Desalination 304 (2012) 41–48

Contents lists available at SciVerse ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Effect of previous coagulation in direct ultrafiltration of primary settled municipal wastewater

Sebastián Delgado Diaz [a], Luisa Vera Peña [a,*], Enrique González Cabrera [a], Marta Martínez Soto [a], Luisa M. Vera Cabezas [b], Luis R. Bravo Sánchez [b]

[a] Department of Chemical Engineering, Faculty of Chemistry, La Laguna University, Av. Astrof, Fco. Sánchez s/n. 38200 La Laguna, Spain
[b] Department of Chemical Engineering, Faculty of Chemistry, Central University of Las Villas, Carretera a Camajuaní km. 5.5 s/n, 54830, Santa Clara, Cuba

HIGHLIGHTS

► Direct ultrafiltration of municipal wastewater can be a feasible option for water reuse.
► Pre-clarification by coagulation permits a significant reduction in cake resistance.
► Cake was detached during backwashing, but only partially dispersed in the suspension.
► High efficiency of COD removal (81–95%) was achieved, regardless of the coagulant dose.

ARTICLE INFO

Article history:
Received 11 April 2012
Received in revised form 31 July 2012
Accepted 1 August 2012
Available online 28 August 2012

Keywords:
Dead-end ultrafiltration
Membrane fouling
Hollow-fibre
Wastewater treatment
Coagulation–flocculation
Primary effluent

ABSTRACT

Direct ultrafiltration of primary settled municipal wastewater can be a feasible option for water reuse in several applications by avoiding biological treatment. This paper discusses the effect of previous clarification by coagulation/sedimentation of raw wastewater on ultrafiltration performance and its relationship with the main operation parameters in dead-end mode (flux and backwashing duration). Reversible and residual membrane foulings in a hollow-fibre bench-scale unit were determined over a broad range of filtration parameters and coagulant doses used in the previous clarification of domestic wastewater. Moreover, results were also compared with those obtained by ultrafiltration of biologically treated effluent from a conventional WWTP. Reversible membrane fouling, which can be described by the cake formation model, seems mainly caused by the colloidal fraction of the wastewater. In fact, under optimal coagulant dosage, the cake resistance values (expressed by $\alpha\omega$, in m^{-2}) were similar to those obtained with the secondary effluent. Direct observation of the membrane surface suggests a solid accumulation in the vicinity of the membrane after backwashing, which could justify the increase in cake resistance during the initial filtration/backwashing cycles until steady-state conditions are reached. Residual fouling resistance decreased exponentially with the duration of backwashing, obtaining a limit resistance value which was independent of the coagulant dose. Moreover, high efficiency of chemical oxygen demand (COD) removal was achieved by the tested treatment train, regardless of the coagulant dose and the initial quality of the primary effluent.

1. Introduction

It is widely known that water resource scarcity is a great problem in many South European regions. Particularly important is the situation in the territories of the Spanish Mediterranean coast (Southern Basins, Júcar, Ebro and Catalonia) and some of the Balearic and Canary Islands. Also, it is a fact that, in these areas, the aquifers are in a critical condition as a result of over-exploitation [1]. As a consequence, wastewater reuse is a common practice in such regions and competent authorities encourage and promote it through legislation. In fact, large investments by the competent authorities of the Canary Islands have been made in regard to infrastructure treatment and reuse for the past two decades. Consequently, Canaries is one of the regions that reuse more water, proportionally to the reclaimed wastewater volume [1,2].

Spanish legislation relative to implementing reclaimed wastewater reuse (RD1620/2007) [3] is very demanding in terms of quality and health security, leading to the development and application of new technologies for wastewater reclamation and reuse. Besides the conventional treatment trains, specially activated sludge plants, many new alternative schemes are feasible options for specific applications. One of these promising technologies is direct membrane filtration, which has been used to treat different types of wastewater—greywater, manure wastewater and municipal wastewater, either raw or after primary sedimentation [4]. The main limitation for its

* Corresponding author. Tel.: +34 922318054; fax: +34 922 318014.
E-mail address: luvera@ull.edu.es (L. Vera Peña).

0011-9164/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.desal.2012.08.005

widespread application is the membrane fouling which decreased process productivity. Therefore, physical–chemical pre-treatments, such as coagulation, flotation, adsorption and ozonisation [5,6], may be added to enhance membrane performance and removal capability.

Ravazzini et al. [7] have suggested the capability of cross-flow ultrafiltration of municipal wastewater for reaching the quality criteria required for regenerated water reuse for agricultural irrigation. During short-term experiments, average fluxes of 120 and 160 l h^{-1} m^{-2} were obtained for raw sewage and primary effluent, respectively, with a filtration cycle of 10 min at cross-flow velocity of 2 m s^{-1} and at a constant TMP of 0.3 bar with 1 min of backflush. However, a deeper investigation in long-term experiments at a pilot-scale should be done in order to confirm the process sustainability.

Ahn and Song, [8] reported that the microfiltered effluent of low-strength wastewater from a resort complex can be used for secondary applications such as toilet flushing. For long term operation, even under subcritical filtration conditions (applied permeate flux of 20 l h^{-1} m^{-2} operating 10–2 min of suction mode), it was impossible to prevent membrane fouling by physical means, although, this fouling was completely removed by chemical cleaning.

Regarding the impact of pre-treatments on membrane performance, in line coagulation is thus preferred if the raw wastewater contains a large amount of colloids and settle-able fractions [9]. Pouet has reported that the previous chemical destabilisation of urban wastewater with $Al_2(SO_4)_3$ can reduce the hydraulic resistance of the membrane by a factor from 10 to 30 according to the wastewater nature [10]. Choo et al. [11] have showed that the addition of coagulants to textile wastewater substantially reduced the fouling of ultrafiltration membranes. Nevertheless, the complexity of the interaction of coagulated particles with membrane makes difficult to pre-establish the optimal coagulant type and dose due to fluctuations in influent characteristics. Also, it has been reported that larger dosages of coagulant can produce a sharp increase of membrane fouling [12]. In addition, even though it is known that the resulting cake layer protects the membrane from internal fouling; it also provides additional resistance to permeation which requires frequent physical cleaning cycles to maintain process productivity. Even though the treatment train is capable of removing coagulated particles by a simple clarification, it seems necessary to test this.

Therefore, the aim of this work is to evaluate the effect of a previous coagulation/sedimentation in direct ultrafiltration of primary settled municipal wastewater for water reuse application. The study has focused on the effect of main operation parameters (coagulant dose, permeate flux and backwashing duration) on membrane fouling, and turbidity and COD removal. Finally, results were also compared with those obtained by ultrafiltration of biologically treated wastewater.

2. Materials and methods

2.1. Bench filtration unit

The experiments have been developed in a laboratory membrane unit, ZW1, which was provided by GE Zenon Environmental, equipped with ZeeWeed® hollow-fibre membranes with 0.03 µm rated pore diameter and 3.4 mm external diameter, assembled vertically. ZeeWeed® is a composite membrane with a polymeric external surface supported by a macroporous polymer. The total filtration area is 0.093 m^2.

The ZW1 unit was operated in closed loop, i.e. at constant concentration, through a 6 litre-tank filled to 21 cm water height. Filtration was achieved by drawing permeate through the membranes under a slight vacuum generated by a pulsing membrane pump Prominent® AlPa-0612 (0–15 l h^{-1}) and measured by an analogical manometer located just after the membranes, on the aspiration line. The hydrostatic pressure was taken into account by compensating the gauge with the atmospheric pressure, just before starting the pump. All the experiments were carried out at constant permeate flux, monitoring transmembrane pressure as a function of time. The unit operated in two modes: filtration and backwashing (Fig. 1). Each filtration cycle finished when a pre-established transmembrane pressure (36 KPa) was reached, beginning the backwash cycle immediately afterwards. The trials were carried out within a period of a few hours, depending on the experimental conditions.

2.2. Feedwater characteristics

Feedwater samples with a wide range of physicochemical characteristics were collected during June 2010 and March 2011 from a conventional wastewater treatment plant (WWTP) located in Santa Cruz de Tenerife (Canary Islands, Spain). It includes grit removal and a primary sedimentation stage followed by activated sludge treatment (designed only for organic matter removal, without nitrification) at a volumetric loading rate of about 1.6 kg BOD_5 m^{-3}·day^{-1}. The main characteristic parameters of the feedwater samples are given in Table 1.

2.3. Operating conditions

The previous coagulation–flocculation treatment was carried out in a conventional jar-test unit (Model JLT6, Velp Scientifica) with aluminium polychloride AlP (10% Al_2O_3, 9.2% Cl^- and 2.7% SO_4^{2-}) as coagulant. The selection of AlP was based on previous coagulation–flocculation studies [13]. The pH of feedwater was not adjusted during the pre-treatment, therefore the system was operated at ambient water conditions (pH = 7–8). This pre-treatment included a 1-min coagulation step at 266 s^{-1} velocity gradient, followed by 9 min flocculation at 50 s^{-1} and 15 min sedimentation. The clarified supernatant was then stored at a constant temperature of +4 °C, and then brought to room temperature (18–20 °C) prior to use as feedwater in the filtration runs.

Filtration tests were conducted with different physicochemical characteristics obtained after coagulation/sedimentation step. The aim of the experiments was to assess the effect of J on cake fouling; therefore different values of J were tested (from 2.5 to 39 l h^{-1} m^{-2}). For this purpose, the TMP evolution during single filtration cycle was registered and the resulting fouling rate was calculated. Afterwards, the dynamic behaviour of the cake formation and the residual fouling development over subsequent filtration/backwashing cycles was studied. Also, different values of backwashing duration (t_b) were tested, ranging from 20 s to 85 s, at a fixed J of 23 l h^{-1} m^{-2}.

After each filtration run, the membrane was chemically cleaned by immersing the module in a 500 mg l^{-1} sodium hypochlorite solution at 35 °C. To assess the cleanliness of the membrane, its resistance was measured by filtering tap water before each feedwater filtration run. Resistance values were in the range of 6.6 to 8.8 · 10^{11} m^{-1}.

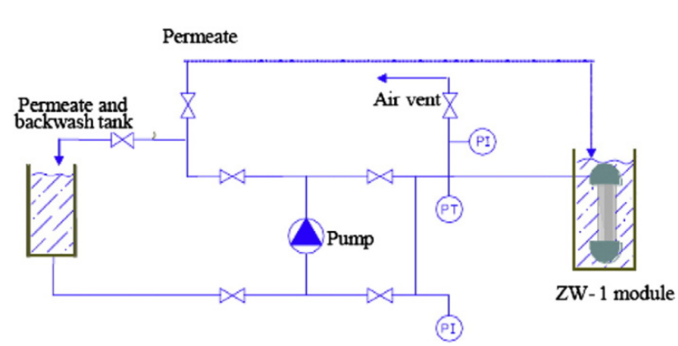

Fig. 1. Experimental filtration unit, ZW1.

Table 1
Main feedwater characteristics.

WWTP sample	Type of water	Turbidity NTU		COD mg l^{-1}		COD_s [a] mg l^{-1}		pH	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range
PE (June 2010)	Primary effluent	128	105–145	368	317–416	186	136–191	7.8	7.6–8.2
PE (March 2011)	Primary effluent	174	150–225	488	400–591	232	158–367	7.9	7.9–8.1
SE (June 2010)	Secondary effluent	7.7	3.1–15	71	55–86	56	51–66	7.8	7.7–7.9
SE (March 2011)	Secondary effluent	17	15–19	82	79–85	58	55–60	8.0	7.8–8.2

[a] Samples were filtered through paper with a nominal pore size of 0.45 μm.

2.4. Analytical methods

COD was determined according to the Standard Methods [14]. For determining soluble COD fraction (COD_s), the samples were filtered through a filter paper with a nominal pore size of 1 μm. Turbidity was measured with a HACH 2100N turbidimeter and pH with a WTW inoLab Level 1 pH metre. Measurements of particle size distribution and the zeta potential were performed with a Zetasizer Nano ZS.

3. Results and discussion

3.1. Turbidity and COD removal by combined processes

The effectiveness of clarification achieved by a combined coagulation/sedimentation process on different types of wastewaters was assessed in terms of turbidity and COD removal at the original water pH (Fig. 2). For the coagulation/sedimentation process, turbidity removal ratio (Fig. 2a) increased with Al^{3+} concentration up to a plateau value at 0.14–0.16 mM. In fact, the increase of turbidity from 128 NTU (June 2010) to 174 NTU (March 2011) caused a significant decrease in the removal efficiency (from 93 to 83%). The asymptotic approach to a steady value has been related to a sweep coagulation mechanism by which colloids are absorbed onto an aluminium hydroxide precipitate [15]. The Al^{3+} dose did not considerably affect the zeta potential, which varied between −15.4 and −12.2 mV. In addition, Fig. 3 displays the particle size distribution at different coagulant doses. With a coagulant dose of 0.04 mM, particle sizes of 4000–7000 nm were effectively coagulated and removed. At higher doses, the supernatant showed a bimodal distribution, which gradually shifted toward smaller sizes. All these facts rule out a mechanism of particle destabilisation by charge neutralisation. Accordingly, it can be concluded that sweep coagulation is the dominant mechanism by which colloids are destabilised in the system. Therefore, for a coagulant dose of 0.14–0.16 mM, the coagulation/sedimentation process is an effective pre-treatment to at least partially remove the colloidal matter from settled raw wastewater and reach an effluent in the range of 7–30 NTU depending on the initial turbidity. These values are not much higher than those (5–15 NTU) reported for a conventional activated sludge treatment [16] or those obtained experimentally for the secondary effluent of the Santa Cruz wastewater treatment plant (SC WWTP) (Table 1).

These results were also compared to those obtained by the combined process (coagulation/sedimentation and ultrafiltration of clarified water) (Fig. 2a). The obtained data showed an excellent removal rate (>99%), regardless of the coagulant dose. As expected, ultrafiltration increased turbidity removal, which became significant at lower Al^{3+} doses.

Analysis of COD removal for the coagulation/sedimentation process shows a similar trend (Fig. 2b) as that found for the turbidity

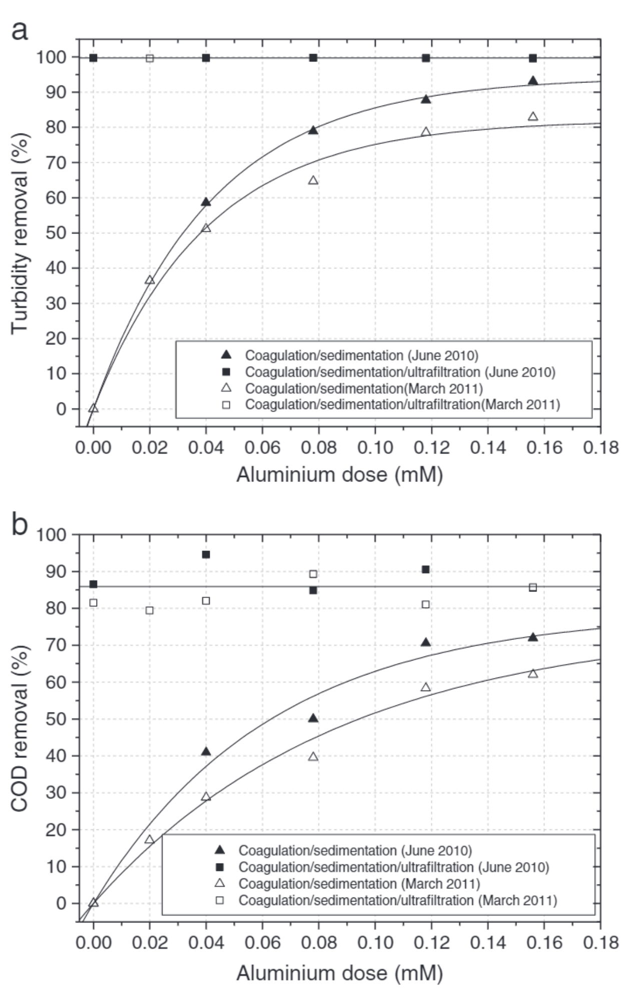

Fig. 2. Effect of coagulant dose on turbidity (a) and COD (b) removal for different treatment trains. SC WWTP primary effluent, $J = 23\ l\ h^{-1}\ m^{-2}$.

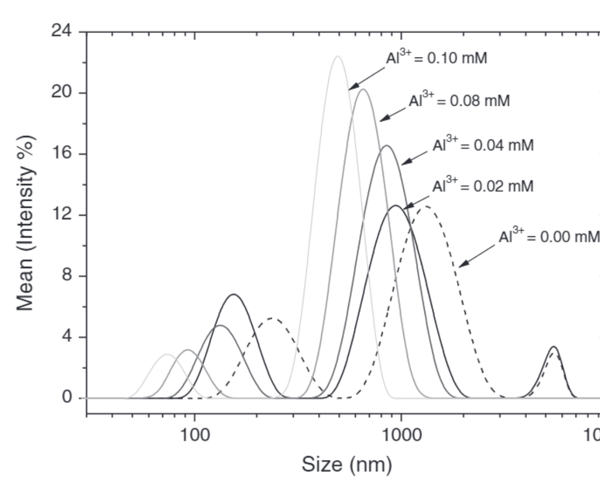

Fig. 3. Particle size distribution in wastewater samples at different coagulant doses. SC WWTP primary effluent (SC_PE) June 2010.

removal, reaching maximum values which varied from 62 to 71%, depending of the inlet COD. It resulted in an effluent COD concentration of 90–210 mg l^{-1}. As expected, an increase in removal was obtained by adding the ultrafiltration step, which ranged from 80 to 95%. Consequently, a COD abatement increase of 5–19% was observed, mainly attributed to the microcolloidal and soluble fraction (measured as COD concentration before filtering the samples through filter paper with a nominal pore size of 0.45 μm (COD_s)) corresponding to the primary effluents (186–232 mg l^{-1}, see Table 1). Notwithstanding, low COD concentrations were achieved in the permeate (18–72 mg l^{-1}). It is important to note that variation in the permeate COD may not be attributed to feedwater COD: it is more dependent on the microparticles and soluble chemicals contained in the feedwater. Therefore, it is expected that increasing COD concentration in the filtration tank does not increase the value of permeate COD. This is also confirmed by Ahn and Song for the microfiltration of domestic wastewater [17], whose unit was operated for more than 220 days with extreme variation in influent COD (from 10 to 622 mg l^{-1}) while the permeate COD remained less than 30 mg l^{-1}.

3.2. Membrane fouling characterisation

In wastewater ultrafiltration with hollow-fibre, it is usual to operate under consecutive filtration/backwashing cycles with a pre-fixed duration of cycles. Recently, several authors have proposed an alternative by monitoring permeability [18]. Based on this approach, the experimental unit was backwashed when a pre-established transmembrane pressure was reached (36 kPa) (Fig. 4).

Transmembrane pressure (*TMP*) evolution was used to characterise membrane fouling. Two types of fouling were assessed: reversible fouling r_f (i.e. cake deposit) and residual fouling R_f (not removed by backwashing) [19].

In all the runs the *TMP* linearly increased with elapsed time: Fig. 4 displays an example. This behaviour could be described by the deposit model with an incompressible cake layer [20].

Despite the simplicity of the cake model, which assumes the building of a uniform cake, the acceptable accuracy and consistency of the experimental data with the model makes it a valid approach. According to the model, the *TMP* evolution in each cycle can be described by the following equation [21]:

$$TMP = TMP_0 + \mu\alpha\omega J^2 t \tag{1}$$

where TMP_0 is the initial transmembrane pressure, μ the solvent dynamic viscosity, α the specific cake resistance, ω the solid concentration in the cake per unit filtrate volume, J the permeate flux and t the elapsed time. The product $\alpha\omega$ can be used to quantify cake fouling.

Finally, TMP_0 can be related to the hydraulic resistance (R_t) using Darcy's law:

$$TMP_0 = J \cdot \mu \cdot R_t \tag{2}$$

where R_t can be considered as the sum of the initial membrane resistance (R_m) and an additional resistance associated with residual fouling phenomena after physical cleaning (R_f):

$$R_t = R_m + R_f. \tag{3}$$

The initial transmembrane pressure increased after each backwashing, which confirms irreversible fouling. However, most of residual fouling was manifested in the first cycle, the increase being much lower in the following cycles.

3.3. Reversible cake fouling

The slope of *TMP* against the elapsed time is proportional to the solvent viscosity, specific cake resistance, solid concentration of the cake and to the square of permeate flux, following Eq. 4:

$$\frac{dTMP}{dt} = r_f = \mu \cdot \alpha \cdot \omega \cdot J^n \tag{4}$$

where n is 2 for cake filtration theory.

Therefore, the coherence between the obtained experimental data and the cake filtration model was analysed by measuring the effect of the coagulant dose and the permeate flux on cake fouling rate during the first filtration cycle. Afterwards, the dynamic behaviour of the cake formation over subsequent filtration/backwashing cycles was studied.

3.3.1. Effect of the coagulant dose and the permeate flux

As an example, Fig. 5 displays the results for the SC WWTP primary effluent at different coagulant doses. The specific parameters fitting the SC WWTP primary and secondary effluents by a non-linear regression are summarised in Table 2. The n values range between 1.95 and 2.05, confirming that the cake filtration model adequately describes the experimental behaviour. Also, $\mu\alpha\omega$ decreases with coagulant dose for all the types of feedwater.

According to the model, the product $\alpha\omega$ depends on the cake properties and, consequently, mainly on the feedwater characteristics.

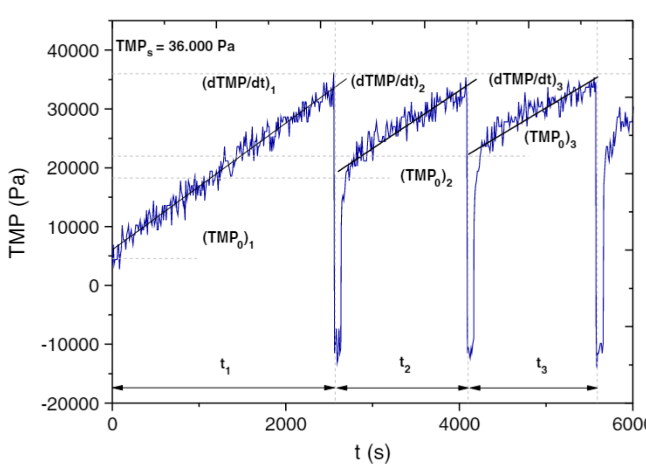

Fig. 4. Typical experimental evolution of TMP against elapsed time under consecutive filtration/backwashing cycle. SC WWTP primary effluent (March 2011). $J = 23$ l h^{-1} m^{-2}. $t_b = 85$ s. $Al^{3+} = 0.16$ mM.

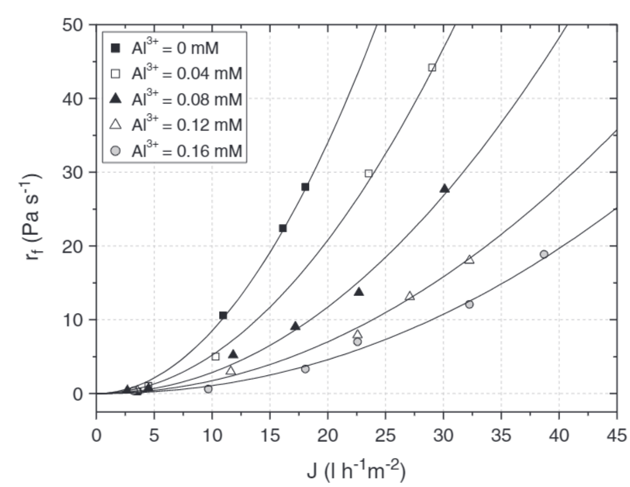

Fig. 5. Cake fouling rate against permeate flux at different aluminium concentrations. Feedwater: SC WWTP primary effluent (June 2010).

Table 2
Fitted parameters of the cake filtration model for different feedwaters and coagulant dose.

WWTP sample	Coagulant dose mM of Al^{3+}	$\mu\alpha\omega$ kg s m^{-4}		n	
		Mean	sd	Mean	sd
SC primary effluent (June 2010)	0	0.099	0.003	1.95	0.009
	0.040	0.052	0.014	2.01	0.082
	0.080	0.026	0.014	2.04	0.161
	0.120	0.017	0.010	2.01	0.187
	0.160	0.012	0.004	2.01	0.109
SC secondary effluent (June 2010)	0	0.018	0.002	2.03	0.038
	0.010	0.016	0.003	2.00	0.060
	0.040	0.011	0.004	2.05	0.096

Moreover, it is also be related to the coagulant concentration in the pre-treatment process. Fig. 6 displays $\alpha\omega$ against the coagulant dose for SC WWTP primary and secondary effluent, which can be well described by an exponential equation. As expected, the effectiveness of coagulant dose was higher for the primary effluent than for the secondary. The optimum doses were in the range 0.14–0.16 mM and 0.03–0.04 mM for the primary and secondary effluents, respectively. Moreover, results showed that, at a high coagulant dose for the primary effluent, the obtained $\alpha\omega$ values were quite similar to those obtained for the secondary effluent.

Consequently, cake fouling can be described in terms of the physicochemical characteristics of the feedwater, essentially linked to its turbidity. In all cases, $\alpha\omega$ linearly increases with turbidity (Fig. 7a and b). The slope of the product $\alpha\omega$ against turbidity ($\alpha\omega/T$) was $9.5 \cdot 10^{12}$ m^{-2} NTU^{-1} for the primary effluent with a 4% error. However, the value obtained from the secondary effluent was $3.8 \cdot 10^{13}$ m^{-2} NTU^{-1} with a 5% error. This variation could be related to the different nature of the feedwater. At these low turbidity values (<10 NTU), the effect of other organic substances (such as polysaccharides and proteins), which cannot be removed by pre-coagulation/sedimentation [22], should probably be taken into consideration along with turbidity. Furthermore, to validate the applicability of the cake formation model, experimental data obtained from the secondary effluent membrane filtration were compared, under the same experimental conditions, with the SC WWTP secondary effluent pre-filtered through 1.00 and 0.45 μm filter papers. This pre-filtration removed the particles and macro-colloids from the liquid-phase. Fig. 7b shows the similar trend in the fouling of clarified samples by coagulation and that caused by previously filtered secondary effluent (white and grey squares) which confirms the physical role of the $\alpha\omega$ product.

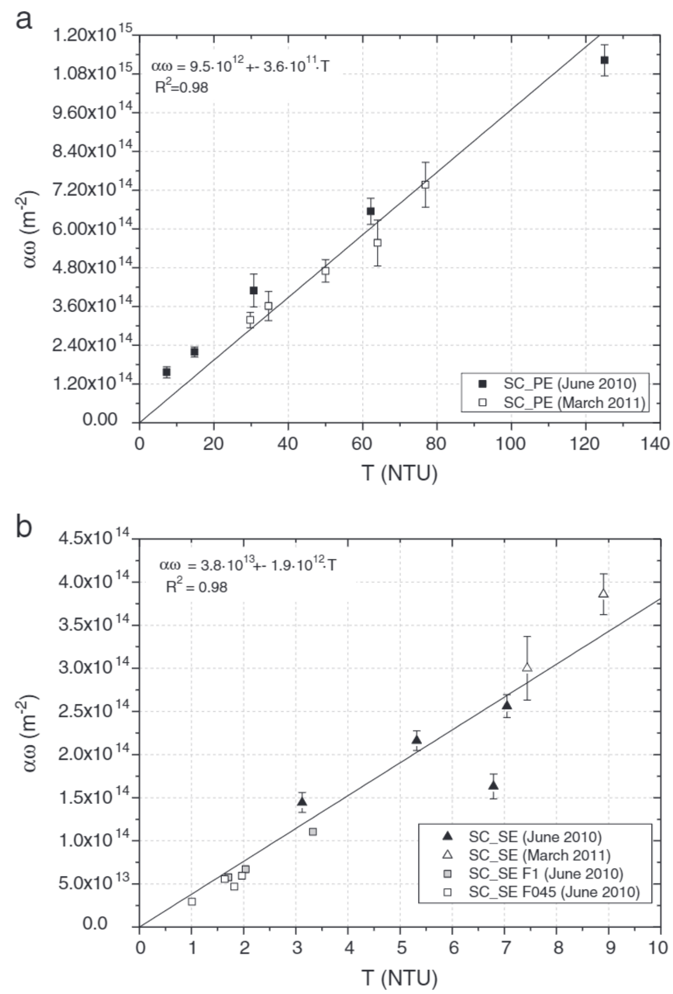

Fig. 7. $\alpha\omega$ against the turbidity of different feedwaters: (a) SC WWTP primary effluent (SE_PE); (b) SC WWTP secondary effluent (SC_SE) and filtered SC WWTP secondary effluent samples by 1.00 μm (SC_SE F1) and 0.45 μm (SC_SE F045).

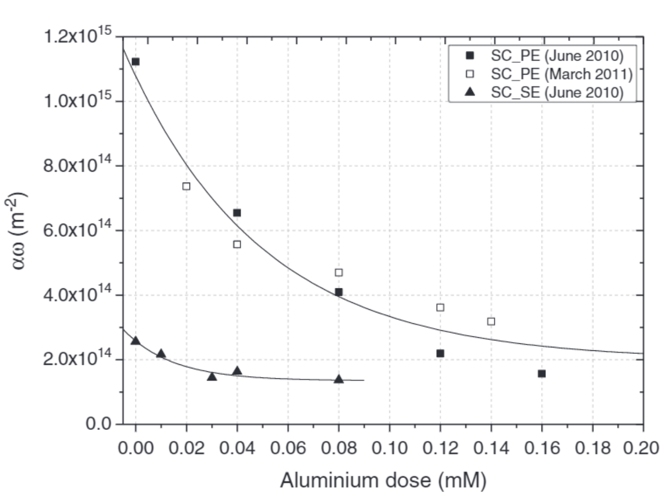

Fig. 6. $\alpha\omega$ against aluminium dose. Feedwaters: SC WWTP primary effluent (SC_PE) and SC WWTP secondary effluent (SC_SE).

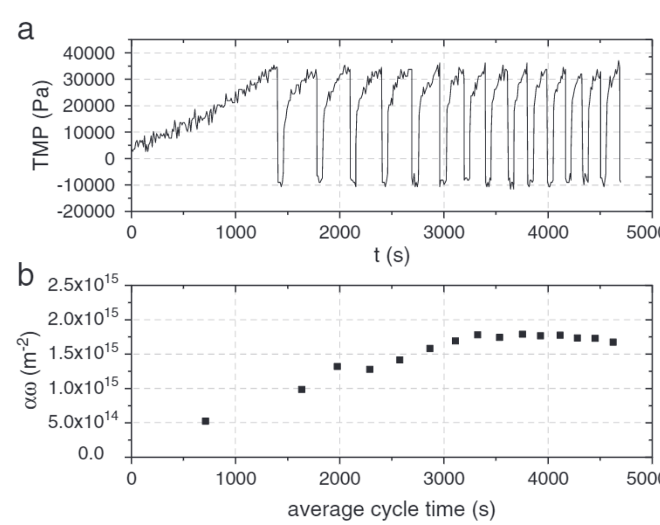

Fig. 8. a) TMP evolution against elapsed time under consecutive filtration/backwash cycles. b) $\alpha\omega$ against average cycle time. SC WWTP primary effluent (March 2011). $J = 23$ l h^{-1} m^{-2}. $t_b = 60$ s. $Al^{3+} = 0.16$ mM.

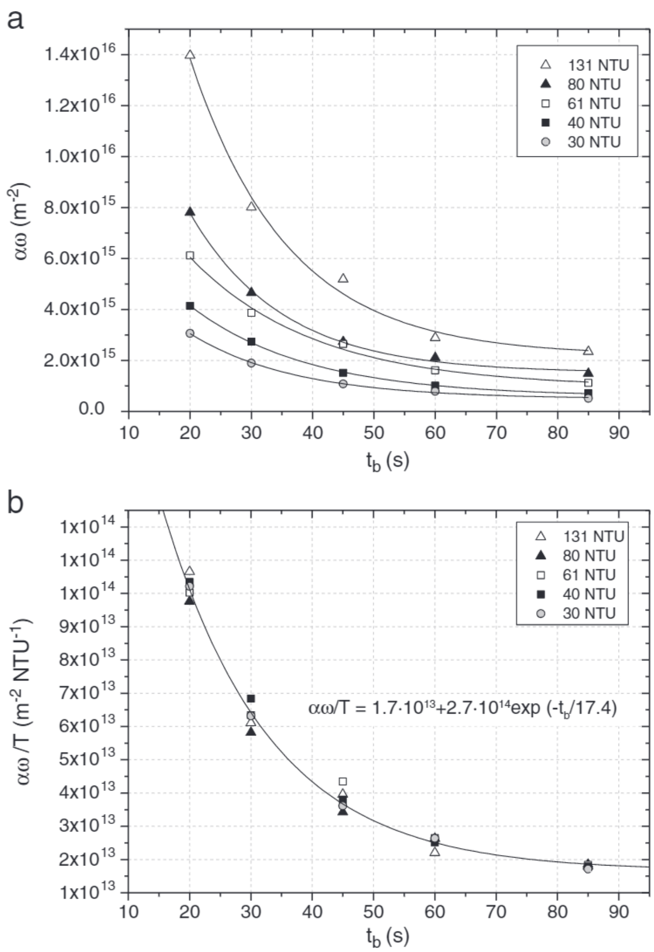

Fig. 9. Effect of the backwash duration on cake fouling: (a) $\alpha\omega$ against t_b; (b) $\alpha\omega/T$ against t_b, in ultrafiltration runs of SC WWTP primary effluent at different coagulant doses.

Finally, another concern should be taken into consideration: the dynamic behaviour in the system with the consecutive filtration/backwash cycles. As previously mentioned, results of *TMP* evolution during the operating time reflect this dynamic behaviour, where the duration of each cycle decreased until it reached a steady state after the initial cycles. Therefore, an increase in $\alpha\omega$ was observed in the first cycles (Fig. 8), this behaviour being more pronounced at higher turbidity values (>40 NTU). At steady state, the duration of the cycles was practically constant at a value that depends on feedwater characteristics and operating conditions, particularly on the backwash conditions.

Fig. 9a displays $\alpha\omega$ at steady state against backwash duration for the primary effluent. The $\alpha\omega$ decreases as backwash duration (t_b) increases. However, it shows that increasing the backwash duration time to over 60 s had no significant influence on decreasing $\alpha\omega$. There was a similar effect of backwash time on $\alpha\omega$ regardless of the feedwater turbidity. Accordingly, the ratio $\alpha\omega/T$ can be used to describe the general behaviour, since it is independent of turbidity (Fig. 9b). In this approach, the asymptotic value of $\alpha\omega/T$ was $1.6\cdot10^{13}$ m^{-2} NTU^{-1}, which is 1.8 times higher than that obtained in analysing the effect of turbidity on $\alpha\omega$ during the first cycle ($9.5\cdot10^{12}$ m^{-2} NTU^{-1}). This behaviour may be due to the fact that the particle cake was detached from the membrane during backwashing, once steady state was reached, but only partially dispersed in the suspension. Therefore, there was a sufficient accumulation of solids in the vicinity of the membrane at the end of the backwash to justify the high $\alpha\omega/T$ ratios, when compared with those obtained for the first cycle. Moreover, a visual examination of the fibres during the backwash phase seems to confirm this interpretation (Fig. 10). The images were captured at backwash durations of 0, 20, 45 and 60 s during one experiment. The effect of these durations on cake dispersion was clearly visible and characterised in terms of dispersed cake height (H_{dc}). It shows that lengthening the backwash positively affected H_{dc} in the range 20–60 s.

From a practical point of view, a continuous operation where filtration cycles will be maintained at a reasonable level (10–15 min) at a moderate flux (20–25 l h^{-1} m^{-2}) is desirable. In fact, similar studies have reported a normal operation alternating 10 min of production with 2 min idle phase at a J of 20 l h^{-1} m^{-2} [8,17]. At optimum doses in pre-coagulation/sedimentation step of 0.14–0.16 mM with a J of 23 l h^{-1} m^{-2} and a backwashing duration of 85 s, the obtained $\alpha\omega$ values where in the range of $5.1\cdot10^{14}$–$7.2\cdot10^{14}$ m^{-2} (Fig. 9a), that implies a filtration duration of approximately 8.5–15 min. However, in order to improve membrane cleaning, it seems necessary to complement the backwashing by other cleaning methods that enhance particle dispersion (such as air sparging on the membrane surface during the backwash).

3.3.2. Residual fouling

Residual fouling has been assessed by studying the influence of backwash duration and feedwater quality on residual fouling resistance. Residual resistance exponentially decreased with t_b until it reached an asymptotic value ($2.9\cdot10^{12}$–$3.1\cdot10^{12}$ m^{-1}), where the effectiveness of the cleaning method tended to stabilise (Fig. 11). This stabilisation was observed for backwash durations of 60–85 s. Furthermore, this permanent residual fouling did not depend on coagulant dose or feedwater characteristics. In fact, the residual resistance varied with the number of cycles at maximum backwash duration (85 s); the results show that most of this fouling was detected in the first cycle and it increased progressively less with subsequent cycles (Fig. 12). This behaviour may be due to a progressive consolidation of the deposit as a consequence of the experimental operating mode for backwash triggering by *TMP* monitoring. Accordingly, backwashing is only initiated when significant fouling (significant mass deposited) is reached during the filtration phase, thus resulting in a growth in residual fouling which backwashing cannot remove. Indeed, Harmant and Aimar [23] have applied the critical

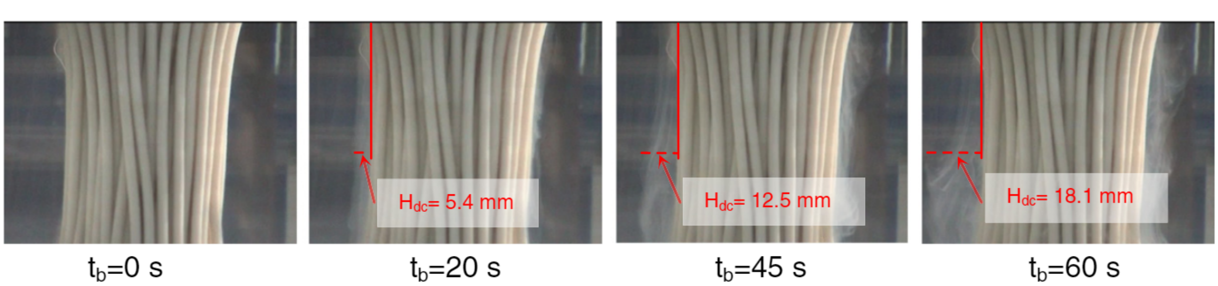

Fig. 10. Effect of the backwash duration (t_b) on dispersed cake height (H_{dc}). Feedwater: SC WWTP primary effluent. J = 23 l h^{-1} m^{-2}. Al^{3+} = 0.16 mM.

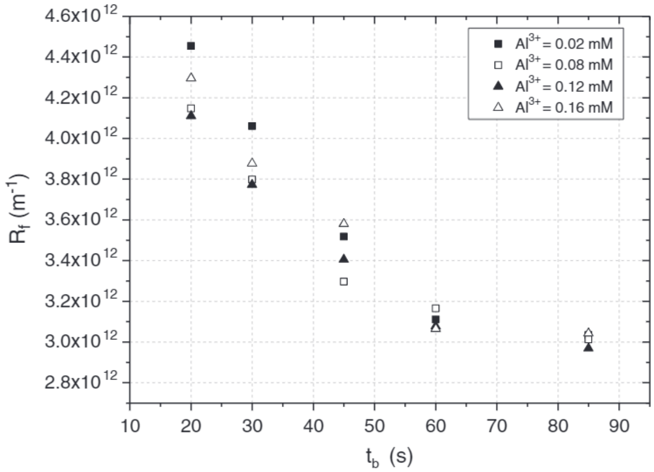

Fig. 11. Residual fouling resistance against backwash duration at different coagulant doses. Feedwater: SC WWTP primary effluent (March 2011). $J = 23$ l h^{-1} m^{-2}.

mass concept for dead-end colloidal filtration, based on the existence of a critical value above which cake consolidation is induced. Furthermore McAdam and Judd [24] reported a similar behaviour in a dead-end denitrification membrane bioreactor. It is suggested that physical cleaning was unable to re-disperse the deposit effectively once it was consolidated. Therefore, it seems necessary to study in a further work the effect of TMP set-point on cake consolidation and thus verify the approach presented here.

4. Conclusions

- High efficiency of COD removal (81–95%) was achieved by combining coagulation and ultrafiltration, regardless of the coagulant dose. Even under optimal coagulant dose, ultrafiltration is needed in order to achieve high COD removal values.
- Pre-clarification by coagulation permits a significant reduction in cake resistance (expressed by $\alpha\omega$, in m^{-2}), but no considerable effect on the residual fouling resistance (R_f) was detected. However, the backwash duration had a significant effect on residual fouling.
- The ratio $\alpha\omega/T$ can be successfully used to quantify reversible cake fouling.
- Direct observation of the membrane surface suggests particle accumulation in the vicinity of the membrane after backwashing, which could justify the increase in cake resistance during the initial filtration/backwashing cycles until steady-state conditions are reached.

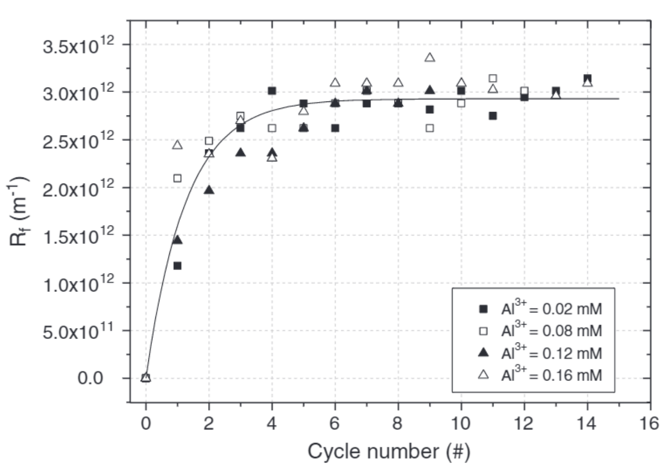

Fig. 12. Residual fouling resistance against the number of cycles at different coagulant doses. Feedwater: SC WWTP primary effluent (March 2011); $J = 23$ l h^{-1} m^{-2}.

Results presented in this work showed that, at a high coagulant dose for the primary effluent, the obtained cake fouling rate values were quite similar to those obtained for the secondary effluent. Therefore, these results suggest the applicability of the process for treating primary effluents and obtaining reusable water. Obviously, a deeper investigation in long-term experiments at a pilot-scale should be done in order to assess process sustainability, mainly focused in residual fouling development and membrane permeability recovery by chemical reagents.

List of symbols

H_{dc}	dispersed cake height, mm
J	permeate flux, l h^{-1} m^{-2}
r_f	fouling rate, Pa s^{-1}
R_f	residual fouling resistance, m^{-1}
R_m	membrane resistance, m^{-1}
R_t	hydraulic total resistance, m^{-1}
t	time, s
T	turbidity, NTU
t_b	backwash duration, s
TMP	transmembrane pressure, Pa
TMP_0	initial transmembrane pressure, Pa

Greek letters

M	solvent viscosity, Pa s
α	specific cake resistance, m kg^{-1}
ω	solid concentration in the cake per unit filtrate volume, kg m^{-3}

Acknowledgements

This work has been funded by the N.R.C. (MAEC project A/030409/10) and CONVAGUA project of the Canary Agency-ACIISI sponsored by the European Commission and the Regional Canary Government. The authors also want to express their gratitude to GE ZENON, to CANARAGUA, to EMMASA and to BALTEN for their support and finally to the Water Analysis Laboratory of the ULL Chemical Engineering Department for the analytical advice. Finally, the authors would like to thank Prof. Elmaleh for his valuable comments and suggestions to this paper.

References

[1] AQUAMAC II Project, Estudio Actualizado de la Situación del Aprovechamiento de Aguas Depuradas en la Macaronesia. Proyecto AQUAMAC II, Técnicas y Métodos para la gestión Sostenible del Agua en la Macaronesia, in: Instituto Tecnológico de Canarias (Ed.), 2008, (in Spanish).

[2] R. Iglesias, E. Ortega, G. Batanero, L. Quintas, Water reuse in Spain: data overview and cost estimation of suitable treatment trains, Desalination 263 (2010) 1–10.

[3] Real Decreto 1620/2007, de 7 de diciembre, por el que se establece el regimen juridico de la reutilizacion de las aguas depuradas, http://www.boe.es/boe/dias/2007/12/08/pdfs/A50639-50661.pdf (in Spanish).

[4] J. de Koning, D. Bixio, A. Karabelas, M. Salgot, A. Schäfer, Characterisation and assessment of water treatment technologies for reuse, Desalination 218 (2008) 92–104.

[5] A.J. Smith, P. Hardy, B.N. Lodge, Alternative water sources and technologies for non-potable re-use, in: 74th WEFTEC Conference, Atlanta, Georgia, 13–17 October 2001, 2001.

[6] D. Abdessemed, G. Nezzal, Tertiary treatment of a secondary effluent by the coupling of coagulation–adsorption–ultrafiltration for reuse, Desalination 175 (2005) 135–141.

[7] A.M. Ravazzini, A.F. van Nieuwenhuijzen, J.H.M.J. van der Graaf, Direct ultrafiltration of municipal wastewater: comparison between filtration of raw sewage and primary clarifier effluent, Desalination 178 (2005) 51–62.

[8] K.-H. Ahn, K.-G. Song, Application of microfiltration with a novel fouling control method for reuse of wastewater from a large-scale resort complex, Desalination 129 (2000) 207–216.

[9] M.H. Al Malack, G.K. Anderson, Use of crossflow microfiltration in wastewater treatment, Water Res. 31 (12) (1997) 3064–3072.

[10] M.F. Pouet, Traitements physico-chimiques associés à une microfiltration d'eau usée urbaine, Thése de doctorat USTL Montpellier, 1994.

[11] K.H. Choo, S.J. Choi, E.D. Hwang, Effect of coagulant types on textile wastewater reclamation in a combined coagulation/ultrafiltration system, Desalination 202 (2007) 262–270.
[12] B.-B. Lee, K.-H. Choo, D. Chang, S.-J. Choi, Optimizing the coagulant dose to control membrane fouling in combined coagulation/ultrafiltration systems for textile wastewater reclamation, Chem. Eng. J. 155 (2009) 101–107.
[13] S. Delgado, F. Díaz, D. García, N. Otero, Behaviour of inorganic coagulants in secondary effluents from a conventional wastewater treatment plant, Filtr. Sep. 40 (7) (2003) 42–46.
[14] APHA, Standard Methods for the Examination of Water and Wastewater, 18 th ed. American Public Health Association/Water Environment Federation, Washington, DC, USA, 1992.
[15] A. Adin, T. Asano, The role of physical–chemical treatment in wastewater reclamation and reuse, Water Sci. Technol. 37 (10) (1998) 79–90.
[16] G. Tchobanoglous, F.L. Burton, H.D. Stensel, Wastewater Engineering: Treatment and Reuse, 3rd ed Metcalf and Eddy, Inc., McGraw-Hill, New York, 2003.
[17] K.-H. Ahn, K.-G. Song, Treatment of domestic wastewater using microfiltration for reuse of wastewater, Desalination 126 (1999) 7–14.
[18] P.J. Smith, S. Vigneswaran, H.H. Ngo, R. Ben-Aim, H. Nguyen, A new approach to backwash initiation in membrane systems, J. Membr. Sci. 278 (2006) 381–389.
[19] S. Delgado, R. Villarroel, E. González, Effect of the shear intensity on fouling in submerged membrane bioreactor for wastewater treatment, J. Membr. Sci. 311 (2008) 173–181.
[20] J. Kim, F.A. DiGiano, Fouling models for low-pressure membrane systems, Sep. Purif. Technol. 68 (2009) 293–304.
[21] S. Delgado, F. Diaz, L. Vera, R. Diaz, S. Elmaleh, Modelling hollow-fibre ultrafiltration of biologically treated wastewater with and without gas sparging, J. Membr. Sci. 228 (2004) 55–63.
[22] K. Kimura, Y. Hane, Y. Watanabe, Effect of pre-coagulation on mitigating irreversible fouling during ultrafiltration of a surface water, Water Sci. Technol. 51 (2005) 93–100.
[23] P. Harmant, P. Aimar, Coagulation of colloids retained by porous wall, AlChE J. 42 (1996) 3523–3532.
[24] E.J. McAdam, S.J. Judd, Optimisation of dead-end filtration conditions for an immersed anoxic membrane bioreactor, J. Membr. Sci. 325 (2008) 940–946.