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Application of NF-RDM (nanofiltration rotating disk membrane) module under extreme hydraulic conditions for the treatment of dairy wastewater

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ABSTRACT

Dairy wastewater can be concentrated by membrane technology to produce reusable water, and the retentate with high concentration could be reutilized as feed supplement for animals or substrate for biohydrogen production. Treatment of a model dairy wastewater (diluted skim milk) by nanofiltration (NF) with a rotating disk membrane (RDM) module was investigated using a NF membrane NF270 (Dow-Filmtec). Compared with the Desal-5 DK (Ge-Osmonics) membrane studied previously, the NF270 membrane gave much higher permeate flux, but same rejection. At a high enough shear rate for reducing concentration polarization, permeate flux and solutes rejection increased while specific energy per m^3 of permeate decreased when transmembrane pressure (TMP) rose from 10 to 40 bar. Because the RDM module could produce very high shear rates, concentration of dairy wastewater by NF could be operated under extreme hydraulic conditions that combined extreme TMP with high shear rate. Process efficiency and permeate quality were improved by operating under these condition, while membrane fouling was a little higher than when operating below critical flux. A real dairy wastewater was also treated by NF270 under same hydraulic conditions, and results were very similar to those of model solution. Membrane fouling was decreased by raising pH above 9. Irreversible fouling was almost removed using a commercial cleaning agent at high shear rate in a short time. The RDM module appeared to be a suitable technology to treat dairy waste without any biological and chemical treatments. Permeate qualities were suitable for reuse or discharge in river. These results from laboratory-scale tests can be very useful in the analysis of membrane process under extreme hydraulic conditions and can serve as valuable guide for process design in industrial application.

1. Introduction

The dairy industry, like most other food industries, generates a large amount of wastewater, essentially composed of diluted milk, which are responsible for a 1–3% loss in milk components (lactose and proteins) [1]. This effluent results in both water eutrophication and nutrients loss when it is discarded without treatment. Therefore, a number of technologies were used to treat dairy wastewater, such as coagulation [2,3], ecological treatment system [4], anaerobic or/and aerobic reactors [5], membrane separation [6]. With the lack of water resources and the promotion of cyclic economy in recent years, water reuse and organic reutilization have become the goals of wastewater treatment. However, effluents treated by coagulation and biological treatments cannot satisfy the requirements for reuse. Membrane technology, especially nanofiltration (NF) and reverse osmosis (RO), provides a promising method to produce reusable water from dairy wastewater [6–8], and the concentrated retentate by membranes can be precipitated by coagulation to obtain feed supplement for animals [9], or can be treated by anaerobic digestion to collect renewable energy sources (H_2 and CH_4) [10], which is regarded as an economical and environment-friendly process for treatment of dairy wastewater.

Several investigations have shown that NF and RO were adequate for concentration of dairy effluents and production of reusable water. Koyuncu et al. [11] found that NF could reject more than 98% of COD (Chemical Oxygen Demand) and conductivity of dairy effluents, and a two-pass RO system could produce water of very high quality. Balannec et al. [6] compared the performance of nine NF and RO membranes for concentration of 1:2 diluted skim milk (COD ≈ 36,000 mg O_2 L^{-1}), to show that high COD rejection (>99%) was achieved with all membranes and feed concentration tested. Using a spiral-wound crossflow module, Turan [12] found

Abbreviations: COD, chemical oxygen demand (mg O_2 L^{-1}); RDM, rotating disk membrane; TMP, transmembrane pressure (bar); VRR, volume reduction rate.

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Nomenclature

List of symbols

- E_c specific energy (kWh m^{-3})
- IF irreversible fouling (%)
- k velocity factor (0.89 for this system)
- L_{pi}, L_{pf} pure water permeability before and after the experiment (L m^{-2} h^{-1} bar^{-1})
- p_c peripheral pressure (bar)
- P_t total power of rotating disk motor and feed pump (kW)
- Q_f permeate flow rate (m^3 h^{-1})
- r radial coordinate (m)
- R module housing inner diameter (m)
- V_o volumes of original feed (m^3)
- V_R volumes of retentate (m^3)

Greek letters

- γ (γ_{max}) membrane (maximum) shear rate at periphery (s^{-1})
- ρ fluid density (g L^{-1})
- μ fluid dynamic viscosity (mPa s)
- ν fluid kinematic viscosity (m^2 s^{-1})
- ω angular velocity (rad s^{-1})

that increase of feed COD concentration and decrease of crossflow velocity could cause flux decline mainly due to concentration polarization, which also induced higher COD concentration in permeate. Moreover, with increase of organic content in feed during a concentration process, a single NF operation could not guarantee the quality of permeate water anymore, and a single RO or NF + RO filtration should be required [13]. Anyway, because of pressure drop and concentration polarization, crossflow membrane modules could not reach a high efficiency level, especially for a concentrated effluent. For example, the transmembrane pressure (TMP) was limited to 20 bar in all articles about crossflow NF treatment of dairy wastewater, while the limit value is normally 40 bar. Thus, operating at the low TMP, permeate flux was low and water recovery and retentate concentration were not high enough. Furthermore, Koyuncu et al. [11] reported that energy consumption per m^3 of permeate decreased with increasing TMP for NF and RO of dairy wastewater.

Our laboratory uses a rotating disk membrane (RDM) shear-enhanced filtration system which can be operated at a TMP of 40 bar and a very high shear rate (4.43×10^5 s^{-1}) [14]. Since this system reduces concentration polarization effectively, permeate flux and solutes rejection keep increasing with TMP up to 40 bar [15]. It not only permits to concentrate wastewater to a very high concentration, but also provides a very valuable tool for process investigation under extreme hydraulic conditions. Akoum et al. [16] and Frappart et al. [17] compared crossflow filtration, vibrating and RDM filtration systems for concentration of model wastewater (1:2 diluted skim milk), and showed that the RDM filtration system was the best with regards to permeate flux and selectivity. Operating under extreme hydraulic conditions with a RDM module (TMP = 40 bar, rotating speed = 2000 rpm, Desal-5 DK), Frappart et al. [14] obtained a highly concentrated retantate (=38% dry mass) and a permeate with low COD concentration (below authorized limit). Here, the extreme hydraulic conditions were defined as using a 40 bar TMP and high shear rate above 10^5 s^{-1}. However, even if the RDM module generates very high shear rate, concentration polarization cannot be eliminated completely at very high TMP. Thus, it is not certain if extreme hydraulic conditions should be applied during the whole concentration process, in order to optimize permeate water quality, process efficiency, energy consumption and membrane fouling.

Table 1
Properties of NF membranes examined [22].

Membrane	NF270	Desal-5 DK
Manufacturer	Dow-Filmtec	Ge-Osmonics
Membrane material	Polyamide	Proprietary polyamide
Molecular weight cut-off	150–200	150–300
L_p[a] (L m^{-2} h^{-1} bar^{-1}) 25 °C	12.0 ± 0.4	4.2 ± 0.3
Max. temperature (°C)	45	50
Max. pressure (bar)	41	40
Isoelectric point (pH)	5.3	4.0–4.1

[a] L_p: pure water permeability, L_p of NF270 was own measurement and L_p of Desal-5 DK was obtained in Ref. [14].

In this study, concentration of dairy wastewater by NF using a RDM filtration system was investigated. In order to promote process efficiency, a new NF membrane was chosen based on flux and selectivity. The focus of this work was to discuss the effect of shear rate and TMP on NF flux and energy consumption, in order to identify whether extreme hydraulic conditions are adequate for treating concentrated feed by NF. Besides, a real effluent was for the first time treated by shear-enhanced filtration system and its result was compared with that of model effluent. The present work should be very useful for understanding membrane processes under extreme hydraulic conditions and to develop shear-enhanced filtration for treatment of dairy wastewater by NF.

2. Materials and methods

2.1. Experimental set-up and membranes

The RDM module, shown in Fig. 1, has been designed and built in our laboratory [18,19]. A flat membrane, with an effective area of 176 cm^2 (outer radius R_1 = 7.72 cm, inner radius R_2 = 1.88 cm), was fixed on the cover of the cylindrical housing in front of the disk. The disk equipped with 6-mm high vanes can rotate at adjustable speeds, ranging from 500 to 2500 rpm, inducing very high shear rate on the membrane. As described previously in the literature [14], the module was fed from a thermostatic and stirred tank containing 12 L of fluid by a volumetric diaphragm pump (Hydra-cell, Wanner, USA). The peripheral pressure (p_c) was adjusted by a valve on outlet tubing and monitored at the top of the cylindrical housing by a pressure sensor (DP 15-40, Validyne, USA), and the data were collected automatically by a computer. The permeate was collected in a beaker placed on an electronic scale (B3100 P, Sartorius, Germany) connected to a computer in order to measure the permeate flux.

The shear-enhanced filtration module should be equipped with a very high flux membrane in order to fully exploit its potential. According to previous studies [20,21], NF270 (Dow-Filmtec) had outstanding advantages in flux and selectivity for treatment of concentrated effluents, so it was chosen to concentrate model wastewater (1:2 diluted skim milk) and compare results with preceding data using Desal-5 DK (Ge-Osmonics) under extreme hydraulic conditions (TMP = 40 bar, rotating speed = 2000 rpm) [14]. Based on the manufactures' data sheet and literature [22], the properties of the two NF membranes are shown in Table 1.

2.2. Test fluid

To keep consistency with a previous study and compare results under same feed compositions, a model effluent was prepared from commercial UHT skim milk (Lait de Montagne, Carrefour, France), diluted 1:2 to one-third of normal concentration with deionized water (Aquadem E300, Veolia Water, France). Real effluents were

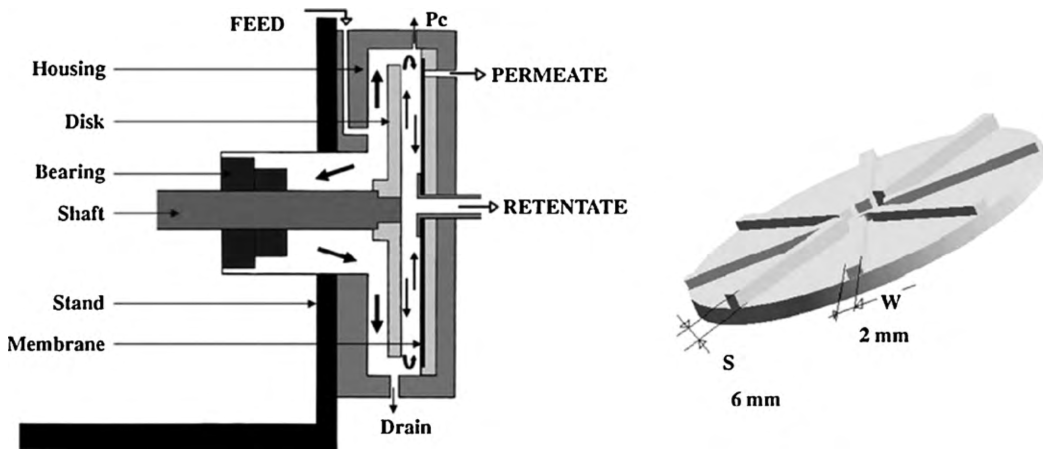

Fig. 1. Schematic of the rotating disk module (RDM) and of the disk with vanes.

collected from a local dairy factory (Lactalis, Clermont, France) in two different batches. The 1st batch contained effluent collected quickly at inlet of treatment tank while the 2nd batch was constituted from 1 L samples collected every hour for 24 h at same inlet in order to average its composition. The main characteristics of model and real effluents are described in Table 2.

2.3. Experimental procedure

A new membrane was used for each series of experiments to ensure the same initial membrane conditions for the entire study. The membranes were soaked in deionized water for at least 48 h prior to use, and pre-pressured with deionized water for 30 min under a pressure of 40 bar at 25 °C. After stabilization, the pure water flux of membranes was measured at five pressures of 20, 16, 12, 8, 4 bar to calculate water permeability (L_p). Before the experiments started, the feed was heated to 35 °C, and then a pre-filtration was conducted for 10 min with full recycling at a feed flow rate of 180 L h^{-1} in all tests as in a previous study [14]. Because using NF270 membrane at high temperature was not recommended [23], all tests were performed at 35 °C. For concentration experiments, a 12 L feed was concentrated to 1.5 L at constant conditions except stated elsewhere. For each series of experiments, with the same membrane and test fluid, feed concentration and TMP were increased in steps, but rotating speed was decreased in steps. This procedure minimized membrane fouling during the tests. Each sample was collected for analyses when permeate flux became stable after changing hydraulic conditions.

After each series of tests, the filtration system was flushed with deionized water until the rinsing water came out clear, and L_p was measured to know the degree of irreversible fouling. Subsequently, a cleaning agent solution (P3 Ultrasil 10, Ecolab, USA) at 0.1% concentration was used to clean the membranes at room temperature (20–25 °C) for 30 min. Then the system was rinsed by deionized water again until the pH of the rinse water was neutral. Finally, the membranes were soaked in deionized water for 30 min to eliminate the effect of swelling by cleaning and then L_p was measured again.

Table 2
Main characteristics of dairy effluents.

Index	Model solution	Real effluents	
		Batch 1	Batch 2
Turbidity (NTU)	NA	101	100
COD (mg O_2 L^{-1})	36,000 ± 2000	297 ± 13	580 ± 16
Conductivity (μs cm^{-1})	2170	1084	1516
pH	6.84	8.72	9.56
Dry mass (g L^{-1})	31.9 ± 0.2	0.8 ± 0.1	1.4 ± 0.1

NA: not available.

2.4. Analytical methods

Turbidities of retentate and permeate were measured with a Ratio Turbidimeter (Hach, USA). COD was measured using Nanocolor Kits (Machery-Nagel, Hoerdl, France) in order to quantify organic matter concentration. The total relative error on COD was estimated to be 8%. Conductivity was measured with Multi-Range Conductivity Meter (HI 9033, Hanna, Italy) and pH was measured with pH Meter (MP 125, Mettler Toledo, Switzerland). Dry mass was determined by measuring the weight loss after drying samples at 105 ± 2 °C for 5 h in an oven. The viscosity was measured by a Falling Ball Viscometer (GV-2100, Gilmont, USA). Powers of rotating disk motor and feed pump under different conditions were measured with Power & Energy Monitors (Metric MX240, Chauvin Arnoux, France).

2.5. Fluid mechanics consideration

Volume reduction rate (VRR) is defined as:

$$\mathrm{VRR} = \frac{V_o}{V_R} \tag{1}$$

where V_o is initial feed volume and V_R is the retentate volume.

According to a previous study [14], the local membrane shear rate (γ) can be calculated by the following equation:

$$\gamma = 0.0296 r^{8/5} (k\omega)^{9/5} \nu^{-4/5} \tag{2}$$

where r is the distance from center and k is the velocity factor (0.89 for this system), ω is the disk angular velocity and ν the fluid kinematic viscosity. The maximum membrane shear rate γ_{max} occurs at the disk rim and is calculated by setting $r = R_1 = 7.72$ cm in Eq. (2).

The mean transmembrane pressure (TMP) is obtained by integrating the local pressure (p_c) over the membrane area as follows [14]:

$$\mathrm{TMP} = p_c - \frac{1}{4} \rho k^2 \omega^2 R^2 \tag{3}$$

where ρ is the density of the fluid at 35 °C and R is the housing inner diameter.

Energy consumption per m^3 of permeate (specific energy, E_c) is represented as:

$$E_c = \frac{P_t}{Q_f} \tag{4}$$

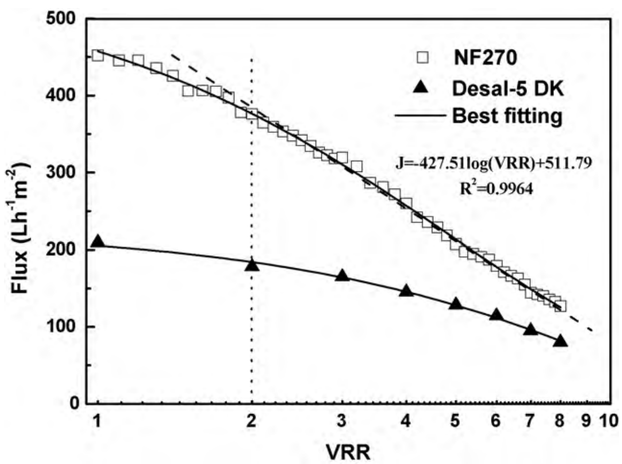

Fig. 2. Permeate flux as a function of volume reduction ratio in semi-log coordinates for different membranes. *Feed*: 1:2 diluted skim milk; TMP = 40 bar; rotating speed = 2000 rpm; T = 35 °C; flux data of Desal-5 DK was obtained from Ref. [14].

where P_t is the sum of power of rotating disk motor and feed pump, and Q_f is the permeate flow rate.

The irreversible fouling index (IF) can be expressed as a percentage of pure water permeability decrease after the experiment:

$$IF \quad (\%) = \frac{L_{pi} - L_{pf}}{L_{pi}} \times 100 \qquad (5)$$

where L_{pi} and L_{pf} are the initial and final pure water permeability, respectively.

3. Results and discussion

3.1. Comparison of membrane performance

As seen in Fig. 2, the NF270 membrane shows a much higher permeate flux than Desal-5 DK, especially at the beginning of concentration. At the same time, COD and conductivity measured in accumulated permeates at each value of VRR for NF270, displayed in Fig. 3, were almost the same as that of Desal-5 DK. This meant that these two NF membranes had similar rejection for solutes but the NF270 had a much higher flux, due to its higher porosity. This is confirmed by permeability data in Table 1, as the pure water permeability flux of NF270 was three times that of Desal-5 DK while their molecular weight cut-off (MWCO) was very close. The mean

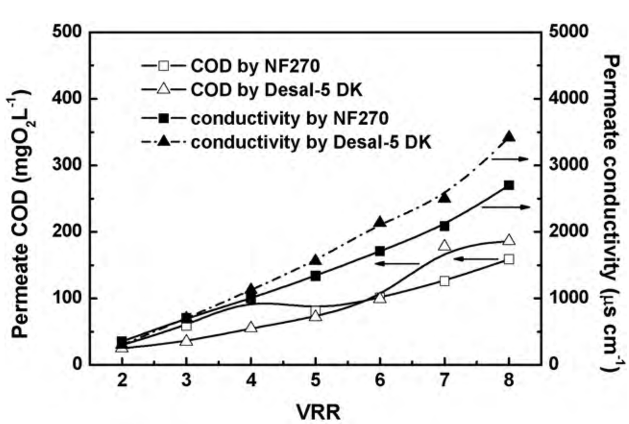

Fig. 3. Mean permeate COD and conductivity in permeate accumulated at each value of VRR as function of volume reduction ratio for different membranes. *Feed*: 1:2 diluted skim milk; TMP = 40 bar; rotating speed = 2000 rpm; T = 35 °C; permeate data of Desal-5 DK was obtained from Ref. [14].

Table 3
Mean COD and conductivity in accumulated permeate at the end of filtration for different membranes.

Index	Mean COD ($mg\ O_2\ L^{-1}$)	Mean conductivity ($\mu s\ cm^{-1}$)
NF270	54 ± 5	685 ± 83
Desal-5 DK	42	974
Authorized limit [14]	125	1000

Feed: 1:2 diluted skim milk; TMP = 40 bar; rotating speed = 2000 rpm; T = 35 °C; permeate data of Desal-5 DK was obtained from Ref. [14].

COD and conductivity in accumulated permeate at a VRR of 8 are shown in Table 3. It can be observed that, although solutes concentration in model solution was much higher than that in real effluent (see Table 2), values of COD and conductivity in permeate are well below the allowed limit for both membranes. Therefore, the NF270 is a better choice for concentrating dairy wastewater due to its higher flux.

From Fig. 2, it can be found that the flux of NF270 falls on a straight line with VRR in semi-log coordinates when VRR is more than 2, which presumably corresponds to the mass transfer limited regime [14]. In this regime, membrane permeability is less important and the flux is governed by membrane shear rate. This is the reason why the flux of NF270 becomes closer to that of Desal-5 DK as VRR increases. Thus, it is necessary to investigate if extreme hydraulic conditions (high TMP and high shear rate) can retain their advantages with increasing solutes concentration and viscosity of feed during concentration by NF.

3.2. Effect of shear rate and TMP

Higher shears rate can reduce solutes accumulation at membrane surface and thus decrease osmotic pressure differences across the membrane, leading to higher permeate flux at constant TMP [24]. However, an excessive shear rate will waste much energy and increase abrasion of equipments. Similarly, if higher TMP increase the driving force for water permeating through membrane, at the same time, they may increase concentration polarization. The variations of permeate flux with rotating speed for different TMP at VRR = 1 and VRR = 4 using full recycling are shown in Fig. 4. The maximum membrane shear rate at different rotating speeds was calculated by Eq. (2). As expected, when TMP was low (10 bar), the effect of shear rate on permeate flux was negligible owing to the very light concentration polarization. That is, the shear rate produced by low rotating speed (500 rpm) could eliminate concentration polarization at low TMP. With increase of TMP, the influence of shear rate on flux was more obvious. As seen in Fig. 4, the highest TMP and shear rate brought the largest permeate flux and this trend was independent of feed concentration. Permeate conductivity at different shear rate and TMP, displayed in Fig. 5, shows that solutes concentration in permeate decreases when shear rate and TMP are raised. This effect is due to a combination of two factors [16]. First, inorganic salt concentration at the membrane decreases as concentration polarization is reduced with increasing rotating speed. Since salt ions transfer through the membrane is mainly controlled by diffusion, it will be smaller at higher shear rate when salt concentration at membrane is lower. The second factor is the dilution of salt ions in permeate as the permeate flux increases with TMP and shear rate (see Fig. 4) while salt ions transfer decreases. These effects were higher when feed was concentrated to VRR = 4 because concentration polarization was higher for concentrated feed with higher viscosity. Thus, extreme hydraulic conditions would also improve the quality of permeate water for reuse.

Although permeate flux and water quality were improved when shear rate and TMP increased, energy cost should also be considered

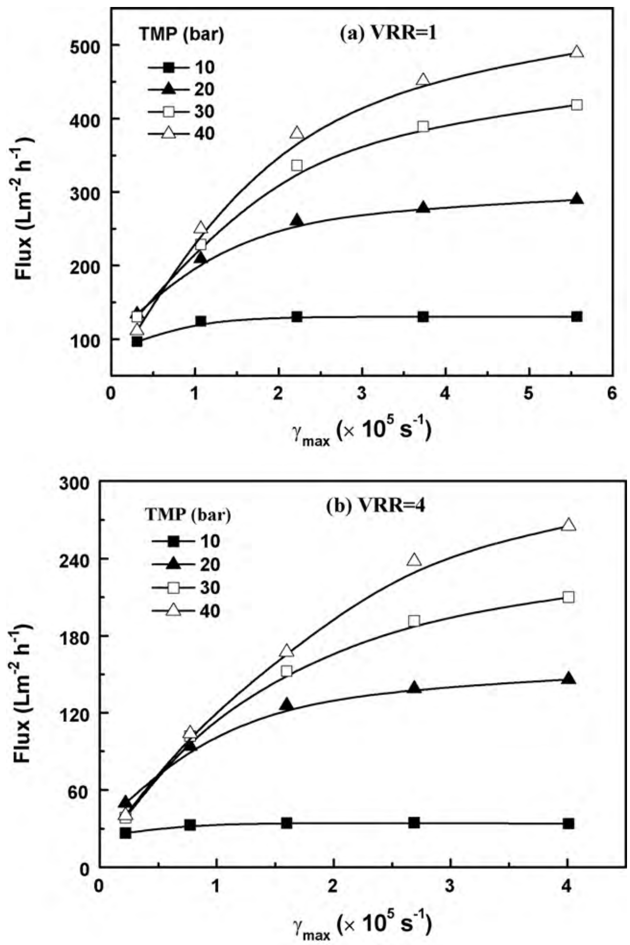

Fig. 4. Effect of shear rate and TMP on permeate flux (a) at VRR = 1 and (b) at VRR = 4.

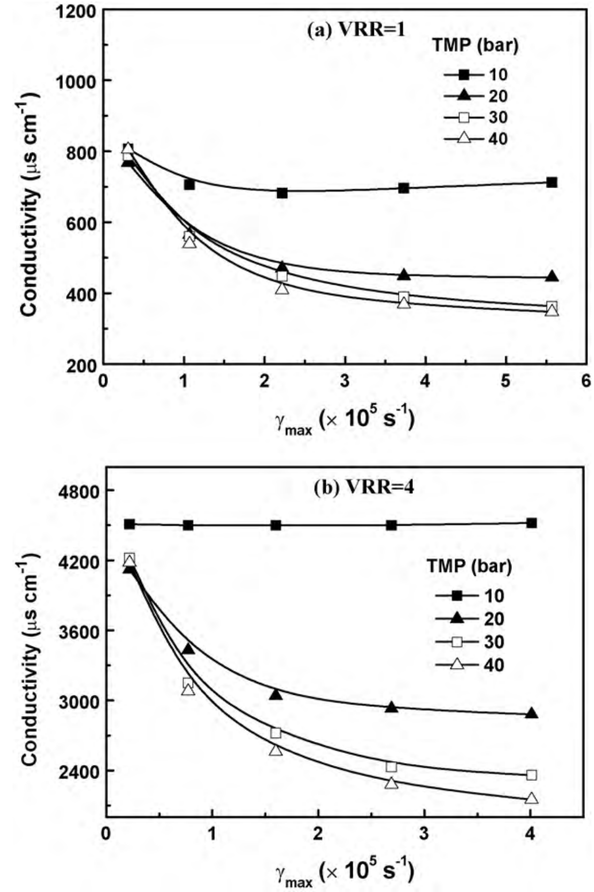

Fig. 5. Effect of shear rate and TMP on permeate conductivity (a) at VRR = 1 and (b) at VRR = 4.

seriously. According to Eq. (4), the specific energy cost for RDM filtration system mainly depended on the permeate flux and power of rotating disk motor and feed pump. Fig. 6 shows the power of rotating disk motor and feed pump. It can be seen that, for rotating disk motor, the power shows an exponential relationship with rotating speed and is independent of TMP, and for feed pump, the power increases linearly with rising TMP but is not affected by rotating speed. In addition, feed concentration and temperature had little influence on the power of rotating disk motor and feed pump in our study range (data not shown). Specific energies for different VRR, TMP and shear rates could be calculated and compared from data in Figs. 4 and 6. As shown in Fig. 7, at TMP = 10 bar, specific energy increases continuously with shear rate because enhanced shear rate does not increase flux; for other higher TMP, specific energy first decreases significantly and then slightly increases or remains almost constant. This could be explained by the fact that, at first, permeate flux increased rapidly at higher shear rate, and according to Eq. (4), specific energy decreased; but at high rotating speed, the rising slope of total power enhanced but the increase of flux with shear rate slowed down or stopped (see Figs. 6 and 4), so the specific energy was no longer reduced above a shear rate of $3 \times 10^5\ s^{-1}$. When the shear rate was low, higher TMP was not recommended, but as shear rate was high enough (more than $2 \times 10^5\ s^{-1}$), extreme TMP (40 bar) was optimum for NF270. Here, the values of specific energy were much higher than that reported previously because this system was not optimized yet. Therefore, taking into account permeate flux, water quality and specific energy, extreme hydraulic conditions, a TMP of 40 bar and rotating speed of 2000 rpm, were optimum for treatment of model wastewater by RDM module with NF270. Using numerical simulation, Torras et al. [25] also found that a rotating speed around 2000 rpm seemed to be a good compromise between energy consumption and permeate flux. However, this result was obtained with full recycling for a short test and should be further confirmed in long-term concentration tests.

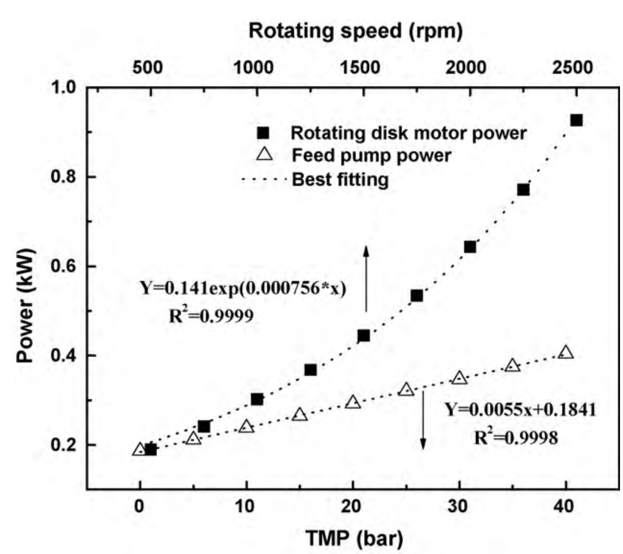

Fig. 6. Power of rotating disk motor and feed pump at different TMP and rotating speed using water as feed (25 °C).

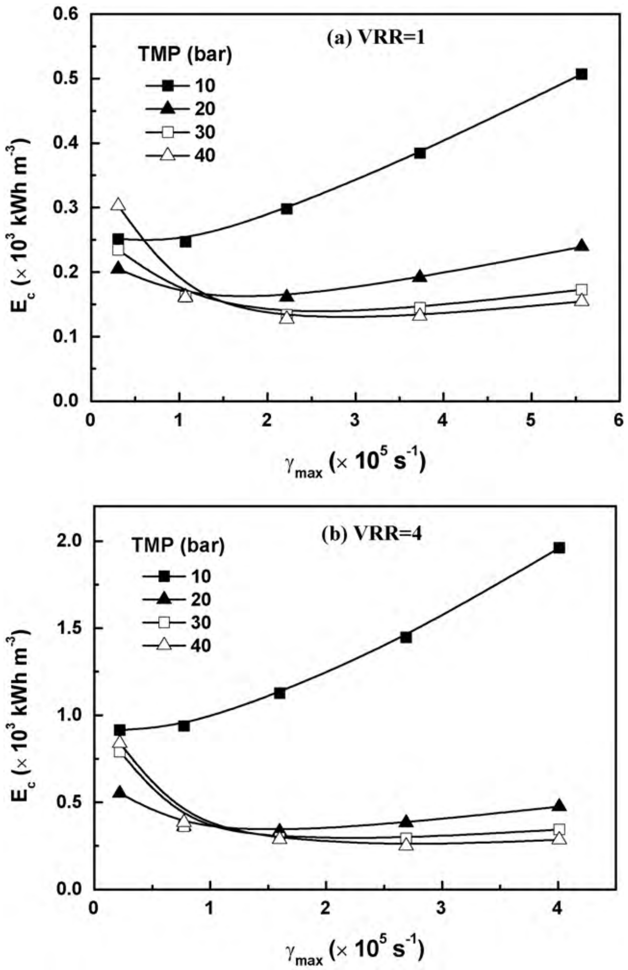

Fig. 7. Effect of shear rate and TMP on specific energy (a) at VRR = 1 and (b) at VRR = 4.

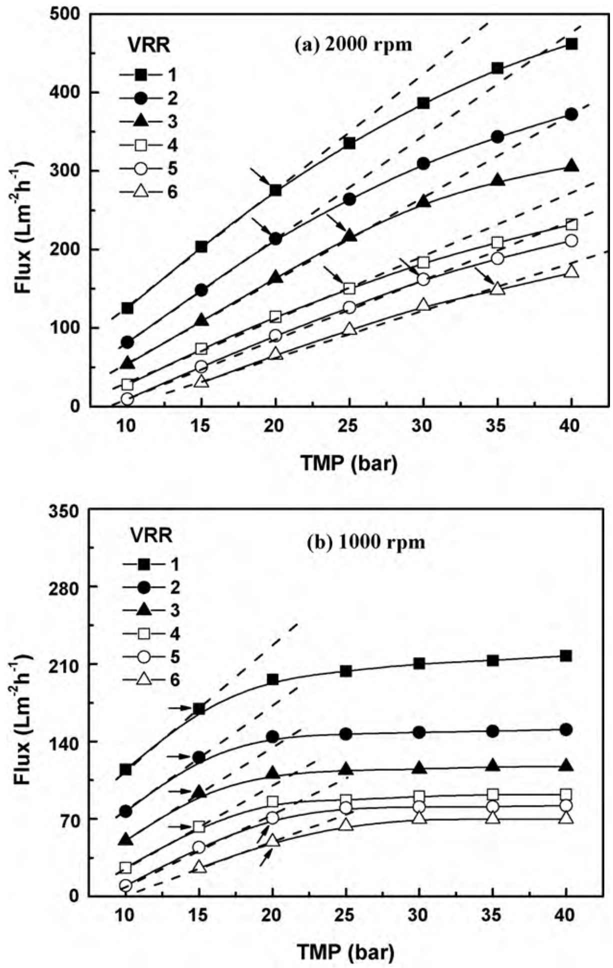

Fig. 8. Variations of permeate flux at different VRR, TMP (a) at 2000 rpm and (b) at 1000 rpm (Arrows indicate values of critical flux and TMP).

3.3. Effect of feed concentration and operating strategy

The concept of critical flux [26] is very important to membrane process for fouling control and energy saving, especially for crossflow filtration module. Manttari and Nystrom [27] found that critical flux was obviously affected by flow velocity and feed concentration for NF of high molecular weight polysaccharides and effluents from paper industry in crossflow mode. For a shear-enhanced NF system, to our knowledge, there was no report yet concerning the critical flux at different shear rates and feed concentrations. In the present study, concentration tests operating below critical flux or under extreme hydraulic conditions were analyzed and compared, and because critical flux varied with shear rate and feed concentration, detailed analyses containing physicochemical properties of feed and permeate flux variation with increase of TMP were carried out at each VRR.

As shown in Fig. 8, permeate flux increases continuously with TMP at all VRR when rotating speed is 2000 rpm, but for the lower rotating speed 1000 rpm, the flux remains almost constant when TMP exceeds 25 bar at most of VRR range because of its relatively low shear rate. The critical TMP corresponding to critical flux at different VRR are shown by arrows and will be used for following investigations of operating strategy (here, the critical value is the point at which the flux ceases to increase linearly with TMP). With increase of VRR, permeate flux decreased due to increased solutes concentration in feed while critical TMP increased. It also can be seen in Fig. 8 that, the critical TMP for RDM filtration system are much higher than that for crossflow module, where it rarely exceeds 15 bar due to limited shear rate. The variations of conductivity in permeate with VRR and TMP, displayed in Fig. 9, shows that for a rotating speed of 2000 rpm, permeate conductivity does not decrease from 30 bar to 40 bar at VRR = 1–3 as expected, although their flux increases with TMP at this range. As we know, a higher flux had two contradictory effects, the first is to increase concentration polarization and thus increasing solutes transfer through membrane by diffusion, and the other is permeate dilution, which decreases solutes concentration in permeate. Therefore, when the dilution effect was counteracted by the concentration polarization effect at higher flux regime for lower feed concentration, the conductivity in permeate did no longer drop with increase of permeate flux. But the trends of permeate conductivity with TMP in other cases were consistent with that of permeate flux due to the dominant dilution effect. Because concentration polarization was well controlled by high shear rate here, increase of solutes concentration in permeate with rise of flux, that usually occurred in crossflow filtration system, was not found in RDM filtration systems. Actually, all phenomena in Figs. 8 and 9 were caused by the effect of concentration polarization at different shear rates and flux, which could be understood from data listed in Tables 4 and 5. With increase of VRR, the dry mass and ions concentration in feed went up, leading to an increase of osmotic pressure at membrane surface rather than an additional hydraulic resistance, thus reducing permeate flux at constant TMP. Shear rate also decreased with increase of VRR due to the increase of fluid kinematic viscosity and reduction of rotating speed.

Table 4
Physicochemical properties of feed at different VRR.

VRR	Dry mass ($g\,L^{-1}$)	ρ ($g\,L^{-1}$)	μ (mPa s)	ν ($m^2\,s^{-1}$)	Conductivity ($\mu s\,cm^{-1}$)
1	31.9	1030	0.9263	0.899E−06	2170
2	63.8	1059	1.0120	0.956E−06	3350
3	98.2	1063	1.1424	1.075E−06	4310
4	135.5	1074	1.4547	1.354E−06	5410
5	174.2	1081	1.7218	1.593E−06	5930
6	217.1	1095	2.1982	2.007E−06	6250
7	258.5	1099	2.6534	2.414E−06	6500

Table 5
Maximum membrane shear rate γ_{max} at different VRR.[a]

Rotating speed (rpm)	γ_{max} ($\times 10^5\,s^{-1}$)						
	VRR = 1	VRR = 2	VRR = 3	VRR = 4	VRR = 5	VRR = 6	VRR = 7
1000	1.07	1.02	0.924	0.773	0.678	0.562	0.486
2000	3.73	3.55	3.22	2.69	2.36	1.96	1.69

[a] Calculated by Eq. (2).

Four concentration tests were separately carried out under different operating strategies according to critical TMP derived from Fig. 8 at each critical flux. They were (1) 2000 rpm at extreme TMP 40 bar (constant TMP); (2) 2000 rpm at critical TMP (variable TMP); (3) 1000 rpm at extreme TMP 40 bar (constant TMP); (4) 1000 rpm at critical TMP (variable TMP). Variable TMP with VRR are shown in Fig. 10a. As seen in Fig. 10b, a constant 40 bar TMP produced higher flux than critical TMP, especially at 2000 rpm. Only at the end of concentration tests, permeate flux at critical TMP was a little higher than that of extreme TMP, confirming that concentration polarization or membrane fouling was lower at critical flux. Table 6 shows permeate water quality, operating time, specific energy and irreversible fouling under different operating strategies. Average values shown in this table are values measured in accumulated permeate collected at the end of the concentration process after stirring. Average permeate turbidity was not much affected by operating strategies because NF could reject almost all feed protein. Permeate conductivity was function of permeate flux due to dilution effect, but permeate COD in strategy (3) was less than that in strategies (2) and (4), and this was mainly attributed to higher membrane fouling for strategy (3), which increased the retention of organic matters. Both operation time and specific energy for operating strategy (1) were lowest because of its highest permeate flux. Nevertheless, specific energy in strategy (4) was less than that in strategy (3) although the latter had a higher flux until VRR = 3.5. This could be caused by excessive TMP at 1000 rpm (shear rate was not high enough) in strategy (3), which induced severe concentration polarization and membrane fouling.

Thus, higher shear rate at critical flux was preferable to minimize the irreversible fouling of membrane, as seen in Table 6. It was seen that, even if concentration polarization and fouling could be greatly reduced by high shear rate in shear-enhanced filtration system, the concept of critical flux was still valid. Anyway, the operating strategy under extreme hydraulic conditions gave higher permeate flux, better water quality, and lower specific energy, but it produced a little more membrane fouling compared with the strategies (2) and (4) at critical flux. Because membrane fouling at high shear rate was not very severe, treatment of model wastewater under extreme hydraulic conditions was still thought to be most efficient, even if additional cleaning time was needed. Thus we apply this new technique to real dairy effluent which had more complex composition and can induce more severe fouling.

3.4. Comparison of model and real effluents

As shown in Table 2, real effluents have much less organic matters and inorganic salt than model solution, but their pH was much higher due to using alkaline solution as cleaning agent. Moreover, the components and concentration of effluents in milk factory changed with time. Two batches of effluents were taken for preliminary experiments under extreme hydraulic conditions in order to compare with the model solution. Fig. 11 shows the permeate flux with VRR for different feed. Unexpectedly, the flux of real effluents was of the same level as that of model solution despite of different compositions. The reason could be that, first, macromolecular proteins that resulted in high COD in model solution had little effect on osmotic pressure at membrane surface, but for real effluents, although its COD was low, the concentration of low molecular weight organic solutes which were responsible for osmotic pressure might be equal with that of model solution; secondly, high shear rate could eliminate the difference of concentration polarization resulted from different viscosity between model solution and real effluents; thirdly, the rejection of inorganic salt was a little higher for real effluents due to higher pH while its salt concentration was slightly lower than in model solution (see Table 7). Moreover, in Fig. 11, flux of Batch 1 becomes constant when VRR is more than 2, while the flux of Batch 2 decreases continuously and becomes lower than that of Batch 1 as VRR exceeds 3. This differ-

Table 6
Effect of operating strategies on treatment results.

Operating strategies/index	Average permeate turbidity (NTU)	Average permeate conductivity ($\mu s\,cm^{-1}$)	Average permeate COD ($mg\,O_2\,L^{-1}$)	Operation time (h)	Mean[a] specific energy ($\times 10^3$ $kWh\,m^{-3}$)	*IF* (%)
(1) 2000 rpm at extreme TMP	0.57	756	58	1.667	0.170	21.88
(2) 2000 rpm at critical TMP	0.62	1000	120	2.633	0.242	15.78
(3) 1000 rpm at extreme TMP	0.55	1160	85	4.417	0.303	33.50
(4) 1000 rpm at critical TMP	0.70	1280	150	5.000	0.277	21.00

[a] Calculated by mean Q_f of the whole concentration process.

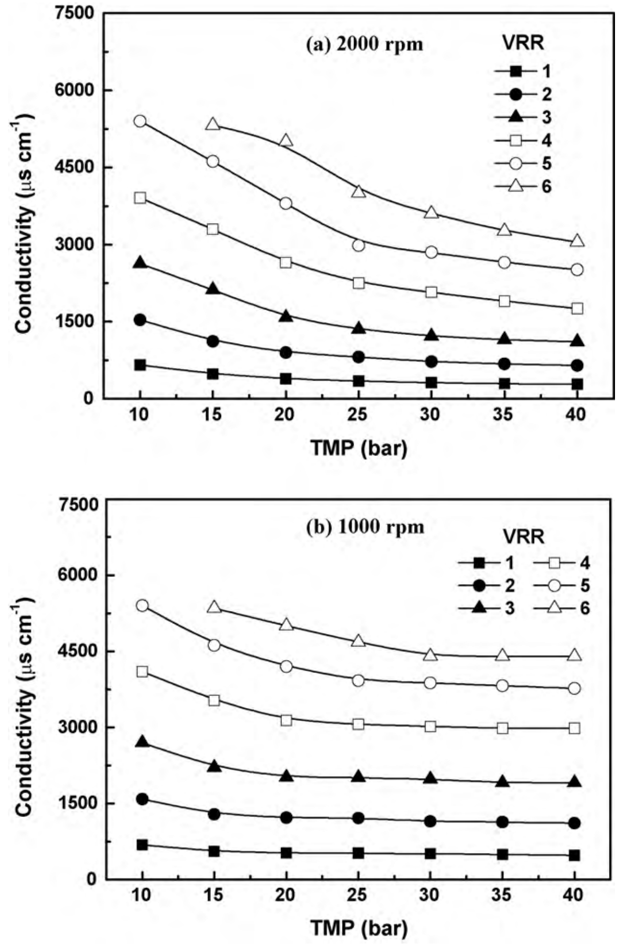

Fig. 9. Variations of permeate conductivity at different VRR, TMP (a) at 2000 rpm and (b) at 1000 rpm.

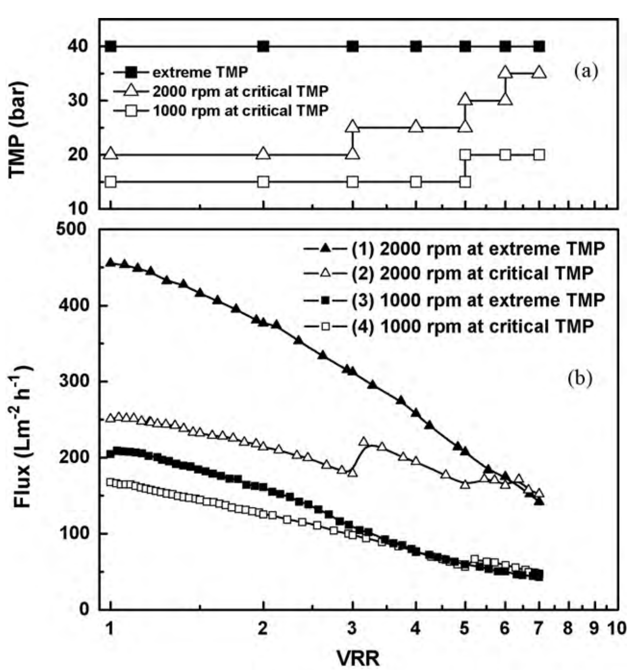

Fig. 10. (a) Variation of TMP with VRR for different strategies; (b) variation of permeate flux with VRR for strategies of (a).

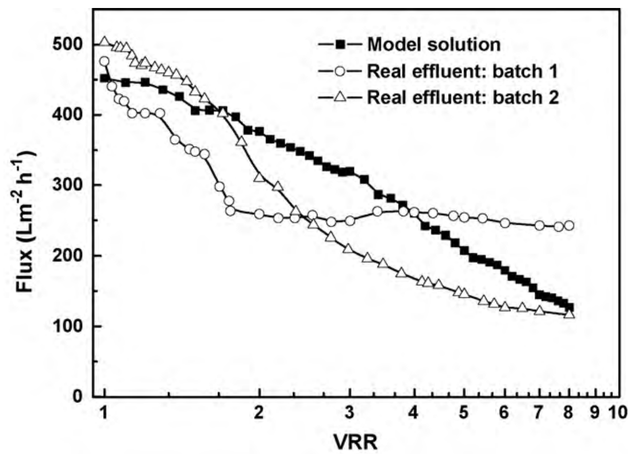

Fig. 11. Comparison of permeate flux between model solution and real effluent under extreme hydraulic conditions. TMP = 40 bar; rotating speed = 2000 rpm.

ence was not caused by membrane fouling because *IF* of Batch 1 was much higher than that of Batch 2, as displayed in Table 7. This could be explained by their different feed pH and pH decrease during concentration process (data not shown). As the concentration process went on, acid radicals (e.g. phosphate) could precipitate with caseins as well as with calcium [28], and thus more hydrogen ions were produced by electrolysis and pH decreased. For Batch 2, its pH might decrease to 9, and Rabiller-Baudry et al. [28] found that the flux of milk by NF dropped sharply when pH was near 9. In addition, due to the stronger electrostatic repulsion between membrane and ions at higher pH, the lower transfer of salt ions through the membrane for Batch 2 (see Table 7) could also explain its lower flux. For Batch 1, as the pH in retentate decreased, salt ions passed through membrane more easily, and thus, flux decline should be reduced, but why the flux remained almost constant at VRR > 2 needs further study. Furthermore, the membrane fouling for Batch 2 was much less than that for Batch 1 due to the pH difference, and this was consistent with the results reported by Rabiller-Baudry et al. [28], that with increase of pH, membrane fouling by milk decreased. However, Rice et al. [29] found that fouling occurred most rapidly at high pH (8.9) when treating dairy ultrafiltration permeates by NF, and this deviation was caused by the different fouling mechanism when casein was removed.

Because real effluents had much lower COD concentration, the permeate COD was less than 15 mg O_2 L^{-1}, indicating that NF treatment of dairy wastewater by RDM module could obtain very high quality water for reuse, and VRR could be raised to a higher level. It also can be seen from Table 7, that membrane fouling for real effluents was not higher than for the model solution, and for Batch 2 with highest pH, *IF* was only 13.40%, much lower than in other tests. It was possible that a higher pH could increase solutes rejection and alleviate membrane fouling [28]. The study about effect of pH on membrane fouling will continue in the future.

Table 7
Comparison of treatment results between model solution and industrial effluent.

Index	Feed/permeate		
	Model solution	Batch 1	Batch 2
Turbidity (NTU)	NA/0.57	101/0.56	100/0.57
COD (mg O_2 L^{-1})	36,000/54	297/<15	580/<15
Conductivity (μs cm^{-1})	2170/685	1084/525	1516/317
pH	6.84/6.62	8.72/7.90	9.56/9.00
Operation time (h)	1.717	1.867	2.083
IF (%)	21.88	26.87	13.40

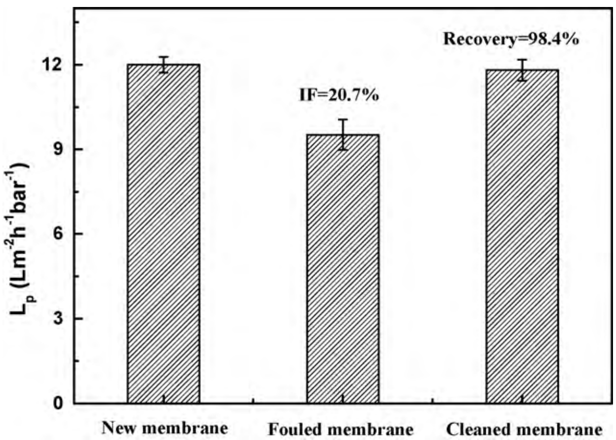

Fig. 12. Membrane permeability for new, fouled and cleaned membranes calculated from Table 7.

3.5. Membrane fouling and cleaning

In preceding section, it was shown that membrane fouling in treatment of real dairy wastewater by RDM module under extreme hydraulic conditions was moderate and that it might be decreased by adjusting the pH of feed. Another approach to offset the effect of fouling under extreme hydraulic conditions was to find an effective cleaning strategy. Using 0.1% Ultrasil 10 with pH = 11 at room temperature, the cleaning process was performed at 1000 rpm for 30 min, and initial membrane permeability could almost be recovered. Fig. 12 shows the L_p of membranes before and after cleaning in the preceding experiments (Section 3.4), indicating that, by using a commercial cleaning agent, NF membrane fouling by dairy wastewater could be removed efficiently in the RDM filtration system.

4. Conclusions

As compared to Desal-5 DK, the NF270 membrane showed much higher permeate flux in treatment of dairy wastewater, and using membranes with high flux permits to fully exploit the potential of the RDM filtration system. Because our filtration system produced very high shear rate to reduce concentration polarization, permeate flux could increase continuously when transmembrane pressure (TMP) rose from 10 to 40 bar. Under extreme hydraulic conditions of highest TMP with high shear rate, the RDM filtration system could produce a better quality permeate and save energy, because of its very high permeate flux. These advantages were present in both full recycling and concentration processes.

With increase of VRR during concentration of dairy wastewater, critical flux decreased but critical TMP increased. Shear rate decline would result in decrease of critical flux and TMP. Flux was higher at extreme TMP except at high VRR when it became equal with the case of critical TMP. Membrane fouling when operating under critical flux (in short tests) was a little smaller than when operating under extreme hydraulic conditions, while specific energy of the former was much higher than that of the latter (~40%) when rotating speed was 2000 rpm. Real dairy wastewater was processed under extreme hydraulic conditions and the results were similar to those of model solution. Thus, diluted wastewater could be concentrated to much higher VRR, increasing water recovery and decreasing the load of following treatment. Higher pH in feed might alleviate membrane fouling and improve rejection of solutes. A routine cleaning procedure could recover membrane permeability so the operation under extreme hydraulic conditions could become practical for treating dairy wastewater. Although wastewater treatment by NF under extreme hydraulic conditions had many advantages, high shear, high TMP and cleaning might deteriorate membrane performance in long-term runs, and future investigations are needed.

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