Ultrasonic irradiation for ultrafiltration membrane cleaning in MBR systems: operational conditions and consequences

L. M. Ruiz, J. I. Perez, A. Gómez, A. Letona and M. A. Gómez

ABSTRACT

Ultrasonic irradiation is one of the most promising membrane cleaning techniques for membrane bioreactors (MBRs) because of several advantages such as high flux-recovery capacity and *in situ* application without interrupting the filtration process. However, significant contradictions may be found and, consequently, this method has not yet been widely developed. In this paper, four MBRs equipped with hollow-fibre polyvinylidene fluoride ultrafiltration membranes were operated continuously. The cleaning method applied consisted of sonication at low power (15 W) with different frequencies (20, 25, 30, and 40 kHz) for each module and aerated backwashing. The different MBRs were analysed comparatively between them and with a conventional MBR in order to check the effects of the irradiated waves on membrane integrity, effluent quality and process performance. Effluent turbidity and chemical oxygen demand, total and volatile suspended solid concentration and activated sludge viscosity were affected by biomass fragmentation or membrane cake removal, mainly at lower frequencies. The best transmembrane pressure control was achieved at the frequency of 20 kHz without a significant effect on membrane integrity. The results showed that under these operational conditions, no negative effects on effluent quality or membrane integrity were found, suggesting that this method was suitable for this type of membrane.

Key words | cleaning methods, ultrafiltration membranes, ultrasonic irradiation

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INTRODUCTION

Membrane fouling, unavoidable in membrane bioreactors (MBRs), constitutes one of the main problems of these systems applied to wastewater treatment (Wang *et al.* 2014) and stands as a major impediment to extensive application of this technology (Wan *et al.* 2013). Therefore, membrane cleaning is an essential part of operating conditions in MBRs, which can be achieved by physical and chemical methods (Wang *et al.* 2014). Usually, physical cleaning methods remove loosely attached material, responsible for the reversible fouling, while chemical cleaning methods eliminate irreversible fouling (Judd 2011). Relax, backwashing or the combination of the two are commonly applied as physical cleaning processes in MBRs, associated with a constant aeration of membranes (Judd 2011). Membrane aeration by coarse bubbles can eliminate concentration polarization, remove reversible fouling, facilitate physical cleaning, and weaken cake resistance (Wang *et al.* 2014) but this process has been considered the largest energy consumer in MBR systems (Fenu *et al.* 2010). In this scenario, for the development of anaerobic applications (Xu *et al.* 2013), other techniques have been developed in recent years to mitigate fouling, such as ultrasonic (US) irradiation (Ahmad *et al.* 2012).

US irradiation consists of sound waves which propagate through a medium with a vast amount of energy dissipation (Patel & Nath 2013). US, which can be performed either *in situ* or *ex situ*, is effective in alleviating the concentration of polarization and removing cake layer (Ahmad *et al.* 2012). Several factors such as US frequency, power density, time of application, irradiation mode or orientation and distance can be considered key parameters influencing cleaning efficiency (Wang *et al.* 2014). Intermittent irradiation must be always used (Xu *et al.* 2013), as it prolongs the lifetime of the membranes and minimizes the

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energy consumption (Wan *et al.* 2013). Transducers must be located facing the fouled membrane surface at not too close a distance to it, to avoid membrane damage (Wang *et al.* 2014) and a control of application time is necessary to avoid clogging due to particle breakage and to ensure an efficient cleaning process (Li *et al.* 2013).

Power density and frequency can be considered the most significant parameters when US irradiation is applied to membrane cleaning (Wang *et al.* 2014). However, both parameters must be optimized in order to get the best results. The most widely applied MBR technology today works with polymeric membranes, made of several materials (Judd 2011) such as polyethylene, polyethersulfone (PES) or polyvinylidene fluoride (PVDF). Some of these materials have been tested with US (Wan *et al.* 2013), making a significant improvement on the recovery of permeate flux with an increase in effectiveness when the power increased from 1 to 15 W and with no improvement for higher powers. However, the application of high power densities might damage the membrane (Wang *et al.* 2014). In this sense, Ruiz *et al.* (2015) revealed that power above 100 W damaged flat PES microfiltration membrane integrity by pore enlargement.

With respect to frequency, relatively low US frequencies are used in order to enhance the cleaning efficiency (Muthukumaran *et al.* 2007). The effect of frequency with respect to cleaning efficiency is more significant in the range from 20 to 100 kHz and it can vary depending on the type of membrane material (Wang *et al.* 2014). Moreover, frequency can also affect membrane integrity (Ruiz *et al.* 2015) and authors such as Masselin *et al.* (2001) reported damage to different polymeric membranes working at a frequency of 47 kHz.

US irradiation can be used as a cleaning method either alone (Muthukumaran *et al.* 2007) or combined with other cleaning procedures (Wang *et al.* 2014) such as aeration, chemical cleaning or backwashing. Wan *et al.* (2013) observed high efficiency in the recovery of permeate flux by combining US cleaning with backwashing, although combining US and hydraulic cleaning methods does not always have a positive effect on permeate recovery.

The advantages of using US irradiation such as high flux-recovery capacity, *in situ* application without requiring the interruption of the filtration process and no generation of chemical by-products (Muthukumaran *et al.* 2007) make this approach a good alternative for use in MBR systems. However, due to the significant contradictions which may be found regarding the effect of sonication over membrane integrity or permeate quality, more thorough studies are needed and special care must be taken in order to analyse and select the most suitable conditions (frequency, power, application time, etc.) to achieve the proposed goals without affecting process performance. In this regard, the present study seeks to help clarify the possibilities of US irradiation combined with backwashing as a cleaning method applied to hollow-fibre ultrafiltration MBR systems.

MATERIALS AND METHODS

Experimental setup

The experimental setup (Figure 1) consisted of four modules working in parallel with a hollow-fibre ultrafiltration membrane immersed inside each one. These membranes were designed and manufactured specifically for this study using adapted hydrophilized commercial membranes of PVDF with a nominal pore size of 0.03 μm. Eighty-three fibres from a commercial membrane manufactured by Zenon-GE, model ZW-10, were cut at the desired length (32 cm) and inserted inside a polymeric resin. Once the resin became solid, the fibres were fixed at the ends. The number of fibres included in each membrane was selected to provide the same membrane surface in each module (around 0.11 m^2).

The modules are made of stainless steel, their capacity is around 32 litres and their external dimensions are $55 \times 39 \times 96$ cm. Every module had a sonicator which provided US irradiation at a different fixed US frequencies (20, 25, 30, and 40 kHz, respectively). The automation and control system allowed the selection of different sonication powers and times of application. For assurance that the US irradiation was evenly distributed over the entire membrane surface, two sheets of transducers ($24 \times 36 \times 0.3$ cm) were vertically mounted on both sides of each module.

Each sonicated module was continuously fed by a peristaltic pump (ESPA, XHM model) with activated sludge taken from a conventional MBR installation, operating in a continuous mode and fed with real pre-treated urban wastewater. During the evaluated period, the sludge retention time in the MBR plant was established at 15 days. The rpm of the peristaltic pumps was selected in order to provide a fixed flux: 10 L/(m^2·h). To keep this flux, if membrane fouling increases, transmembrane pressure (TMP) will also increase; so, as the TMP is measured every second, the evolution of this parameter gives information about the evolution of fouling. On the other hand, at the top of each module, an overflow has been included in order to collect the excess of sludge. These rejected streams from the sonicated modules were

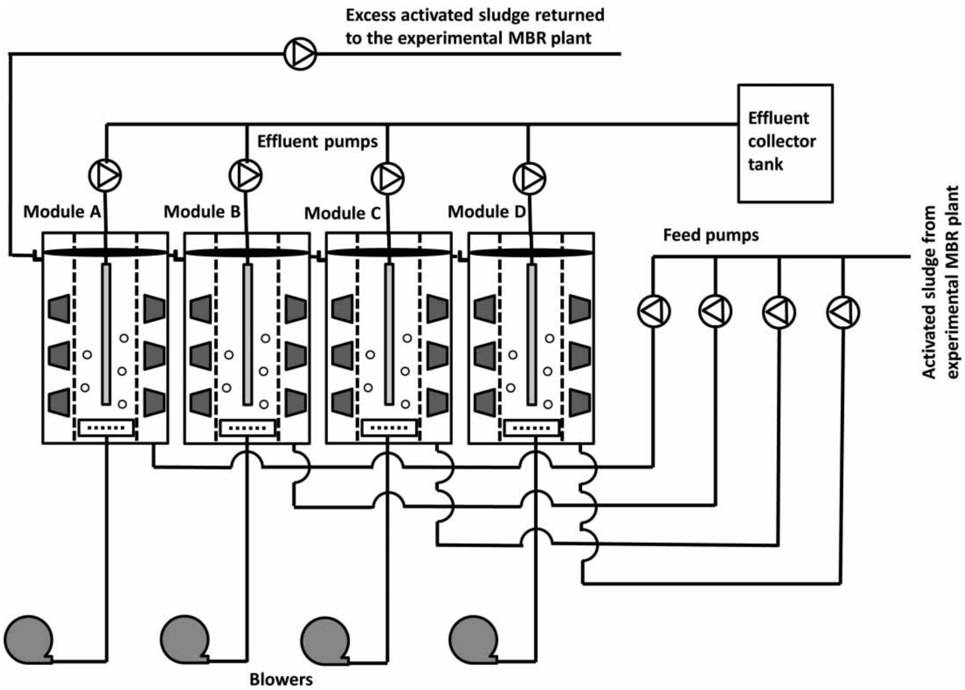

Figure 1 | Schematic experimental setup.

collected in a common pipe and returned continuously to the conventional MBR plant. The effluents were also pumped by vacuum with peristaltic pumps and discharged after sampling for laboratory analyses. Moreover, the experimental installation was also equipped with four blowers (MEDO LA-60B) and perforated pipes at the bottom of each module to provide aeration to membranes.

These modules were fully automated and controlled with a programmable logic controller. Parameters such as flow rates, maximum allowable TMP, filtration, backwash periods, aeration on/off cycles or other set points were selected in the SCADA (supervisory control and data acquisition) system before each experiment. The bioreactor level, activated sludge temperature, TMP, and flow rates were continuously measured by sensors and registered each second in a database in order to assess the effectiveness of the treatment process as well as to compare the performance of each module with that of the experimental MBR installation. All experiments were performed at constant sonication power of 15 W (power density around 0.014 W/cm^2). Operational conditions consisted of cycles of 5 min filtration followed by 1 min backwashing. Intermittent US pulses were applied for 3 seconds every 3 minutes, and during the backwashing periods 5 seconds aeration was applied. The experimental period was 4 months and the estimated retention time inside these modules was around 30 minutes.

Physical–chemical analyses

During the experimental period, activated sludge, influent and effluent samples were collected daily from the conventional MBR system and from each sonicated module. Effluent chemical oxygen demand (COD) concentration was determined using the acid oxidation method (APHA *et al.* 2012). Results were recorded after comparing spectrophotometric (Heλios) measurements at 600 nm with a standard solution of potassium acid phthalate. Total (mixed liquor suspended solids: MLSS) and volatile (VSS) suspended solids concentrations in activated sludge were determined by filtration (0.45 μm), drying at 105 °C and weighing of the samples according to the standard method (APHA *et al.* 2012). For VSS, the dried filters were heated again at 550 °C for 15 min. Colour at 436 nm and absorbance at 254 nm (Abs_{254}) analyses were performed by spectrophotometry following the standard UNE-EN ISO 7887:1995 (AENOR 1995) and turbidity was determined by the spectrophotometry method UNE-EN ISO 7027:1999 (AENOR 2001). Particle-size distribution (PSD) analyses were made by a LiQuilaz HW E20 particle counter (particle measuring systems) based on laser light extinction caused by particles from 2 to 125 μm with a resolution of 1 μm. The system was calibrated using inert latex particles of a defined size.

Activated sludge and effluent viscosities were measured at standard temperature (20 °C) using a rotational viscometer (Fungilab, SMART model) with a low-viscosity adaptor.

At the end of the experimental period, a piece of each membrane (1 cm^2) was cut and analysed using scanning electron microscopy (SEM) with a GEMINI (Carl Zeiss SMT) instrument in order to check whether the membrane surface was damaged due to the exposure to US irradiation. All pieces were preserved with glutaraldehyde (3%) in PBS (phosphate buffered saline) buffer. The pieces were dehydrated using critical-point drying (Polaron CPD 7501) and covered by a conductor metal mixture (60% gold and 40% platinum).

Statistical analysis

The data were analysed by computer-assisted statistics, using SPSS software package (IBM-SPSS v22). Differences between systems were determined using analysis of variance (ANOVA) with a significance level of 1% ($p < 0.01$) and the Student–Newman–Keuls test. A regression fit was performed to TMP data and PSD in order to compare the performance of each system assayed.

RESULTS AND DISCUSSION

TMP evolution

The evaluation of TMP evolution in conventional MBR systems provides useful information on the performance of filtration, and it advises about clogging or fouling problems and membrane integrity (Judd 2011). The same information can be gained in MBR systems with US as the cleaning method, and even TMP can be used as indicator for the efficiency of fouling removal using sonication (Li *et al.* 2013). In the present study, TMP was evaluated continuously in the four sonicated modules and the daily average values were calculated for each module (Figure 2). In consideration of TMP evolution of a similar hollow-fibre ultrafiltration MBR (Parada-Albarracín *et al.* 2012), the data revealed that despite the above sonicated frequency, TMP can be considered highly stable, and no chemical cleanings were required in any module during the period evaluated, showing no negative effects on the membrane permeability due to sonication after a 4 month period.

When the data on the TMP time course were fit to a linear regression, each system was found to perform differently. Data from the membranes in which a US frequency of 20 kHz was applied showed the best fit to a linear regression (Table 1). Coefficients of the regression (Table 1) showed the higher initial TMP values, but the positive value of the slope indicated that membrane fouling was not only controlled, but also reduced. For 25 and 30 kHz, the trend of the data was similar. No linear fit was achieved, but in both cases the slope registered a negative value, signifying a progressive fouling of the membrane. The data found for the system working with 40 kHz can be considered anomalous. Linear fit was non-existent with continuous falls in TMP.

Statistically significant differences (Table 1) were found when comparing the average TMP values for the four modules (p-value 0.0001). These results confirm that lower US frequencies resulted in more effective membrane cleaning

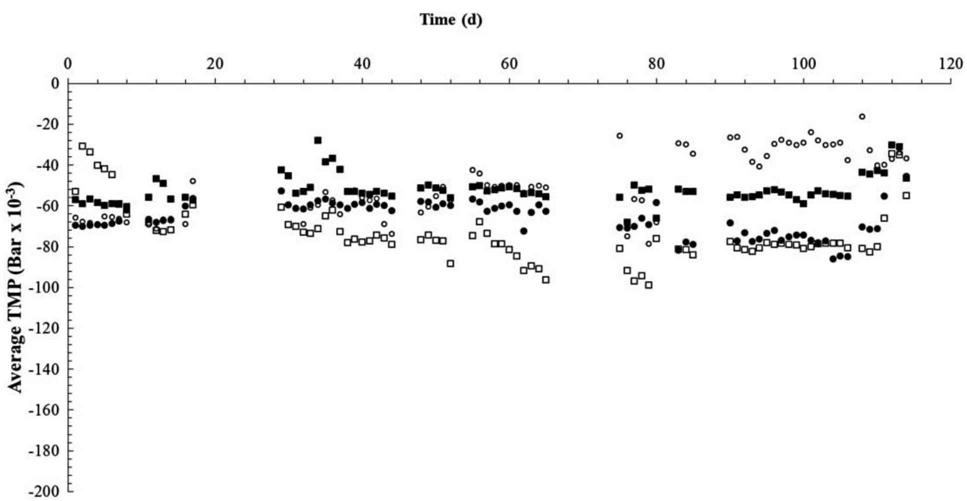

Figure 2 | Time course of average daily TMP in sonicated modules at 20 (○), 25 (●), 30 (□) and 40 (■) kHz of sonication frequencies.

Table 1 | Statistical analysis of TMP data

Frequency	Average	TMP_{min}	TMP_{max}	r^2	Slope (a)	(b)
20 kHz	−0.052	−0.028	−0.068	0.645	4×10^{-4}	−0.072
25 kHz	−0.073	−0.031	−0.099	0.211	-1×10^{-4}	−0.060
30 kHz	−0.067	−0.046	−0.086	0.177	-2×10^{-4}	−0.063
40 kHz	−0.050	−0.016	−0.079	0.031	3×10^{-5}	−0.054

(Muthukumaran *et al.* 2007). However, notably, the TMP values for the module sonicated at 20 kHz tended to decline with time, according to the positive value of the slope. This trend may be due to an increase in the membrane pore size because of structural damage, as reported by several authors (Masselin *et al.* 2001; Ruiz *et al.* 2015). However, to make conclusions regarding this effect, longer periods and other tests are required.

The performance of the systems working at 25 and 30 kHz can be considered normal, with a slow change in TMP with time, which requires the application of other cleaning methods such as membrane chemical cleaning to restore TMP values. The performance of the system working at 40 kHz did not follow the pattern of the other systems, showing a slight upward trend in the TMP values, but with specific recoveries of TMP values (Figure 2).

Effluent quality

Of the parameters selected to evaluate the effluent quality, turbidity and COD concentration registered statistically significant differences (Table 2) between the sonicated systems and the conventional MBR plant. Other parameters evaluated, such as effluent absorbance at 254 nm (Abs_{254}) and effluent colour at 436 nm ($Colour_{436}$), showed similar values for all the effluents. The resulting values showed a great stability, and no statistically significant differences were found regarding these parameters (Table 2).

Effluent turbidity (Figure 3) registered similar values for all the effluents from sonicated modules (Table 2), which ranged from 1.0 to 3.5 NTU without statistically significant differences. However, statistically significant differences resulted when comparing these results with those found for the conventional MBR system (Table 2), in which the values were lower than those from sonicated modules. Trends were not detected in the time course of data (Figure 3).

With respect to COD concentration, values reveal that the organic matter was properly removed in all the systems assayed. However, statistically significant differences were found between the results for the module operated at 20 kHz and the other three modules (p-value 0.0247). On the other hand, no statistically significant differences were found between the modules which operated at 25, 30, and 40 kHz and the conventional MBR system (p-value 0.2168). The time course of the data for sonicated modules showed a negative trend, which was most pronounced in the module sonicated at 20 kHz (Figure 3).

Several authors (Li *et al.* 2013; Ruiz *et al.* 2015) have observed that effluent turbidity obtained from sonicated membranes increased at higher US power and lower frequencies because of the pore-size enlargement. Loderer *et al.* (2013) also noted greater effluent turbidity in relation to US irradiation exposure time due to the deflocculation effect of US irradiation over activated sludge biomass. However, although the turbidity values were higher in the effluent from sonicated systems, influence of the applied US frequency was not detected during the 4 month period, contradicting the observations of previous authors.

Among the analysed parameters, only COD showed statistically significant differences at the lowest US frequency

Table 2 | ANOVA and Student–Newman–Keuls test ($p < 0.01$) for effluent quality parameters

		40 Hz	30 Hz	25 Hz	20 Hz	MBR	*p*-value
Turbidity	NTU	2.08 ± 0.45^a	1.93 ± 0.59^a	1.97 ± 0.65^a	1.85 ± 0.54^a	1.23 ± 0.48^b	0.0001
Abs_{254}	m^{-1}	0.142 ± 0.017	0.138 ± 0.015	0.138 ± 0.014	0.133 ± 0.017	0.136 ± 0.015	0.2170
$Colour_{436}$	m^{-1}	0.014 ± 0.005	0.013 ± 0.003	0.014 ± 0.003	0.014 ± 0.004	0.014 ± 0.004	0.8440
COD	mgO_2/L	$25.85 \pm 12.90^{a,b}$	$27.24 \pm 11.96^{a,b}$	$26.08 \pm 13.94^{a,b}$	32.71 ± 16.25^a	21.05 ± 14.66^b	0.0250

[a,b]Different letters mean significant statistical differences between groups.

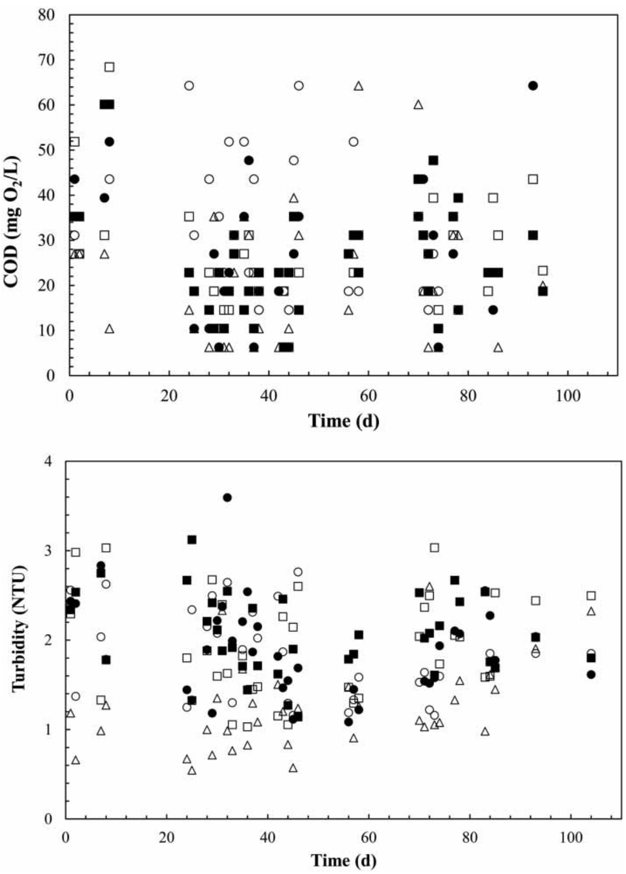

Figure 3 | Effluent COD concentration and turbidity in the conventional MBR system (Δ) and in sonicated modules at 20 (○), 25 (●), 30 (□), and 40 (■) kHz of sonication frequencies.

compared with the other sonicated effluents, although higher concentrations were observed in all the sonicated effluents in comparison with the conventional MBR system. Given the downward trend of COD concentrations over time, enlarged pore size did not appear to be the main cause of the differences in effluent quality. In contrast, this trend may be explained according to the deflocculation process which occurs in sonicated systems and the organic matter released during cell breakage. The observations of Ruiz *et al.* (2015) for a longer period of time and microfiltration membranes showed an increase in the effluent COD concentration and turbidity with time, but in the present work this effect has not been found. Dissimilarities such as the use of a different type of membrane (microfiltration or ultrafiltration) or the lower US power applied may influence the best system performance observed in the present study.

Deflocculation can occur in a system where ultrasound is applied (Feng *et al.* 2009; Loderer *et al.* 2013). Ultrasound can cause acoustic cavitation, agitation, and local heating, thus altering the floc structure or breaking up cell walls. Consequently, PSD is affected (Feng *et al.* 2009; Loderer *et al.* 2013) and smaller particles can more easily pass through the membrane. Deflocculation depends heavily on the energy applied (Feng *et al.* 2009). In this case, the experimental system worked at a power as low as 15 W with 3 sec of application time and variable frequencies in order to avoid membrane damage.

The analyses of PSD (Figure 4) showed that the amount of particles passing through the membrane was clearly higher in sonicated effluents compared with the conventional MBR system and that the highest values were found at the lowest ultrasound frequency (20 kHz), showing statistically significant differences (Table 3). Comparing these results with those for sonicated microfiltration membranes at higher power (Ruiz *et al.* 2015), it can be concluded that the effect was significantly less aggressive in the present study, presumably due to the different energy applied and to the different type of membrane used.

Total particle count (2–125 μm) reveals that deflocculation occurred when US was applied at 15 W of power, regardless of the frequency used. However, the values found for maximum detected particle size were contradictory (Table 3) as not only small particles but also longer ones were also detected in the effluent from sonicated modules. Longer periods of study are required in order to get better conclusions about the mechanisms involved in the evolution of PSD and other effluent quality parameters.

PSD between 2 and 125 μm of all systems under study followed a logarithmic X-model regression (Figure 5), with different values on the slope and at the intersection with the ordinate axis. These differences were not very significant, but they do represent a distinguishing factor between the modules tested, as the conventional MBR module had a lower large-particle content. The coefficient reveals that modules working at 25 and 30 kHz produce a similar effluent quality with respect to particle-size count and modules working at 20 and 40 kHz produce effluent with similar quality, too. This statistical association was similar to that found with respect to the values of TMP and may be due to different phenomena.

One mechanism that improves the quality of effluent passing through the membrane systems is cake formation (Farahbakhsh & Smith 2004). This phenomenon is more significant in microfiltration membranes as their way of working allows cake formation, to improve the quality of the effluent (Arévalo *et al.* 2012). When US irradiation is applied, cake formation can be retarded, contributing to the production of a lower-quality effluent. Moreover, deflocculation occurs due to the US irradiation, generating a higher amount of smaller particles that may cross through

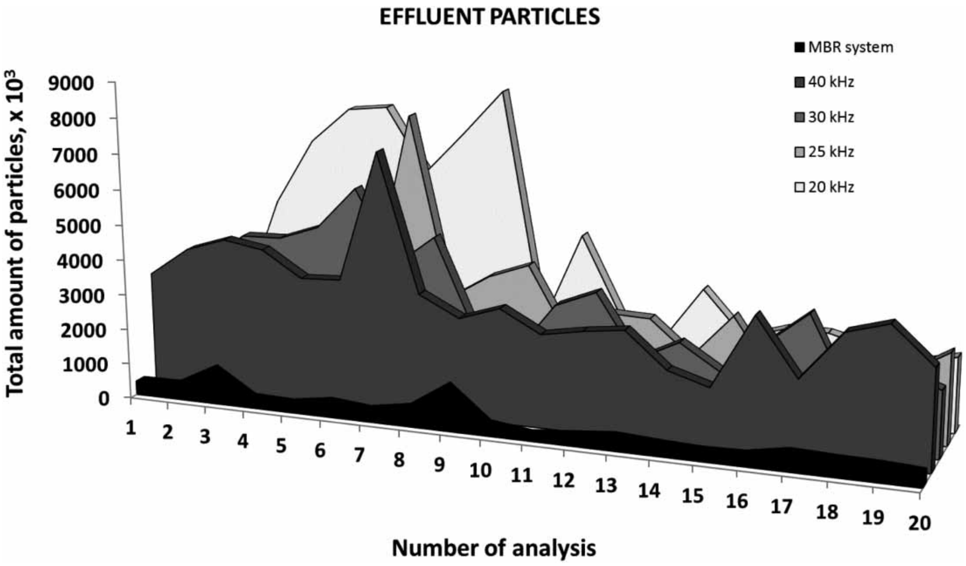

Figure 4 | Presence of particles in the effluent of the conventional MBR system and sonicated modules at different sonication frequencies.

Table 3 | ANOVA and Student–Newman–Keuls test ($p < 0.01$) for PSD analysis

	40 Hz	30 Hz	25 Hz	20 Hz	MBR	*p*-value
Maximum detected size						
Max.	43	43	43	43	29	0.0001
Min.	28	35	25	26	22	
Aver.	38.6[a]	41.2[a]	38.3[a]	39.2[a]	26.5[b]	
SD	5.9	2.3	5.7	5.9	2.1	
Total particle count 2–125 μm						
Max.	17,301	12,335	15,094	30,077	5,419	0.0001
Min.	3,353	3,047	2,874	3,353	394	
Aver.	8,645[a]	6,507[a]	6,772[a]	9,596[a]	1,057[b]	
SD	3,974	2,648	3,043	7,470	1,062	

[a,b]Different letter means significant statistical differences between groups.
SD: standard deviation.

the membrane. Both phenomena may explain the presence of more particles in the effluent from sonicated modules than in the effluent from conventional MBR systems and the downward trend with time of COD concentration in effluent from sonicated modules. Both phenomena were more significant for low US frequency.

Activated sludge properties

Statistically significant differences were found when comparing MLSS and VSS concentrations for activated sludge recirculating from each module and the MBR system (Table 4). According to these results, it can be concluded that in those modules where sonication was supplied at lower frequencies, the MLSS and VSS concentration increased in a more significant way. Discrepancies can be found about this issue. Authors such as Na *et al.* (2007) observed mineralization of organic matter by sonication with the consequent decrease of suspended solid concentration. Other authors such as Feng *et al.* (2009) also showed that ultrasonication strongly affected the amount of soluble matter due to deflocculation and solubilization of particulate material, in agreement with the result of PSD analysis. This phenomenon would imply a loss of biomass because smaller particles could pass through the membrane, as seen with COD for lower assayed US frequencies, which is favoured by the efficiency in removing cake layers with lower US frequencies because pores remain free. However, these results showed that when lower frequencies were applied, higher MLSS and VSS concentrations resulted.

US cleaning is effective in alleviating the concentration polarization and removing cake layers on the membrane (Ahmad *et al.* 2012), so that the portion of biomass retained on the surface of membranes turns again to activated sludge if effective sonication is applied. When lower frequencies are applied, the cake layer removal is more effective than at higher frequencies or in conventional MBR systems. This phenomenon could affect the different MLSS and VSS concentration.

Regarding the viscosity results, these values are directly related to the previous ones, following the same trends as the MLSS concentration. The higher MLSS and VSS concentration in activated sludge is related to viscosity (Sun

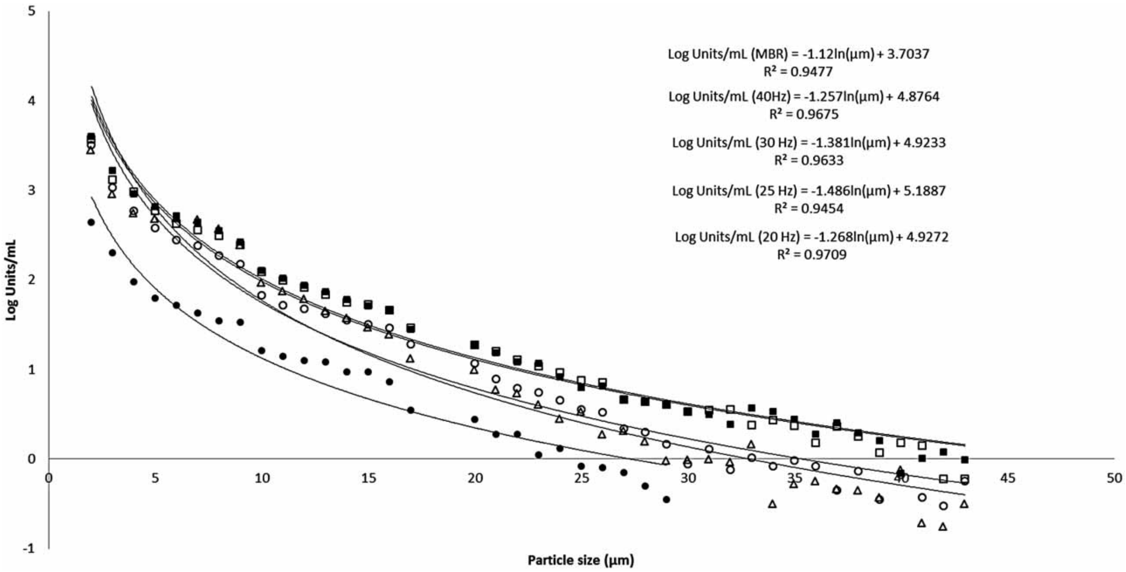

Figure 5 | PSD (2–125 μm) for effluent from conventional MBR (●), and Ultrasound MBR at 40 Hz (□), 30 Hz (○), 25 Hz (Δ) and 20 Hz (■).

Table 4 | ANOVA and Student–Newman–Keuls test ($p < 0.01$) for activated sludge parameters

		40 Hz	30 Hz	25 Hz	20 Hz	MBR	*p*-value
MLSS	g/L	4.71 ± 0.67^a	5.32 ± 0.94^a	6.16 ± 1.10^b	6.41 ± 0.97^b	5.13 ± 0.76^a	0.0001
VSS	g/L	3.61 ± 0.50^a	4.06 ± 0.74^a	4.70 ± 0.86^b	4.90 ± 0.74^b	3.92 ± 0.58^a	0.0001
Viscosity	cP	3.11 ± 0.43^a	3.56 ± 0.56^a	4.29 ± 0.68^b	4.05 ± 0.61^b	3.60 ± 0.40^a	0.0001

[a,b]Different letters mean significant statistical differences between groups.

& Li 2011), so that the same tendency could be expected. In this case, differences were found also for the different modules (Table 4), showing that at higher sonication frequencies, obtained values for these parameters were in the same range as for the conventional MBR system, but at lower frequencies statistically significant differences were found. On the other hand, deflocculation and biomass disintegration observed by Feng *et al.* (2009) affect the protein and polysaccharide concentrations in activated sludge, this being related to viscosity. Increases in MLSS and VSS concentrations and activated sludge viscosity are supposed to negatively affect MBR membrane filtration (Judd 2011). However, no problems with membrane filtration were observed in these systems during the evaluated period.

Membrane integrity

SEM micrographs for several fragments of hollow-fibre membranes (Figure 6) reveal that membrane integrity was not compromised after their exposure to US irradiation under the conditions selected for this study. Membrane surfaces were intact after 4 months of continuous operation and pore-size enlargement was not detected. On the other hand, Ruiz *et al.* (2015), using SEM micrographs, found a significant enlargement of pore size in microfiltration membranes irradiated with US continuously over a period of 6 months under different operational conditions. Flat-sheet microfiltration membranes made of PES were used in the studies of Ruiz *et al.* (2015) and they were irradiated with powers from 100 to 400 W. Although the US frequencies applied and the time of application were similar, several factors, such as applied power, membrane material or membrane configuration, did influence the different performance of the membranes.

Porcelli & Judd (2010) found that some membranes were more susceptible to integrity failure than others, highlighting the fragility of PES materials to US, which failed after 5 min of exposure at US frequency of 47 kHz. Masselin *et al.* (2001) analysed the evolution of the structure of several polymeric

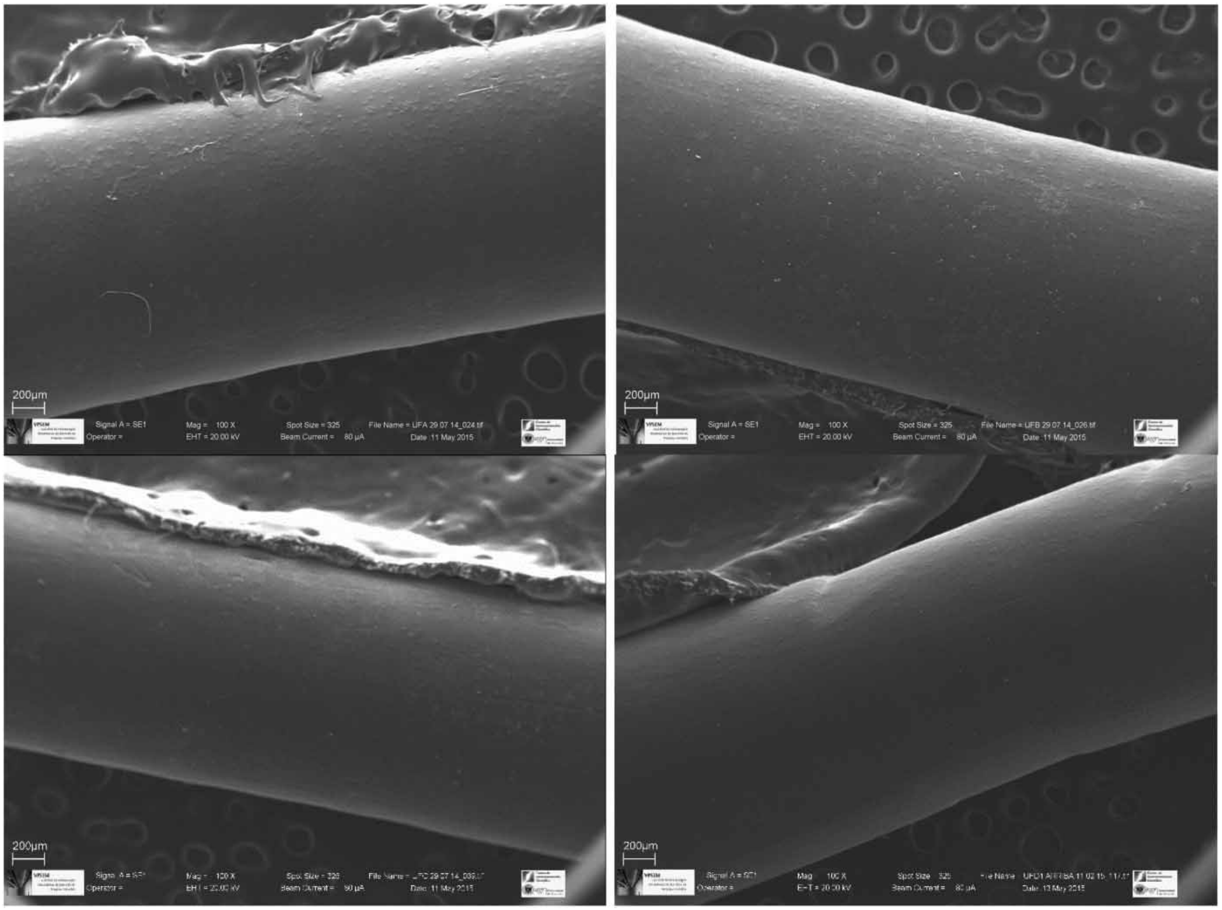

Figure 6 | SEM images of the membrane surfaces after sonication at 40 kHz (top left), 30 kHz (top right), 25 kHz (bottom left) and 20 kHz (bottom right).

materials exposed to US and showed that PES membranes were strongly affected by a significant pore-size enlargement (30%) due to interconnection of neighbouring pores. However, PVDF material has shown higher resistance than PES in comparison to the results of Ruiz *et al.* (2015), showing that sonication powers lower than 15 W have demonstrated to be sufficient for membrane cleaning (Wan *et al.* 2013) and, thus, higher powers do not seem to be adequate for polymeric membranes due to the negative consequences they might cause.

According to this, one of the most remarkable results of Ruiz *et al.* (2015) was the inefficiency of high power to clean membrane surfaces, with significant increases in TMP after a few days of operation. Hence, power does not appear to be the only cause of membrane failure. The effect on the microfiltration flat-sheet membranes was significantly more aggressive than on hollow-fibre ultrafiltration membranes, presumably because flat-sheet membranes were fixed and they could not vibrate when sonication was applied, and therefore the irradiated energy was continuously focused on the same point. However, ultrafiltration membranes are fixed only at the ends and they are able to vibrate, and thus the irradiated energy is distributed in different points of the membrane surface. However, more experiments are necessary to evaluate the influence of the membrane-module configuration on the resistance against US irradiation.

According to these results, the consequences of selecting inappropriate membrane types or materials, ultrasound powers, frequencies and/or exposure times are extremely important when sonication is applied for membrane cleaning, and special care must be taken in order to select the best values for all these parameters. These results highlight the need for gathering more information to determine the overall effects of sonication in MBR systems and carrying out new experimental studies in order to find the correct values for correct membrane cleaning and recovery of TMP and permeability without affecting activated sludge characteristics and effluent quality.

CONCLUSIONS

In general, the results show that under the operational conditions evaluated, hollow-fibre PVDF ultrafiltration membranes performed well and gave satisfactory results regarding the effluent quality or membrane integrity, without significant negative effects due to ultrasound application combined with backwashing. US power of 15 W and frequencies from 40 to 20 kHz with intermittent application of 3 seconds irradiation every 3 minutes and 1 min of backwashing with 5 seconds aeration are proper operational conditions for cleaning this type of membrane. More specific conclusions can be highlighted:

- According to the PSD of the effluent, a biomass fragmentation can be expected when US is applied as the cleaning method of membranes in MBR systems. This phenomenon is more significant at lower frequencies and can affect the quality of the effluent.
- US frequencies of 20 kHz retard cake formation on the membrane surface, favouring the membrane TMP control but lowering effluent quality.
- MLSS and VSS concentration and activated sludge viscosity in the recirculate of the membrane module were influenced by sonication, mainly at low frequencies, although effects on membrane fouling were not detected.

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