



## Analysis of the effects of ultrasound irradiation over effluent quality and membrane integrity in flat sheet microfiltration MBR systems

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Received 15 July 2014; Accepted 13 October 2014

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### ABSTRACT

Membrane fouling is one of the main problems regarding the performance of membrane bioreactors (MBRs) and it constitutes an important impediment to the increasing application of this technology. One of the most promising alternative cleaning methods is ultrasound irradiation and for that reason, in this study, the performance of four pilot-scale MBR modules using flat sheet microfiltration membranes working in parallel was evaluated and compared with a conventional MBR system. In these modules, sonication at different frequencies (20, 25, 30 and 40 kHz) and powers (100, 200, 300 and 400 W) was simultaneously applied during the filtration process and parameters such as total suspended solids (TSS) and volatile suspended solids activated sludge concentrations and effluent chemical oxygen demand (COD) concentration, turbidity, viscosity, colour or particle size distribution were analyzed. Moreover, operational parameters such as temperature or TMP were also evaluated, and scanning electron microscope (SEM) and Fourier transform infrared spectroscopy (FTIR) analyses were carried out once the membranes were replaced. Although parameters such as effluent COD concentration, absorbance at 254 nm, colour at 436 nm, viscosity and activated sludge TSS did not show significant differences at different US frequencies, other parameters such as effluent turbidity or particle size distribution reached values too high compared with those obtained for the effluent from the microfiltration MBR system, especially at lower frequencies (20 kHz) and higher powers (400 W). Moreover, SEM images demonstrated that membrane integrity was negatively affected especially at these conditions and membrane pore size was enlarged due to sonication.

*Keywords:* Cleaning methods; Fouling; Membrane bioreactors; Microfiltration; Ultrasound

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*Presented at the IX Ibero-American Congress on Membrane Science and Technology (CITEM 2014), 25–28 May 2014, Santander, Spain*

## 1. Introduction

Membrane fouling is one of the main problems regarding the performance of membrane bioreactor (MBR) systems applied to wastewater treatment [1,2] and it constitutes an important impediment to the increasing application of this technology because the replacement of a membrane due to fouling is one of the largest operating costs in full scale WWTPs [3]. For that reason, many techniques have been developed in recent years to mitigate fouling [4,5]. On the one hand, chemical cleanings in which detergents, acids or alkalies used are usually employed, but chemicals may damage the membrane materials and cause secondary pollution. Moreover, filtration must be interrupted for membrane cleaning. On the other hand, physical methods such as relax or backwashing are also commonly used, but these methods increase separation time and costs because of successive shut-downs for membrane cleaning. Other methods such as sparging of gas bubbles into the feed [6,7], enzymatic cleaning [8] or applications of electric fields [9] are also alternative techniques, but they are still in an initial stage. Among all these methods, one of the most promising ones is ultrasound irradiation, which can be used as a unique method or combined with other cleaning methods such as chemical cleaning or backwashing [10–12].

Ultrasound irradiation is defined as the acoustic energy or sound waves with frequencies above 20 kHz. These waves propagate through a medium with a vast amount of energy dissipation and generate gas and vapour bubbles which grow and collapse violently at high velocity (“acoustic cavitation”), leading to localized high temperatures and pressures and releasing highly reactive free radicals [13]. Although higher frequencies may lead to the collapse of more cavitation bubbles, these bubbles are smaller in size and collapse less energetically, so they may not be capable of detaching particles from the cake layer as readily as lower frequencies. For that reason, cavitation occurs more readily at frequencies in a range from 20 to 40 kHz and it is known that the effect of sonication on flux recovery and fouling mitigation is more significant at lower ultrasonic frequencies [14–17].

In recent years, many researchers employed low frequency (<100 Hz) and low intensity (<2 W/cm<sup>2</sup>) sonication not only for fouling mitigation but also for excess sludge reduction. Intermittent application was usually selected because it prolongs the lifetime of the membranes and minimizes the energy consumption [18,19]. In general, due to the significant contradictions which may be found in literature regarding the effect

of sonication over membrane integrity and cells destruction, special care must be taken in order to select the most suitable conditions (frequency, power, application time, etc.) which enable the achievement of the proposed goals without affecting the process performance. For that reason, in this paper, different conditions have been tested. Moreover, ultrasound irradiation in this study was simultaneously applied during the normal operation of MBR modules, although many authors have stated the effectiveness of sonication as a promising membrane cleaning method which greatly reduces fouling [13,19], few papers have been published in which US irradiation was applied simultaneously, instead of separately, once the process was interrupted [20,21].

## 2. Materials and methods

### 2.1. Experimental setup

The experimental setup used in this study is schematically illustrated in Fig. 1. It consisted of four modules working in parallel with a flat sheet microfiltration membrane immersed inside each one. These modules were made of stainless steel and they had a single capacity of 32 L. Each module had a sonicator which provided ultrasonic irradiation at a fixed US frequency (20, 25, 30 and 40 kHz, respectively) and it allowed the selection of different US powers, times of application or even power rise ramps. In order to ensure that ultrasound irradiation was homogeneously distributed along the whole membrane surface, two sheets of transducers were vertically mounted in both sides of each module. Their dimensions were 240 × 360 × 3 cm.

Activated sludge taken from an experimental microfiltration MBR plant was used to feed these modules using peristaltic pumps (ESPA, XHM model). This microfiltration experimental plant was manufactured by KUBOTA and it was constituted by an anoxic bioreactor for pre-denitrification, an aerobic bioreactor where organic matter was degraded and ammonium was removed and an MBR module. It operated in a continuous mode and was fed with real urban wastewater previously pretreated to remove sand, solids and oils. Rejected streams from the US modules were collected and recycled again to the aerobic bioreactor of the MBR system and the effluents from each module were also pumped by peristaltic pumps and collected in a permeate tank after sampling for laboratory analyses. The experimental installation was also equipped with four blowers (MEDO LA-60B) and perforated pipes at the bottom of each module to provide aeration and membranes were

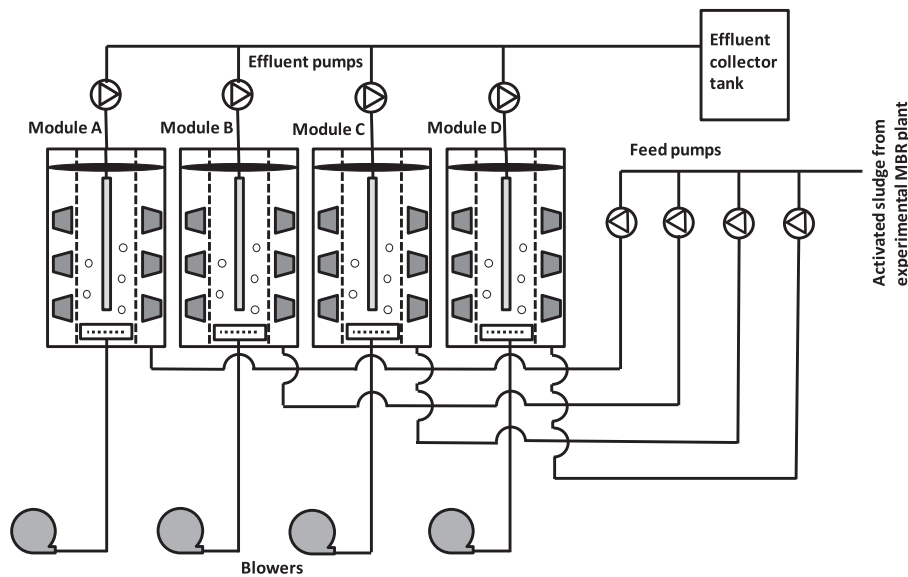


Fig. 1. Schematic experimental setup.

two-faced flat sheet microfiltration membranes made in PE, with an average nominal pore size of 0.4  $\mu\text{m}$ .

These modules were fully automated and controlled with a PLC. Bioreactor levels, temperature, transmembrane pressure (TMP) or flow rates values were continuously measured by sensors and their values were continuously registered in a database in order to assess the effectiveness of the treatment process as well as to compare the biological performance with that of the experimental microfiltration MBR system. Moreover, liquid levels were controlled in each module in order to ensure that the membranes were always completely immersed in the liquid. Parameters such as flow rates, maximum allowable TMP, filtration, relax and backpulse periods, aeration on/off cycles or other set points were selected in the SCADA before each experiment depending on the research purposes.

Table 1 summarizes the operational conditions tested during this study and the different phases in which this period were divided. Aeration cycles varied depending on the TMP data and the specific requirements of each module.

## 2.2. Physical–chemical analyses

During the filtration experiments, activated sludge and effluent samples were daily collected and effluent chemical oxygen demand (COD) concentration, turbidity, absorbance at 254 nm ( $A_{254}$ ), colour at 436 nm and activated sludge total suspended solids (TSS) and volatile suspended solids (VSS) concentrations were analyzed.

Effluent COD concentration was determined using the acid oxidation method [22] with potassium dichromate ( $\text{Cr}_2\text{O}_7\text{K}_2$ ) and closed reflux. Results were obtained after comparing spectrophotometric measurements at 600 nm (spectrophotometer Helios) with a standard solution of potassium acid phthalate ( $\text{HOOC-C}_6\text{H}_4\text{COOK}$ ). TSS and VSS concentrations were also determined according to the standard method [22] by filtration (0.45  $\mu\text{m}$ ), drying at 105  $^\circ\text{C}$  and weighing of the samples. For VSS, the dried filters were heated again at 550  $^\circ\text{C}$  for 15 min. Colour and absorbance analyses were performed by spectrophotometry following the standard UNE-EN ISO 7887:1995. Turbidity was evaluated using a nephelometric turbidimeter

Table 1  
Operational conditions

Phase	Time filtration, min	Time relax, min	US Power, W	US time on, s	US time off, s	Sampling range
1	9	1.5	400	1	10	1–30
2	9	1.5	200	2	5	31–74
3	9	1.5	300	4	5	75–113
4	9	1.5	100	4	2	114–155

(Dinko) and for the particle size distribution analyses, a LiQuilaz HW E20 instrument was used. This equipment measures the dispersion of the light beam caused by particles from 2 to 125  $\mu\text{m}$ . Finally, activated sludge and effluent viscosities were measured using a rotational viscometer (FUNGILAB, SMART model) based on the measurement of the resistance of the liquid to the rotating movement of a spindle at a constant velocity. Due to the characteristics of the samples analyzed in this study, a low viscosity adaptor was used. Samples were introduced in a water bath before the analyses in order to get comparable results at standard temperature (20°C).

At the end of the filtration experiments, once the maximum allowable TMP was reached and no recovery flux was observed after intensive aeration, a piece of each membrane was cut and analyzed using scanning electron microscopy (SEM). The chemical nature of the foulants was also examined by Fourier transform infrared spectroscopy (FTIR). On the one hand, SEM analyses allowed the observation of physical changes in the membrane surface using a GEMINI (FESEM) Carl Zeiss SMT instrument. Previous sample preparation was required for these analyses. First of all, biofilm attached to the membrane surface was fixed using glutaraldehyde and later, samples were dried and covered by a conductor metal mixture (60% gold and 40% platinum). On the other hand, FTIR allowed the identification of the functional groups attached to the membrane surface. In this study, pieces of 1  $\text{cm}^2$  of the membranes were dried before being analyzed in a JASCO 6200 instrument for ATR-FTIR analysis.

### 3. Results and discussion

#### 3.1. Effluent quality and activated sludge characterization

One of the main parameters evaluated in this study was the effluent COD concentration, which allowed the estimation of the amount of soluble organic matter which leaves the system. Obtained results (Fig. 2) showed that the COD concentrations of the effluent leaving the experimental microfiltration system and the effluents leaving the US-MBR modules were all under the discharge limits established in the Spanish legislation (R. D. 509/1996) and, in general, there were no significant differences in the results obtained for each module working at different US frequencies. However, it can be observed that effluent COD concentrations were higher at the lowest frequencies (20 and 25 kHz), especially in the second half of the period.

Regarding the effluent turbidity (Fig. 3), it can be observed that this parameter was almost constant around 2 NTU for the effluent from the experimental MBR plant, but effluents from the sonicated modules reached values as high as 20 NTU and, in general, their values showed important fluctuations. The most significant variations were obtained again for the effluent of the module which operated at 20 kHz, especially in the second half of the evaluated period. In general, these results were in the same range than those obtained by authors such as Loderer et al. [14], who tried to get a relationship between the effluent quality and the US exposure time and got values from 3 to 5 NTU when the exposure time was low (from 1 to 5 min) and above 11 NTU when the exposure time

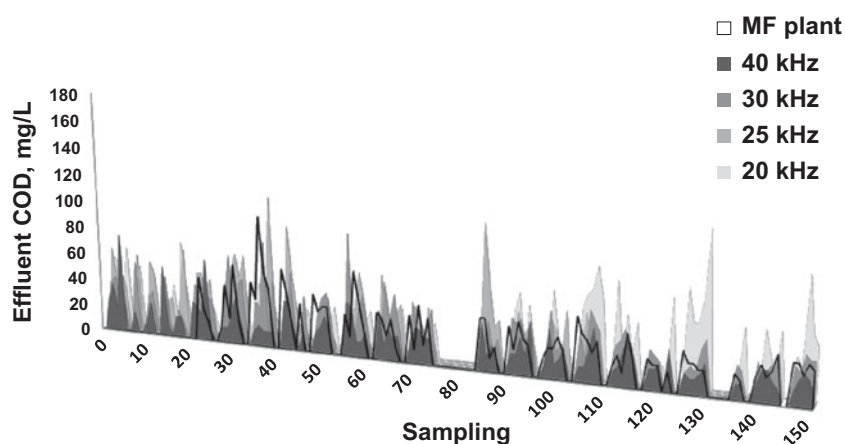


Fig. 2. Effluent COD evolution.

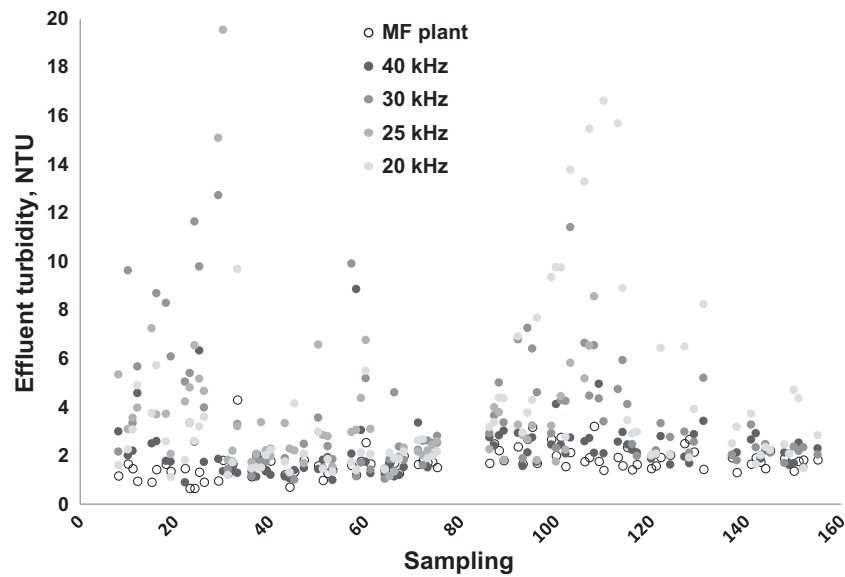


Fig. 3. Evolution of the effluent turbidity.

was 60 min. These authors attributed turbidity to the deflocculation effect of the ultrasonically treated sludge, as the size of the flocs decreased with increased exposure times or US powers and much smaller particles were able to cross the membrane. Li et al. [23] also stated that effluent turbidity significantly increased at higher US power and lower US frequencies, but they attributed it to the fact that, at these conditions, membranes might be damaged and the pore size might be enlarged. Obtained results agreed with those obtained by these authors and higher turbidity values were obtained at higher US powers (300 and 400 W) and lower frequencies (20 kHz).

Abs<sub>254</sub> (Fig. 4 left) gives useful information regarding the presence of organic matter in the effluent, especially aromatic organic compounds with double bonds which absorb at this wavelength. On the other hand, colour (Fig. 4 right) is associated with the presence of humic substances. In both cases, results showed a high stability for all the effluents except for some values obtained during the initial start-up period and no differences were observed at different US conditions. Moreover, the results obtained for the effluents of the sonicated modules were similar to those obtained for the effluent of the microfiltration plant during the whole period. According to Yoon [24], the effluent colour density in sonicated systems was higher than that in conventional MBR systems because protoplasmic polymers such as DNA, proteins and carbohydrates were released from disintegrated cells and these substances caused the increase in colour

density. Generally, colour causing matter is slowly a biodegraded matter and it is detected as COD, so these parameters are expected to be related and follow the same trends. However, in the evaluated conditions, no differences depending on the ultrasonic conditions were observed for these parameters, so it can be stated that no cells disintegrate due to sonication taking place in the sonicated modules.

The evaluation of viscosity is also useful to evaluate the effect of sonication over the activated sludge and over the filtration process. A great stability was observed both for activated sludge and effluent viscosities (Fig. 5). Moreover, the results obtained for each sonicated module were similar to those obtained for the microfiltration plant except for some values of the initial start-up period. Pham et al. [25] analyzed this parameter when they evaluated sonication as a technique for increasing activated sludge solubility and biodegradability. According to these authors, US irradiation reduced activated sludge viscosity, so mass transfer and subsequently biodegradability were improved. This decline of viscosity was due to the higher temperatures, reached, inside the bioreactor when sonication was applied and led to the disintegration of sludge flocs, cell lysis and cleavage of interactions due to shear forces from acoustic cavitation and the partial hydrolysis of EPS. Patel and Nath [13] also stated that the internal and elastic forces created by the acoustic waves caused changes in the interfacial phenomena of the solid phase, leading to a reduction of viscosity and improved dewatering and flux enhancement. However, the results obtained in the

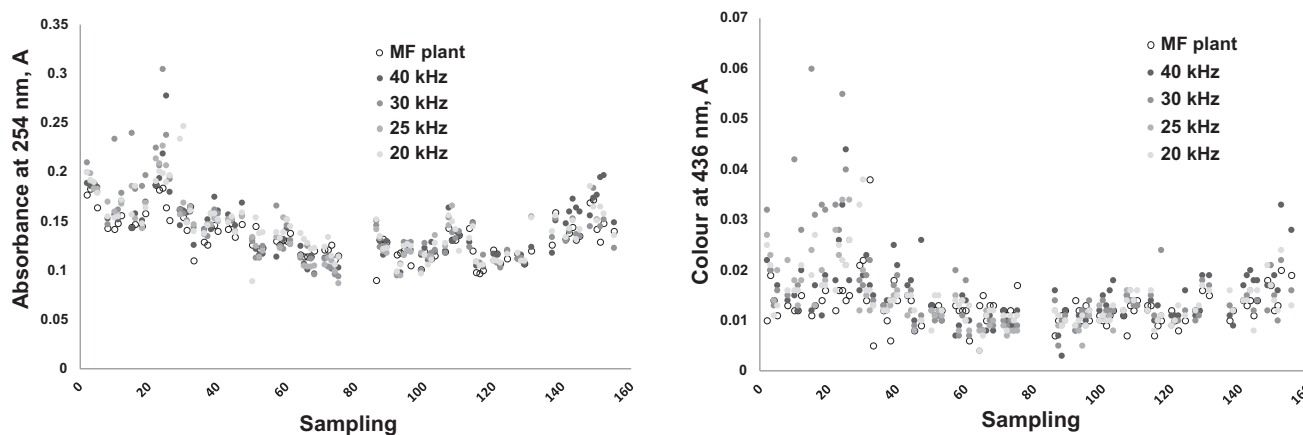


Fig. 4. Evolution of the effluent absorbance at 254 nm (left) and colour at 436 nm (right).

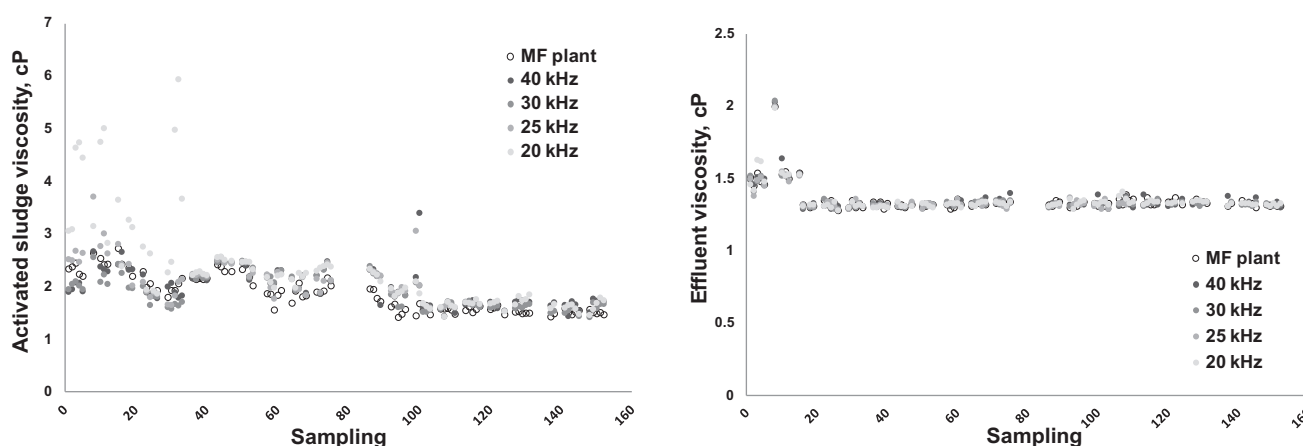


Fig. 5. Evolution of the activated sludge (left) and effluent (right) viscosities.

present study did not show differences when sonicated sludge was compared with activated sludge from the experimental MBR plant, even though the hydraulic retention time inside the sonicated bioreactors was 1 h approximately during the whole research period.

With respect to the activated sludge TSS and VSS concentrations (Fig. 6), except for the initial phase, a great similarity was observed not only among the values obtained for the different sonicated modules but also for the activated sludge coming from the microfiltration plant. VSS values were, in general, high indicating that the amount of mineralized matter was low in all the MBR modules. Authors such as Krzeminski et al. [26] observed that membrane filterability decreased and ratios VSS/TSS increased with sonication due to the internal matter released as the flocs are broken. However, these results show again that no

differences were obtained depending on the applied frequency.

Effluent particle size distribution is, probably, one of the parameters, which is a more useful information regarding the effect of the ultrasound irradiation over the activated sludge and the membrane integrity. Fig. 7 shows the evolution during the evaluated period of the total number of particles (from 0.2 to 125  $\mu\text{m}$ ) in each effluent in order to provide a general view of the differences among the effluent from the microfiltration plant and those from the sonicated modules. First of all, it must be mentioned that, during this period, 34 analyses were carried out for each effluent and the number of particles obtained in those analyses, carried out for the effluent, from the microfiltration plant showed a high stability and significantly lower values than the results obtained when sonicated effluents were analyzed. On the other hand,

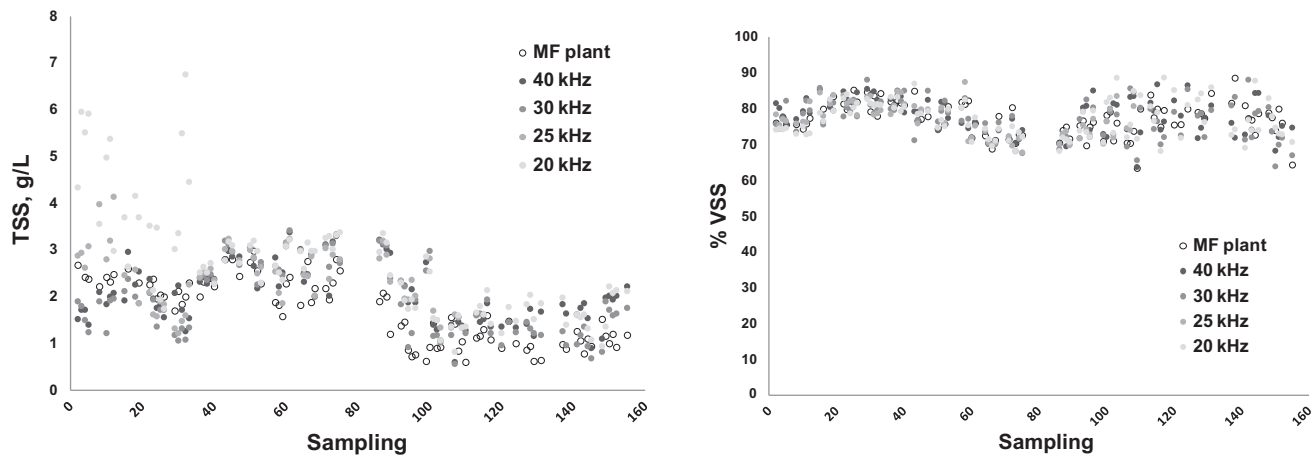


Fig. 6. Evolution of the activated sludge TSS concentration (left) and the fraction of VSS (right).

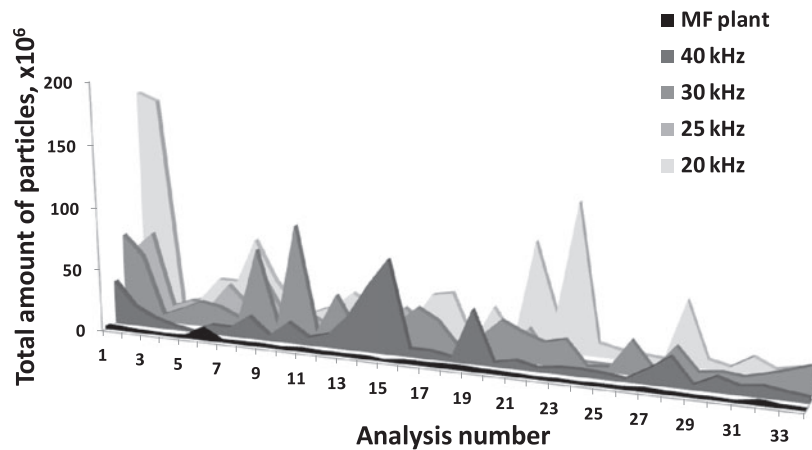


Fig. 7. Evolution of the total number of particles in the effluents.

effluents from the four sonicated modules showed values too high compared with those obtained for the conventional MBR system.

Fig. 8 (top left) shows the results of particle size distributions obtained for the analyses carried out using the effluent from module A, which operated at 40 kHz. It can be observed that most of the particles found in this effluents were smaller than 10  $\mu\text{m}$  and that, in some cases, the amount of particles reached values as high as  $52 \times 10^6$ . These extremely high values are not usual for effluents from conventional microfiltration MBR plants (Fig. 8 bottom). Similar results were obtained for module B (Fig. 8 top right), which operated at 30 kHz. In this case, most of the particles were also in a range from 2 to 10  $\mu\text{m}$  and values as high as  $50 \times 10^6$  particles were also observed.

Values obtained for the effluent from module C which operated at 25 kHz (Fig. 8 centre left) were in agreement with those obtained for the previous modules, although no punctual peaks as high as those obtained in the other effluents were observed. Regarding module D (Fig. 8 centre right), which operated at the lowest frequency of 20 kHz, values were in general higher than the previous ones, although no punctual peaks were observed. Moreover, in this module, there were significant amounts of particles whose size was in a range from 2 to 20  $\mu\text{m}$ , indicating that the particle size distribution was broader and particles with higher pore sizes passed through the membrane.

If previous results are compared with those obtained when the effluent comes from the conventional microfiltration plant (Fig. 8 bottom),

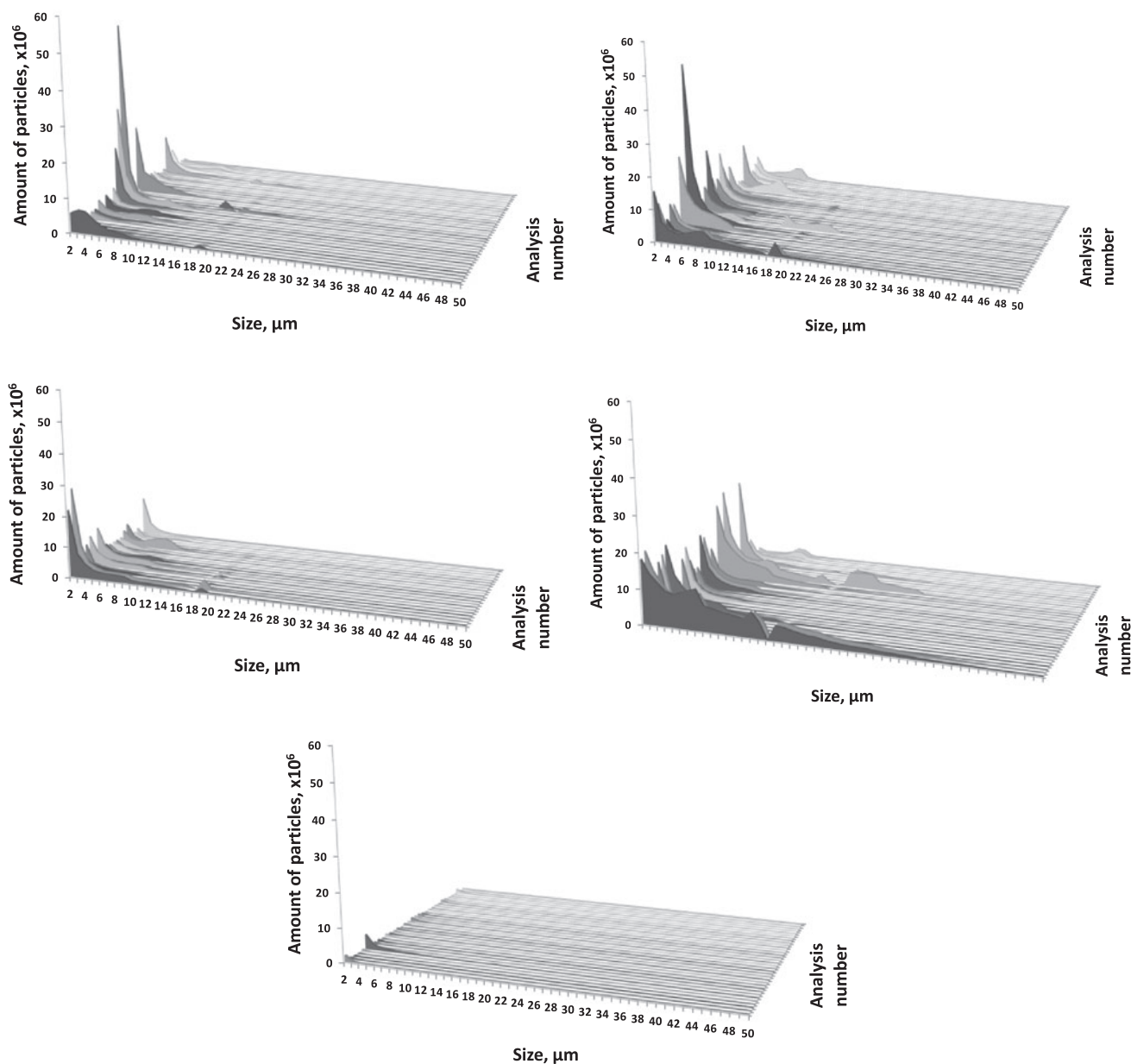


Fig. 8. Particle size distributions for the effluents from the module which operated at 40 kHz (top left), 30 kHz (top right), 25 kHz (centre left), 20 kHz (centre right) and from the microfiltration plant (bottom).

great differences were observed regarding the stability of the results, the efficiency of the system and the effluent quality. In summary, effluents from all sonicated modules contained an amount of particles higher than those expected for conventional MBR systems and higher than the membrane nominal pore size. These results, together with those obtained for other parameters such as effluent COD concentration or turbidity showed that sonication probably caused membrane damages. Again, the effluent which

showed the highest differences with respect to the conventional microfiltration plant was the effluent from module D, which operated at the lowest frequency (20 kHz).

### 3.2. Analysis of operational parameters

Besides the evaluation in the lab of the analytical parameters described above, the most important operational parameters involved in the performance of



MBR systems were also analyzed in this study. The first one was the temperature, which is one of the most influential operational parameters over the performance of biological MBR systems [27]. An increase in the temperature due to the ultrasonic irradiation was observed inside the sonicated bioreactors, reaching values as high as 43°C in summer while ambient temperature did not exceed 35°C. These data agreed with authors such as Caia et al. [28], who observed that at ultrasound irradiation of 180 W, the heat generated could not be removed by the cooling system, resulting in a temperature raise up to 55°C. Several advantages may be derived from an increase in the temperature inside the bioreactors, as it can be found in literature that the biological activity of micro-organisms usually increases with the temperature [29] and fouling formation decreases as the temperature increases due to the lower activated sludge viscosity and higher diffusivity and solubility [13]. Regarding the effect of sonication as a cleaning method, Chai

et al. [30] evaluated different temperatures (20, 30 and 40°C) and they observed that ultrasonic cleaning was faster at higher temperatures. However, other authors stated that the influence of the temperature over the effect of ultrasounds in removing foul was negligible [16,17]. Besides the discrepancies found in literature regarding this topic, according to Krzeminski et al. [26], the optimal range of temperatures for the most common bacteria in WWTPs is from 25 to 35°C, so in these modules, sonication may help to increase the temperature of the activated sludge and to maintain it inside the optimal range, even in cold winter periods.

Fig. 9 shows the TMP in each module during the period in which the selected ultrasounds power was 200 W. It can be observed that this parameter was stable in all the modules during the first 20 d but then, it started to increase and it readily reached the maximum allowable TMP value in modules A, B and C operating at 40, 30 and 25 kHz, respectively. These results were in agreement with authors such as

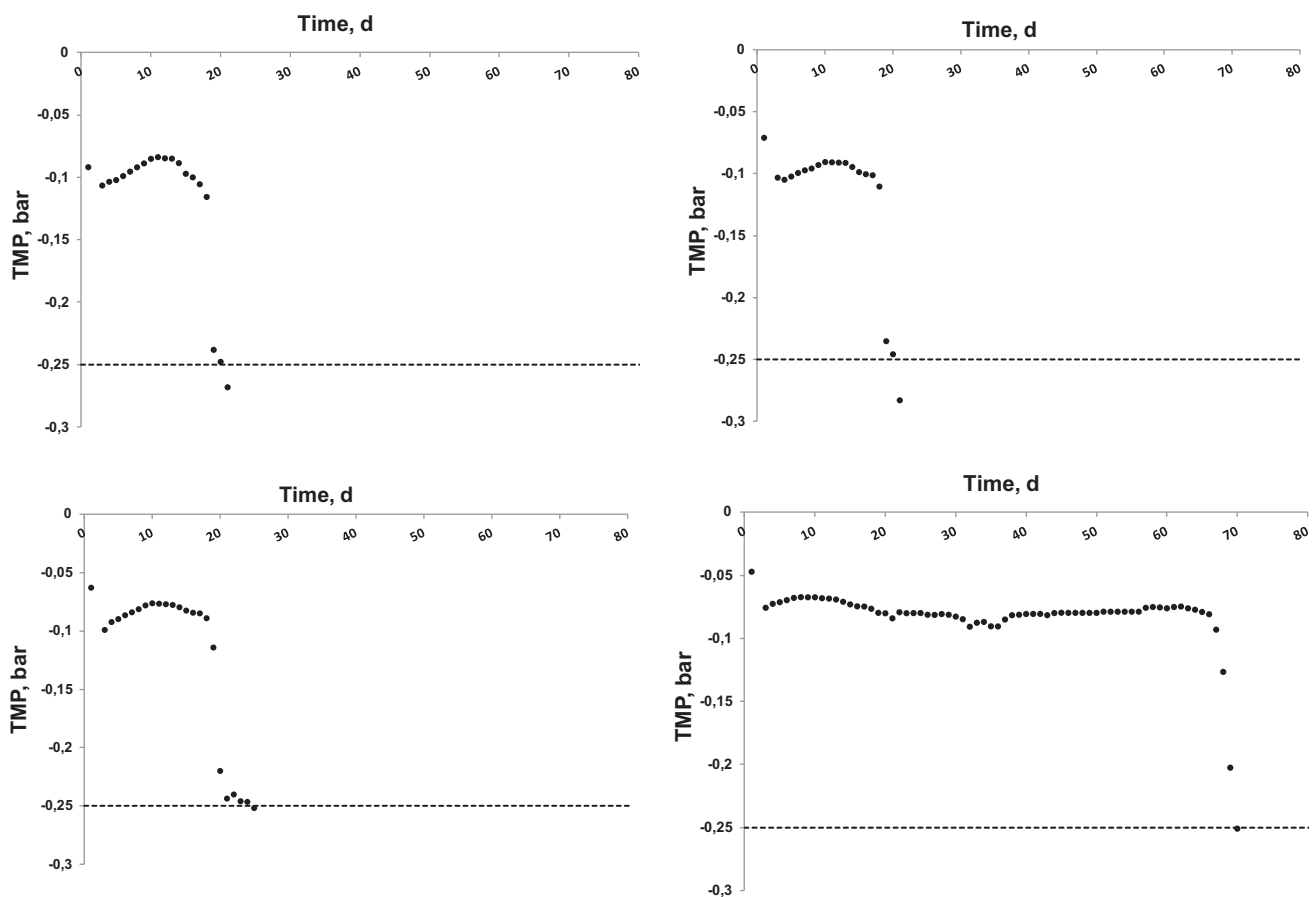


Fig. 9. Evolution of the TMP in the module which operated at 40 kHz (top left), 30 kHz (top right), 25 kHz (bottom left) and 20 kHz (bottom right).

Pendashteh et al. [31] or Loderer et al. [14], who, respectively, stated that the effluent flux readily decreased when ultrasounds were applied and it was reduced to half, in more than 15 d, and that after 20 d, an extensive membrane clogging was observed and TMP rapidly increased. On the contrary, the module which required more time for membrane clogging and whose TMP was constant for almost 70 d was the module which operated at 20 kHz, so confirming the conclusions obtained by other authors such as Li et al. [21], who stated that sonication at higher intensity and lower frequency substantially reduced membrane fouling and in turn enhanced the permeate flux of the filtration process or Cai et al. [15], who compared results at 28, 45 and 100 kHz and stated that the lower the ultrasonic frequency, the higher the recovery flux in the process. However, it is remarkable that according to the membrane manufacturers, these microfiltration membranes are supposed to be able to operate for more than 6 months without requiring chemical cleanings, so these results showed that sonication at the tested conditions, as the only cleaning method, was not effective for membrane maintenance and flux recovery, although sonication was able to remove the cake layer from membrane surface, i.e., the reversible resistance, which usually contributes to more than 50% of the total resistance [15], it leads to a decrease in the average particle size in the activated sludge [21] and destroys the cake in such a way that narrow pathways are built through these pathways, small particles can easily pass and block the pores [14], so it may

accelerate blocking of solute particles into the membrane pores and lead to even more serious irreversible resistance [28].

### 3.3. Membrane integrity

Once the membranes were removed from the bioreactors, analyses such as scanning electron microscope (SEM) or FTIR were carried out in order to get information regarding the damage caused by the exposure of the membrane to sonication or differences in the cake composition which causes membrane clogging. Fig. 10 shows the results of the FTIR analyses for the membranes of the four modules and for a clean membrane in order to compare the resulting profiles. In general, some differences in the presence of some specific molecular groups can be observed after 2 months in operation. These profiles showed absorption peaks around  $1,010\text{ cm}^{-1}$ , which indicated the presence of hydroxyl groups (O–H bounds), i.e. polysaccharides. Peaks found at  $1,529$  and  $1,664\text{ cm}^{-1}$  corresponded to the presence of the amides structures of proteins. Both peaks indicated the presence of EPS in the membrane surface [32,33]. Peak at  $1,664\text{ cm}^{-1}$  was the only one in which the absorption was higher for the sonicated membranes than for the clean one, but values were similar for all the sonicated membranes. On the other hand, peaks around  $2,300\text{ cm}^{-1}$  and the bands of absorption observed in a range from  $1,400$  to  $1,600\text{ cm}^{-1}$  corresponded to aromatic compounds, but these bands were overlapped by those

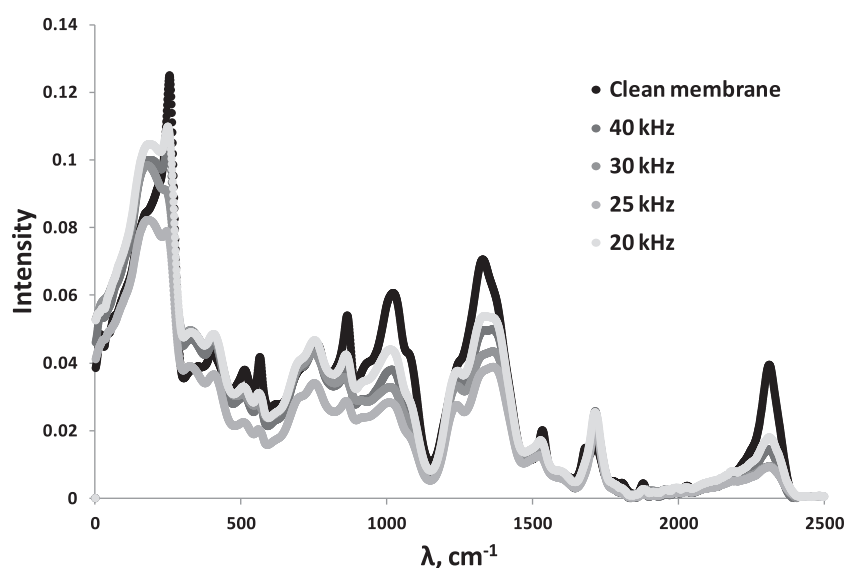


Fig. 10. Comparison of the FTIR spectrum for sonicated and clean membranes.

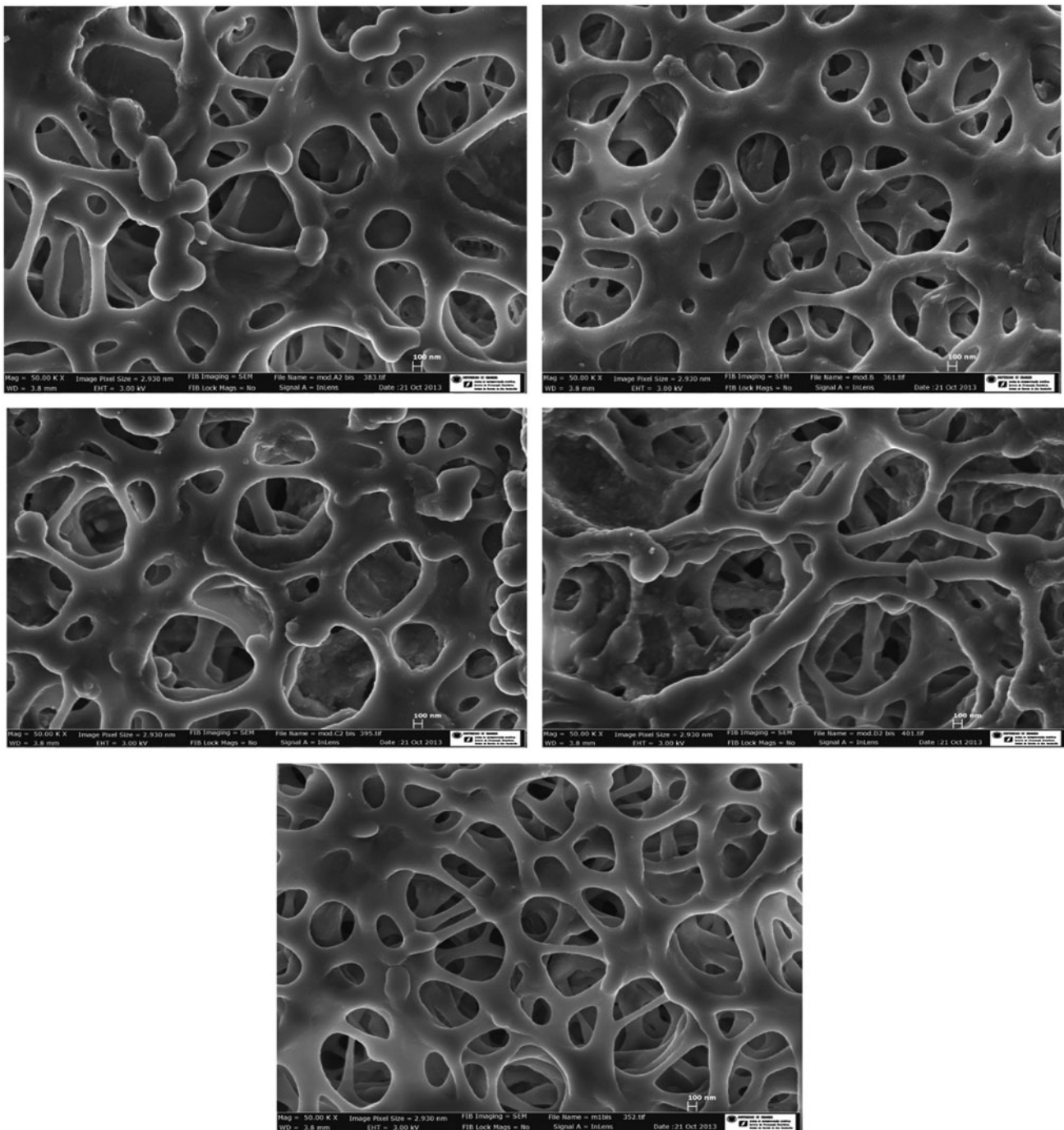


Fig. 11. SEM images of the membranes corresponding to 40 kHz (top left), 30 kHz (top right), 25 kHz (centre left), 20 kHz (centre right) and a clean membrane (bottom).

bands corresponding to the amides contained in proteins. Another peak found at  $860\text{ cm}^{-1}$  indicated the presence of carbonate and the formation of  $\text{CaCO}_3$ . The C–H absorption of alkanes ( $1,450\text{--}1,500\text{ cm}^{-1}$ ) was also overlapped by proteins absorption bands. Finally, several bands were observed in a range from 900 to

$1,100\text{ cm}^{-1}$  and in a range from 560 to  $1,320\text{ cm}^{-1}$ . The formers indicated the presence of C–C chains and the second ones were characteristics for aliphatic linear chains [34]. These results were similar to those published by authors such as Pendashteh et al. [31], who stated that the cake was mainly constituted by

EPS (proteins, polysaccharides, etc.), hydrocarbons and inorganic compounds.

Finally, membrane surfaces were observed using a SEM in order to evaluate if they were damaged because of the ultrasonic irradiation. According to the observed micrographs (Fig. 11), membrane integrity was considerably damaged after their exposure to ultrasound irradiation and the pore enlargement and, subsequently, the effect of the irradiation over the membrane surface was more significant at lower frequencies. Authors such as Masselin et al. [35] also analyzed the evolution of the polymeric structure of the membrane exposed to sonication and showed that important variations occurred on the polyethersulfone membranes after 2 h of ultrasonic irradiation. According to these authors, PES membranes were strongly affected as the permeability was more than 10 times higher after the exposure and the mean pore radius undergoes a 30% increase, resulting from the interconnection of neighbouring pores. On the other hand, Porcelli and Judd [5] found that some membranes were more susceptible to integrity failure than others and PES materials failed after 5 min of exposure at US frequency of 47 kHz. On the contrary, other researchers concluded that the integrity and microstructure of the membranes was maintained throughout sonication at similar frequencies and powers than those evaluated in this study (20 kHz and 156 W) [36]. These contradictory results regarding the effect of sonication over the effluent flux enhancement and the membrane integrity indicate that an inappropriate selection of membrane materials, ultrasound powers, frequencies and/or exposure times may damage the membranes and caution must be taken in order to select the best values for these parameters.

#### 4. Conclusions

According to these results, the following conclusions may be derived:

- (1) Most of the analytical parameters evaluated in the lab showed a high stability, with no differences due to sonication. Parameters such as activated sludge TSS concentration, effluent COD, viscosity,  $Ab_{5254}$  or colour showed a high stability and similar values for all the effluents coming from the sonicated modules which operated at frequencies of 20, 25, 30 and 40 kHz.
- (2) On the contrary, effluent turbidity showed higher instabilities, reaching values as high as 20 NTU at higher powers (300 and 400 W) and lower frequencies (20 kHz). Significant

differences when these results were compared with those obtained for the effluent from a conventional MBR system (always below 2 NTU) were observed.

- (3) The amount of particles found in the effluent coming from the conventional microfiltration system showed a high stability and lower values than the sonicated effluents, which contained an amount of particles significantly higher than the expected for conventional microfiltration MBR systems. Significant amounts of particles higher than the membrane pore size appeared in the effluent, especially in those effluents from the modules which operated at lower ultrasound frequencies, indicating that sonication greatly affects membrane integrity.
- (4) An increase in the temperature due to the ultrasonic irradiation was observed.
- (5) Sonicated membranes operated at stable TMP just during 20 d except for the module which operated at 20 kHz, whose TMP was stable for almost 70 d.
- (6) FTIR analyses showed some differences in the presence of specific molecular groups in the membrane surface after 2 months in operation. These results also showed that the cake was mainly constituted by EPS (proteins, polysaccharides, etc.) and hydrocarbons.
- (7) SEM images showed that ultrasonic irradiation damaged membrane integrity and that at lower frequencies, the effect of the irradiation over the membrane surface was more significant and the pore enlargement was higher.
- (8) Care must be taken in order to select the most appropriate operational conditions which are able to reduce membrane fouling without affecting membrane integrity.

#### Acknowledgements

This study was supported by funds from the European Union and the Spanish Ministry of Economy and Competitiveness within the framework of the program INNPACTO (IPT-2011-1078-310000). It was conducted at the University of Granada with the collaboration of CADAGUA S.A.

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