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Effect of Ultrasonic Frequency on the Performance and Cleaning of UF Membranes Used in Sea Water Desalination Pretreatment

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Abstract
This study aims to evaluate the impact of ultrasound (US) on the performance and cleaning of ultrafiltration membranes (UF) used in the pretreatment of sea water desalination plants. The effects of ultrasonic frequency on flux and fouling during the ultrafiltration process to saline solutions contaminated with Humic Acid (HA) were investigated. Ultrasonic application had a strong impact on the normal ultrafiltration process, especially at low frequency. The estimated increase in the flux were 67% and 38% when applying ultrasonic irradiation on dead end flat sheet ultrafiltration cell at 300 W and frequencies of 28 kHz and 40 kHz, respectively, but there was no significant increase in the flux at the upper frequency 100kHz where the flux nearly identical to that was obtained by stirring only. Upon ultrasonic application, total filtration resistance and reverse resistance, which include concentration polarization and cake layer, were sufficiently reduced as revealed by the quantitative resistance analysis using resistance in series model. The flux recovery ratio was increased to 72% when 28 kHz frequency was applied during mechanical and chemical cleaning. Ultrasonic enhancement factor (E_{US}) was higher by 11% in the online application of US on UF process than that of cleaning process. The application of ultrasound is an effective and promising way to enhance the ultrafiltration and cleaning of fouled membranes used in the pre-treatment of the seawater desalination process.
Keywords: ultrasound frequency, ultrafiltration, pretreatment, desalination of sea water

1.Introduction

The main drawback of membrane pre-treatment in Sea Water Reverse Osmosis desalination plants (SWRO) is membrane fouling associated with particulate matter/colloids, organic/inorganic compounds, and biological growth [1]. Various studies have been carried out to find the factors affecting organic fouling. The dissolved organic matter in sea water is not a single substance but a mixture of many aliphatic and aromatic compounds. However, among the total dissolved organic substance in seawater, 90% of them are represented by the humic materials or substances [2]. Humic substances (HS) are generated from the degradation of organic matter and they are mostly constituted of humic acids (HA) and fulvic acids (FA). Humic and fulvic acids possess a significant negative charge density and a bulky macromolecular shape [3].

The commonly hydraulic methods used to reduce membrane fouling such as forward flushing and backward flushing are time consuming due to the interruption of operation process for cleaning. Chemicals applied in chemical cleaning sometimes are more likely to erode membrane materials and result in secondary pollution. Electrical techniques used for permeate flux enhancement has the potential danger of electrolysis taking place at the electrodes [4]. The use of magnetically assisted filters has not yet been commercially adopted. Therefore, development of effective method to address membrane cleaning problems is a critical step for sea water desalination plants.

Recent studies have shown that ultrasound provides an alternative technology for membrane fouling control [5]. Ultrasound has significant advantages over traditional methods to control membrane fouling. For example, there is no chemical use in the membrane cleaning process. The ability to maintain overall higher permeate fluxes through continuous and pulsed use of ultrasound during filtration reduces filtration driving pressure and total membrane area. Moreover, ultrasound is considered as a novel technology able to reduce the total processing cost while maintaining or enhancing product quality in an environmentally benign manner [6].

Ultrasound is longitudinal wave with a frequency above 20 kHz. This frequency is above the sonic range (20 Hz to 20 kHz) at which humans can hear and below the megasonic region (>600 kHz) [7].

For the purposes of ultrasonic cleaning the main method to generate ultrasound is through the use of piezoelectrics. Piezoelectric materials convert electrical energy into mechanical energy and vice versa. This behavior was discovered in 1880 by Jacques and Pierre Curie [8]. They demonstrated that several natural minerals are able to generate electrical signals as result of mechanical stresses. They later found that the reverse would also be possible to apply an electrical charge and get a mechanical response. Transferring this mechanical response from one medium to another results in energy transfer. In membrane cleaning systems this would be transfer of energy from the piezoelectric material to a solution.

Most ultrasonic cleaning devices work on the principle of cavitation. As a result of the propagation of ultrasonic waves, the medium is subjected to alternating rarefaction and compression cycles. If during the rarefaction stage of the cycle, the tensile stress exceeds the tensile strength of the fluid (i.e., water), a bubble cavity is formed. This cavity may: (1) dissipate back into the liquid, (2) grow to a resonant size and fluctuate about this size, or (3) grow to a size at which the surface tension forces of the liquid cause it to collapse on itself. The latter is termed cavitational collapse. Cavitational collapse results in extreme conditions producing light emission, shock waves and localized high temperatures (up to ~5000 K) and pressures (up to ~1000 atm). These high temperatures and pressures dissociate water into hydrogen atoms (H•) and hydroxyl radicals (OH•). More importantly with respect to membrane cleaning, cavitational collapse also produces a number of phenomena that result in high velocity fluid movement. Cavitation creates micro turbulence and creates high shear velocities in its near vicinity and can also create shock waves that travel radially from cavitation collapses. These forces are able to dislodge particulate matter from surfaces and enhance the dissolution of substances due to the increased mass transfer of liquid to surfaces [7]. Important variables of ultrasound are the frequency (the number of cycles per time) and the power intensity (the amount of power put into the system per unit area).

Recently, a number of researchers have demonstrated the effective use of ultrasound for cleaning fouled membranes or for increasing permeate flux of water through membranes.

Allen and Shippey [9] were the first researchers to investigate ultrasonic cleaning of membranes. They tested a number of different cleaning techniques and found that ultrasound was the most effective for cleaning reverse osmosis and ultrafiltration membranes.

In 1981, Thompson [10] patented a system to use ultrasound to scour off particles from reverse osmosis and ultrafiltration membranes. However, the system was to be operated below the cavitation threshold, possibly limiting mechanisms leading to the cleaning of fouled membranes. Kobayashi and Chai et al. [11, 12, 13, 14] continue work in this area, in these papers, they investigated fouled membrane cartridges submerged into an ultrasonic bath for treatment. They performed a number of experiments studying membrane type, frequency of the ultrasonic bath, and power intensity of the ultrasonic bath. They found that the ultrasound enhanced the filtration through the membrane. Lower frequencies and higher power intensities increased the membrane cleaning. They performed the first systematic experiments of ultrasound on membrane filtration. However, they lacked the ability to observe mechanistically how ultrasound affected membranes and whether or not there was any damage to the membrane.

Utilizing inorganic membranes, Kokugan et al. [15] found that ultrasound increased the mass transfer coefficient of water across membranes, but determined that ultrasound was not effective at removing fouling material inside pores. The rejection of fouling material was marginally decreased because of increased permeate flux. They utilized a probe system which irradiated approximately 1/8 of the membrane surface.

Li et al. [16] used a probe ultrasonic system to clean membranes fouled with paper mill effluent. They found that they were able to increase permeate flux and restore the original structure of the membrane surface. SEM images did not reveal any damage to the membrane surface.

Juang and Lin [17] used a low frequency probe to clean ultrafiltration membranes. They studied regenerated cellulose membranes and suspensions of organic foulants in the system with a 20 kHz probe aimed at the membrane surface. They found that the membrane experienced increased fluxes and could maintain high fluxes with ultrasound.

Masselin et al. [18] found significant damage to several types of polymeric membranes as a result of cleaning the membranes in an ultrasonic bath at 47 kHz. These membranes were not in a membrane module during the experiment rather they were placed in a beaker which was then put into an ultrasonic bath.

Most recently, Muthukumaran et al. [19, 20] looked at the use of ultrasound for cleaning ultrafiltration membranes in the dairy industry. The setup that they used was similar to that used by Kobayashi et al. [11-14] in that they had an enclosed membrane cell suspended in an ultrasonic bath. They found that the recovery in flux was independent of the sonication time and that there was an increase in flux with increases in ultrasonic power. They also used the membrane system in the ultrasonic bath for over a month and found that there was no change in the permeate flux, which indicated to them that there was no damage to the membrane.

Simon et al. developed an US-assisted dead-end UF system using ultrasonic probe at 20 kHz [21]. Obvious improvement of flux was obtained due to a decrease of the boundary layer resistance on the membrane surface.

In 2004, M. O. Lamminen et al. [7] itemized all possible ultrasound cleaning Mechanism and factors affecting particle-fouled ceramic membranes. With the assistance of Scanning electron microscope, they observed that mircojets, microstreaming and microstreamers are important in detaching particles from the membrane surface.

Most recently, H. Kyllonen et al. [22] found that the flux improvement was significant by low frequency ultrasound irradiation when using a cross-flow alumina-based ceramic membrane system for membrane filtration of industrial wastewater.

D. Feng et al. [23] employed on-line ultrasound for cleaning reverse osmosis membrane fouled by carboxyl cellulose solution. They demonstrated that permeate flux of the membrane increased significantly in the presence of ultrasound. SEM confirmed the beneficial effect of ultrasonication on membrane permeate flux.

D.Chen studied the effect of ultrasonic factors on the cleaning of particle- fouled membranes and found that the most effective control of membrane fouling occurred at high pH, low ionic strength, and the absence of divalent cations [24].

Cai M studied mechanisms to enhance ultrafiltration and membrane cleaning at different ultrasonic frequencies [25], and found that the flux could be recovered by 200% at 28 kHz.

Li X found that the flux can be restored by 95% at 12kW/m^2 and 40 kHz when considering the possibility of reducing the fouling of UF hallow fiber membrane [4].

2. Research Aim

The primary mechanisms by which ultrasound aids membrane filtration and cleaning may be affected by a number of factors, such as orientation and position of the ultrasonic field, ultrasonic power intensity and frequency, membrane material, membrane housing, operating pressure, and fouling material.

The aim of this research is to investigate the influence of ultrasonic frequency on the performance and cleaning of ultrafiltration membranes used in pretreatment of sea water reverse osmosis desalination plants.

3. Material and Methods

A model synthetic solution with constant specifications similar to sea water in salinity level and organic material content was used in all experiments. The solution was prepared by dissolving 30 mg of Humic Acid (Sigma Aldrich, UK) in 1L of sodium chloride solution (25000 mg/L NaCl).

A stirred Amicon dead-end ultrafiltration cell model 8200 (Millipore) was used for UF and membrane cleaning experiments. The maximum operating pressure of the cell is 5.17bar. The cell has a volume capacity of 200mL and could hold a membrane disc of 63.5mm in diameter. The effective membrane area is 28.7cm^2. The stirrer had a diameter of 48mm, the design of the body allowed the stirrer to be as close as possible to the membrane. After each run the cell is flushed with de-ionized water.

The polyethersulfone (PES) UF membrane (Nadir Filtration GmbH-Germany) with Molecular Weight Cut Off (MWCO) of 20 kDa was installed as flat sheet in the UF cell. Operating pressure required for the membrane cell was induced using oxygen-free nitrogen cylinder. The operating pressure was manipulated using a pressure regulating valve.

The experimental setup is shown in figure (1), the UF cell was immersed in ultrasonic bath (W-118, Honda Electronics Co., Japan), operating at 28, 45 and 100 kHz frequencies and adjustable power of 600W (50-100%).

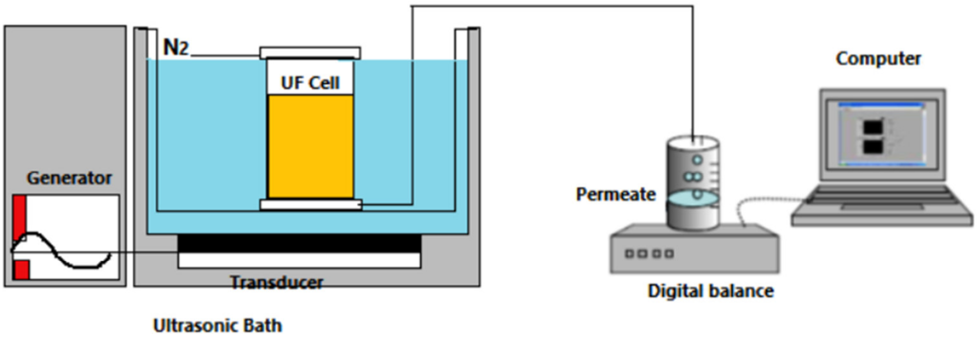


Figure 1: Experimental set up

200 ml of HA saline solution was transferred into the UF cell and the filtrations were operated at the constant trans-membrane pressure (TMP) mode by controlling TMP at 1 bar. The stirring speed was set at 300 rpm in each run. The permeate fluxes at various experimental conditions, namely frequency of 28, 45 or 100 kHz at a fixed power of 300 W were measured at a 5 min interval by an electronic balance.

Pure water flux J_w and permeate flux J are calculated using the formula [1, 26,27]:

$$J = \frac{V}{At} = \frac{\bar{Q}}{A} \qquad (1)$$

Where J is the pure water flux or permeate flux (L/m^2.h), V is the volume collected (L), A is the effective membrane area (m^2) and (t) is the time taken to collect the filtrate (hours).

According to resistance-in-series model [25,27,28] the permeate flux can be calculated by Darcy's Law:

$$J = \frac{\Delta P}{\mu R_{tot}} \qquad (2)$$

Where J is the permeate flux ($L.m^{-2}.h^{-1}$), ΔP the TMP (bar), μ the viscosity of solution (bar.h), and R_{tot} the total resistance (m^{-1}).
The total resistance R_{tot} can be defined as:

$$R_{tot} = R_m + R_{rev} + R_{irr} \quad (3)$$

Where R_m is the intrinsic membrane resistance (m^{-1}), R_{rev} the reversible resistance (m^{-1}) including concentration polarization and cake/gel layer resistances, and R_{irr} the irreversible resistance (m^{-1}) which cannot be removed by water flushing.
Each resistance can be determined based on experimental measurements, according to the procedures described by Simon with modifications as follows [21]

1) the flux of DI water was measured to obtain R_m
2) the permeate flux of HA saline solution was measured during the UF process to obtain R_{tot}
3) the HA saline solution was removed and the reversible resistance was washed away by 200 mL DI water;
4) the flux of DI water was measured again in the UF process at the initial 5 minutes to obtain the value of R_m+R_{irr}
5) the membrane was chemically cleaned by 0.1 M NaOH for 30 minutes.
6) The value of R_{rev} can be obtained based on the Equation (3).

Cleaning process of fouled membrane
200 mL of HA saline solution was used in each fouling experiment. The fouling process was operated at a constant TMP of 1 bar till the accumulated volume of permeate solution reached 160 ml. When the fouling completed, the retentate of the HA saline solution was emptied out, followed by mechanical cleaning with fresh DI water. 200 mL DI water was put into the cell and filtered at mild flow velocity with US irradiation. Instead of DI water, 200 mL sodium hydroxide of 0.1M solution was used in the chemical cleaning process with or without US after mechanical cleaning. DI water flux was determined right after every on-site cleaning stage.
Flux recovery, used as an index for the efficiency of the cleaning method is defined as follows [4,19,20,25]:

$$\Phi = \frac{J_{ac}}{J_w} \quad (4)$$

where Φ is flux recovery, J_{ac} the water flux after cleaning, J_w the water flux of original membrane.
The effect of ultrasound also determined by an ultrasonic enhancement factor [4,19,20] which specifically evaluates the effect of ultrasound on improving permeate flux in case of online application of ultrasound and ultrasound cleaning.

$$E_{US} = \frac{J_u}{J_n} \quad (5)$$

where, J_u is the permeate fluxes achieved with the presence of ultrasound and J_n is the permeate fluxes achieved without ultrasound

4.Results and discussion

4.1Effect of ultrasound frequency on UF Performance

The influence of ultrasonic waves application at frequencies of 28, 45 and 100 kHz, on the ultrafiltration process of HA saline solution is shown in Figure (2). The flux decreased gradually during the first stage of filtration when applying ultrasound at 28 kHz and 40 kHz, while the flux decreased rapidly and sharply at 100 kHz and without the application of ultrasound, this could be due to the faster occurrence of pore blocking which in turn increases the concentration polarization and forming thick compact fouling layer on the membrane surface in the second case, while the opportunity of pores blocking was less and concentration polarization was weak in the first case and thinner and less dense fouling layer was formed.

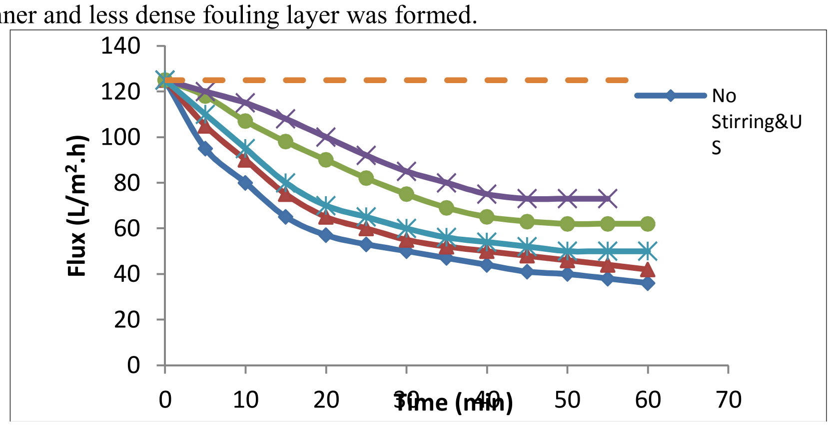


Figure 2: Effect of US frequencies on the flux of the P020F UF membrane at TMP of 1 bar to filter a solution with salinity level 25000 mg/L NaCl and 30 mg/L HA at 300W US power

It is generally observed that the flux is increased when applying the ultrasonic waves at all frequencies compared to the flux obtained by mixing only or without it, that is due to permeability increasing and preventing the formation of fouling layer on the membrane surface, but the flux enhancement was inversely proportional to the frequency increase. The flux at 28 kHz increased by 74% from the flux obtained by mixing only and by 48% for the 40 kHz frequency. There was no significant increase in the flux at the highest frequency 100 KHz where it was almost identical to that obtained by mixing as the flux increased by only 19%.

This corresponds to the findings of Kobayashi in his studies using typical solutions such as dextran and peptone, where the flux with the 28 kHz and 45 kHz ultrasound application increased continuously during the entire filtration process [11-14].

Although higher ultrasonic frequencies cause more cavitation bubbles collapsing with time, but the bubbles at these frequencies are smaller in size and release less energy. They may not be capable to detaching particles from the membrane surface and cake layer as they do at lower frequencies where the bubble collapse violently.

According to Mason's results [29], an increase in ultrasonic frequency reduces the production and intensity of cavitation in the liquid. At very high frequency the rarefaction (and also compression) cycles are too short to allow the bubble to grow to a size sufficient to cause its disruption of the liquid.

As the ultrasonic frequency changes, the fouling resistances change. Figure (3) shows the effect of ultrasound on the reversible and irreversible fouling resistances with respect to total resistance.

The membrane self-resistance remained constant at all frequencies indicating that the ultrasound technique did not affect the original membrane resistance. The reversible resistance decreased by 64% and 55% at 28 kHz and 45 kHz respectively, while the reduction was 25% only at 100 kHz. This indicates that lower frequency may generates stronger vibration and greater energy due to the violent collapse of the cavitation bubbles, reducing the occurrence of concentration polarization and prevents the particles from accumulating and forming the cake layer and that is one of the US enhancement mechanisms, but increasing vibration with frequency stimulates Humic Acid particles to penetrate across the membrane and increase the adsorption on its pores which speeds up the clogging of pores and thus increasing irreversible resistance but it remains less than this obtained by mixing only and without the application of ultrasound case.

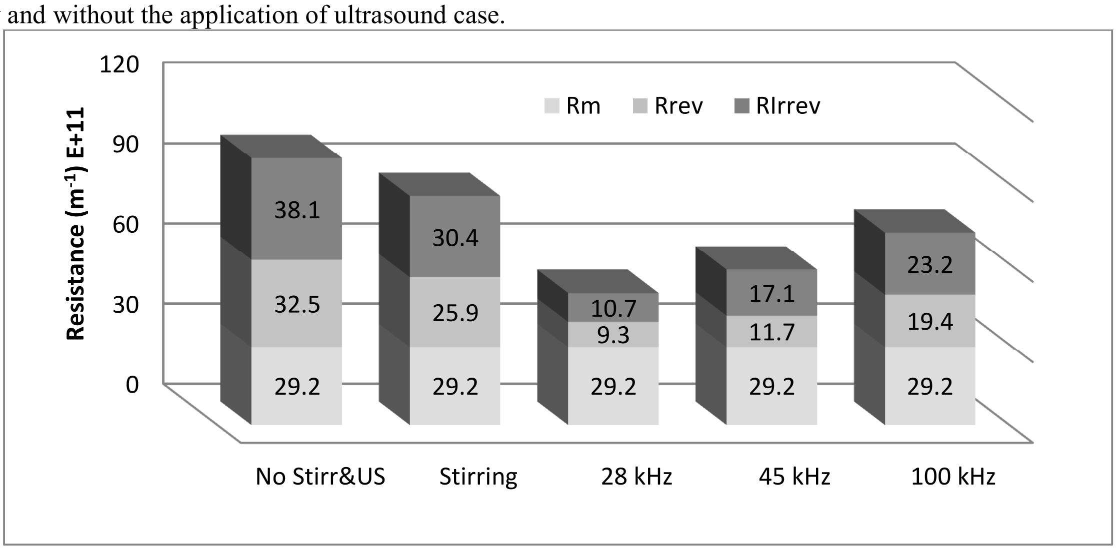


Figure 3: Effect of ultrasonic frequency on resistances of the P020F UF membrane at TMP of 1 bar to filter a solution with 25000 mg /L NaCl salinity and 30 mg /L HA at 300W US power

4.2 Effect of ultrasound frequency on UF cleaning

Mechanical cleaning was performed after fouling occurrence in the ultrafiltration process by running the deionized water through the fouled membrane, which resulted in a 30% flux recovery. The chemical cleaning was then applied with sodium hydroxide solution and the flux recovery ratio was increased to 36%.

Mechanical and chemical cleaning experiments were repeated by applying Ultrasonic waves at 28, 45, 100 kHz at constant US power of 300 W, the results were presented in Figure (4). The flux recovery ratio was increased to 72% when 28 kHz frequency were applied, ie 36% higher than without the application of ultrasound , While the frequency of 100 kHz did not have much effect on mechanical and chemical cleaning, as the flux recovery was the same in the case without the application of the ultrasound. This result indicates that the lower ultrasonic frequency was more effective in enhancing the cleaning process. The effect of low ultrasonic frequency may correspond to cleaning with its effect on the UF process.

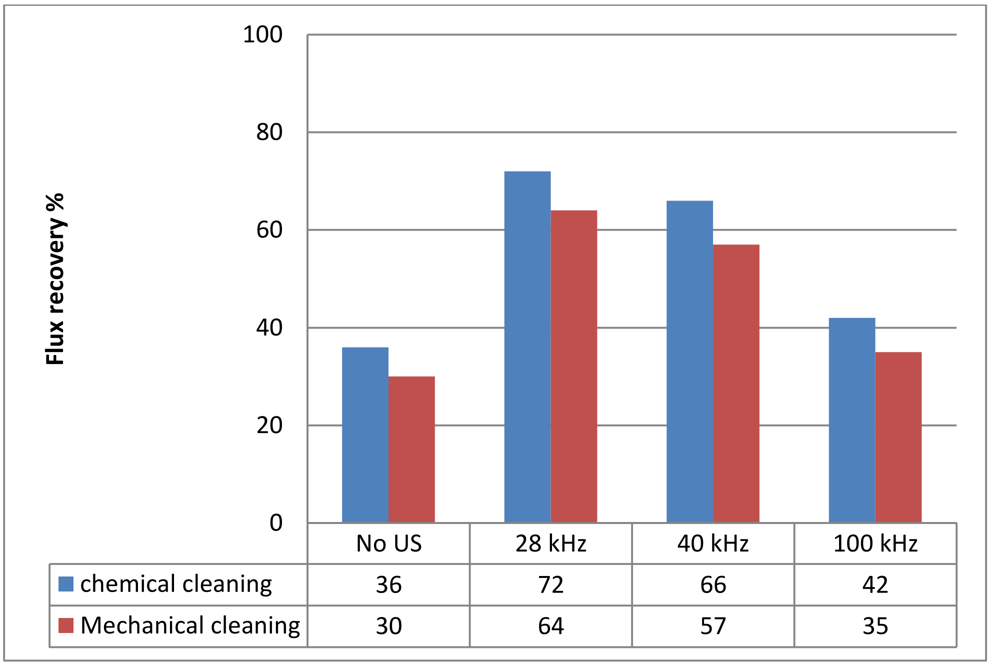
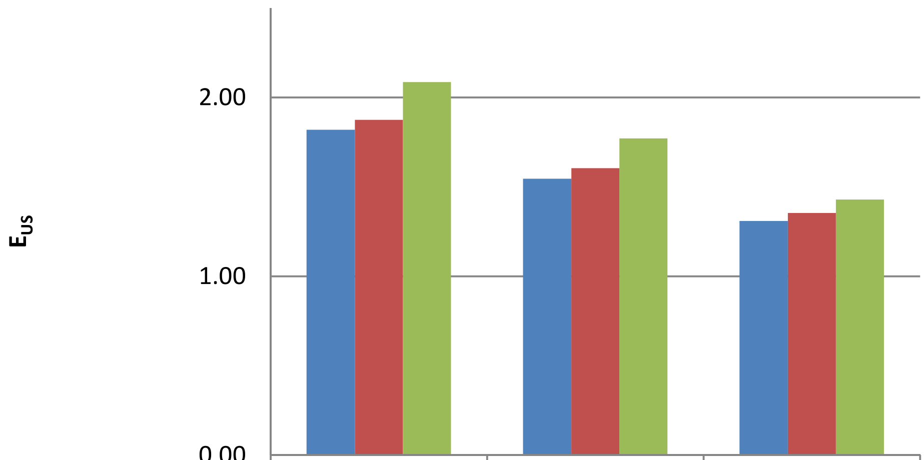


Figure 4: Effect of ultrasonic frequency on flux recovery after mechanical and chemical cleaning of the P020F UF fouled membrane at 300W US power.

Altering the frequency of ultrasound input to a medium changes both the wave interactions with the fluid and the characteristics of the cavitation bubbles formed. For example, at higher frequency, sound is attenuated more readily [30]. Therefore acoustic energy may not be available for cleaning throughout the system. Also, the maximum size bubbles attain is smaller than with lower frequencies. However, there are more collapses per time, although the bubbles tend to collapse less violently producing lower temperatures and pressures [31,32].

Based on the results of this research, which show that the flux increase was better at lower frequencies, it can be suggested that the cleaning mechanism is more dependent on characteristics of bubble collapse and the intensity of shear forces with bubble vibrations than on the number of bubbles collapsing over time. The violence of collapse at lower frequencies and associated disturbance is more important than the increasing number of weaker collapses observed at higher frequencies and mechanical forces (such as shear efforts due to attenuation) are controlled at lower frequencies and greatly contribute to the cleaning of membrane surfaces [33].

Ultrasonic enhancement factor (E_{US}) was used to compare the efficiencies of US application as presented in figure (5), it was clear that the online application of US power during filtration has higher enhancement factor than that of US application during cleaning. The online application of US prevent accumulation of HA particles on membrane surface so reduce concentration polarization and thin cake layer will be formed, while more power was needed to detach the particles from fouling later and from membrane surface in the case of US application during mechanical and chemical cleaning.



	28 kHz	45 kHz	100 kHz
chemical cleaning	1.82	1.55	1.31
Mechanical cleaning	1.88	1.60	1.35
On line	2.09	1.77	1.43

Figure 5: Effect of ultrasonic frequency on US enhancement factor E_{US} after mechanical and chemical cleaning and online filtration of the P020F UF membrane at 300W US power

5.Conclusions

The effects of US frequencies on UF and membrane cleaning processes were studied in this work. Dead-end stirred cell with PES UF membrane was used to filtrate saline water polluted with humic acid with similar condition to sea water pretreatment process.

US irradiation can efficiently enhance the permeate flux, reduce the filtration resistances in UF process, and improve the flux recovery in fouled membrane cleaning.

The results showed that the US at 28 kHz has stronger effect than that at 45 kHz on both UF and membrane cleaning processes, whereas 100 kHz has almost no effect.

According to the analysis of the three resistances, R_m, R_{rev} and R_{irre}, the main effect induced by the US was on the reduction of the reversible resistance, R_{rev}, caused by concentration polarization and cake layer.

Ultrasonic enhancement factor (E_{US}) was higher by 11% in the online application of US on UF process than that of cleaning process.

This study suggests that US irradiation at low frequency is a promising approach that can be applied in both UF and fouled membrane cleaning processes used in sea water pretreatment desalination plants.

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