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Ultrafiltration of biologically treated wastewater by using backflushing

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Abstract

Membrane technologies offer a possibility to further improve the quality of the wastewater. In this study a biologically treated wastewater from a municipal wastewater treatment plant was ultrafiltered with monotubular ceramic membranes of nominal pore size 20 and 50 nm. Backflushing was introduced with different levels of frequency and duration to reduce the formation of fouling during filtration. The results showed good retentions of main wastewaters components as BOD_5, COD, TKN, TP, total coliform and TDS. Moreover with membrane of nominal pore size 20 nm the results showed that the higher permeate flux was obtained when backflushing was carried out for 0.5 s every 1 min without an excessive worsening of permeate quality (improvement of flux about 9%). With membrane of nominal pore size 50 nm, backflushing caused a higher improvement of flux (17%) but also a decrease in permeate quality. The increase in permeate flux decreased slightly the retention of main quality parameters with a small contamination of the permeate with total coliform.

Keywords: Ultrafiltration; Wastewater; Backflushing; Fouling; Ceramic membrane

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1. Introduction

Membrane processes can be utilized to meet the challenge of increasingly stringent drinking water regulations, to produce high purity water as well as to develop new water resources beyond those traditionally used. Typical applications include: drinking water purification, municipal and industrial wastewater treatment applications, brackish water and seawater desalination, ultrapure water for industries, boiler feed for power stations, process water for food industry and wastewater reclamation and reuse. Membrane filtration can be also used as a good technique for water disinfection. Experimental studies carried out on natural and synthetic surface waters

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showed that the microbial load was completely removed with ultrafiltration membrane [1].

Membranes are providing more viable and cost-effective opportunities for water reclamation. Cooling water makeup, irrigation, fire protection and process washing/cleanup are only a few of the potential uses which are gaining popularity [2]. The principal advantages of membrane processes when compared to other separation processes are low energy consumption, simplicity, environmental friendliness and high quality product independently on feed quality.

As for municipal wastewater treatment, ultrafiltration membranes have been used for secondary and/or tertiary effluent purification, with good rejections of many wastewater substances; however, few data are available on single molecule rejections and a good agreement on researchers on membrane performance has been not found. Table 1 reports the results of a literature analysis on different retentions found by using ultrafiltration membranes (cut-off in the range 1–100 nm): the analysis of the table confirms the uncertainty of the results at disposal. Moreover, Table 1 shows that even with membranes with the same nominal pore size, retentions are very different; as for example, total phosphorus retentions range from 26% to 78% for the same cut-off of 4 nm, while BOD_5 percentage removals are in the range 18% to about 90%.

The main problem in membrane filtration processes is membrane fouling which causes a decrease in permeate flux with time and a limitation of the separation efficiency. The performance of membrane in crossflow filtration is strongly influenced by the build up of a fouling layer that finally may completely plug the porous membrane surface. Fouling occurs when some contaminant coats the membrane surface and decreases the rate of water transport through the membrane. As a result, greater pressure (i.e., energy) is required to produce the same amount of water. Common types of fouling include colloidal deposition, scaling, biofouling, and organic adsorption.

It is possible to classify the methods useful for reducing concentration polarization and fouling effect [3] as follows:

- chemical cleaning methods including strong acid and basic solutions or oxidizing agents;
- physical methods such as backflushing and the use of turbulence promoters;
- hydrodynamic methods related to module design.

A way to minimize fouling is to use an in situ cleaning technique such as backflushing or backpulsing. These techniques are particularly effective in minimizing external fouling, since the external fouling layer is lifted off the membrane by the reverse flow and swept out by the cross flow. Should severe pore plugging occur, backpulsing or backflushing will most likely be ineffective in preventing precipitous flux decline. This type of irreversible fouling may only be corrected by chemical cleaning [4].

In these techniques the permeate is periodically forced back through the membrane in the reverse direction to normal permeate flow in order to flush out the accumulated material from the membrane pores and the membrane surface. The reverse flow can be realized by using pressurized air, water or permeate. Depending on the frequency and the time of the flow reversal, we can have backflushing, backpulsing or backshock. In particular in a backflushing cycle, the permeate flow is applied through the membrane in the reverse direction to the filtration for a few seconds once in every several minutes or longer, leading to the removal of the deposited gel layer. The optimization of backflushing parameters is important because if pulses are too weak or infrequent, the membrane will not be adequately cleaned while too high frequency pulsing and too long pulses cause an excessive lost of permeate. Therefore it is advantageous to

Table 1
Percentage retentions of ultrafiltration membranes for municipal wastewaters

	Alonso et al. [5]	Ravazzini et al. [6]	Kim et al. [7]	Tchobanoglous et al. [8]	Ahn et al. [9]
Nominal pore size (nm)	4	30	Not specified	10	4
BOD_5	46	43	18	87.5	—
COD	81	40	16	78.8	70
Total phosphorous	26	15	26	—	78
Total nitrogen	12	10	20	—	33
Total coliforms	100	—	—	—	—
Electrical conductivity	—	18.8	18	—	—

optimize backflushing condition to maximize net flux.

Several researchers have observed net flux maxima with varying backflushing duration, frequency and pressure. Ramirez and Davis [10] demonstrated the effectiveness of rapid backpulsing in enhancing the permeate flux in crossflow microfiltration of a wastewater. They observed flux improvements from three times to over ten-times for suspensions of bentonite clay of various concentrations in water. During filtration of oil/water emulsions through ceramic membranes, Srijaroonrat et al. [11] obtained an optimum forward filtration time of 1 min and an optimum backflush duration of 0.7 s. Sondhi et al. [12] achieved up to a five-times increase in flux with backpulsing in experiments filtering $Cr(OH)_3$ suspensions through porous alumina ceramic membranes; they used an optimum forward filtration interval of 1.5 min and an optimum backpulse duration of approximately 1 s. During filtration experiments for dilute yeast suspensions with the 0.2 μm membrane, Sondhi and Bhave [13] found that high flux can be sustained at backpulse interval of 1 min. In the absence of backpulse the flux decreased rapidly in the first 15 min of filtration. Laitinen et al. [14] studied the effect of backflushing on ultrafiltration of board industry wastewaters. They found that backflushing can improve the flux of 11–28% when done for 1 s every minute at a pressure of four bars from the permeate side while the retentions were slightly lower than without backflushing. More recently, Mores and Davis [15] studied the microfiltration of yeast suspension; they experimented an optimum backflush time of t_b= 0.5 s.

In this study, in order to prevent or minimize the deposition of foulants on the surface of the membrane and thus to maintain high permeate fluxes during the operation, the backflushing technique was introduced to improve the efficiency of a ceramic membrane in the ultrafiltration of a biologically treated municipal wastewater. Backflushing has been achieved by using small amounts of permeate through the membrane. The permeate flux was increased by varying the time between two backflushes (t_f) and pulse length (t_b) in order to investigate the relative importance of these two parameters. The backflushing duration and frequency were varied for fixed values of the transmembrane pressure and cross-flow velocity to find the best backflushing conditions, which give maxima flux without worsening permeate quality excessively.

2. Materials and methods

2.1. Apparatus description

Experimental studies have been carried out in

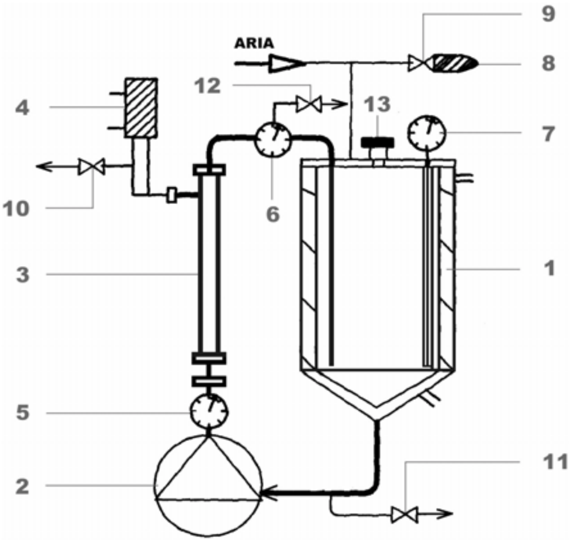

Fig. 1. Experimental apparatus employed for ultrafiltration; 1: jacketed feed tank; 2: pump; 3: membrane module; 4: back–flush device; 5, 6: manometers (0.4 bar); 7: temperature gauge; 8: muffler; 9, 10, 11: valves; 12: air purge valve.

a tangential flow laboratory pilot plant Membralox® XLAB 3 (Exekia, Bazet,France) with a single tube ceramic ultrafiltration membrane Membralox® Tl-70 (Fig. 1), whose characteristics are reported in Table 2. Recirculation pump gives a fixed tangential velocity of about 7 m/s. Temperature is controlled by the tank jacket, which is connected to a thermostat Crioterm 10–80. The plant is equipped with a backflush system BF3, controlled by an electrovalve (pressure 7 bar, re-injected volume 3 ml). Backflush intervals and lengths were regulated manually. The pore size of membranes used in experimental test were 20 and 50 nm. Operative conditions are reported in Table 2 as well.

Table 2
Summary of parameter values used for the backflush experiments

Membrane characteristics
Tubular ceramic
Material: zirconium oxide
Specific membrane area = 50 cm^2
Nom. Pore size: 20 nm, 50 nm
I.D. = 7 mm, L = 25 cm
Process parameters:
Forward filtration transmembrane pressure: 1.8 bar
Cross-flow velocity: 7 m s^{-1}
Backflush duration: 0.5, 1 s
Backflush intervals: 1, 2 min

Table 3
Average feed water characteristics

Analysis	Units	Value
pH	-	7.8
Electrical conductivity (T=20°C)	(µS/m)	400
Total dissolved solid	(mg/L)	300
COD	(mg/L)	12
BOD$_5$	(mg/L)	4.5
Total phosphorous	(mg/L)	1.2
Total nitrogen	(mg/L)	4
Total coliforms	(UFC/100ml)	250

2.2. Feed water characteristics

In this work, ultrafiltration experiments with and without backflushing were performed with biologically treated wastewater. The studied water is the effluent discharged from the wastewater treatment plant of Ponte Rosarolo in L'Aquila (Italy) after the chlorination section. Before ultrafiltration tests, water was microfiltered at 0.45 µm to remove suspended fine material causing an excessive fouling of the ultrafiltration membrane. This first microfiltration also reduces the feed total coliform concentration.

During filtration tests, samples were taken from the feed and permeate in order to evaluate the efficiency of the membrane to withhold pollutants. The average characteristics of the feed water are presented in Table 3.

The permeate was analyzed for the presence of total suspended solids (TSS), total dissolved solid (TDS), total nitrogen (N), total phosphorous (P), total coliform, biological oxygen demand (BOD_5), chemical oxygen demand (COD) and electrical conductivity. These parameters were measured according to the Standards Methods [16].

2.3. Testing procedure

The effect of backflush frequency and pulse length on the permeate flux was studied. The backflushing was achieved by using pressurized nitrogen to push small amount (3 ml) of permeate through the membrane. For each membrane flux decay and concentration tests were executed with and without backflush, investigating all combinations of backflushing frequency and pulse length.

The backflushing parameters and other operating conditions used in tests are presented in Table 4.

Table 4
Operating conditions during ultrafiltration tests with treated wastewater; $P_T = 1.8$ bar; $v = 7$ m/s; $T = 20$°C

	Backflush Parameters		Membrane
	Frequency (min^{-1})	Time (s)	Nominal pore size (nm)
Test 1	—	—	20
Test 2	1	0.5	20
Test 3	2	0.5	20
Test 4	1	1	20
Test 5	2	1	20
Test 6	—	—	50
Test 7	1	0.5	50
Test 8	2	0.5	50
Test 9	1	1	50
Test 10	2	1	50

The testing procedure for the evaluation of the effect of backflushing on permeate flux was the following: at the beginning of each test, distilled water flux was measured for different values of transmembrane pressure to control if the membrane was clean. After this measurement, the feed tank was drained, refilled with the feed water and the backflush was stored for a particular combination of frequency and pulse length. During the flux decay tests, the transmembrane pressure was adjusted to 1.8 bar and temperature was controlled by the water jacket and kept constant to the value T=20°C.

The used apparatus doesn’t allow to work in a continuous way, so during each flow decay test, filtration equipment was stopped periodically (every 500 ml of permeate) and permeate was recirculated back to the feed tank to ensure constant feed concentrations (steady-state conditions). In this way for each combination of backflushing frequency and length, permeate flux was measured for 3 h manually and concentration tests were executed on feed and permeate samples.

After each test, the equipment and membrane were washed with alkaline detergents (P3-Ultrasil 25) and rinsed with distilled water until pH returned to the value of about 7. Chemical cleaning is necessary, in order to get outlet fluxes similar to those obtained with distilled water; the cleaning procedure is described in Table 5. After the washing procedure, membrane was soaked in a distilled water with little amount of hydrogen peroxide.

3. Results and discussion

Concentration tests were carried out to evaluate the membrane retentions towards different compounds present in municipal wastewaters; in order to investigate the effect of backflushing, two different levels for reverse filtration time (t_b) and forward filtration time (t_f) were studied with two membranes of nominal pore size (n.p.s) 20 nm and 50 nm. Moreover, flux decay tests were conducted with and without backflush to quantify

Table 5
Cleaning procedure

Cleaning solution	Concentration	Backflush	P_T (bar)	Time (min)	Temperature (°C)
P3-Ultrasil 25	2%	—	1.3	30	room
P3-Ultrasil 25	2%	yes	1.3	30	room
Distilled water	-	—	1.3	30	room
Distilled water	-	yes	1.3	30	room

membrane fouling in different operating conditions.

3.1. Concentration tests

Results obtained without backflush (see Fig. 2 for membrane n.p.s. 20 nm and Fig. 3 for membrane n.p.s. 50 nm) show that ultrafiltration works well for the removal of COD, BOD_5, and total coliforms, while as for nutrients (total phosphorus and total nitrogen) retention is very variable, and for total conductivity is very low. This confirms that UF is a reliable technique for disinfection of wastewaters, while it doesn't work for dissolved salts. A comparison with other retentions available in literature and resumed in Table 1 shows a good agreement with some of the data (e.g. total coliforms, BOD_5 and nitrogen), but as a matter of fact a serious comparison is unreliable due to data dispersion and different membrane cut-off used. As concerns the effect of backflushing on retentions, these tests did not show a well-defined trend but it is clear that backflush slightly decreases the retention. This effect was more evident with membrane of 50 nm n.p.s. as a consequence of the higher flux obtained with backflush. For the membrane of 20 nm n.p.s., it seemed that an increase in backflushing time causes a stronger decrease in retentions while with the membrane of 50 nm n.p.s. higher backflushing time with higher backflushing intervals give a lower decrease in permeate quality.

However with both membranes, retentions appeared to be lower when lower backflushing intervals and longer pulses were used (t_f=1 min and t_b=1 s). In particular with membrane of 20 nm n.p.s., backflushing conditions giving the lowest reduction of permeate quality were for t_f=1 min and t_b=0.5 s while with membrane of 50 nm n.p.s. these conditions shifted to t_f=2 min and t_b=1 s.

Retentions of substances obtained during concentration tests with and without backflushing are reported in Figs. 2 and 3.

The obtained results showed that without backflushing, ultrafiltration membranes removed completely microbial load (retention of 100%) while forcing back permeate flux during backflush, there was a contamination of permeate (total coliform retentions range from 96% to 99%).

The lower retentions caused by backflushing can be explained by considering that backflush removes the cake layer which retains some of the substances acting as a secondary membrane. When this secondary membrane layer is removed more material go through the membrane causing a decrease in permeate quality, as already observed in the ultrafiltration of board industry wastewater [17].

3.2. Flux decay tests

Relevant decreases of flux with time were observed with both membranes. After three hours of filtration without backflush, there was a flux

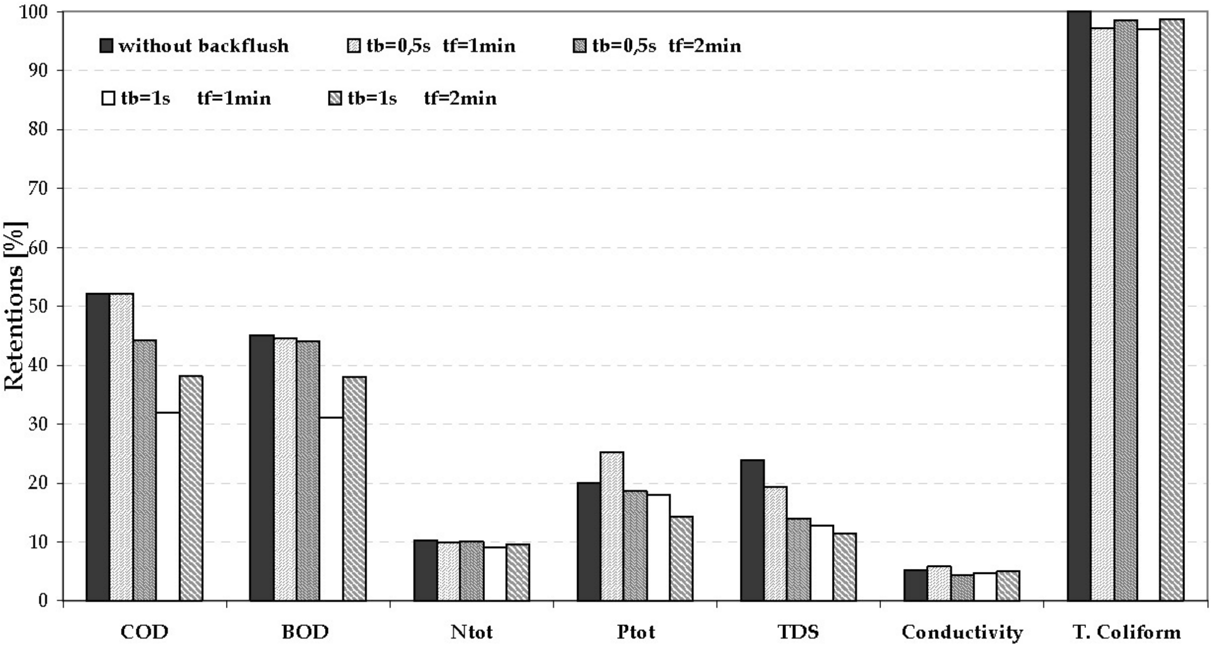

Fig. 2. Retentions of different substances with and without backflush for different levels of backflush frequency and length with membrane of n.p.s 20 nm; P_T=1.8 bar, v = 7 m/s; T = 20°C.

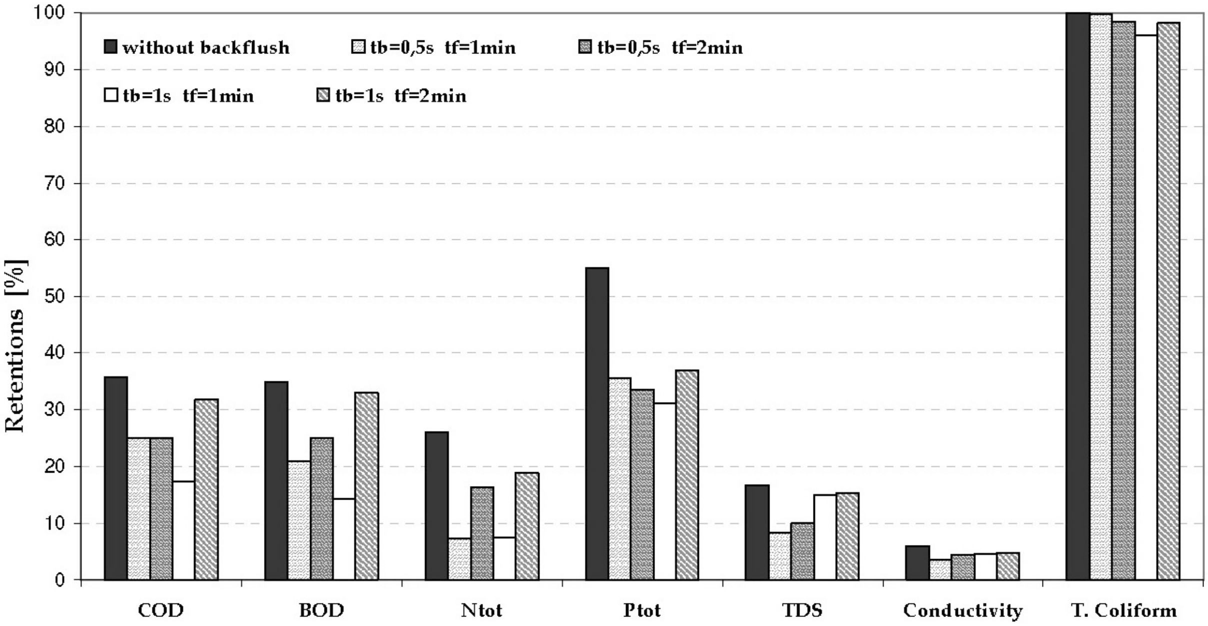

Fig. 3. Retentions of different substances with and without backflush for different levels of backflush frequency and length with membrane of n.p.s 50 nm; P_T = 1.8 bar, v = 7 m/s; T = 20°C.

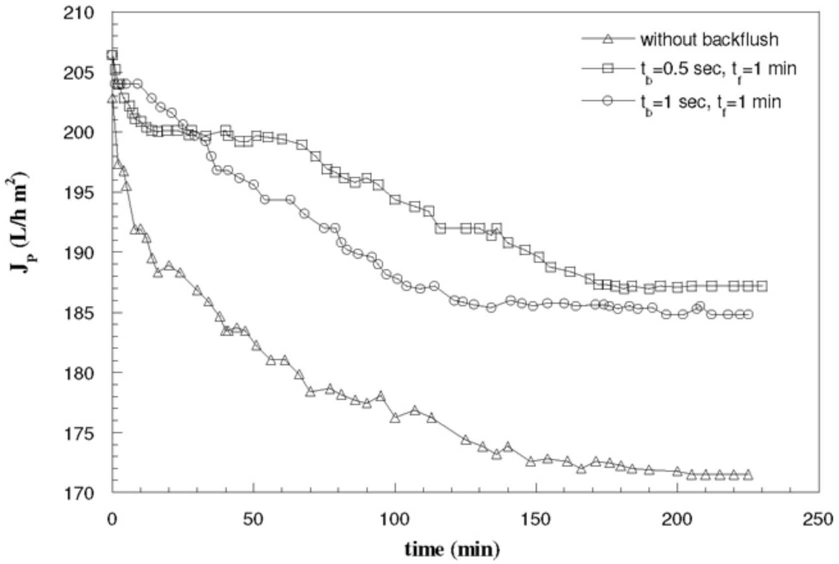

Fig. 4. Flux decay tests with and without backflush at t_f=1 min and t_b=0.5 s and 1 s with membrane of n.p.s 20 nm; P_T = 1.8 bar, v = 7 m/s; T = 20°C.

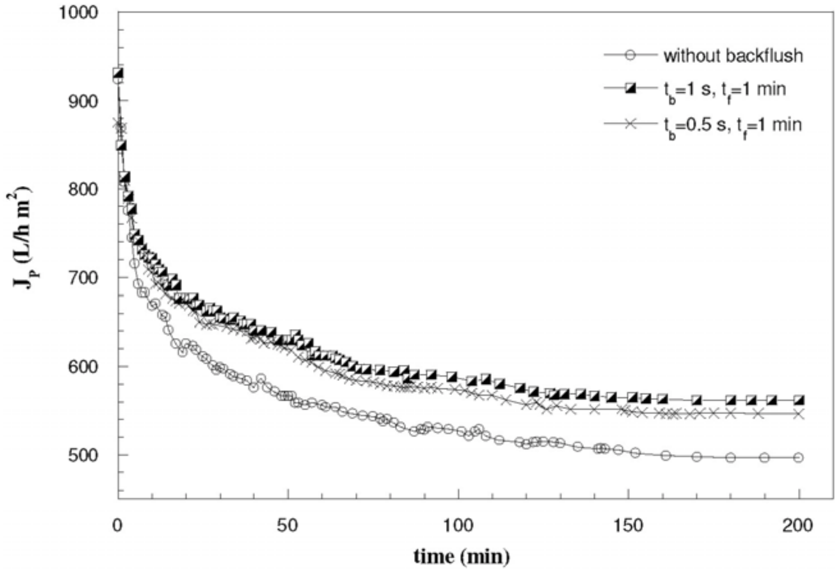

Fig. 5. Flux decay tests with and without backflush at t_f=1 min and t_b=0.5 s and 1 s with membrane of n.p.s 50 nm; P_T = 1.8 bar, v = 7 m/s; T = 20°C.

decay of 15% and 46% for membrane of 20 nm and 50 nm n.p.s., respectively (Figs. 4 and 5).

Obviously the backflush could not prevent the fouling totally, thus during flux decay tests the permeate flux also decreased.

With membrane of 20 nm n.p.s., flux decay tests showed that reverse backflush time played an important role in flux improvement when applied for shorter intervals and for a shorter time (Fig. 4). Since the amount of permeate used in each backflushing cycle was the same (3 ml), the shorter t_b resulted in a higher ability to push back the fouling effect at membrane surface, while the longer the t_f, the greater the development of the gel-layers on the membrane surface [11].

Table 6
Results of flux decay tests with and without backflush with membrane of n.p.s 20 nm and 50 nm; P_T = 1.8 bar; v = 7 m/s; T = 20°C

t_b (sec)	t_f (min)	Steady state flux (L/hm^2)	Reduction of initial flux (%)	Increase of steady state-flux (%)
Membrane 20 nm				
—	—	171.54	15.4	—
0.5	**1**	**187.2**	**9.3**	**9.13**
0.5	2	182.35	11.1	6.30
1	1	184.80	10.1	7.73
1	2	184.85	10.5	7.75
Membrane 50 nm				
—	—	480	46	—
0.5	1	546.12	37.6	13.77
1	**1**	**561.48**	**34**	**17**

As reported in Fig. 4 with membrane of 20 nm n.p.s., the maximum value of steady-state flux of 187.2 L/hm^2 was found for t_f =1 min and t_b= 0.5 s. The obtained results showed that when backflush was applied for t_b=1 s, backflushing frequency has no influence thus after three hours of filtration, steady-state fluxes were the same.

With membrane of 50 nm n.p.s. (Fig. 5) the backflushing effect was more evident because the larger the pore diameters, the greater the effectiveness of backflush [13]: flux improvements of 17% were obtained for t_f=1 min and t_b=1 s.

In this case, flux decay tests were tested for a single forward filtration time t_f (1 min) because the previous tests with membrane of 20 nm n.p.s., have showed that longer backflushing intervals couldn't control fouling adequately.

The obtained results for flux decay tests are reported in Table 6 in which the best combination of t_f and t_b are highlighted in bold.

The best backflush duration increases with increasing forward filtration time as observed by Mores et al. [18] and with increasing membrane nominal pore size (Fig. 6). Probably at lower cross-flow velocity, the effect of the backflushing on permeate flux enhancement would be higher

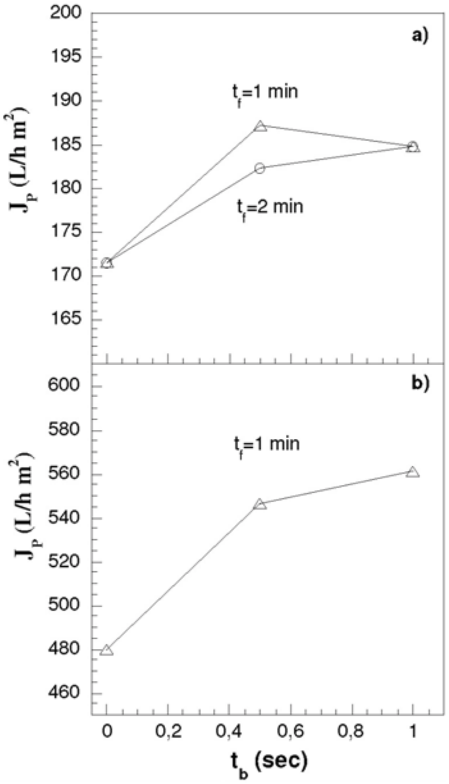

Fig. 6. Effect of forward filtration time t_f and backflush duration t_b on steady-state permeate flux with membrane of n.p.s 20 nm (a) and 50 nm (b); P_T=1.8 bar, v=7 m/s; T=20°C.

because backflushing is more effective for shear rates [19].

4. Conclusions

Experimental results carried out on biologically treated wastewater by using a laboratory scale apparatus, equipped with an ultrafiltration membrane, have shown that ultrafiltration is a reliable technique for secondary or tertiary wastewater filtration since good retentions of main wastewaters components as BOD_5, COD, total nitrogen, total phosporus, total coliforms and TDS were obtained. Moreover, backflush used as a cleaning procedure has a major effect on permeate flux and on permeate quality by decreasing fouling formation on membrane surface. When backflushing was applied, whatever the conditions and nominal pore size of membrane, the flux was found to increase. The obtained results show that with membrane of 20 nm n.p.s., the shorter the forward and reverse filtration times, the higher the permeate flux. The highest permeate flux was obtained for backflushing realized for 0.5 s every 1 min, with a quality permeate similar to that without backflush. For what concerns permeate quality, it worsens mainly with low backflushing intervals and long duration, this effect being more evident with membrane of n.p.s. 50 nm. For both membranes, conditions giving best flux also give the lower retention for total coliform. This problem could be resolved realizing reverse flux with a mixture of permeate and oxidizing agents. Since backflushing is more effective al low shear rates, should be interesting to use backflushing at lower cross-flow velocity obtaining also a decrease of operating costs. Further experiments should therefore use backflushing at lower reverse filtration time with the membrane of 20 nm n.p.s. while higher backflushing duration with the membrane of 50 nm n.p.s.

5. Symbols

t_f	forward filtration time, min
t_b	reverse filtration time, s
v	cross flow velocity, m/s
P_T	transmembrane pressure, bar
J_P	permeate flux, l/m^2 h

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