powder in the SGBR. The real textile wastewater was fed into the system after reaching a 90% COD removal rate.

The SGBR was operated at room temperature (18–20°C) and a hydraulic retention time of 24 h with an organic loading rate of 1.7 kg COD m^{-3} d. COD and colour removal efficiencies were found to be 74% and 57%, respectively. The total gas production of the anaerobic reactor was about 1327 ml d^{-1}, and the methane gas content of the released biogas was approximately 68%. The characteristic of the effluents to the anaerobic treatment process is given in Table 1.

2.2. Equipment

The membrane system was supplied by Osmonics® Inc. and consisted of a GE Sepa™ CF2 membrane cell [22]. The concentrated stream flowed back to feed the vessel, while the permeate stream was collected separately, as shown in Fig. 2. A cartridge filter (10-µm pore size) was used as a pre-filter to remove coarse particulates from the wastewater prior to entering the membrane cell. All membrane experiments were performed at 23–25°C with a heat exchanger that was located in the feed vessel. Biologically pre-treated textile wastewaters were filtered using UF and NF processes at operating conditions of 3 and 10 bar, respectively.

NF and UF membranes were supplied from Macrodyn® Nadir as flat sheets. Each experiment was performed with a new membrane sheet. The characteristics of the membranes are given in Table 2.

2.3. Analytical methods

The wastewater characteristics were determined by analysing the pH, chemical oxygen demand (COD), suspended solids (SSs), ammonia nitrogen (NH_3-N), total phosphorus (TP) and sulphate parameters according to Standard Methods [23]. The permeate flux was determined gravimetrically.

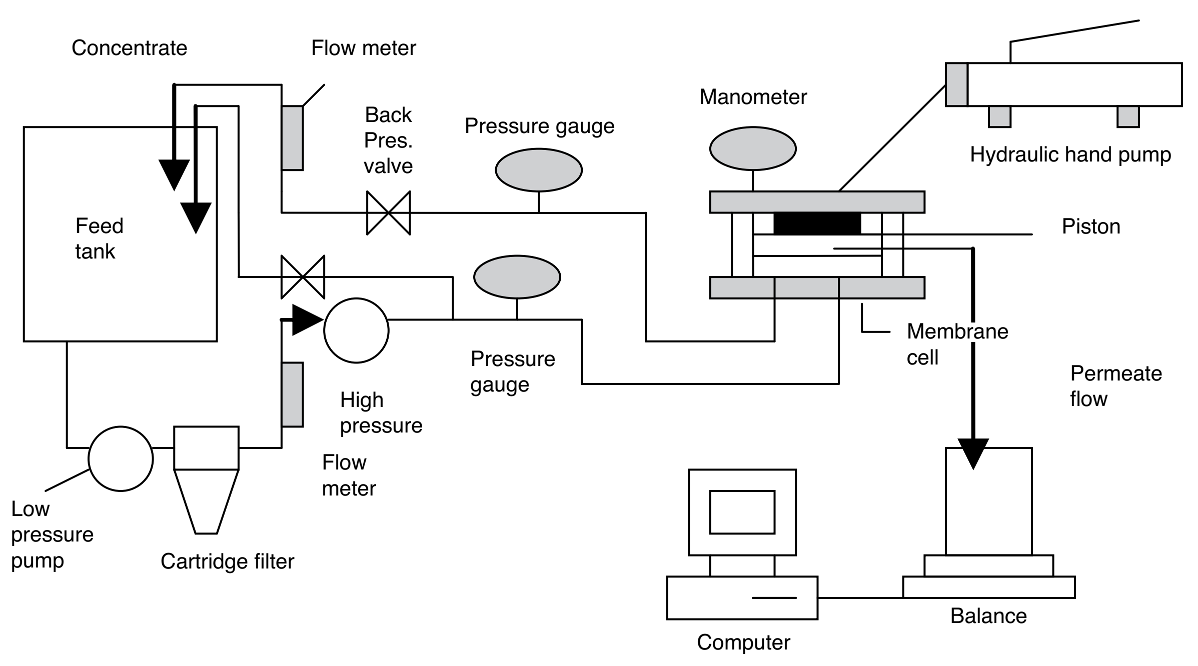

Fig. 2. Schematic diagram of the membrane process [15].

Table 2
Characteristics of the NF and UF membranes used in experiments

Membrane type	Manufacturer	Material	Membrane property	[a]MWCO, kDa	[b]M.O.P., bar	[c]M.O.T, °C
NP010	Macrodyn® Nadir	Polyethersulfone	Hydrophilic	[d]NA	40	95
NP030	Macrodyn® Nadir	Permanently Polyethersulfone	Hydrophilic	[d]NA	40	95
UC010	Macrodyn® Nadir	Cellulose	[d]NA	10	3	55

[a]Molecular weight cut-off.
[b]Maximum operation pressure.
[c]Maximum operation temperature.
[d]Not available.

3. Results and discussion

In this study, treatment of aerobically and anaerobically pre-treated wastewater was examined using NF and UF membranes in a laboratory-scale unit. The performance of the membranes was determined by measuring the permeate flux at 10 bar membrane pressures and long-term experiments.

Total flux decline, fouling and concentration polarisation were evaluated by using long-term experiments with NP010 and NP030 membranes in a laboratory-scale anaerobic reactor and effluents of a full-scale aerobic treatment plant. Experiments were performed at constant flow (300 l h^{-1}), pressure (10 bar) and temperature (23–25°C). NP010 and NP030 membranes were used in applications of effluents collected after UC010 experiments.

Flux measurements were carried out in each 24 h period during the long-term experiments. Fluxes obtained within 96 h were compared during the long-term experiment in Fig. 3.

High fluxes were obtained for both types of wastewaters at the beginning of the UC010+NP010 experiments. Initial fluxes for aerobically and anaerobically pre-treated wastewaters were obtained as 138.9 l m^{-2} h^{-1}, 133.7 l m^{-2} h^{-1}, and 62.4 l m^{-2} h^{-1} and 58.5 l m^{-2} h^{-1} with UC010+NP010 and UC010+NP030, respectively. According to the obtained fluxes, aerobically pre-treated wastewater had higher fluxes in both applications as compared to anaerobic wastewater. However, the obtained initial fluxes with NP010 seriously declined after 24 h. Permeate fluxes for aerobically and anaerobically pre-treated wastewaters decreased to 39.3 l m^{-2} h^{-1} and 33.4 l m^{-2} h^{-1}, respectively.

The decline of the permeate flux was also observed with NP030 experiments within 24 h, but this decline was smaller than in the NP010 experiments. Flux decline was observed at the end of 48 h in all applications. However, the reduction obtained in NP010 experiments was significantly higher than in NP030 experiments. This situation can be explained by the possibility of a higher accumulation in pores of NP010 membranes than in pores of NP030 membranes. No other sharp flux decline was detected in membrane experiments by 72–96 h. According to flux conditions, at 96 h, higher fluxes were obtained with NP030 membranes. The anaerobically pre-treated wastewater fluxes derived from NP010 and NP030 were 20.3 l m^{-2} h^{-1} and 44.4 l m^{-2} h^{-1}, respectively, while the aerobically pre-treated wastewater fluxes derived from NP010 and NP030 were 25.2 l m^{-2} h^{-1} and 52.7 l m^{-2} h^{-1}, respectively.

To determine the extent of membrane fouling using distilled water, the membranes were tested after 96 h, before and after washing. Membrane washing processes were carried out by holding the membranes at low pH with HNO_3 and in high pH with NaOH solutions for 1 h. Equations used to determine the total flux decline, the concentration polarisation and the fouling are summarized in Table 3 [24]. J_{cwi}, J_{cwf}, J_{cwc} and J_{ww}, given in Table 3, are clean water flux of clean membrane, clean water flux of fouled membrane, clean water flux of cleaned membrane and wastewater flux, respectively.

The results of long-term experiments are given in Figs. 4 and 5 for aerobically and anaerobically pre-treated wastewaters, respectively. According to Figs. 4

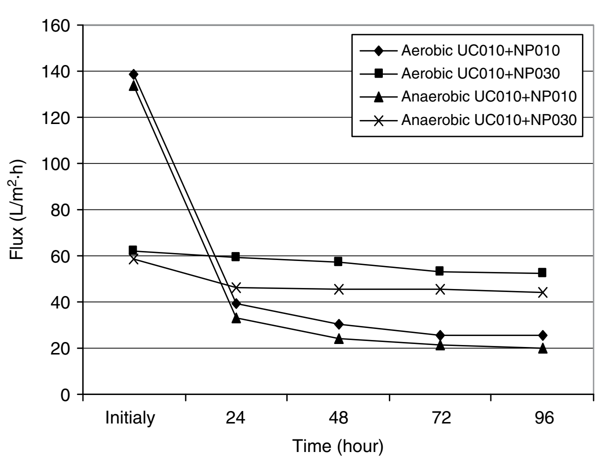

Fig. 3. NP010 and NP030 experiments at 96 h compared to the flux decrease.

Table 3
Equations used to determine the membrane performance in long-term membrane experiments [24]

Permeate flux (l/m^2.h)				Flux decline (%)				
Pure Water			Wastewater	Total	Con. Pol.	Fouling		
Initial	Final	Cleaned				Total	Reversible	Irreversible
J_{cwi}	J_{cwf}	J_{cwc}	J_{ww}	$(J_{cwi} - J_{ww})/J_{cwi}$	$(J_{cwf} - J_{ww})/J_{cwf}$	$(J_{cwi} - J_{cwf})/J_{cwi}$	$(J_{cwc} - J_{cwf})/J_{cwc}$	$(J_{cwi} - J_{cwc})/J_{cwi}$

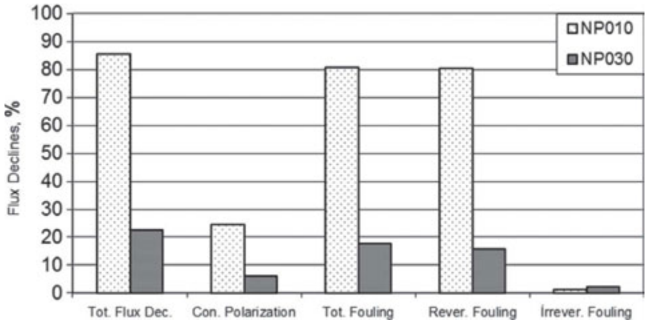

Fig. 4. Comparison of obtained flux declines for aerobically pre-treated wastewater with NP010 and NP030 membranes during long term experiments.

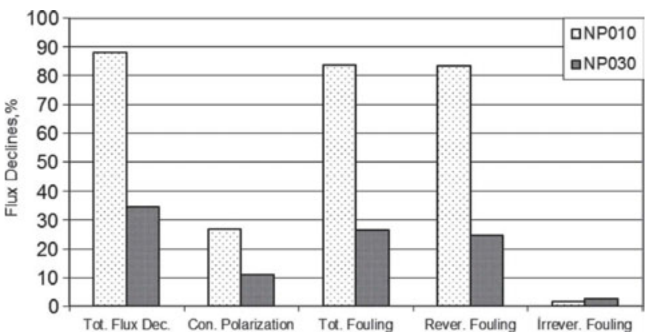

Fig. 5. Comparison of obtained flux declines for anaerobically pre-treated wastewater with NP010 and NP030 membranes during long term experiments.

and 5, higher permeate fluxes were obtained with aerobically pre-treated wastewater than with anaerobically pre-treated wastewater because of the dissolved organic matter produced by the anaerobic reactor.

According to the flux results, the high flux decline for both wastewaters was observed for the NP010 membrane. A flux decline was obtained with NP010 at a rate of 85.4% and 88.2% for aerobically and anaerobically pre-treated effluents, respectively. On the other hand, flux decline obtained with NP030 for aerobically and anaerobically pre-treated wastewaters were determined for 22.7% and 34.9%, respectively. As the flux value is one of the most important parameters for the operation and design of membrane systems, the high flux decrease occurring for the NP010 membrane is problematic.

Increased concentrations occurred on the membrane surface, and the movement of dissolved substances through the membrane pores is called concentration polarisation. The concentration polarisation values with NP010 membranes for aerobically and anaerobically pre-treated wastewaters were determined to be 24.6% and 27.0%, respectively. The concentration polarisation for the NP030 membrane was lower than the NP010 membrane and was calculated for aerobically and anaerobically pre-treated wastewaters to be 6.2% and 11.0%, respectively. The flux decline due to membrane fouling for NP010 membranes was higher than for NP030 membrane. The total flux decline due to fouling was 80.6% and 83.9% with NP010 membranes for aerobically and anaerobically pre-treated wastewaters, respectively, whereas they were 17.6% and 26.8% with NP030, respectively. Reversible fouling obtained after membrane washing for NP010 and NP030 were determined to be 80.3% and 83.6% for aerobically pre-treated wastewater and 15.9% and 24.7% for anaerobically pre-treated wastewaters, respectively. Irreversible fouling for NP010 and NP030 were calculated at 1.4% and 1.8% for aerobically, and 2.1% and 2.8% for anaerobically pre-treated wastewaters, respectively.

The results of the membrane flux and fouling rates with aerobically and anaerobically pre-treated textile wastewaters by long-term experiments showed that aerobic treatment was slightly better. This situation can be explained by the presence of dissolved organic matter in anaerobically pre-treated wastewater. SGBR effluents consisting of dissolved organic matter, humic matters, polysaccharides, amino acids, proteins, fatty acids, phenols, carboxylic acids, quinine, lignin, carbohydrates, alcohol and resins must be considered [25]. The escape of fragmented anaerobic granules also occurs in the SGBR. Moreover, it is known that dyes are converted to aromatic amines in anaerobic treatment systems [12,26–28]. Aromatic amines that are produced in anaerobic treatment processes have smaller dimensions than dyestuffs. Therefore, it is easier to retain particles on NF membranes that are not decomposed in aerobic treatment. To further validate this statement, dissolved organic matter analysis should be made on effluent samples and on the organic matter at the membrane surface.

4. Conclusions

Textile industry wastewater is usually treated by using aerobic treatment processes in Turkey. However, colour removal efficiencies are the lowest with this method, and the resulting effluents are not appropriate for reuse facilities. Further treatment is necessary to reuse the effluents. In the study, UF and NF membranes were used and tested to determine their condition during long-term experiments, which was performed for 96 h, with aerobically and anaerobically pre-treated textile wastewater. The fluxes obtained with NP010 were higher than with NP030 in all experiments.

Initial fluxes with NP010 seriously declined after 24 h during the long-term experiments. According to the membrane flux decline and fouling rates, aerobically pre-treated textile wastewater had better results than anaerobically pre-treated textile wastewater, which suggests that dissolved organic matter formed during the anaerobic treatment process.

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