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How a Microfiltration Pretreatment Affects the Performance in Nanofiltration

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ABSTRACT

The use of a well-chosen pretreatment system is a key element to avoid fouling in nanofiltration (NF). Among the different possibilities for pretreatment systems, microfiltration (MF) emerges as the most compatible with NF. This article explores the influence of a MF pretreatment by comparing the performance of three NF membranes (UTC-20, Desal 51 HL,

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and NF-PES-10) with and without MF pretreatment for the purification of two types of wastewaters from the brewery of Westmalle, Belgium. The wastewaters studied were bottle-rinsing water and rinsing water from the fermentation tanks, respectively. The results indicate that MF pretreatment had no influence for UTC-20, a considerable influence for Desal 51 HL, and a dramatic influence for NF-PES-10. No correlation with the membrane roughness, determined by AFM, was found, but the results support the assumption that particle fouling is mainly determined by the hydrophobicity of the membrane. The effect of pretreatment was largest for the hydrophobic NF-PES-10 membrane, smallest for the hydrophilic UTC-20 membrane, and intermediate for Desal 51 HL. The effect of the composition of the feed solution was considerably smaller than the effect of the membrane used, although, small differences where found depending on the size and concentration of the particles.

Key Words: Nanofiltration; Microfiltration; Pretreatment; Fouling; Brewery wastewater.

INTRODUCTION

As it allows the removal of components with relatively low molecular weight, nanofiltration (NF) is a process with numerous applications in drinking water production, process water recovery in industry, and wastewater treatment.[1] In addition to the frequently reported problems for concentrate discharge, the major limitation for the implementation of nanofiltration is the occurrence of fouling. In general, fouling can be defined as "irreversible precipitation or sorption of retained particles, bacteria, colloids, macromolecules, salts, or organic components onto the membrane surface or into the membrane bulk structure." Thus, different types of foulants can be distinguished; the most important are inorganic precipitates, organic components, bacteria, and particles.[2–6] Depending on the type of the dominant foulant, a strategy or a combination of strategies can be chosen to maintain the performance of the membrane. Inorganic precipitation, usually referred to as scaling, can be controlled by a periodic chemical cleaning with acid solutions (H_3PO_4 and citric acid). Organic fouling can be minimized by using low-fouling membranes, which consist of a hydrophilic polymeric top layer, so that interactions between organic components and the membrane (e.g., adsorption) are minimized. Additionally, a chemical cleaning procedure helps to remove adsorbed organics; chemicals used for this purpose are, e.g., NaOH, detergents, and complexing agents (EDTA, polyacrylates, and sodium hexametaphosphate). These chemicals, along with disinfectants, are also useful to reduce the effect of biofouling. Particle fouling can be controlled by hydraulic rinsing methods (e.g., forward flushing or periodic changes in the flow direction) or mechanical rinsing methods using slightly oversized sponge balls. The latter can only be used in tubular systems. Newer methods include electrical cleaning by applying an electrical field that removes charged particles or molecules from the interphase. Obviously, the membranes used should be sufficiently conductive, which limits the application to nonpolymeric systems, and special modules have to be used with built-in electrodes.

In addition to these specific remediation methods, the design of the membrane module and the operational parameters also have a significant influence. General measures to prevent fouling aim at decreasing concentration polarization by an increase of the mass resistance coefficient. The use of turbulence promoters and a suitable flow pattern inside the module are also important.[7,8] Furthermore, a low recovery decreases the risk of scaling and organic fouling; however, high recoveries are usually preferred in view of increasing the permeate yield and minimizing the concentrate fraction.

Regardless of the method(s) used to control fouling, a pretreatment is usually applied to prevent fouling.[9] The most important pretreatment methods are biological degradation, coagulation/flocculation, microfiltration (MF), and ultrafiltration (UF). Aerobic biological pretreatment is frequently used as a pretreatment method for historical reasons: NF is usually a later addition to the treatment sequence, in view of obtaining the required quality for water reuse. Existing installations, such as an activated sludge system, are usually maintained and serve as a pretreatment system for the NF unit. Experimental results[10,11] show that in NF the water flux and the (overall) rejection of organic compounds are higher when a biological pretreatment is used.

An alternative to biological pretreatment is coagulation/flocculation using inorganic electrolytes, organic polymers, or synthetic polyelectrolytes.[12] The effect of a coagulation/flocculation pretreatment depends on the pH, the type and concentration of the additives used, and the characteristics of the feed water. This method is mainly used for the removal of organic substances (macromolecules and organic particles).[13–16] However, a negative influence on the permeate flux in NF has been reported;[17] furthermore, components leading to biological growth are not efficiently removed,[18] which may result in biofouling of the NF membranes. Other negative aspects are the cost of chemicals and the further treatment of the generated sludge.[17]

MF reduces the concentration of bacteria, colloids, turbidity, and dissolved compounds associated with particles.[14,19–23] A decrease of chemical oxygen demand (COD) or total organic carbon (TOC) is only obtained when a significant organic fraction is associated with particles larger than the pore size.[24,25] However, instead of contributing to the removal of contaminants, the main effect of a MF treatment seems to be the prevention of decrease in

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the NF flux on a long-term basis,[13,26] which has significant implications on cost factors.[27] A similar effect is obtained with UF, which has the same operating principles as MF, but with smaller pores and a lower permeate flux per unit-membrane area. A higher removal efficiency of organic compounds is obtained because high molecular weight dissolved components are also retained by the UF membrane; however, a recycle of UF permeate is not feasible.[26,28,29] Given the large difference in water permeability between UF and MF, the latter process is usually advantageous if it is used only as a pretreatment method. However, it is unclear to what extent particles contribute to fouling in NF, and thus, to what extent a pretreatment improves the NF performance. Apart from some empirical observations, no information about the necessity of pretreatment as a function of membrane characteristics and the composition of the feed solution is available. Based on observations of fouling by particles in NF,[30,31] it can be stated that the membrane properties have a significant influence. This article evaluates the effect of particles in NF on fouling and rejections by comparing fluxes and rejections in NF with and without MF pretreatment, obtained for two types of wastewater from a brewery of specialty beers.

MATERIALS AND METHODS

Feed Solution

Two types of wastewater were used in this study. Both were obtained from the Westmalle brewery, Malle, Belgium. The Westmalle brewery produces high-quality “Trappist” beers with secondary fermentation and long maturation in the bottle. This brewing method has significant implications on the composition of the wastewaters, which are very different from wastewaters from breweries specializing in lager beers. The first type of wastewater was rinsing water used for the returned bottles, which contains small fractions of beer and nondissolved fractions originating from labels, sand, etc. The suspended solids concentration ranged from 60 to 100 mg/L; the ion conductivity from 950 to 1600 μS/cm; and the COD from 380 to 480 mg/L. The second type of waste water was rinsing water from the fermentation tanks, which contains large concentrations of nonsettleable yeast particles with a size of approx. 10 μm. The suspended solids concentration ranged from 760 to 2300 mg/L (10–20 times higher than for the first type); the ion conductivity from 580 to 1100 μS/cm (slightly lower than for the first type); and the COD from 2800 to 8700 mg/L (10–20 times higher than for the first type).

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Microfiltration Experiments

An Amicon 8200 dead-end stirred cell (Millipore, Billerica, MA) was used for the MF experiments. The volume of the cell was 200 mL; a connection to a 2-L reservoir was made in order to allow filtration of a larger volume in a single operation. The active membrane area was 28.7 cm^2. The membranes used were 0.45 μm cellulose ester membranes (Millipore, Billerica, MA).

Nanofiltration Experiments

A Test Rig PSSITZ NF unit (Amafilter, The Netherlands) was used for the NF experiments. The module had an effective membrane area of 44 cm^2. A batch recirculation system was used in which the retentate is recycled to the feed solution. The pressure in the experiments was set at 9 bar and the temperature was set at 25°C. The feed velocity was 6 m/sec in all the experiments. Three membranes were used in the experiments: Desal 51 HL (Osmonics, Minnetonka, MN), UTC-20 (Toray Ind. Inc., Tokyo, Japan), and NF-PES-10 (Nadir GmbH, Wiesbaden, Germany). The water flux and the rejection of suspended solids and organic matter were taken as the main indicators of the membrane performance. In all experiments, the water flux was evaluated as an absolute value and as a fraction of the initial water flux (at the start of the experiment).

Analysis

Organic matter was measured as COD using a standard procedure.[32] Conductivity was measured using an Orion Model 160 conductivity meter. Measurement of pH was carried out with a Orion Model 420A pH meter. The suspended solids concentration was determined by filtering the solution with a 0.22 μm membrane and measuring the weight change of the membrane after drying.

Atomic Force Microscopy

Atomic force microscopy (AFM) images were obtained using a M5 AFM System (Park Scientific Instruments) in tapping mode. Silicium supports were used (Sharp Ultralevers, Park Scientific Instruments) and a cantilever with a spring constant of 3.2 N/m and a nominal apex radius of 10 nm.

RESULTS AND DISCUSSION

The pH, COD, conductivity and volume of the feed solutions, the NF permeates (without pretreatment), the MF permeates, and the NF permeates (after pretreatment) are summarized in Table 1 for the bottle rinsing water and in Table 2 for the fermentation tank rinsing water, together with the rejections in NF and in the combined MF–NF process; the volume of each fraction obtained during the experiments was also indicated. The COD rejections were as expected from the molecular weight cut-off (MWC) of the membranes: NF-PES-10 has a large MWC (ca. 1000) and subsequently relatively low COD rejections; Desal 51 HL (MWC 150-300) and UTC-20 (MWC ca. 180) have

Table 1. pH, COD, conductivity, and obtained volumes for the three NF membranes UTC-20, Desal 51 HL, and NF-PES-10 with bottle rinsing water as feed.

	pH	COD ($mg\ O_2/l$)	Conductivity ($\mu S/cm$)	Vol. (l)
Feed				
UTC-20	8.07	1005	479	9.8
Desal 51 HL	7.47	946	388	9.4
NF-PES-10	6.98	1581	413	10.1
NF (no pretreatment)				
UTC-20	8.16	406	106	6.9
Desal 51 HL	7.51	283	160	6.5
NF-PES-10	6.64	1352	66	0.7
Rejection NF (%)				
UTC-20		59.6	77.9	
Desal 51 HL		70.1	58.8	
NF-PES-10		14.5	84.0	
MF				
UTC-20	8.21	960	212	8.4
Desal 51 HL	7.57	859	292	9.7
NF-PES-10	7.39	1478	260	9.4
MF + NF				
UTC-20	8.08	276	99	5.9
Desal 51 HL	8	183	16	7
NF-PES-10	7.65	1254	47	3.6
Overall rejection MF + NF (%)				
UTC-20		72.5	79.3	
Desal 51 HL		80.7	95.9	
NF-PES-10		20.7	88.6	

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Table 2. pH, COD, conductivity, and obtained volumes for the three NF membranes UTC-20, Desal 51 HL, and NF-PES-10 with fermentation tank rinsing water as feed.

	pH	COD (mg O_2/l)	Conductivity (μS/cm)	Vol. (l)
Feed				
UTC-20	5.92	580	2846	10
Desal 51 HL	6.38	1111	6452	10.2
NF-PES-10	5.77	776	8650	9.9
NF (no pretreatment)				
UTC-20	5.97	85	556	5.9
Desal 51 HL	6.24	322	2512	3
NF-PES-10	6.09	386	4080	0.3
Rejection NF (%)				
UTC-20		85.3	80.5	
Desal 51 HL		71.0	61.1	
NF-PES-10		50.3	52.8	
MF				
UTC-20	6.46	625	1600	9.6
Desal 51 HL	6.58	1164	4010	8.1
NF-PES-10	5.38	641	5032	8.6
MF + NF				
UTC-20	6.46	106	404	4.9
Desal 51 HL	5.96	208	1236	3.1
NF-PES-10	6.4	372	3772	0.8
Overall rejection MF + NF (%)				
sUTC-20		81.7	85.8	
Desal 51 HL		81.3	80.8	
NF-PES-10		52.1	56.4	

significantly higher COD rejections. Less difference in ion rejections was found; the surface charge of the three membranes is similar. For UTC-20 only size exclusion effects may further increase the ion rejection. Changes in pH are relatively small.

It was observed that the overall rejections in the combined MF + NF process are generally somewhat higher than the rejections in NF without pretreatment, although in general the difference is small. This is due to a small but significant removal of COD and ionic species in the MF pretreatment. Although, the final NF barrier is identical, the rejections are different because a small fraction was already removed in the pretreatment step.

Comparison of the COD rejection for bottle rinsing water and fermentation tank rinsing water shows that the rejections are always larger in the latter

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case. This confirms the assumption that the fermentation tank rinsing water contains larger suspended particles and dissolved organic compounds with high molecular weight than the bottle rinsing water. The final permeate, however, still has a considerable COD, presumably due to small dissolved organic compounds such as ethanol from the returned bottles or from remaining fractions in the fermentation tanks.

Table 3 summarizes the pure water flux measured with distilled water, the water fluxes with both bottle rinsing water and fermentation tank rinsing water and the observed flux decline for the three NF membranes used (UTC-20, Desal 51 DL, and NF-PES-10), with and without MF pretreatment. The water flux after 8 hr of filtration is taken as a reference value in this table. A large variation of pure water fluxes was observed. The pure water flux is generally higher than the initial water flux, so that flux decline relative to the pure water flux is more important than flux decline relative to the initial water flux. However, both parameters reflect the same tendencies; therefore, the initial water flux will be used as a reference value when possible. The evolution of the water flux as a function of time with the three NF membranes is given in Fig. 1 for the bottle rinsing water and in Fig. 2 for the rinsing water from the fermentation tanks. Open symbols refer to NF fluxes without MF pretreatment, filled symbols refer to NF fluxes after MF pretreatment of the feed solution.

In all cases, the MF pretreatment resulted in a similar or higher water flux during NF, although, the difference is sometimes small. For NF-PES-10, a temperature increase was observed, which resulted in a decrease of the viscosity and consequently, a flux increase. The effect of the MF pretreatment depends on (a) the membrane used and (b) on the feed solution.

(a) *Influence of the membrane material.* The effect of a MF pretreatment (see Table 3) is small for UTC-20: for the bottle rinsing water, no improvement of the performance was found; for the fermentation tank rinsing water, a small increase of the relative fluxes ($J_{8\,\mathrm{hr}}/J_0$ or $J_{8\,\mathrm{hr}}/J(0)$) was found after MF pretreatment (max. ca. 7%). A somewhat larger effect was observed for Desal 51 HL: a 6–8% increase when compared to the initial water flux and a 14–24% increase when compared to the pure water flux. A large difference in flux decline with and without MF pretreatment was observed for NF-PES-10. Without pretreatment, the water flux decreases to ca. 25% of the initial value for both bottle rinsing water and rinsing water from the fermentation tank, and to 3–8% when the pure water flux is taken as a reference. After MF, the water flux was significantly higher: 85.0% of the initial water flux (67.7% of the pure water flux) for the bottle rinsing water, and 59.8% of the initial water flux (24.2% of the pure water flux) for the fermentation tank rinsing water.

Because, the pore size of the MF membrane used is ca. 0.45 μm, MF removes particles larger than ca. 0.45 μm. Therefore, the improvement of the NF performance is due to membrane fouling by particles. The suggestion

Table 3. Water fluxes and flux decline obtained with the NF membranes UTC-20, Desal 51 HL, and NF-PES-10 and for bottle rinsing water ("bottle") and fermentation tank rinsing water ("tank").

Membrane	Feed	Pretreatment	J_0 (L/m^{2}hr)	$J(0)$ (L/m^{2}hr)	$J_{8hr}/J(0)$ (%)	J_{8hr}/J_0 (%)	$J(0)/J_0$ (%)
UTC-20	Bottle	No MF	104.1	107.1	98.1	100.9	102.9
Desal 51 HL	Bottle	No MF	150.5	103.1	67.8	46.5	68.5
NF-PES-10	Bottle	No MF	163.0	52.4	25.3	8.1	32.1
UTC-20	Bottle	MF	130.5	100.5	99.1	76.3	77.0
Desal 51 HL	Bottle	MF	109.8	101.6	76.2	70.5	92.5
NF-PES-10	Bottle	MF	136.1	108.3	85.0	67.7	79.6
UTC-20	Tank	No MF	103.1	94.8	84.2	77.5	91.9
Desal 51 HL	Tank	No MF	111.4	75.6	69.2	47.0	67.9
NF-PES-10	Tank	No MF	171.4	20.0	25.8	3.0	11.7
UTC-20	Tank	MF	141.2	125.1	90.8	80.4	88.6
Desal 51 HL	Tank	MF	103.2	83.7	75.2	61.0	81.1
NF-PES-10	Tank	MF	66.6	27.0	59.8	24.2	40.5

Note: J_0 = pure water flux measured with distilled water; $J(0)$, initial water flux measured with feed solution; flux J_{8hr} is measured after 8 hr of filtration.

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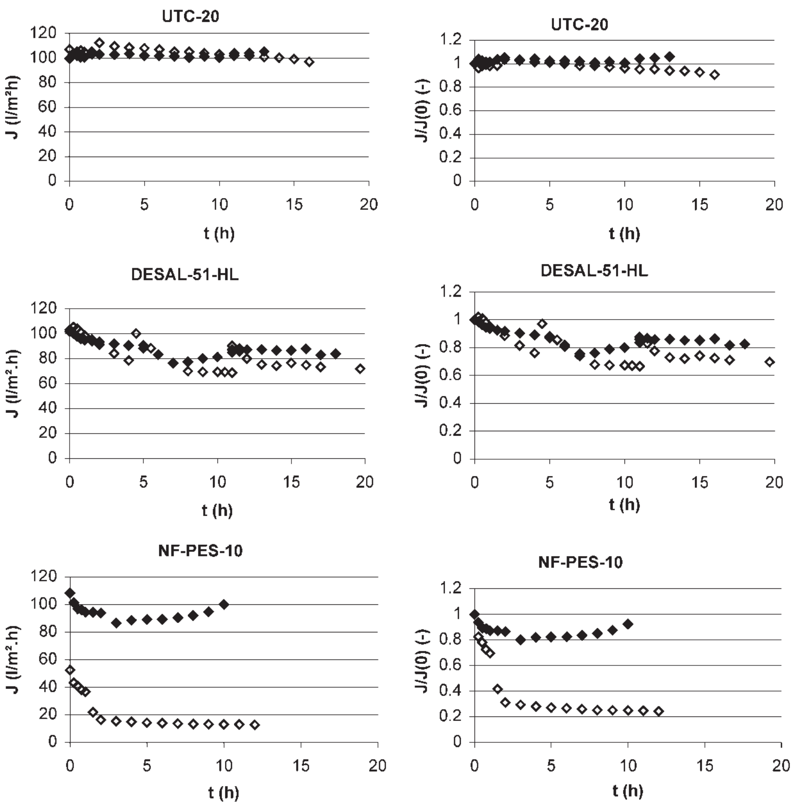

Figure 1. NF flux as a function of time with and without MF pretreatment, for NF membranes UTC-20, Desal 51 HL, and NF-PES-10, with bottle rinsing water as feed (open symbols: NF fluxes without MF pretreatment, filled symbols: NF fluxes after MF pretreatment of the feed solution).

that flux decline caused by particles is related to the surface roughness of the membrane[30] was tested by calculating the surface roughness from AFM images, given in Fig. 3. The average roughness, measured as the mean average of the peaks and valleys to the average position of the surface (for a $3 \times 3\,\mu m^2$ membrane surface area), was 2.86 nm for UTC-20, 14.7 nm for Desal 51 HL, and 0.69 nm for NF-PES-10. No correlation between flux decline and surface roughness was found; the largest flux decline effects correspond to the smoothest membrane (NF-PES-10).

An alternative explanation is that interactions between particles and the membrane material are caused by hydrophobic interactions; hence, the particles present in brewing waste water have an organic nature. Interactions

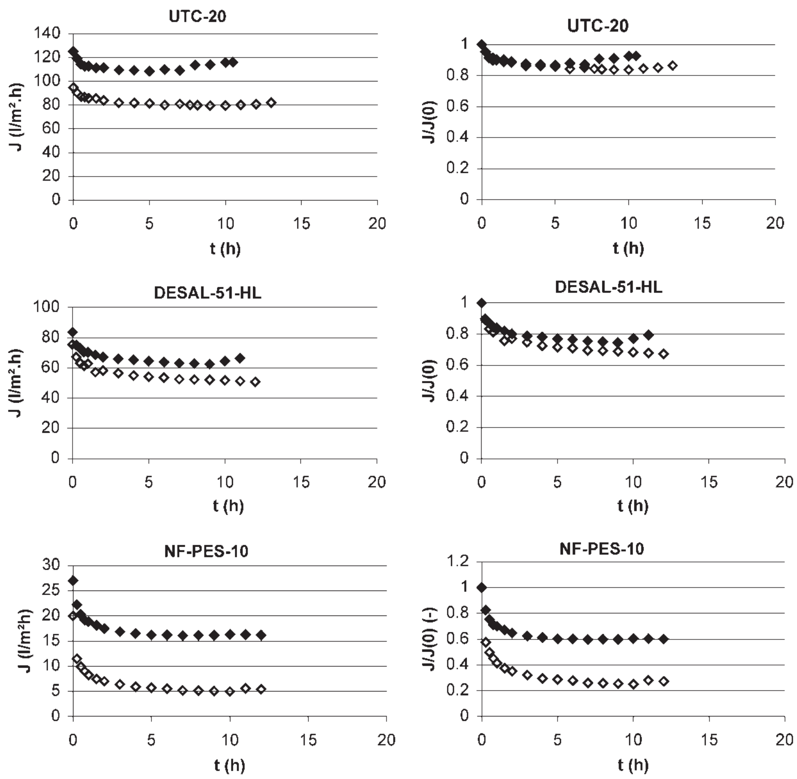

Figure 2. NF flux as a function of time with and without MF pretreatment, for NF membranes UTC-20, Desal 51 HL, and NF-PES-10, with rinsing water from fermentation tanks as feed (open symbols: NF fluxes without MF pretreatment, filled symbols: NF fluxes after MF pretreatment of the feed solution).

between these particles are thought to depend on the hydrophobicity of the membrane, resulting in a larger flux decline with the most hydrophobic membrane. Table 4 summarizes the contact angles water-membrane measured for the three NF membranes. A correlation was found between the flux decline and the hydrophobicity of the membrane: NF-PES-10 is a rather hydrophobic membrane, whereas Desal 51 HL and particularly UTC-20 are made of hydrophilic materials. The correlation between the water flux relative to the initial water flux (%) and the contact angle membrane–water is given in Fig. 4(a) ($r^2 = 0.9998$). It can be concluded that the hydrophobicity of the membrane is the most important factor determining membrane fouling by (organic) particles and dissolved organic compounds. Furthermore, Fig. 4(b) indicates

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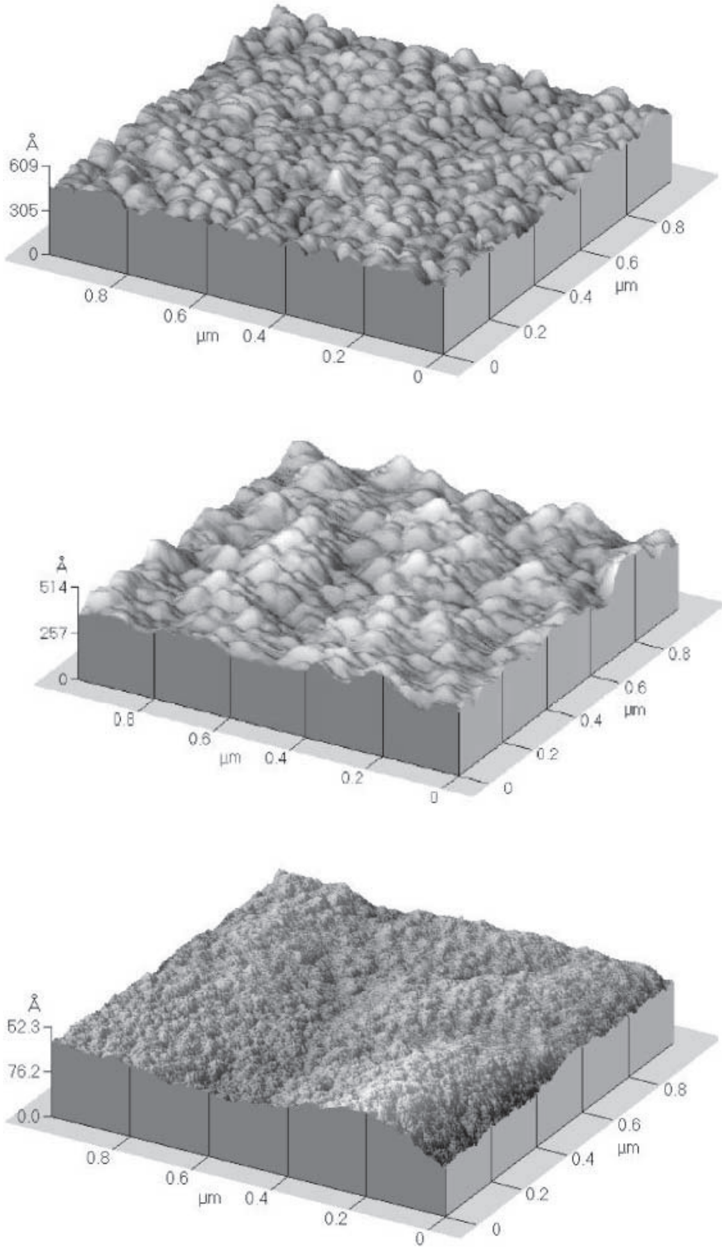

Figure 3. AFM images of NF membranes UTC-20 (a), Desal 51 HL (b), and NF-PES-10 (c).

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Table 4. Contact angles membrane-water measured for the three NF membranes UTC-20, Desal 51 HL, and NF-PES-10.

	Contact angle (°)	Standard deviation (°)
UTC-20	34.8	3.3
Desal 51 HL	45.5	3.6
NF-PES-10	74.6	4.7

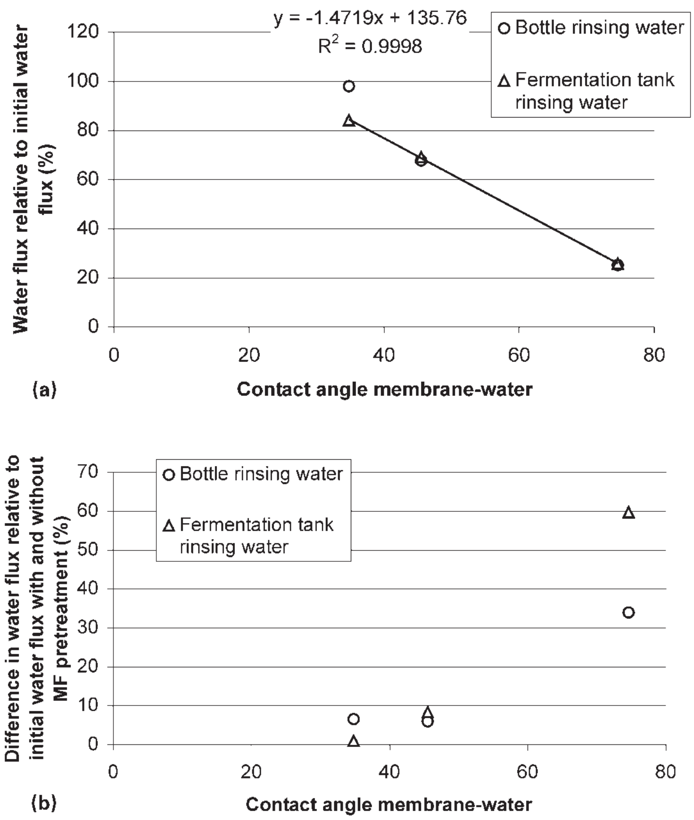

Figure 4. Correlation between water flux relative to the initial water flux (%) and the contact angle membrane-water, for NF membranes UTC-20, Desal 51 HL, and NF-PES-10, using bottle rinsing water and fermentation tank rinsing water as feed: (a) Total flux decline caused by particles and organic compounds in solution; (b) flux decline specifically caused by particles.

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that membrane fouling specifically caused by particles also increases when more hydrophobic membranes are used: the difference in flux decline with and without MF pretreatment is the largest for the most hydrophobic membranes.

(b) *Influence of the feed solution.* The effect of flux decline is more important for the rinsing water from the fermentation tank than for the bottle rinsing water. This is obviously due to the higher concentrations (COD and suspended solids) in the latter solution. The ratio of the pure initial water flux (J_0) and the pure water flux (Table 3) is generally lower for the fermentation tank rinsing water. Thus, the immediate fouling effect is larger when the concentrations in the feed solution are higher. Because this was observed for feed solutions with and without pretreatment, the immediate effect is related to fouling by particles as well as dissolved organic matter.

The ratio of the water flux after 8 hr of operation and the initial water flux indicates the effect on long terms, without taking the immediate effect into account. From a comparison between bottle rinsing water and fermentation tank rinsing water (Table 3), no general conclusions can be made. For UTC-20, the relative flux for the fermentation tank rinsing water is lower both with and without pretreatment. For NF-PES-10, the relative flux for the fermentation tank rinsing water is only lower after MF pretreatment (no particles present). No effect was observed for Desal 51 HL.

The conclusion from these observations is that the feed concentration has a significant influence on the initial water flux for all membranes studied, but the feed solution influenced the further flux decline as a function of time only for some membranes.

CONCLUSIONS

The influence of a MF pretreatment on the performance in NF depends mainly on the hydrophobicity of the NF membrane material, not on surface roughness. For hydrophilic membranes such as UTC-20, the influence of a MF pretreatment on the NF water flux is small. On the other hand, relatively hydrophobic membranes such as NF-PES-10 require a MF pretreatment for an optimal operation. If the pretreatment is left out, the water flux decreases dramatically because of membrane fouling by suspended solids.

A higher concentration in the feed solution has a negative effect on the water flux; further study of this effect revealed that it is mainly an immediate effect; only in some cases was a further long-term effect of fouling observed.

The MF pretreatment has only a small influence on the rejection characteristics; a small improvement of the quality of the final permeate is obtained because of the additional removal in the MF step.

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