Study of ultrasonically enhanced chemical cleaning of SWRO membranes at pilot plant scale

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Fouling accumulated on reverse osmosis (RO) membranes during operation is one of the main problems affecting seawater desalination processes. This phenomenon causes a deterioration of the permselective properties of the membranes, which turns into a loss of performance of the process and costs increase. Conventionally, recovery of the process performance in desalination plants is carried out periodically by means of physical and chemical cleaning stages. However, conventional cleaning does not manage to recover completely the membrane properties and eventually can damage the membrane materials. New membrane cleaning techniques have been developed in order to improve this recovery. Ultrasound (US) radiation, which has shown to be an interesting technique during filtration since it avoids fouling deposition and allows to lengthen the period between cleaning stages, is proposed as an alternative technique to remove membrane fouling. This work investigates the effect of the combination of chemical cleaning methods and US application to clean a RO membrane from a desalination plant. The experiments performed were able to determine the best operating conditions to carry out the US cleaning protocol. Sodium hydroxide 2% w/v and sodium dodecyl sulphate 4% w/v solutions at 25º C were used, as they showed the highest recovery of the membrane properties in the chemical cleaning tests. Results showed that cleaning by US had a positive effect on the membrane selectivity (increase by 15.2%), and a low significant effect on its permeability. The utilization of the chemical cleaning combined with US improved the permeate flux considerably, without modifying salt rejection index in a significant way. Among the two cleaning solutions tested, the best results in terms of permeability and selectivity of the cleaned membrane, were those obtained by the US procedure using NaOH 2% w/v cleaning solution at 25 ºC.

1. Introduction

Membrane processes for water treatment have experienced great development in recent decades, as they represent a suitable solution to the growing demand for potable water and the reduction in availability of clean freshwater resources. One of the main difficulties affecting the membrane processes is membrane fouling [1]. It accumulates on the membrane surface during operation, causing worsening of its permselective properties and a reduction of the process performance [2]. Required pressure increases due to fouling and the power costs increase consequently. In high pressure processes, such as SWRO, these costs represent a significant part of the operation costs [3].

In order to prevent and minimize fouling, improvements in the water intake and the pretreatment can be adopted, such as implement subterranean intakes that minimize biofouling or the inclusion of a unit of UF membranes that improves the pretreatment. Even these strategies could improve the quality of the feed stream, periodic cleaning procedures are nowadays compulsory in order to keep the process performance and to remove the membrane fouling. The cleaning efficiency of these procedures depends on factors such as the fouling nature, membrane nature and the cleaning stage conditions. The optimization of these procedures in every specific case is advisable [4], since fouling phenomenon varies considerably depending on the water origin and it is of complex nature, being composed of organic, inorganic and microbiological compounds. Therefore this recommendation needs to be considered for seawater reverse osmosis desalination, on which this work focuses.

Conventionally, cleaning procedures applied in seawater desalination plants are composed of physical and chemical cleaning stages. The physical cleaning methods most widely used are flushing (pumping clean water for the feed stream) and backflushing (pumping clean water for the concentrate stream) for spiral wound modules and also backwashing (pumping clean water for the retentate stream) only for hollow fiber membranes. Chemical cleaning usually employs alkaline, acid, surfactant, chelating or enzymatic solutions, and it is known to be more detrimental to the membrane if concentrated solutions are used, especially when cleaning frequency needs to be increased [5].

In these cases, new techniques that work against fouling in the most efficient way are being developed for that reason, by either preventing it, minimizing it, or improving its removal. The application of magnetic and electric fields, cleaning by hypersaline solutions or cleaning with ultrasounds (US) are some of those novel techniques.

US application was developed many decades ago and it has been successfully implemented in different areas like medicine, physiotherapy o civil engineering. In the industry, it is currently applied in leak detection, cleaning of components, solid-liquid extraction in the alimentary industry [6, 7] and many other applications. In membrane technology, its application is still under research. Although there are numerous works to improve either membrane cleaning [5, 8-18] or membrane filtration [1, 19-22] and some of them focus on RO membranes [23, 24], all of them are bench-scale or pilot plant scale.

The efficiency of US to minimize fouling deposition is due to the ability of the ultrasonic waves to transmit substantial amounts of mechanical power through small mechanical movements [1]. Transmitted waves through a liquid with enough power may be able to exceed the attractive forces among the liquid molecules and cavitation bubbles will form. Cavitation bubbles can be as large as 100-200 µm and collapse quickly. The collapse has significant mechanical and chemical effects in aqueous systems, since each bubble can generate temperatures of about 4000-6000K and pressures of 100-200 MPa [1].

Ultrasonic radiation can be used in the submergible water intake to prevent biofouling. Sonication used in this way precludes the need to use other biofouling elimination procedures such as water/air jets, chemical treatments, or biocides and reduces the cleaning requirements of the membrane [25].

Membrane filtration enhanced by ultrasound has been investigated in order to minimize fouling and biofouling during filtration, by soaking the membrane module in the ultrasound bath. This technique does not affect the intrinsic permeability of membranes

[8, 9], but it increases permeate flux by minimizing the concentration polarisation effect and avoids fouling deposition, so the time period between cleaning steps can be prolonged [19]. These results were also confirmed for RO membranes in a work about wastewater filtration enhanced by US, where the US radiation allowed to recover significantly the permeate flux with no decrease in rejection [23].

Ultrasonic radiation may also be of great interest when it is applied as a cleaning step, either by itself or combined with other methods. Several works have focused on the combination of US with physical cleaning methods (forward flushing [5], backwashing [20], electric fields [21]), as well as others have analysed its combination with chemical cleaning methods for MF, UF and NF membranes [10-14~~6~~]. Some ~~of these~~ works [15, 16, 26] indicate that ultrasonic radiation can improve the cleaning efficiency of conventional cleaning methods for these types of membranes.

Ultrasonically enhanced chemical cleaning of RO membranes had not been so much widely studied as other type of membranes, but there are some works that focused on it. One of them studied acid and alkali cleaning enhanced with US and concluded that the best procedure to remove fouling of used RO membranes was oxalic acid cleaning plus US radiation. It achieved a defouling rate of 91% and it would shorten cleaning time and reduce reagents costs if compared with other commonly used acids [24].

Several parameters can affect the influence of the US in the cleaning efficiency, such as frequency, power, temperature, pressure or cross-flow velocity, as the previous works on US cleaning of MF, UF and NF membranes have reported. According to literature, lower US frequencies achieve better results than higher frequencies, as well as higher cleaning efficiencies have been obtained by mean of high US powers [1]. Cleaning efficiency increases linearly with sonication power and cross-flow velocity, whereas it decreases with the transmembrane pressure applied, as it was reported in a recent work about the combination of US and chemical cleaning applied on ceramic UF membranes [16]. The effect of temperature on the UF cleaning efficiency is not straightforward, since some works have shown a positive effect [17, 18] in contrast to other ones that suggest the opposite [5]. Other works focused on UF membranes indicated that the temperature effect was not significant when chemical cleaning was enhanced by US [15].

The present work investigates the application of an US cleaning procedure to clean SWRO membranes used in a seawater desalination plant. It also compares this procedure with the combination of US and chemical cleaning. In comparison to other works that employ artificial solutions composed of proteins or other compounds, the fouling phenomenon observed in this work is considerably more complex, since it derives from real seawater and foulant deposition has occurred during the whole membrane lifetime. Thus the effect on the permselective properties of the membrane is expected to be more pronounced and, consequently, more difficult to remove.

2. Materials and methods

2.1. Membranes

A spiral-wound SWRO membrane module SWC3 model (Hydranautics), which came from a seawater desalination plant, was used in this work. The membrane was removed from the plant after several years of operation, so it presented severe fouling.

2.2. Chemical cleaning agents

Sodium dodecyl sulphate (SDS) and sodium hydroxide (NaOH), both from Panreac (Spain), were used as cleaning agents in the chemical cleaning tests, because they achieved the best results during the static cleaning performed in a previous work by the authors [27]. The selected solutions (NaOH 2% p/v and SDS 4% p/v) maximized the recovery of membrane properties in static cleaning at 25 ºC.

2.3. US module and equipment

Pieces of 500x100 mm of the membrane, permeate collector and flux distributor were cut out to carry out the experimental tests of US cleaning. They were spiral-wounded together in order to simulate the original configuration and packed in a polyethylene module with 45 mm diameter x 165 mm length. The module was soaked in distilled water inside of the US device. US equipment was a USC500D model from VWR (Belgium), whose irradiation frequency was 45 kHz, its maximum power was 200 W and whose bath temperature and US power were adjustable.

2.4. Experimental methodology

2.4.1. Preliminary tests to define the US cleaning procedure

Preliminary tests were carried out in order to set the appropriate parameters for the US cleaning stage. Bath temperature evolution during cleaning time was monitored in these tests. Two different US power values (70 and 100% of the maximum US power) and two starting temperatures (25 and 40 ºC) were evaluated. The values of cleaning time, US power and starting temperature considered to perform the US cleaning procedure were selected according to the following criteria:

- maximizing cleaning time, as long as it is included in the range of values of the literature [1, 5, ~~8-20,~~ 22], so the effect of the cleaning agent on the fouling can be seen.

- maximizing US power, as long as the selected value is included in the range of the literature reviewed, since higher power achieves higher flux recovery [8, 9, 20, 22].

- not exceeding the maximum temperature tolerated by the membrane (45 ºC), because it might be irreversibly damaged.

2.4.2. Cleaning tests

A cleaning and characterization procedure to analyse the influence of US and the combination of US and chemical cleaning in the cleaning efficiency was defined. It was composed of three stages: cleaning, rinsing and characterization of the membrane properties (permeate flux, J_P ($L·h^{-1}·m^{-2}·bar^{-1}$); and salt rejection index, SRI (%)). During the cleaning stage, the chemical solution was recirculated through the US module at 26 L/h with no pressure by means of a peristaltic pump, while the US module was soaked in the US bath and irradiated at 45 kHz, as figure 1 shows. After the cleaning stage, the membrane was rinsed with distilled water following the same procedure. Finally, permeate flux and salt rejection index of the membrane were characterized in the pilot plant attending to the specifications of the membrane manufacturer (NaCl 32000 ppm, 25 ºC and 55 bar), similarly to the previous works by the authors [27, 28].

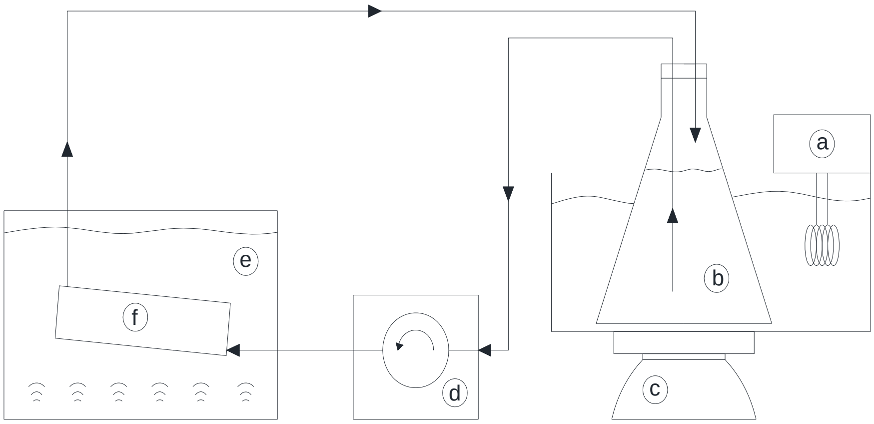

Figure 1. Experimental equipment used to perform the cleaning tests. a) immersion thermostat; b) cleaning solution; c) magnetic stirrer; d) peristaltic pump; e) US equipment; f) membrane module.

Additionally, the fouled membrane and membrane cleaned with US (fouled membrane submitted to US radiation using distilled water as cleaning solution) were included in all the experiments performed. Every cleaning protocol was tested on eight-membrane samples and the results displayed are the average values (mean relative error was lower than 10 %). Figure 2 shows a diagram of the experimental methodology followed to evaluate the effect of the US radiation and the addition of the chemical cleaning to the US on the cleaning efficiency.

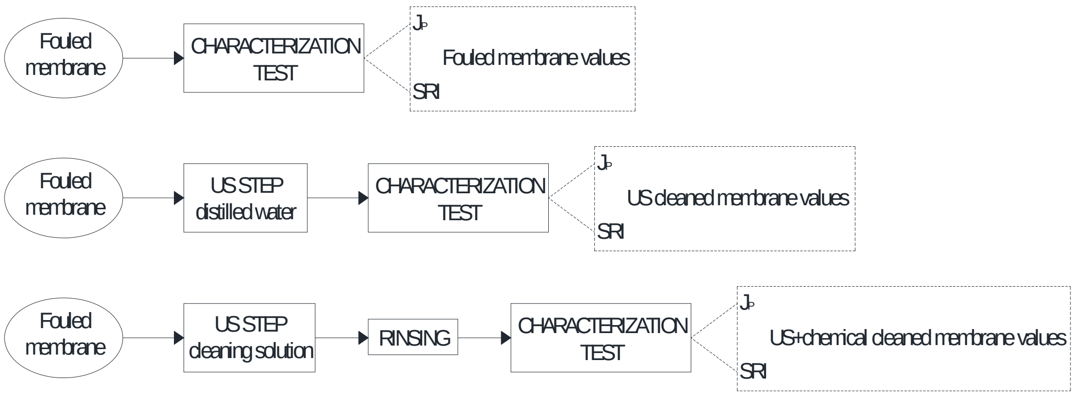

Figure 2. Experimental methodology followed to evaluate the effect of the US radiation and the addition of the chemical cleaning to the US on the cleaning efficiency.

Permeate flux (J_P) and salt rejection index (SRI) values were calculated from the experimental values obtained in the pilot plant tests, according the equations 1 and 2.

$$J_P\ (L/m^2 \cdot h \cdot bar) = \frac{V_P}{A_m \cdot t \cdot P_r} \qquad \text{[Eq. 1]}$$

$$SRI\ (\%) = \frac{\Lambda_0 - \Lambda}{\Lambda_0} \cdot 100 \qquad \text{[Eq. 2]}$$

where V_P is the permeate volume collected, A_m is the effective area of the membrane samples (0.003 m^2/sample), t is the sample time (15 min), P_r is the operating pressure (55 bar), Λ_0 is the conductivity at 25 ºC of the feed and Λ is the permeate conductivity at 25 ºC.

3. Results

3.1. Preliminary tests to define the US cleaning procedure

Ultrasonic power is transmitted to the water contained in the US bath as heat [17], so water temperature increases during the cleaning stage. In order not to exceed the maximum temperature recommended by the membrane manufacturer, preliminary tests were performed to evaluate two different values of US power, called P (70 and 100%) and two different starting temperatures, called T_i, (25-40ºC). Results are shown in figure 3.

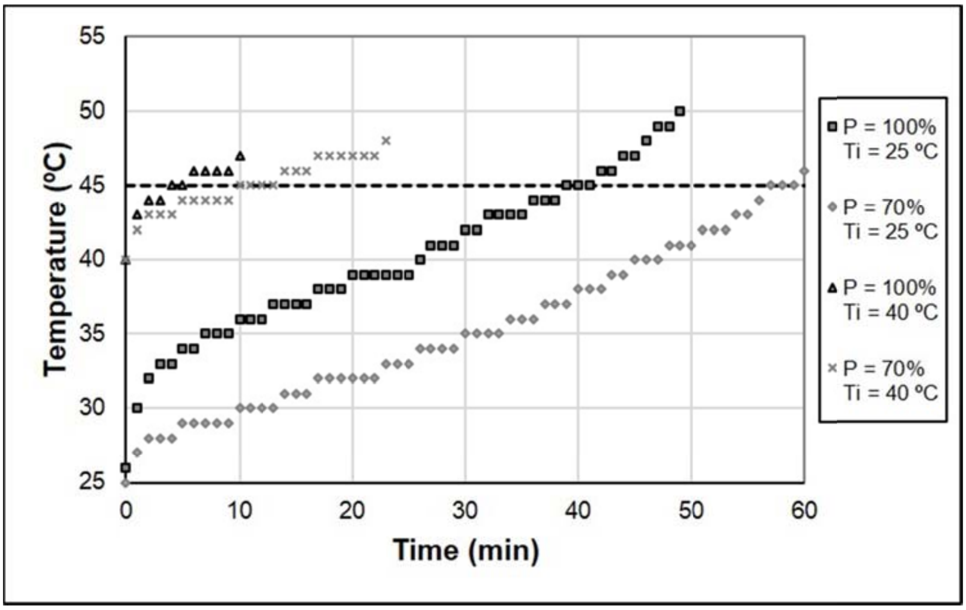

Figure 3. Temperature evolution with time at different US powers and starting temperatures.

As it can be observed in figure 3, the US bath temperature reached 45 ºC in less than 10 minutes for both US power values tested when the starting temperature was 40 ºC. Therefore starting the cleaning step at 40 ºC was dismissed, since cleaning times greater than 10 minutes must be considered in order to be able to appreciate the effect of the chemical cleaning.

When the starting temperature was 25 ºC, the limit temperature was reached in 39 minutes when US power applied was 100% of the nominal value of the equipment, whereas this time could be prolonged up to 57 minutes when 70% of the nominal US power value was applied.

Considering the temperature evolution and according to the previously exposed criteria, the cleaning time was decided to be 30 minutes, the US power was fixed at 70% of the maximum US equipment power and the starting temperature selected was 25 ºC. Higher values of US power and cleaning time were dismissed in order to ensure that there is no risk of damaging the membrane due to the increase of the temperature.

Finally, it can be stated that the increase of temperature of the solution caused by the application of US can become beneficial as it could allow to reduce the duration of the chemical cleaning, since an increase of temperature of the cleaning solution have been proved in a previous work [27] to have a positive effect on its efficiency.

3.2. Cleaning tests

Permeate flux and SRI values obtained in the tests performed for the fouled membrane, the US treated membrane and the membranes cleaned by combination of US and chemical cleaning are shown in figure 4. Bars represent mean value and error bars standard deviation, respectively, for the eight tested membranes under each experimental condition.

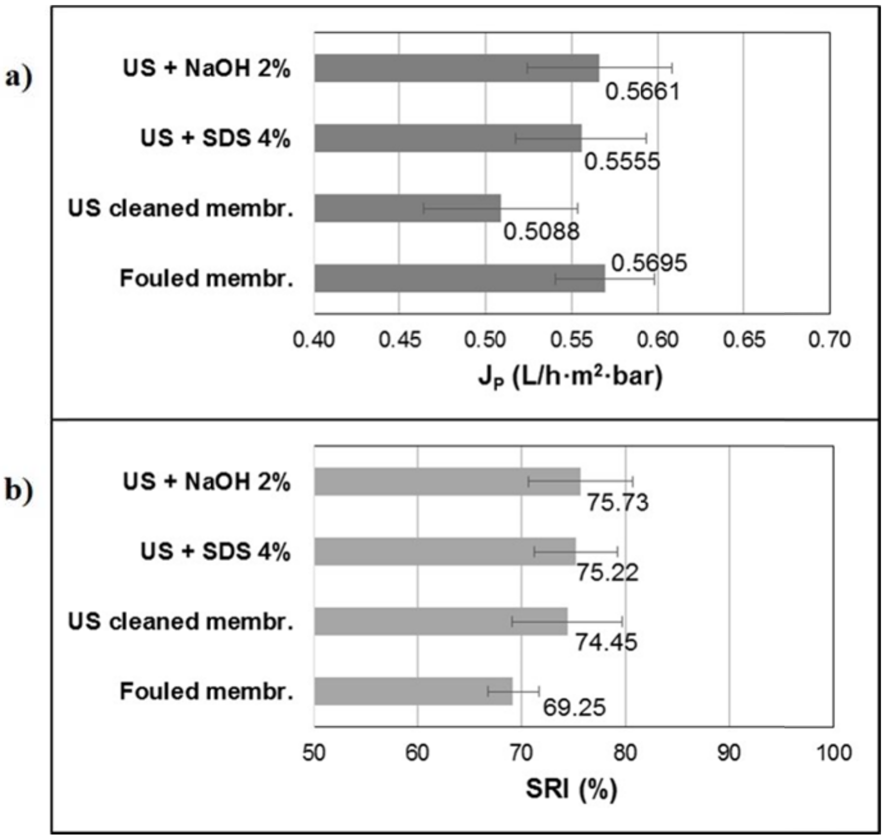

Figure 4. Results of the characterization tests performed in the pilot plant. a) Permeate flux values; b) SRI values.

A) Effect of US cleaning

Results of the pilot plant tests indicated that the fouled membrane presented a permeate flux of 0.5695 $L \cdot h^{-1} \cdot m^{-2} \cdot bar^{-1}$ and a salt rejection index of 69.25%, which pointed to a severe fouling affecting mainly to the selectivity of the membrane.

US cleaned membrane showed a J_P of 0.5088 $L \cdot h^{-1} \cdot m^{-2} \cdot bar^{-1}$ and a SRI of 74.45%. In comparison to the fouled membrane values, it can be observed that US cleaning recovered 5.2% of the SRI (fig 4b), which is the property most affected by fouling. However, the permeate flux decreased up to 10.66% when the fouled membrane was treated with US (fig 4a).

B) Effect of cleaning with US and chemical cleaning

As it can be observed in figure 4, the cleaning with US irradiation simultaneous to the circulation of the alkaline solution (NaOH 2% w/v) was the combination that achieved the best results. It achieved the maximum value of SRI, 75.73%, and the second maximum value of J_P, 0.5661 $L \cdot h^{-1} \cdot m^{-2} \cdot bar^{-1}$. In comparison to the fouled membrane, it obtained a SRI recovery of 6.48% and a trivial decrease (0.6%) of J_P.

The combination of US with the surfactant cleaning (SDS 4% w/v) also had good results, since it obtained a SRI of 75.22% and a permeate flux of 0.5555 $L \cdot h^{-1} \cdot m^{-2} \cdot bar^{-1}$. In comparison to the fouled membrane, these values meant a SRI recovery of 5.97% and a trivial decrease (2.46%) of J_P.

Therefore, the values of SRI obtained by combination of chemical cleaning and US were slightly better than the values obtained by US (fig 4b). Moreover, when US was combined with chemical cleaning the permeate flux did not decrease significantly, whereas it decreased up to 10.66% when US was applied without combination of chemical cleaning.

However, as the final values of SRI obtained by combination of US and chemical cleaning were not as good as they could be expected, it will be considered to extend the chemical cleaning and US irradiation time or cleaning procedure (static and static-dynamic cleaning) in order to valuate these effects in a future research work.

3.3. Industrial application of the ultrasonically enhanced chemical cleaning

Once the experimental results of the application of US combined with chemical cleaning have been shown, the possible application at industrial scale in order to improve the efficiency of the chemical cleaning or fouling prevention during filtration in desalination plants is considered. In these plants, membrane modules are placed in the interior of pressure vessels, which are supported on a metallic holder that holds a great amount of vessels. The vessels are placed in several rows and columns and the ensemble is called rack.

The application of US in SWRO desalination plants would imply a modification of the racks distribution, and the main challenges to make this viable are the space requirements and costs related to the inclusion of new equipment. In the current racks, space between rows and columns of vessels is quite limited and there are pipes and auxiliary equipment also placed next to them. This would be a problem to include and operate new equipment such as US. The design for the application must consider the current arrangement of the rack and should be adapted specifically. In this work, a draft of the design is suggested, considering the increase of the rack surface to allow the placement and movement of the new US equipment and allowing its accessibility and compatibility to the current system. However, an analysis to determine the increase of the space between vessels and the increase of the plant surface needed is subsequently required to ensure the viability of the application.

Besides the analysis of the space requirements previously mentioned, an evaluation of costs of the new equipment and power consumption versus the benefits of US on the cleaning process performance would be also required in order to ensure the viability of the implementation of US in the system.

Three elements per rack would be necessary to implement US in a rack: holders or guides, mechanical arms and US emitter as it is shown in Figure 5. These three elements would operate always under control of an automaton that will be programmed in such a way that US radiation is applied in all vessels of the rack consecutively during its application.

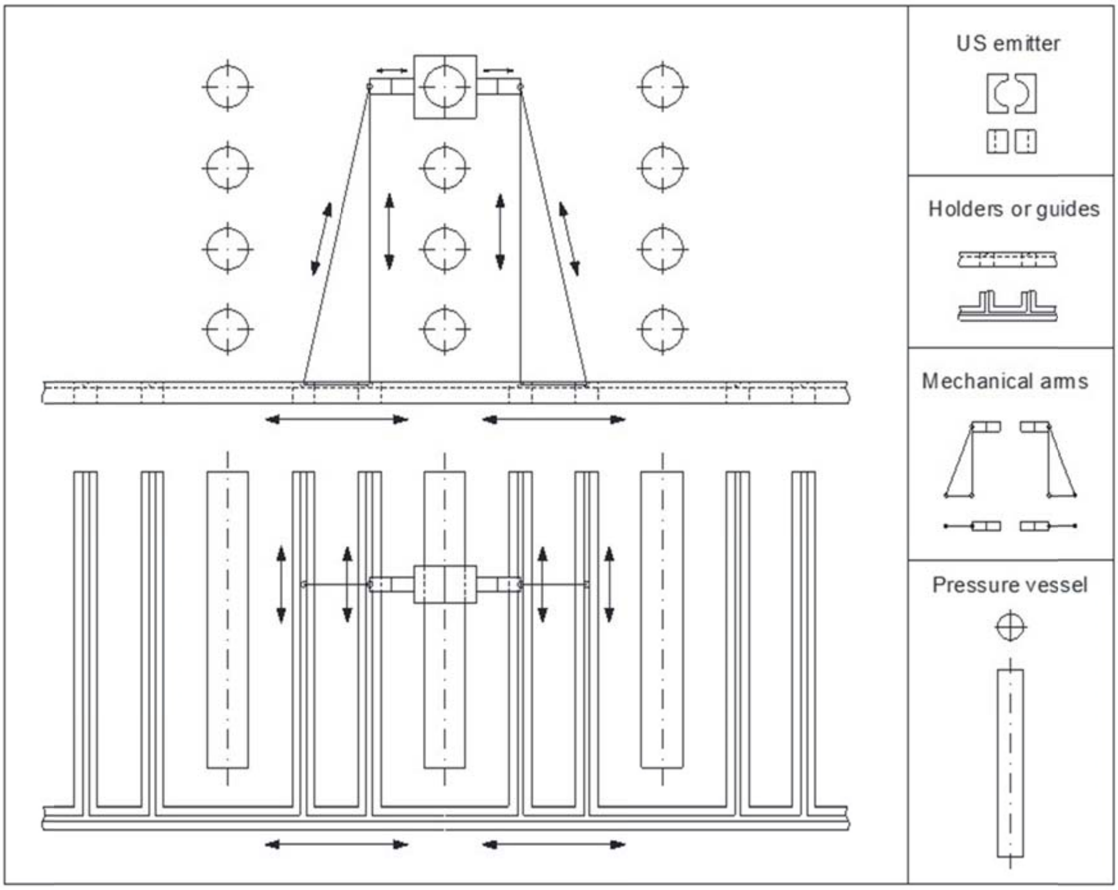

Figure 5. Draft design of the application of US equipment in a current rack of a SWRO desalination plant.

The holders or guides would be placed on the floor under the rack and their function would be to lead the mechanical arms along the whole rack, allowing the access of the US emitter to all columns and rows of vessels. There would be two guides parallel to the vessels between columns and two more at both sides of the rack. The mechanical arms would move through these guides to apply the US along the whole vessel. When the US is applied on a vessel, the mechanical arms would move to the extreme of the vessels, where they would have free space to change the height and have access to vessels placed in different rows of the same column. Besides these guides, there would be other perpendicular guide placed at the extreme of the vessels, which would connect all the other guides and would make possible to change the column of operation.

There would be two symmetric mechanical arms, which would operate at both sides of the pressure vessel, following the guides. Each mechanical arm would hold a half of the US emitter and allow its vertical movement and closeness to the vessel. Their functions would be: firstly, when they are at the extreme of the vessels, to lead the guides to the column target and put the US emitters at the height of the row of the vessel target; secondly, to direct the two parts of the US emitter until they completely enclose the vessel target; and thirdly, to move axially the US emitter along the vessel at a speed that ensure US radiation has effect on all the membrane modules contained in the interior of the vessel.

The US emitter would be composed of two symmetric parts whose shape is shown in Figure 5. The radius of the semicircular shape of the US emitters should be equal to the external radius of the pressure vessel, so they can enclose the vessel. US emitter should be able to regulate wave frequency and power to optimize the effect of the US on the membranes.

The installation of these new equipments in the SWRO desalination plant would allow to apply US in the process in three different ways at least: US application during membrane filtration to prevent fouling deposition on the membrane, US application during daily physical cleaning as a part of the physical cleaning procedure, and US application during chemical cleaning to improve the cleaning efficiency.

In case the industrial application in SWRO became viable, several parameters such as the US power and frequency, the periodicity of application or the scanning speed of the mechanical arms should be determined from pilot plant tests and real scale tests to optimize the effect of US radiation on the process performance.

4. Conclusions

The SWRO membrane used in the present work presented severe fouling after several years of operation, affecting mainly the SRI values.

Cleaning by US had a positive effect on the membrane selectivity (increase of 5.2%), but a negative effect on its permeability (decrease of 10.66%).

US application causes an increase of the solution temperature, which might improve the efficiency of the chemical cleaning and reduce the cleaning time required.

The utilization of chemical cleaning combined with US allowed to improve the salt rejection index without modifying the permeate flux in a significant way.

The best results obtained in terms of both permeability and selectivity of the cleaned membrane, were those that corresponded to the US cleaning procedure combined with chemical cleaning by means of $NaOH$ 2% w/v solution at 25 ºC, achieving an increase of SRI of 6.48% without significant variation of permeate flux (decrease of 0.6%).

A draft design for the industrial application of US radiation in SWRO desalination plants was considered. However, exhaustive analysis of space requirements and costs versus the benefits of including the new equipment would be needed to evaluate the viability.

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