Ultrasound-assisted membrane technologies for fouling control and performance improvement: A review

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Abstract

Membrane separation is widely used in wastewater treatment and desalination due to its high performance and ability to handle feed solutions of different qualities. Despite vast history of success, membrane fouling remains a major system deficiency that imposes substantial process limitations by reducing permeate production and increasing energy demand. Besides, chemical cleaning-in-place (CIP) adversely affects membrane integrity and generates an extra waste stream. Ultrasound (US) is a relatively new cleaning technique that improves process performance by mitigating fouling accumulation at a membrane surface and improving permeate flux by promoting mass and heat transfer. US-assisted membrane processes is an efficient method for fouling reduction and significant flux improvement. This study comprehensively reviews US applications in pressure-, thermally- and osmotic-driven membrane technologies and their impact on process performance. It also explores the impact of US operating conditions on membrane separation properties and how these parameters can be tuned to achieve the desirable outcome. To date, the application of US in membrane technologies is limited to laboratory tests. In the authors opinion, there is a niche market for

US-assisted membrane technology in heavily contaminated water such as wastewater and brine. After critical analysis of the literature, we found that there are still several aspects of the process need to be scrutinized carefully to make an adequate evaluation of its feasibility on an industrial scale. The most urgent one is the techno-economic evaluation of the technology based on large-scale and long-term tests. The study proposed a set of recommendations for future research directions of US applications in membrane technologies.

Keywords: Ultrasound, Pressure-driven membrane technologies, Emerging membrane technologies, Fouling mitigation, Flux improvement.

1. Introduction

Population increase and rapid industrial development imposed additional demand on freshwater resources [1, 2]. Although developed countries enjoy good quality water provided by centralized municipal water supply systems, safe drinking water remains scarce in developing countries. Contaminants in drinking water are among the most significant issues, and millions of people suffer from their hazardous effects. Different filtration processes and adsorption processes were applied for water cleaning and contaminants removal [3]. Membrane-based processes are increasingly applied to overcome water shortage and produce high-quality drinking water by separating water molecules from contaminants. Different types of pressure-driven membrane processes are commercially available for water treatment, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [4-6]. Recently, membrane distillation (MD) is introduced as an emerging technique that combines thermal and membrane separation [7-19]. Membrane distillation (MD) relies on a partial vapour pressure gradient generally caused by a temperature difference across the membrane [20, 21]. Although MD was suggested decades ago, it is still in the developmental stages. One of the major reasons behind its late commercialization is the low recovery rate, severe temperature polarization, and, to a lesser extent, fouling of the membrane, especially when treating concentrated feed solutions [22, 23].

A range of cleaning techniques was used to control membrane fouling, including physical [24] and chemical [25, 26] cleanings. The advantages of these techniques include removing fouling materials from the membrane surface and increasing water flux by reducing concentration polarization. On the other hand, disadvantages are mainly i) reduced membrane lifetime [16, 27], ii) generation of contaminated wastewater [27], and iii) changes in membrane hydrophobicity and surface morphology [28, 29].

Recently, ultrasound (US) was proposed among other innovative cleaning techniques for water treatment processes, such as CO_2 nucleation, which was tested for ultrafiltration [30] and reverse osmosis (RO) [31]. In water treatment context, US can be defined as the application of sound waves in frequency range higher than the human hearing limits. The detailed definition of terminologies used in ultrasound field and the parameters affecting its throughput will be discussed in the following section. US was integrated successfully with pressure-driven membrane separation [32-36] and emerging technology such as MD [37-42] to remove foulants from the membrane surface. The US-assisted membrane processes can significantly improve membrane performance. For example, water flux increase of up to 600% can be achieved with US help [5, 7]. In addition, US technology was applicable for fouling mitigation for various feed solutions such as surface water [4], milk solution [5], soybean [7] and oil wastewater [12]. The advantages of this cleaning technique are no chemical usage [43], no system shutdown and no need for membrane removal from the system for *ex situ* cleaning so that possible membrane contact with the air is minimized. Ultrasound removes deposited particles from the membrane surface as a result of it shaking. As a result, permeate flux through the membrane is increased. Ultrasound can also increase a membrane's operation time by reducing the occurrence of fouling events. Several concerns are associated with applying US for mitigating membrane fouling, such as high energy requirements [44], adverse effects on membrane integrity, and the selection of best system configuration that suits large-scale applications [45]. On the other hand, the advantages of US application for fouling mitigation are immense. They include reduced or no chemical usage [46], no system shutdown [39, 40], minimal effects on the environment and human health [39, 40, 47, 48] and high potential of scaling and biofouling removal [49, 50].

There are several studies that presented reviews on the application of ultrasound for improving membrane filtration technologies [35, 36, 51-54]. While these studies offer a comprehensive analysis of the commonly studied parameters in ultrasound-assisted membrane system such as power, frequency, medium pressure and temperature, membrane materials and flow conditions, this work discusses further the effect of other system parameters such as ultrasonic waveform, techniques for producing ultrasonic waves (piezo-electric and magneto-strictive) and system configuration on the overall performance of the system. In addition, most of these studies focused on the cleaning effects of ultrasound particularly at cavitational level, whereas this work addresses also the of effect ultrasound on flux enhancement with special attention paid to the potency of non-cavitational ultrasound effects. The other unique future of the current work is the attempt to establish connection between ultrasound energy output and the required energy to achieve the desirable change in the membrane separation process (flux enhancement and fouling removal). This could motivate further research to utilize advanced computational tools to fine tune energy usage in ultrasound-assisted membrane technology, which is the main challenge for scaling-up the process. This paper provides concise discussion for the impact of ultrasound effects on fouling deposition onto membrane surface and mass and heat transfer phenomena in membrane separation processes. The effect of US technology on the performance of different membrane processes is also reviewed presenting up-to-date literature data and recent development in systems configurations. Recommendations and future research directions are also proposed based on literature research findings and authors own views of the process.

2. Ultrasound effects on membrane processes

Prior to reviewing and analysing the reported applications of ultrasound technology with membrane filtration it is essential to briefly discuss the fundamentals of ultrasound technology. Ultrasound is a term commonly used to refer to sound waves with frequency higher than the human hearing limits ≥ 16 kHz [45]. The introduction of ultrasound waves to liquid medium such as water generates negative (rarefication phase) and positive (compression phase) pressure swings. When the ultrasonic amplitude pressure surpasses the tensile strength of liquid, bubbles are formed [51]. These bubbles grow in the negative cycle of pressure and collapse during the positive swing of the pressure. Bubbles produced during ultrasonic waves propagation are generally categorised into transient bubbles that collapse violently and stable bubbles that collapse gently [55]. In addition to the bubbles generated in the liquid phase, bubbles can also be produced at the liquid-solid interface. The process of bubbles generation in liquid phase is termed as homogenous cavitation, while bubbles generated in the liquid-solid interface is known as heterogeneous cavitation [56]. Pre-existing bubbles in the liquid can also grow to transient or stable bubbles depending on their sizes. The movement of ultrasound waves in the liquid medium and bubbles oscillation and collapse generate a range of physical effects that have been harnessed to enhance membrane technology performance. The impact of these effects on the dynamics of membrane separation processes on one hand and their influence by ultrasound operation parameters on the other hand will be discussed succinctly in later parts of this section.

To maximise the benefits of ultrasound application with membrane-based technologies, it is imperative to understand the enhancement mechanisms of ultrasound and how the operating parameters and process environment influence these mechanisms. Ultrasound impacts membrane filtration through three pathways: detaching deposited foulants and driving particles and molecules away from the membrane interface (i.e. reducing concentration polarization) (cleaning effects), improving water transport across the membrane (mass transfer effects) and boosting heat transfer of water for thermally-driven membrane processes [45].

The propagation of ultrasound waves results in several effects such as acoustic streaming, microstreaming, micro-streamers, micro-jets and shock waves generated from transient bubbles collapse [45]. The definition and detailed explanation of these phenomena are well documented in the literature [24, 49, 57, 58]. The occurrence and intensity of ultrasonic effects depend on factors such as power, frequency, environmental conditions of the treatment (i.e. pressure and temperature), nature of the irradiated water, operation mode, mechanical vibration, and excitation wave shapes.

The ultrasound effects can be classified into cavitational and non-cavitational, depending mainly on power and frequency, as demonstrated in Figure 1. Apart from acoustic streaming, Figure 1 shows that other events can only occur if the applied acoustic pressure exceeds a threshold pressure and frequency is lower than MHz range. Blake pressure threshold is commonly applied to estimate the minimum ultrasonic power required for generating cavitation in given conditions [59]. Ultrasound power higher than cavitation threshold can overcome the cohesive forces of the medium and generate bubbles. The higher the applied power, the more violent ultrasonic effects are expected to occur. For ultrasound-assisted membrane technology, high power may damage the membrane. Hence, if more energy required to improve fouling detachment or fluid dynamics in the adjacent area to the membrane, longer treatment time applied.

Contrary to the power, increasing frequency reduces the intensity of acoustic events except for acoustic streaming. A study conducted by Costalongaet al. [60] demonstrated that acoustic streaming velocity increases with frequency. The fluid pattern changes with frequency, and rotational flow diminishes as the frequency increases. A linear motion occurs, especially in the middle of the irradiating surface, as shown in Figure 1. When it comes to the cleaning effects of ultrasound, the linear motion can be problematic as it may push the fouling particles deeper into the membrane pores instead of pushing them away, as observed in the circular motion.

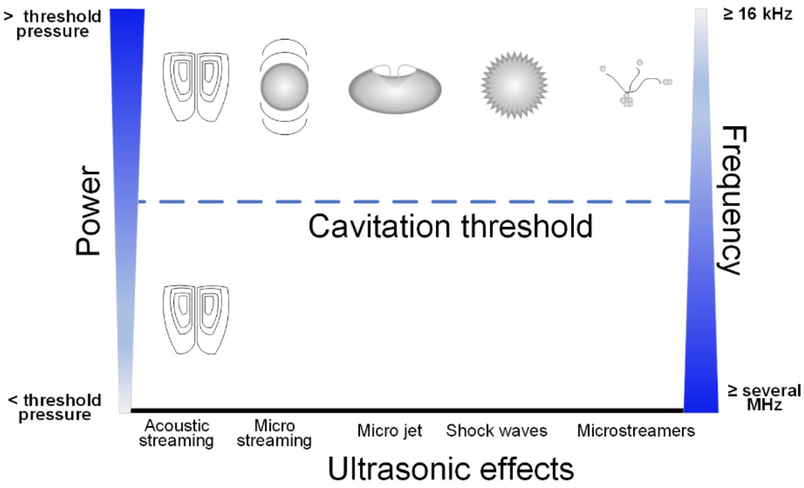

Figure 1: a) Influence of power and frequency on ultrasound effects.

Temperature and pressure of the medium can influence ultrasonic events through their effects on medium properties and bubble dynamics. For instance, higher power is required for the ultrasonic wave to propagate and generate bubbles in a pressurized medium. The opposite is also true in a medium under high temperature [61]. Increasing the temperature reduces the medium viscosity and surface tension facilitating the generation of cavitating bubbles. However, such action can also generate bubbles with less violent collapse [62].

Fluid properties may also impact the nature of its interaction with ultrasonic waves. For example, the type of dissolved gas affects the thermal product of the collapse. Gases with a high adiabatic ratio result in bubble collapse with high temperature [63]. Heavy gases can produce high collapse temperatures, but they have low thermal conductivity and convey the heat from collapse sites to the bulk slower than light gases [63]. The fluid content of dissolved and suspended solids can also influence ultrasound effects. It was found that both the number of bubbles and their size decreases with an increasing salt concentration in water [64].

The operation details of ultrasonic devices can also play an important role in controlling ultrasonic effects. The effect of such details on ultrasound performance in assisting membrane filtration is scarcely investigated in the literature. These details include the operation mode (continuous or pulsed), vibration generation techniques (piezoelectric or magnetostrictive) and the excitation wave (sinusoidal, square, triangle etc.). Applying pulsed mode was more effective in utilizing energy and producing more cavitational effects [65]. In terms of the operation mode on non-cavitational effects (i.e. acoustic streaming), it was reported that applying this mode reduces the acoustic streaming velocity[66]. Therefore, depending on how vigorous the acoustic streaming needs to achieve treatment performance, such as removing a fouling layer or improving mass/heat transfer phenomena, one can decide whether to apply continuous or pulsed mode. Some studies found continuous mode more beneficial for improving membrane filtration flux [67], while others found that pulsed mode is more effective [68]. The techniques used to generate mechanical vibrations in the transducer impact both the efficiency and durability of ultrasonic devices. Magneto-strictive transducers are reported to be more resistant to mechanical impact, more tolerant to high temperatures and have longer working life compared to piezoelectric transducers [69]. The latter type of transducers is commonly used in membrane filtration studies due to its availability as an off-the-shelf product in the market. This may be one reason that makes the ultrasound technique perceived to be costly. Kyllönen et al., [35] concluded that the main reason that hinders the commercialisation of ultrasonic-assisted membrane technology is the absence of active efforts for developing transducers that cater for this application. The effect of the excitation wave on the transducer's electrical output and the cavitational chemical yield (measured by $OH^{\cdot}$ and H_2O_2 production) was evaluated by Al-juboori et al. [69]. The results showed that among the tested waveforms, square wave resulted in the best transducer displacement and the highest concentration of $OH^{\cdot}$ and H_2O_2. A numerical study by another team Kerboua, and Hamdaoui [70], on bubble dynamics under different excitation waveform showed that a square wave produces the highest pressure and temperature inside the bubble compared to triangle and sinusoidal waves.

2.1. Effects of ultrasound on fouling

Applying the US for removing/preventing fouling layer formation requires an adequate understanding of the forces acting on the particle in a dynamic system. There are mainly four forces exerted on a particle at the membrane/water interface, as depicted in Figure 2. These forces are the lubrication force (F_L), the adhesion/repulsion force ($F_{A/R}$), the tangential drag force (F_T) and the friction force (F_F) [71, 72]. The roughness variation of the membrane surface is represented by δ in Figure 2. For additional details regarding forces affecting a particle deposition onto a membrane surface, readers are referred to existing literature [71, 72] and references presented therein.

The impact of US on deposited particles is mainly governed by power intensity and the effective distance from the membrane surface. From the force balance presented in Figure 2, it can be inferred that the particle adhesion condition is satisfied when $F_T = F_F$ and $F_A \geq F_L$. Hence, the forces generated by US effects need to tip the balance in favour of tangential force and lubrication. For instance, the hydrodynamic force (F_s) generated by the acoustic streaming (eq. 1 [73]) or the shock wave energy (E_{SW}) generated from bubble collapse (eq. 2 [74]) need not only to exceed the friction force but to also move the particle away from the membrane.

$$F_s = \frac{P_{US}}{c} e^{-2\alpha x} \tag{1}$$

$$E_{SW} = \int \frac{\Delta P^2}{(\rho c)^2} dV \tag{2}$$

where P_{US} is the US power (W), c is the speed of sound (m/s), α is the attenuation coefficient of the acoustic pressure in water (m^{-1}), x is the distance between the irradiating surface and the membrane (m), ΔP is the pressure difference across bubble wall, and V is the cavitating bubble volume (m^3).

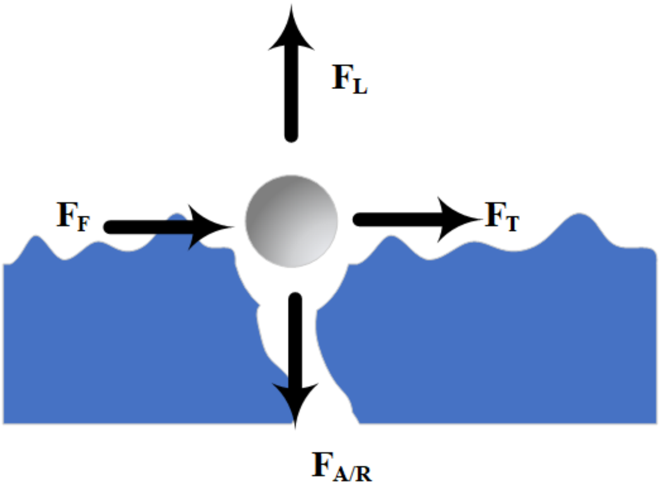

Figure 2: Forces acting on a particle being deposited onto a membrane pore.

As for the effective range of ultrasonic events, an illustrative representation is provided in Figure 3 [24, 57, 75]. It is clear that except for acoustic streaming, other ultrasonic effects need to occur close to the membrane-water interface to remove particles from the membrane surface. Several studies [68-70] reported these effects are more intense than acoustic streaming, raising concern of possible membrane damage. Strong forces such as those generated by the cavitational effects are only needed when the fouling layer is already established. This also depends on the fouling type: cake layer or pore blocking. The US was found to be less effective in removing pore-blocking fouling as opposed to the cake layer fouling [76]. Given the fact that the US is not effective in removing all forms of developed fouling on the membrane and the potential damage cavitation effects may cause, one can deduce that the most efficient way to apply the US for alleviating the fouling problem is by utilizing low power non-cavitational effects to prevent/reduce fouling formation at early stages of filtration.

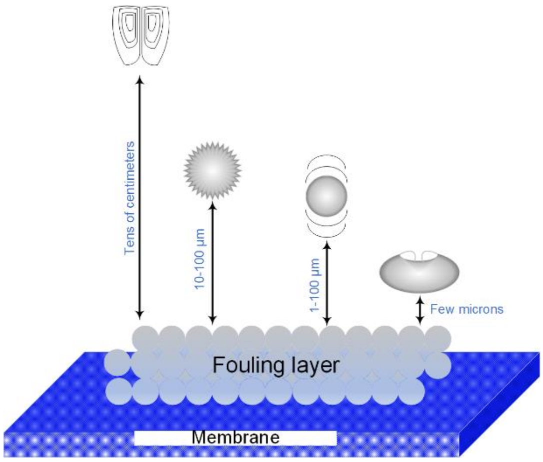

Figure 3: Active ultrasonic effects' distance.

2.2. Mass and heat transfer enhancement

The US can affect mass transfer through its influence on flow nature by generating turbulence in the membrane's vicinity. However, the direction of turbulences needs to be in the same direction as the flow; otherwise, it may slow down the water near the membrane surface, promoting fouling. The velocity of the turbulences can be estimated using dedicated equations such as the maximum acoustic streaming velocity formula (eq. 3) [77], where v is the vibrating velocity (m/s), k is the wavenumber, δ is the boundary layer thickness (m), y is the distance to the membrane surface (m), and a is the transducer radius (m). The direction could also be identified based on the mounting of the emitting surface onto the membrane module. Species diffusion coefficient being a function of pressure and temperature [78], US can affect diffusion through pressure and temperature increase that results from US effects.

$$u_{as} = \frac{3v^2\delta a^6 k^3}{8cx^4}\left\{1 - \frac{y}{\delta} - \left(1 - \frac{y}{\delta}\right)^3\right\} \quad (3)$$

The effect of the US on heat transfer is mainly related to its impact on the convective heat transfer coefficient on the feed side. The US increases the convective heat transfer coefficient by a component (h_{as}, W/k·m^2) presented in eq. 4, where C_p is the specific heat of water (kJ/kg·K). Knowing the velocity of the acoustic streaming and the feed water properties, one can estimate the extent of enhancement expected with a chosen set of operating conditions.

$$h_{as} = \rho v_{as} C_p \quad (4)$$

3. Integration of US with pressure-driven membrane technology

There are two types of US connections in the membrane module, i.e. *in-situ* (internal) and *ex-situ* (external) [39]. The advantage of the *in-situ* connection is that it requires low US power to remove the fouling layer from the membrane surface as the transducers can be close to the membrane [40]. Compared to the *in-situ* connection, the *ex-situ* connection requires high US power as the transducers are located far from the membrane surface. Most of the attempts if not all, on using US-assisted pressure-driven membrane technologies adopted the *ex-situ* configuration to avoid membrane damage [79]. However, as stated earlier, this requires high-energy consumption to convey the effects to the membrane surface. The purpose of applying to the US could also vary. Some studies applied ultrasound as an offline cleaning technique, while others applied it as an online cleaning technique that could simultaneously enhance water flux. The following sections discuss the coupling of US with various pressure-driven membrane processes.

3.1. MF-US.

Microfiltration (MF) is considered one of the most common membrane technologies used for water and wastewater treatment. The MF technology showed great potential in treating various wastewaters. However, membrane fouling is a critical issue in MF, which significantly affects process performance. Among techniques used for cleaning MF, US technology has captured considerable attention, and the majority of ultrasound applications for membranes cleaning was trailed using MF setups.

Table 1 shows a summary of the studies conducted using the US with pressure-driven membrane technologies. It should be noted that the authors tried to include all relevant information available in the reported studies with the focus on US effects alone membrane performance. Some information such as the thickness of the tested membranes before and after filtration and cleaning processes are rarely reported in the literature. Hence, they have not been covered in this review.

A range of synergistic techniques has been reported to improve membrane throughput when combined with the US. Sanderson et al. [80] found that combining forward washing with ultrasound for offline cleaning of MF membrane fouled with paper mill wastewater improved permeate flux by 750% compared to only 300% with ultrasound alone. Another study reported that adding ethylenediaminetetraacetic acid (EDTA) to feed solution while applying ultrasound on the fouled membrane with milk solution enhanced the flux further. A mixed frequency of 28, 45 and 100 kHz was the least affected by EDTA addition than individual frequencies [81]. There is other possible synergestic processes that incorbrate ultrasound with membrane technology and adsorption in a hybrid system as a combination of filtration and adsorption has proven to be effective for treating wastewater [82]. As an example for such hybrid system is the combined UF, US and activated carbon processes tested by Mona et al. [83] for removing industerial dyes. The ourcome of these studies is summaries in Table 1. Another synergy that can benefit from ultrasound application is the hybrid electro-chemical and adsorption system such as the one reported by Kadhum et al. [84] if combined with membrane technolgy assited by electro-chemical techniques [85]. Although such combination has not been reported in the literature, one can postulate the potential benefit of ultrasound. For instance, ultrasound can improve the adsorption capacity of adsorbents [83] and allivate the impact conencertation polarization on membrane and electrodes [86]. However, ultrasound physical and chemical effects can lead to the destruction of electrodes just as it is the case with posible membrane surface deterioration [87]. The other possible risk with such combinesd systems is that if the adsorbents are immoblised on membrane surface, ultrasound effects could detach them rendering the membrane structure weak and more prone to serious damage.

The compiled information in Table 1 is useful to gain an in-depth understanding of the effects of membrane and ultrasound operating conditions on the overall performance of the US-assisted membrane process. There are three ways for pressure-driven membrane processes through which the US is applied: online flux enhancement, pretreatment and offline cleaning. Online flux enhancement appears to be the most effective form of US application. Examples of common US-membrane design systems are illustrated in Figure 4. In addition to the 3 designs mentioned above, there are self-cleaning US-vibrated piezoceramic membranes that have recently been developed and found to increase the flux by about 30% when the vibration is in operation [88]. It appears that increasing the input US energy either through increasing the applied power or the irradiation time affects the permeate flux negatively. This is likely to occur due to high power density, resulting in the breakdown of particles leading to severe pore-blocking fouling. For the case of MF, two studies [67, 89] showed the adverse effect of ultrasonic energy on membrane flux used a high power density of 200 W/l - 300 W/l. Similarly, the increasing frequency seems to result in lower permeate flux enhancement. This has been attributed to the negative effect of frequency on cavitation threshold and bubble growth [90]. Evaluating the effect of frequency of flux with non-cavitational effect has not been addressed in the literature. In this case, a higher frequency may be useful as more wave cycles are generated. A mix of low and high frequency was more powerful than the low frequency alone [81]. It appears that the pressure has an inverse correlation with permeate flux enhancement. The latter is expected since pressure increase raises the resistance against the propagation of the sound wave.

Regarding the effect of membrane materials on the efficiency of US cleaning, a study conducted by Wang et al. [91] tested polyethersulfone (PES), mixed ester of cellulose nitrate with cellulose acetate, PVDF and nylon six and found that the latter exhibits the highest permeate flux improvement. For more details on membrane materials effect on US performance, readers are referred to the study by [51]. However, this study pinpointed that it was hard to conclude from the literature regarding the effect of membrane materials on US effectiveness.

Some researchers have investigated other parameters, such as the distance between the emitting surface of the ultrasound and the membrane surface. Mirzaie and Mohammadi [67] observed a drop in flux enhancement of MF-US from 228% to 145% when the distance between the US horn and the membrane surface was increased from 2.6 cm to 4.4 cm. However, increasing the distance between the ultrasound source and membrane surface does not always have a negative impact on flux enhancement. Thus, [92] showed that increasing the distance between ultrasonic transducer and membrane from 4 cm to 8 cm increased the flux from 5.8×10^{-5} $m^3/m^2 \cdot s$ to 7.5×10^{-5} $m^3/m^2 \cdot s$. However, when the distance was further raised to 12 cm, permeate flux declined to 7.1×10^{-5} $m^3/m^2 \cdot s$. The observed effect was attributed to the uniformity and intensity of the ultrasonic field governed by the applied power, the reactor design and the nature of the irradiated fluid. As stated earlier, the content of the water being irradiated could influence ultrasound performance. It was reported that increasing particles concentration in water from 0.1 g/L to 1.8 g/L resulted in a decrease in permeate recovery of US-assisted membrane technology by ~ 60% [93].

Table 1: Summary of the effects of operating conditions on flux enhancement for US-assisted pressure-driven membrane processes.

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
MF	Nylon	Feed: paper mill effluent, configuration: flat-sheet, Flowrate: 18 L/h, Pressure: 50 kPa, Temperature: 23°C, pH: 4.96.	Power: 375 W, Frequency: 20 kHz, Time: 10 mins, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 34-38 %.	Not reported.	[94]
	Nylon	Feed: pure water, configuration: flat-sheet, Flowrate: 18 L/h, Pressure: 50 kPa, Temperature: 23°C, pH: 7.	Power: 375 W, Frequency: 20 kHz, Time: 10 mins, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 300 %.	Not reported.	[94]
	Polyvinylidinefluoride (PVDF)	Feed: 1% milk solution, configuration: ,flat-sheet, Flowrate: 300 ml/min, Pressure: 60 kPa, pH: 11, effective area: 30 cm^2.	Power: 300 W, Frequency: 28 kHz Time: 30 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 400 %, reduced resistance: 77 %.	Not reported.	[81]
	Polyvinylidinefluoride (PVDF)	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 300 ml/min, Pressure: 60 kPa, pH: 11, effective area: 30 cm^2.	Power: 300 W, Frequency: 45 kHz Time: 30 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 280 %, reduced resistance: 71.6 %.	Not reported.	[81]
	Polyvinylidinefluoride (PVDF)	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 300 ml/min, Pressure: 60 kPa, pH: 11, effective area: 30 cm^2.	Power: 300 W, Frequency: mixed 28, 45, 100 kHz Time: 30 min, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 160 %, reduced resistance: 60.6 %.	Not reported.	[81]
	Polyvinylidinefluoride (PVDF)	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 300 ml/min, Pressure: 60 kPa, pH: 11, effective area: 30 cm^2.	Power: 300 W, Frequency: 28 kHz Time: 30 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 579 %, reduced resistance: 80.7 %.	Not reported.	[81]
	Cellulose	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 200 ml/min, Pressure: 60 kPa, Temperature: 25°C, effective area: 30 cm^2.	Power: 295 W, Frequency: 28 kHz, Time: 60 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 310 %.	Not reported.	[90]
	Cellulose	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 200 ml/min, Pressure: 60 kPa, Temperature: 25°C, effective area: 30 cm^2.	Power: 295 W, Frequency: 45 kHz, Time: 60 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 127 %.	Not reported.	[90]
	Cellulose	Feed: 1% milk solution, configuration: flat-sheet, Flowrate: 200 ml/min, Pressure: 60 kPa, Temperature: 25°C, effective area: 30 cm^2.	Power: 295 W, Frequency: 100 kHz, Time: 60 min, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 36 %.	Not reported.	[90]

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Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative	Ref.
MF	PVDF	Feed: 1% peptone solution, configuration: flat-sheet, Flowrate: 325 ml/min, Pressure: 30 kPa, Temperature: 20°C, , effective area: 69 cm^2.	Power: 295 W, Frequency: 45 kHz, Time: 20 min, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 6 %, reduced resistance: 30 %.	Not reported.	[90]
	Ceramic filter	Feed: Synthetic oil field wastewater, configuration: hollow tubes, Vacuum pressure: 133 Pa.	Power: not reported, Frequency: 40 kHz, Time: 1 min, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 150 %.	Not reported.	[95]
	PVDF	Feed: Biologically treated wastewater, configuration: flat-sheet, Pressure: 70 kPa, Temperature: 20 °C, , effective area: 13.4 cm^2.	Power: 107 W, Frequency: 45 kHz, Time: 20 min, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 30 %.	Not reported.	[96]
	Ceramic filter	Feed: Yeast suspension in NaCl, configuration: hollow tubes, Flowrate: 8.3 L/s, Pressure: 40 kPa.	Power: 120 W, Frequency: 28 kHz, Time: 4 h, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 400 % reduced resistance: 78 %.	Not reported.	[97]
	Mixed cellulose ester	Feed: milk, configuration: Flat-sheet, Pressure: 500 kPa, Temperature: 22°C, Flowrate: not available (dead end filtration), effective area: 78.6 cm^2.	Power: 20 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 228 %.	None (confirmed by SEM analysis).	[67]
	Mixed cellulose ester	Feed: milk, configuration: Flat-sheet, Pressure: 800 kPa, Temperature: 22°C, Flowrate: not available (dead end filtration) effective area: 78.6 cm^2.	Power: 20 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 184 %.	None (confirmed by SEM analysis).	[67]
	Mixed cellulose ester	Feed: milk, configuration: Flat-sheet, Pressure: 1400 kPa, Temperature: 22°C, Flowrate: not available (dead end filtration) effective area: 78.6 cm^2.	Power: 20 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 27 %.	None (confirmed by SEM analysis).	[67]
	Mixed cellulose ester	Feed: milk, configuration: Flat-sheet, Pressure: 500 kPa, Temperature: 22°C, Flowrate: not available (dead end filtration) effective area: 78.6 cm^2.	Power: 40 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 490 %.	None (confirmed by SEM analysis).	[67]
	Mixed cellulose ester	Feed: milk, configuration: Flat-sheet, Pressure: 500 kPa, Temperature: 22°C, Flowrate: not available (dead end filtration) effective area: 78.6 cm^2.	Power: 50 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 274 %.	None (confirmed by SEM analysis).	[67]

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Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
MF	Polyethylene (PE)	Feed: 1% milk solution, configuration: hollow fiber, Flowrate: 54 ml/min, Pressure: 60 kPa effective area: 500 cm^2.	Power: 300 W, Frequency: 28 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 317 %.	None (confirmed by SEM analysis).	[92]
	γ-alumina membranes	Feed: 0.5 g/L of 1.56 µm silica solution, configuration: flat sheet, Flowrate: 500 ml/min, Pressure: 34.5 kPa, Temperature: 20 °C, pH: 5.6, effective area: 17.4 cm^2.	Power: 19 W, Frequency: 20 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 96 %.	Membrane pitting and visual cracking.	[93]
	Polyethersulfone	Feed: 1% isolated soybean protein, configuration: flat-sheet, Pressure: 20 kPa, Temperature: 20 °C, pH: 6, effective area: 9.6 cm^2.	Power: 238 W, Frequency: 40 kHz, Time: 60 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 77 %.	Pores enlargement and cracks occurrence.	[91]
	Nylon 6	Feed: 1% isolated soybean protein, configuration: flat-sheet, Pressure: 20 kPa, Temperature: 20 °C, pH: 6, effective area: 9.6 cm^2.	Power: 238 W, Frequency: 40 kHz, Time: 60 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 634 %.	Not reported.	[91]
	Cellulose nitrate with cellulose acetate	Feed: 1% isolated soybean protein, configuration: flat-sheet, Pressure: 20 kPa, Temperature: 20 °C, pH: 6, effective area: 9.6 cm^2.	Power: 238 W, Frequency: 40 kHz, Time: 60 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 88 %.	Not reported.	[91]
	PVDF	Feed: 1% isolated soybean protein, configuration: flat-sheet, Pressure: 20 kPa, Temperature: 20 °C, pH: 6, effective area: 9.6 cm^2.	Power: 238 W, Frequency: 40 kHz, Time: 60 mins, Mode: continuous, Connection: *In-situ.*	No flux enhancement. observed	None (confirmed by SEM analysis).	[91]
	PVDF	Feed: 1% isolated soybean protein, configuration: flat-sheet, Pressure: 90 kPa, Temperature: 20 °C, pH: 6, effective area: 9.6 cm^2.	Power: 400 W, Frequency: 40 kHz, Time: 90 mins, Mode: intermittent, Connection: *In-situ.*	Flux enhancement: 500 %.	None (confirmed by SEM analysis).	[91]
	PE	Feed: surface water, configuration: hollow fibre, Temperature: 20 °C, pH: 6.	Power: 1500 W, Frequency: 20 kHz, Mode: continuous, Time: 4 min, Connection: *Ex-situ.*	Flux enhancement: 50 %.	Not reported.	[89]
	Polysulfone (12% wt.) + PVD (1% wt.) + N-methylpyrrolidone (87% wt.)	Feed: whey solution, configuration: flat-sheet, Pressure: 300 kPa, Temperature: 15 °C, pH: 7, effective area: 12.56 cm^2.	Power: 200 W, Frequency: 24 kHz, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 150 %.	Cracks formation.	[98]

Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
MF	Nylon	Feed: Grape pomace extracts, configuration: Flat sheet, Pressure: 20 kPa, effective area: 31.65 cm^2.	Power: 160 W, Frequency: 24 kHz, Time: 30 mins, Mode: continuous, Connection: *In-situ.*	Flux enhancement: 340 %.	None (estimated through permeability and selectivity).	[99]
UF	Ceramic filter	Feed: 50 mg/L azo dye solution, configuration: tubular, Flowrate: 39.6 L/s, Pressure: 110 kPa, Temperature: 25 °C, pH: 6.5, effective area: 132 cm^2.	Power: 158 W, Frequency: mixed 37 and 80 kHz, Mode: continuous, Connection: *Ex-situ*.	Reduced resistance: 32.2 %.	None (estimated through permeability and selectivity).	[100]
	Polysulfone	Feed: Real wastewater, configuration: hollow fiber, Flowrate: based on fixed flux of 75 $L/m^2 \cdot h$, Pressure: 20 kPa, Temperature: 25 °C, pH: 7.9, effective area: 6.6 cm^2.	Power: 175 W, Frequency: 35 kHz, Mode: continuous, Connection: *Ex-situ.*	Reduction in fouling rate (kPa/h): 36.54 %, increased turbidity removal:~3%, increased UV_{254} removal: ~20%.	Not reported.	[101]
	Polysulfone	Feed: Real wastewater, configuration: hollow fiber, Flowrate: based on fixed flux of 75 $L/m^2 \cdot h$, Pressure: 20 kPa, Temperature: 25 °C, pH: 7.9, effective area: 9.6 cm^2.	Power: 145 W, Frequency: 130 kHz, Mode: continuous, Connection: *Ex-situ*.	Reduction in fouling rate (kPa/h): 15.38 % increased turbidity removal:~14%, increased UV_{254} removal: ~60%.	Not reported.	[101]
	Polyethersulfone	Feed: *Radix astragalus* extract, configuration: flat sheet, Pressure: 80 kPa, Temperature: 25 °C, effective area: 41.8 cm^2.	Power: 120 W, Frequency: 28 kHz, Mode: continuous, Time: 60 min, Connection: *Ex-situ*.	Flux enhancement: 12-15 %, reduced resistance: 40%.	Increased irreversible fouling.	[102]
	Polyethersulfone (PES)	Feed: *Radix astragalus* extract, configuration: flat sheet, Pressure: 80 kPa, Temperature: 25 °C, effective area: 9.6 cm^2.	Power: 120 W, Frequency: 20 kHz, Mode: continuous, Time: 60 min, Connection: *In-situ.*	Flux enhancement: 68 %.	Rapid temperature increase of up to 55°C.	[102]
	Polysulfone	Feed: *Radix astragalus* extract, configuration: hollow fibre, Flowrate: 40 mL/min, Pressure: 60 kPa, Temperature: 40 °C, effective area: 150 cm^2.	Power: 120 W, Frequency: 45 kHz, Mode: continuous, Time: 20 min, Connection: *Ex-situ*.	Reduction in fouling degree: ~6 %, reduction of process duration: ~18 %.	Not reported.	[103]
	Polyacrylonitrile (PAN)	Feed: wt%,dextran (2×10^6 MW), configuration: flat sheet, Flowrate: 325 mL/min, Pressure: 30 kPa, Temperature: 25 °C, effective area: 300 cm^2.	Power: 248 W, Frequency: 45 kHz, Mode: continuous, Time: 30 min, Connection: *Ex-situ*.	Flux enhancement: ~100 %.	Not reported.	[104]

Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
UF	PES	Feed: dextran solution (3g/L), configuration: Flat sheet, Pressure: 80 kPa, Temperature: 20°C, effective area: 41.8 cm^2.	Power: 100 W, Frequency: 28 kHz, Time: 30 mins, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 78 %, Reduced resistance: 44 %.	None (confirmed by SEM analysis).	[105]
	Polysulfone	Feed: whey solution (6% wt/wt), configuration: Flat sheet, Flowrate: 550 mL/min, Pressure: 55 kPa, Temperature: 25°C, pH: 12, effective area: 30 cm^2.	Power: 300 W, Frequency: 50 kHz, Time: 10 mins, Mode: continuous, Connection: *Ex-situ.*	Increased cleaning efficiency: 17 %.	Not reported.	[106]
	Polysulfone	Feed: real wastewater, configuration: hollow fibre, Flowrate: 127mLL/min, Pressure: 31 kPa, pH: 7.2, effective area: 8.48 cm^2.	Power: 0.75 W, Frequency: 35 kHz, Time: 6 h, Mode: continuous, Connection: *Ex-situ.*	Reduction in trans-membrane pressure by ~7 kPa, increased UV_{254} removal: ~280%.	Not reported.	[107]
	Polysulfone	Feed: real wastewater, configuration: hollow fibre, Flowrate: 127 mL/min, Pressure: 31 kPa, pH: 7.2, effective area: 8.48 cm^2.	Power: 0.75 W, Frequency: 130 kHz, Time: 6 h, Mode: continuous, Connection: *Ex-situ.*	Increased UV_{254} removal: ~400%.	Not reported.	[107]
	PAN	Feed: dextran solution 2×10^6 MW (% wt), configuration: hollow fibre, Flowrate: 132 mL/min, Pressure: 30 kPa, Temperature: 25°C, effective area: 96 cm^2.	Power: 150 W, Frequency: 28 kHz, Time: 680 min, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 100 %.	Not reported.	[108]
	Regenerated cellulose	Feed: Cu^{2+}–polyethylenimine ([Cu^{2+}]/[polyethylenimine]: 0.2) , configuration: flat sheet, Pressure: 69 kPa, Temperature: 25°C, effective area: 176.7 cm^2.	Power: 30 W, Frequency: 20 kHz, Time: 60 min, Mode: continuous, Connection: *In-situ*.	Flux enhancement: 70 %.	Reported membrane structure damage with high power and small horn tip.	[109]
	Polysulfone	Feed: 10 ppm mixture of diclofenac, carbamazepine, and amoxicillin, configuration: flat sheet, fixed flux of 150 L/m^2.h, Pressure: 20 kPa, Temperature: 25°C, effective area: 6.6 cm^2.	Power: 175 W, Frequency: 35 kHz, Time: 4 h, Mode: continuous, Connection: *Ex-situ*, Podwer activated carbon (PAC) dose of 0.75 g/m^2 was added .	Increased removal of all studied contaminants from ~92% to ~100 % likely due to increasing adsorption capacity of PAC and chemical effects of US.	Not reported.	[83]
	Polysulfone	Feed: 10 ppm mixture of diclofenac, carbamazepine, and amoxicillin, configuration: flat sheet, fixed flux of 150 L/m^2.h, Pressure: 20 kPa, Temperature: 25°C, effective area: 6.6 cm^2.	Power: 145 W, Frequency: 130 kHz, Time: 4 h, Mode: continuous, Connection: *Ex-situ*, Podwer activated carbon(PAC) dose of 0.75 g/m^2 was added.	Increased removal of all studied contaminants from ~92% to ~100 % likely due to increasing adsorption capacity of PAC and chemical effects of US.	Not reported.	[83]

Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
UF	Polysulfone	Feed: Real wastewater spiked with 10 ppm mixture of diclofenac, carbamazepine, and amoxicillin, configuration: flat sheet, fixed flux of 150 L/m^2.h, Pressure: 20 kPa, Temperature: 25°C, pH 7.6-8.3, effective area: 6.6 cm^2.	Power: 145 W, Frequency: 130 kHz, Time: 4 h, Mode: continuous, Connection: *Ex-situ*, Podwer activated carbon(PAC) dose of 4.5 g/m^2 was added.	Increased removal of all studied contaminants from ~90% to ~100 % likely due to increasing adsorption capacity of PAC and chemical effects of US.	Not reported.	[110]
NF	Hydrophilized polyamide (HPA) membrane	Feed: mixture of reactive black and Reactive yellow dyes and 100 mg/L NaCl, configuration: flat sheet, Flowrate: 4.8 L/s, Pressure: 490 kPa, Temperature: 25°C, effective area: 160 cm^2.	Power: 145 W, Frequency: 34 ± 3 kHz, Time: 60 min, Mode: continuous, Connection: *Ex-situ*.	Recovered flux to initial value and increased it further by 16.5%, Small improvement in COD and TDS removal (3-4%).	Observed pores enlargement.	[111]
	Aromatic polyamide (NF3A)	Feed: synthetic arsenic-rich brackish water, Configuration: flat sheet, Flowrate: 1 L/s, Pressure: 1 MPa, Temperature: 20°C, effective area: 20 cm^2.	Power: 20 W, Frequency: 40 kHz, Time: 8 min, Mode: continuous, Connection: *Ex-situ*, citric acid was used in combination with US.	Increased flux from ~25 L/m^2.h to ~60 L/m^2.h, Increase As rejection from ~55% to ~100%.	Not reported.	[112]
	Not specified	Feed: oil field water, Configuration: flat sheet.	Power: 70 W, Frequency: 25 kHz, Time: 4 h, Mode: continuous, Connection: *Ex-situ*, SSA-I scale-dissolving agent was used with US.	Increased the flux recovery from 49.1% with only anti-sealant to 95.6%, decreased the treatment time from 6 h to 4 h.	Not reported.	[113]
	Polyamide	Feed: mixture of Direct Black 155 and Direct Blue 150 dye (3000 ppm), Configuration: flat sheet, Pressure: 1500 kPa, effective area: 14.6 cm^2.	Power: 135 W, Frequency: 24 kHz, Time: 1 min, Mode: continuous, Connection: *Ex-situ*.	Flux recovery: 78.51 %.	None (confirmed by re-testing the treated membrane), however, cracks and cervices were observed at frequency of 40 kHz.	[79]
RO	Polyamide	Feed: $CaSO_4$ solution (500 mg/L), Configuration: flat sheet, Flowrate: 5 mL/minPressure: 100 kPa, Temperature: 20°C, effective area: 56 cm^2.	Power: 2016 W, Frequency: 20 kHz, Time: 3 h, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 50.8 %.	Slight decrease in rejection, temperature increase due to US decreases $CaSO_4$ solubility.	[114]
	Polyamide	Feed: $FeCl_3$ solution (16 mg/L), Configuration: flat sheet, Flowrate: 5 mL/minPressure: 100 kPa, Temperature: 20°C, pH: 4.5, effective area: 56 cm^2.	Power: 2016 W, Frequency: 20 kHz, Time: 3 h, Mode: continuous, Connection: *Ex-situ*.	Flux enhancement: 215 %.	Slight decrease in rejection.	[114]

Table 1-continued

Membrane process	Membrane materials	Membrane operating conditions	US conditions	Improvements achieved	Reported negative impacts	Ref.
RO	Polyamide	Feed: carboxymethyl cellulose (1000 mg/L), Configuration: flat sheet, Flowrate: 5 mL/min, Pressure: 100 kPa, Temperature: 20°C.	Power: 2016 W, Frequency: 20 kHz, Time: 3 h, Mode: continuous, Connection: *Ex-situ.*	Flux enhancement: 264 %.	Slight decrease in rejection.	[114]
	Polyamide	This study used post cleaning of heavily contaminated RO membrane from pharmaceutical industry.	Power intensity: 0.64 W/cm^2, Frequency: 50 kHz, Time: 30 min, Mode: continuous, Connection: *Ex-situ.*	Increased organic and inorganic defouling indicated by rise in UV absorbance from 0 to ~0.009 and conductivity form ~2.3 to ~ 3.0 µS/cm.	Long exposure to ultrasound lead to membrane damage especially for fouled membranes stored dry.	[115]
	Polyamide layered on polysulfone	Feed: *E.coli* suspension (10^6 CFU/mL), Configuration: flat sheet, Pressure: 450 kPa, Temperature: 25°C, pH: 7.3, effective area: 50 cm^2.	Power: 81.7 W, Frequency: 60 kHz, Time: 4 min, Mode: continuous, Connection: *Ex-situ*, US was combined with heating at 48°C (thermosonication).	Flux enhancement: 100 %, development of sparse and weak biofilm structure.	None (confirmed by microscopic analysis).	[116]
	Not specified	This study used post cleaning of a spiral-wound SWRO membrane module SWC3 model (Hydranautics) used for seawater desalination for several years.	Power: 140 W, Frequency: 45 kHz, Time: 30 min, Mode: continuous, Connection: *Ex-situ*, US was used with NaOH 2% w/v at 25°C.	Increased rejection by 6.4%.	Trivial reduction in flux by 0.6%.	[117]

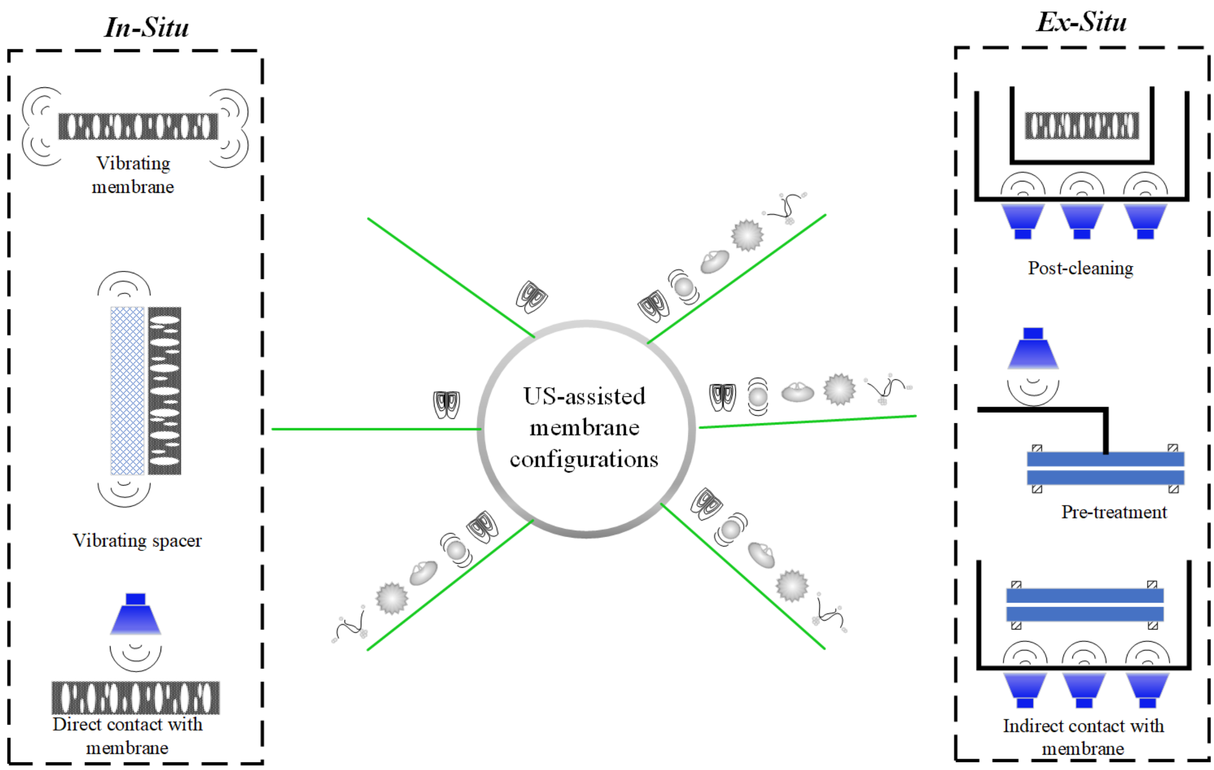


Figure 4: US-assisted membrane configurations with their effective mechanisms for membrane performance improvement.

3.2. UF-US

Ultrafiltration (UF) is a promising separation process that covers a wide range of industrial processes, including concentration, fractionation, water treatment and macromolecular species elimination or macro-solutes elimination from various industrial effluents [118, 119]. Membrane fouling is a serious issue with UF membranes, causing a decrease in permeate flux and increasing process and maintenance costs. Different chemical and physical methods have been used for UF membrane cleaning [100, 120]. Physical cleaning might change the membrane's hydrodynamics, while chemical cleaning can be expensive [121]. US technology is considered for cleaning the UF membrane using a bath configuration, as illustrated in Figure 5.

Table 1 shows a summary of the studies conducted using the US combined with pressure-driven membrane technologies. Various sonication modes have been used in US-assisted UF, such as continuous, pulsed, sweeping, and degassing, to improve process performance.

Shahraki et al. [68] studied the effect of different sonication modes (continuous, pulsed, sweeping, and degassing) on permeate flux and fouling of flat sheet UF polyethersulfone membrane. The optimum UF process was achieved at 37 kHz using a pulsed mode, which corresponds to a percentage of fouling and effect of sonication factor of 10.53% and 187.4%, respectively. The US pulsed mode is more energy-efficient than the continuous mode. This is critical for implementation because one of the main US disadvantages is the energy cost. The US energy cost is high and would be only useful for a laboratory test [109]. Another reason for this optimum filtration process is the US's low frequency, which could increase the removal of the fouling layer from the membrane surface. The low US frequency results in 1) localized turbulence and stronger vibration [101, 122] and 2) lower concentration polarization and the cake layer resistance [90, 102, 105, 123].

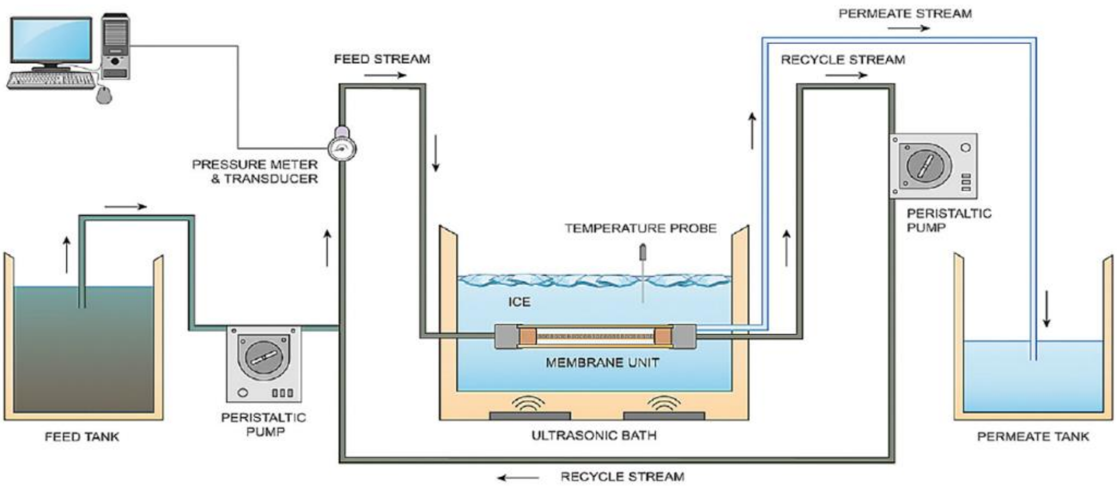


Figure 5: Schematic of UF-US system [83].

Even though US technology can remove the fouled layer from the membrane surface and increase the permeate flux Latt and Kobayashi [92], Matsumoto et al., [97] Muthukumaran et al., [124], its effect also depends on the feed solution matrix. For example, Chai et al. [104] used the US for cleaning polyacrylonitrile membrane after UF of dextran solution and found that the US technology could not remove the fouled layer from the membrane surface. This might be due to membrane vibration caused by the US waves, which increased bulk mass transfer [104]. Yu et al. [125] pointed out that applying the US for 10 mins could separate more organic matter from the membrane surface. The US removed predominantly hydrophilic, high molecular weight organic matter from the UF membrane. Concurrently, the US process had a low effect on the accumulation of smaller molecular weight organic matter. Chen et al. [126] studied the impact of particle characteristics on the ultrasonic control of membrane fouling. The authors found that US cleaning was affected by the particle size and higher permeate flux recovery was observed when feed water contained large particles. Interestingly, the authors reported membrane damage when the US was applied at a short distance from the membrane surface under high pressure.

Some researchers have also investigated the effect of distance between the emitting surface of the US and the membrane surface. The effect of US technology is fundamentally mechanical (i.e., largely rely on the US transducer connection methods), so the highest permeate flux could be achieved when the system is properly connected. Hengl et al. [127] found that at 8 mm distance between the membrane surface and the blade, permeate flux increased by seven folds without apparent damage to the membrane surface. It is believed that as the US transducer was close to the membrane surface, the acoustic streaming could break down the polarization layer formed at the surface of the membrane [127]. Mackley and Sherman [128] used a direct connection of the US as a cleaning technique and monitored particle deposition during UF sub-millimeter particles. The authors found that the development of a cake fouling layer has virtually ceased. In some cases, the close distance between the membrane surface and the US emitting surface may cause damage to the membrane surface, especially with high US power. Juang and Lin [109] found out that the polymeric membrane could get slightly damaged when the US power was 80 W, especially when the emitting horn surface was 10 mm below the membrane surface.

The US power is considered an important parameter due to the high effect on fouling removal and water flux recovery [92, 97, 124]. , In general, permeate flux increases linearly with US power up to a certain limit, after which no significant permeate flux improvement is observed. Furthermore, operating at elevated US power could also damage membrane material [109].

Researchers also evaluated the coupling of US with backwash techniques. Chai et al. [34] utilized an ultrasonic bath at 45 kHz and 2.73 W/cm^2 to clean UF and MF membranes fouled by peptones using a cross-flow filtration cell. The study revealed that cleaning fouled membrane by combining US and backwash was better than the US alone. Furthermore, Secondes et al. [83] and Naddeo et al. [129] reported removing emerging contaminants from wastewater by activated carbon adsorption was about 90%, but decreased over time. However, the removal increased to almost 100% by applying the US, especially with a low frequency of 35 kHz.

The cleaning process by the US is also affected by the type of membrane material. Thus, membranes made from the mixed ester of cellulose nitrate with cellulose acetate, nylon 6, and polyvinylidene fluoride materials could be affected strongly by the US. In contrast, PES material was only slightly affected [91]. The observed effects may be due to depolymerization reactions enhanced by US irradiation via temporarily dispersing aggregated or permanently breaking chemical bonds in polymeric chains [34]. Using low-frequency US, the polyvinylidene fluoride is more resistant, and less change occurs on the surface [34].

3.3. NF-US

NF membrane fouling is a critical issue, as it is responsible for the deterioration of the membrane performance [130, 131]. It was mentioned that the cost of fouling control is almost 30% of the total operating cost [132]. US technology was proposed by many researchers as an alternative cleaning technique to control NF membrane fouling [79, 112, 113]. US frequency is one of the main parameters that significantly affect the cleaning process. Tejal and Kaushik [111] studied low-frequency US effects with two different modes (continuous and intermittent) to remove the fouling accumulated on the membrane surface. They found that permeate flux increased by 3% - 4% when the US was applied continuously or intermittently for 160 min. Continuous ultrasonic irradiation mode was more effective than the intermittent mode, but the intermittent mode is still a better option when energy efficiency is considered [133, 134].

The second main parameter is the US's power, which could have a massive impact on the treatment process. Some researchers investigate the effect of high power on cleaning efficiency and permeate flux enhancement. In a study by Renata et al. [135], high ultrasonic power of 240 W was applied to clean the NF membrane used in treating artichoke's solid wastes no significant effect on the fouling layer was observed. Still, the highest chlorogenic acid recovery was achieved when the US power was at 240 W. Thombre et al. [79] used US technology for cleaning fouled NF membranes. An ultrasonic power of 135 W achieved the best cleaning process, while with a higher power of 150 W, pitting and corrosion was detected on the membrane surface. These results agree with a study by Muthukumaran et al. [106], who used 300 W of US power. The authors also mentioned that permeate flux recovery of the NF membrane increased by 90% in only 4 min of US.

It should be noted that applying a high power US increases the energy required for UF process and the major parts of US waves would be wasted [136]. Many researchers used the US to assist other cleaning techniques such as chemical and physical to avoid more energy waste. Liu et al. [113] used the US-assisted chemical cleaning at a frequency of 25 kHz and a power of 70 W. They found that US technology is a more effective way to improve chemical cleaning. They reported that the recovery rate reached up to 95.6% by applying US-assisted chemical cleaning. Also, Jian et al. [112] used US-assisted chemical cleaning for fouling removal caused by inorganic scales in arsenic-rich brackish water. Despite the increase in permeate flux, which reached 80% when the membrane was cleaned only by chemicals, the NF membrane water flux reached 99.99% when the US power intensity of 1 W/cm^2 was applied.

3.4. RO-US

Reverse osmosis (RO) is a well-established conventional desalination and water purification technology that uses a semipermeable membrane. RO technology is successfully used for the treatment of seawater and groundwater. Despite the advantages of RO technology in water purification [87, 88], it presents some disadvantages, such as sensitivity to pH and ionic strength, high energy consumption, and requirements for pre-treatment and membrane fouling [137]. The RO process requires high pressure (usually 0.2-1.7 MPa) for fresh and brackish water and 4-8.2 MPa for seawater treatment [138, 139]. The high-pressure demand translates into a higher pressure drop inside the module and reduced membrane permeability, which increases the pumping cost and alters rejection [140]. By applying high pressure, the membranes also become susceptible to fouling which clogs their pores [141] and reduces the permeate flux.

Researchers tested different chemical, physical, and US technology techniques to improve the permeate flux and reduce membrane fouling [39, 91, 109]; the latter technique is the subject of interest. Most US applications for alleviating RO fouling and improving permeate flux have been implemented in ultrasonic bath configuration on a lab-scale, as shown in Figure 5. Rarely, the US could cause damage to the RO membrane during the treatment process, which required more attention when US parameters were selected. Yong et al. [115] compared US application with acid and alkali agents for RO membrane cleaning while treating pharmaceuticals wastewater loaded by organic compounds. It was found that 50 kHz frequency and 0.64 Wcm^{-2} power were the most effective US cleaning parameters. However, membrane damage occurred when the US power was 0.636 Wcm^{-2} and applied for 60 min. The study highlighted two observations from the membrane damage test, including lengthy treatment time and dry storage of membrane make it more susceptible to structural damage. Feng et al. [114] tested a combined RO-US system to reduce the fouling layer on the membrane surface and increase the permeate flux without causing any damage to the membrane. Permeate flux improvement of the RO process was attributed to the US cleaning. However, the increase in permeate flux of the RO process by the US technology was not high enough. The authors explained this by the deposition of $CaSO_4$ due to hot spots created by US cavitation leading to a reduction in the $CaSO_4$ solubility [142]. A slight improvement is likely due to the advanced crystallisation stage such that complete dislodgement was not possible, especially that US effects are contactless with the membrane.

Sanderson et al. [80] suggested that integrating US treatment with the RO system during operation could remove quickly built $CaCO_3$ from the membrane surface, which facilitated permeate flux improvement. The reason behind the quick fouling is that the $CaCO_3$ might be transformed into more stable calcite crystals from a meta-stable aragonite form after 7 h of operation due to the unstable ambient temperatures and pressure [143]. After fouling accumulation, the membrane was cleaned with DI water backwash for 3 h. This exercise did not clean the membrane surface efficiently, as seen in Figure 6b. Hence, US irradiation was used after 7 h of operation and was found to be efficient in almost complete removal of $CaCO_3$ (Figure 6c). Although permeate flux increased after US application, it has never returned to the permeate flux of a virgin membrane. The study also found that the cavitation of the US reduced concentration polarization and the clogging of the membrane pores during the operation of the RO system [144]. Using the US with biofouling remediation in membrane filtration, Raed et al. [145] used a combination of US and heat (thermosonication) to remove biofilm developed by *E. coli* from the RO membrane. The study showed that using thermosensation, the developed biofilm was less dense with a smaller number of active microbes due to the biocidal effects where some cells were killed, while others survived but remained injured, which in turn caused starvation.

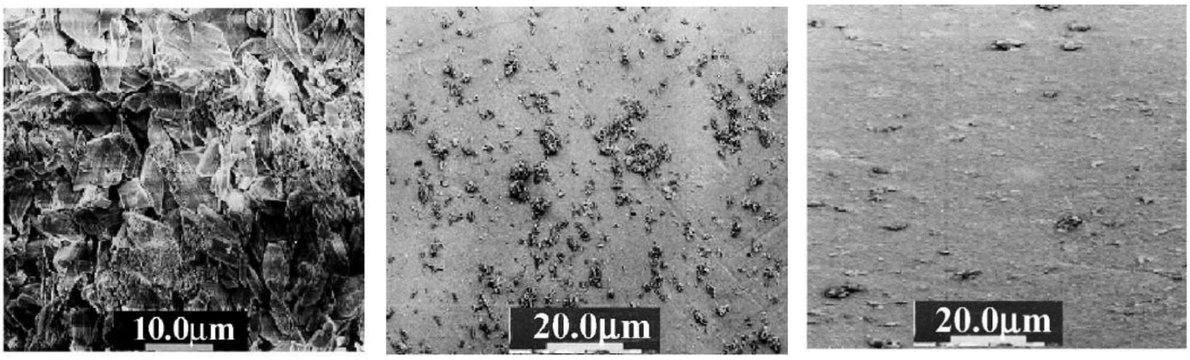


Figure 6: SEM images of a membrane surface: (a) after 7 hrs of operation; (b) after 3 hrs of cleaning with water; (c) after 0.5 h US treatment with dilute HCl [80].

4. US application with membrane bioreactor systems (MBR)

The MBR can be categorized into two types, namely aerobic membrane bioreactors (AeMBRs) and anaerobic membrane bioreactors (AnMBRs) [146-148]. Even though air (in AeMBR processes) can reduce the membrane fouling by scouring the membrane surface [149], the membrane bioreactor system was stressed due to fouling deposition on the membrane surface. As such, integrating the US with an MBR was introduced prominently to tackle the fouling problem. Jai et al. [150] suggested and tested catalytic US oxidation (CUO) with membrane bioreactor for treating real wastewater. Integrating the US with catalytic oxidation resulted in a high removal of total organic carbon (TOC) and improved biodegradability of recalcitrant contaminants in wastewater at US frequencies of 35-65 kHz [151]. The study of Pendashteh et al. [152] utilized the US process for cleaning the MBR system, which was used for treating synthetic hypersaline oily wastewater samples. The US cleaning removed the fouled layer and recovered the permeate flux for a long time.

In a study carried out by Xu et al. [153], an integrated AnMBR-US system (Figure 7) was applied for the digestion of sludge under high volatile solids (VS) at a loading rate of (3.7 gVS/L d) for 54 days. Although the US process could successfully control the cake layer formation on the membrane surface, it had only a slight effect on the gel layer removal formed by the adsorption of proteins and humic compounds. Sui et al. [154] tested intermittent US applications with an MBR system to reduce fouling development on the membrane surface. This study found that increasing the sludge concentration in the reactor increased the need for longer ultrasonic irradiation. The study pointed out that the US irradiation had a small negative impact on the anaerobic bacteria activity; however, this did not affect chemical oxygen demand (COD) removal. Ruiz et al. [155] studied the effect of ultrasonic frequencies in a range of 20 kHz - 40 kHz on membrane integrity, process performance and effluent quality using four different UF modules. The fouled membrane received two different cleanings: the US for 3 s every 3 min with the power of 150 W and various frequencies or backwash for 1 min with 5 s of aeration. The highest cleaning effect was observed with a frequency of 20 kHz with no sign of damage to the membrane surface.

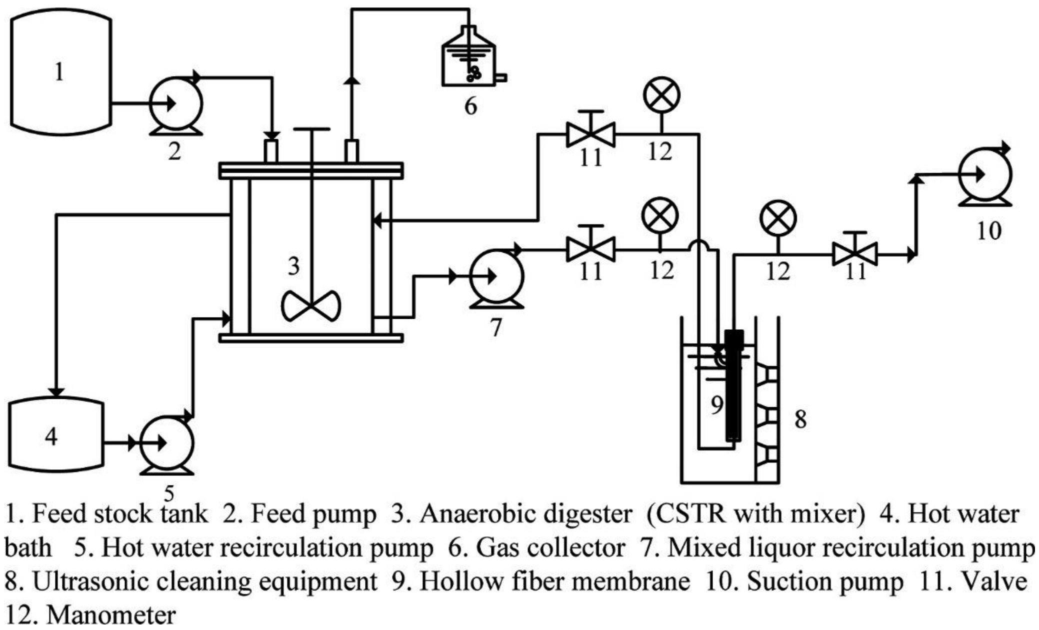


Figure 7: The flow diagram of the US-AnMBR [153].

Another study by Ruiz et al. [156] found that even though the high US power of 300 W and 400 W increased the turbidity of the effluent from 2 NTU to 20 NTU, other parameters like viscosity, colour, effluent COD and total suspended solids concentration did not change. This could be due to the deflocculation of the sludge under ultrasonic irradiation [157]. Li et al. [158] used US for cleaning the fouled membrane in a submerged membrane bioreactor under different US frequencies of 25 kHz - 90 kHz, and applied power of 200 W – 300 W. The results of this study showed that the US could reduce the quantity of the sludge produced with the MBR system. The higher the transmembrane pressure, the higher the fouling layer on the membrane surface. Hence, fouling removal by the US would reduce the transmembrane pressure in the filtration process.

Sui et al. [159] applied US technology to clean the membrane used in an anaerobic membrane bioreactor. It was noted that the US effect on fouling diminished when the crossflow velocity was greater than 1 m/s as the fouled membrane could be cleaned by hydrodynamic forces. On the other hand, the total filtration resistance was drastically improved when applying the US with a crossflow velocity of less than 1 m/s, and the membrane fouling rate was 8.33×10^{6} $m^{-1}s^{-1}$ and 3×10^{7} $m^{-1}s^{-1}$. The study reported a stable low total filtration resistance for one week with ultrasonic power of 150 W. Abdurahman and Azhari [160] tested US-AnMBRs to treat oil mill effluent with multi-frequency ultrasonic transducers. The study found that this system could achieve COD removal of 98.7% with a hydraulic retention time of 4 days and maximum methane production of 0.47 L/g COD·day. Similar findings were also reported by Shafie et al. [161]. The authors indicated that the violent mechanical effects of the US are responsible for membrane damage and the interaction of the strong oxidants produced due to bubbles collapse with membrane materials. Liu et al. [162] investigated the effect of the online US-MBR system on removing organic pollutants from the membrane surface. The study found that the activity of the biological process was increased when the US was applied with the power of 10 W. This increment in the activity of biological was due to the turbulences accompanying propagation of US waves and the cavitational effects, which can increase the mass transfer by moving the particles in a fluid and increase the production of the extracellular enzyme.

Yoon et al. [163] studied the effect of *ex-situ* US on removing sludge production in the MBR system using submerged hollow fibre membrane with the power of 600 W and a frequency of 20 kHz. This study found that the mixed liquor suspended solids (MLSS) were kept constant in the range of 7000 mg/L - 8000 mg/L when the US was applied, while the range of the MLSS increased from 7000 mg/L to 13700 mg/L without US application. This is attributed to the virtue of the US in preventing excess sludge production. Joshi and Parker [164] used the US as a pretreatment with hydrogen peroxide to treat waste stream before digesting in a submerged anaerobic membrane bioreactor. The study showed that COD solubilization increased by about 40% when the hydrogen peroxide dose was 50 g/kg TS and sonication operated for 60 min. Wu et al. [165] investigated the US irradiation effect on the liquor properties of activated sludge using a power range of 40 W - 300 W, volatile suspended solids concentration of 6 g/L, and a concentration range of mixed liquor suspended solids of the activated sludge of 10 g/L -12 g/L. The results showed that US treatment with an intensity of up to 2 kJ/mL could increase the width of particle size distribution and the biodegradability of the activated sludge. Pardo et al. [166] used US combined with ozonation (O_3-US) to treat wastewater prior to the submerged MBR. The study found that the decomposition of the organic compounds was increased due to the O_3-US treatment, resulting in a decrease in the microorganism's growth. Hence, the concentration of extracellular polymeric substances was reduced by around 50%. Overall, US technology could improve MBR performance by increasing nutrients degradability. Improving mass transfer across the membrane and reducing membrane fouling. However, the positive effects can only be achieved if the proper US parameters are selected.

5. Integrating US with emerging membrane technologies

5.1. Ultrasound- Forward Osmosis (US-FO)

The FO process uses natural osmotic pressure difference of feed solutions of different concentrations to transfer water through a semipermeable membrane from the higher solute concentration side to the lower solute concentration side. FO is far more energy-efficient and lower membrane fouling than the RO process [167]. However, the FO process also suffers from membrane fouling, especially when treating a low-quality feed solution. Integrating US technology with FO could be an attractive solution to this problem. Heikkinen et al. [168] tested the US-assisted FO process and found that permeate flux ofFO system was increased after applying US technology. The US irradiation improves the FO process performance by reducing both internal (ICP) and external (ECP) concentration polarization effects (Table 2).

Choi et al. [169] found that US combined with FO decreased the concentration polarization occurrence and membrane fouling and improved membrane efficiency. However, the US effect on the membrane's durability is not obvious, and the outcome of previous studies on this issue has been contradictory. Chanukya and Rastogi [170] investigated the US effect on FO membrane concentration polarization while treating fruit juice and natural colorant. The authors found that US applications can increase permeate flux due to ECP mitigation on the feed side and ICP in the support layer. Despite the permeate flux of the FO membrane enhanced by US technology when the concentration of sucrose was up to 5%, the authors found that US was not able to mitigate the ECP and prevent fouling layer formation when pectin was present in the feed solution which resulted in a reduction in the permeate flux. Chio et al. [171] also studied the effect of US on ICP during the FO process with flat sheet membrane by utilizing different US frequencies of 25 kHz, 45 kHz and 72 kHz and power of 10 W 70 W. The authors found that US technology could significantly mitigate the ICP by improving the diffusion rate of a draw solution. The authors also reported that membrane damage was observed at the US frequency of 25 kHz and 50 W of the applied power, leading to a 3000% increase in permeate flux. This damage is likely caused by changes in membrane properties which were significantly affected by the US. The low-frequency US irradiation was proposed by Wang et al.[172] to improve the permeate flux of the FO process with TFC PES-based polymeric membranes. The authors found that the US significantly improved the FO process permeate flux via mitigating ECP effect. Lee et al. [173] studied the effect of US cleaning on the FO membrane fouled by activated sludge was investigated. An effective fouling removal was also observed when the US was combined with flushing. Thus, 40 % of permeate flux was recovered when the US was used for 60 s, while with flushing only, the permeate flux of the FO was recovered only by 29% [153]. Nguyen et al. [174] used the US to control the fouling on the FO membrane. The study found that the sludge concentration reached 20,400 mg/L and 28,400 mg/L from the initial sludge concentration of 3,000 mg/L and 8,000 mg/L with 40 kHz after 22 hours. However, from an energy requirements perspective, this method is not an energy-efficient technique.

5.2. US-MD

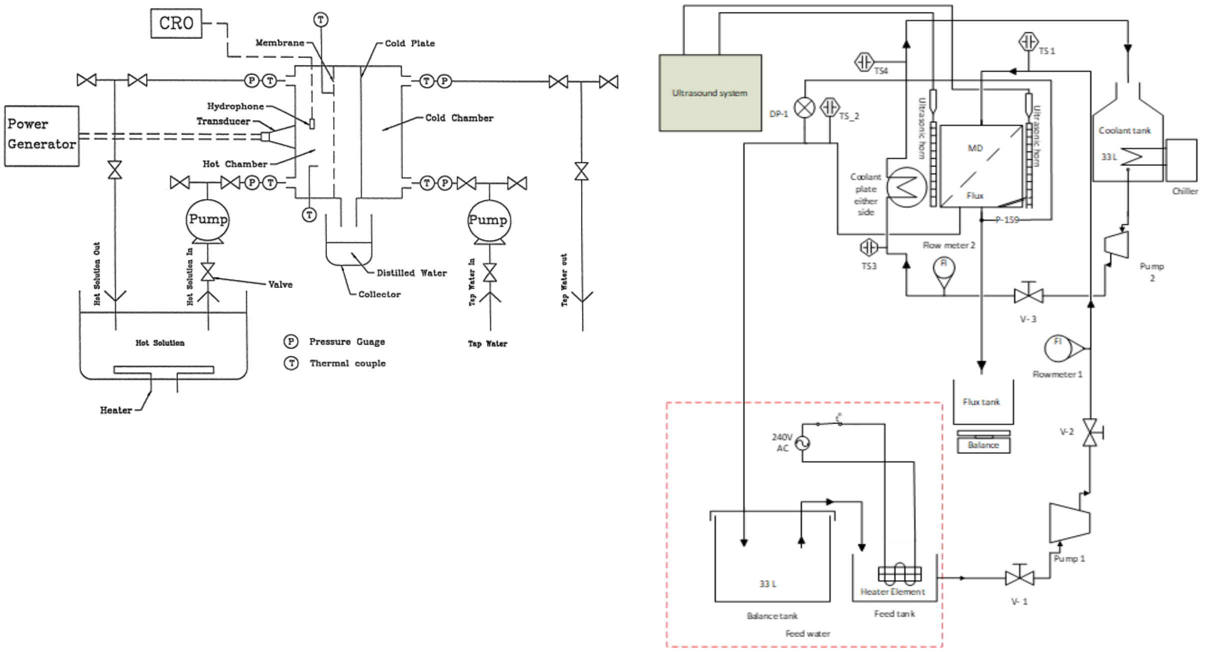
5.3. Ultrasound- Air Gap Membrane Distillation (US-AGMD)

AGMD has many advantages, including cost efficiency [1], lower chemical demand [175], no feedwater pretreatment [40] and low membrane damage [176, 177]. Moreover, AGMD is capable of separating all non-volatile matter under moderate operating conditions that ensure system reliability and durability with no requirement for additional complex condensers [178, 179], which are needed in vacuum membrane distillation (VMD) and sweeping gas membrane distillation (SGMD) [180]. Although AGMD has witnessed many physical developments, the permeate flux of the AGMD is still low compared to some other membrane separation processes. Another limitation of the AGMD system is that the fouling layer is quickly built on the membrane surface due to the relatively big pore size of the membrane (0.2 μm – 1.0 μm) [181]. This layer can prevent water vapour from crossing the membrane, resulting in low permeate flux. The AGMD process can be integrated with US technology to overcome fouling and improve water flux, as seen in Figure 8 [35, 59, 180]. Technically, the US energy could be converted to heat which can reduce the heat transfer loss across the membrane and therefore increase the permeate flux of the AGMD). Table 2 shows a summary of studies which utilized US for mitigating fouling problem in MD processes, including AGMD. Although the US has a benign environmental effect, it can increase the water treatment energy consumption. Also, applying high power of US, waves may damage the membrane surface [39].

Table 2. Effect of US technology on FO and MD performance.

Connection type	**Module type**	**Parameters**	**Feed concentration** **Units should be here**	**Initial water flux,** **kg/m^2·h**	**US water flux,** **kg/m^2·h**	**Percentage increase**	**Rejection, %**	**Ref**
In- situ	AGMD	Feed temp:50°C, coolant temp: 20°C, feed flow: 60L/h, coolant flow: 200L/h, US power: 24W/m^2, US frequency: 20kHz	natural groundwater 12960µS/cm	0.6	1.2	100%	99.98	[39]
			RO reject water 3790µS/cm	0.5	1.0	100%	99.98	
Ex-situ	AGMD	Feed temp: 35 to 75°C, coolant temp: 25°C, feed concentration: feed flow: 0.063m/s, US power: 30W, US frequency:20kHz	sodium chloride 0.5 wt.%, 1 wt.% and 5 wt.%	N/A	1.06	5%-30%	N/A	[180]
			Tap water	N/A	1.15	5%-30%	N/A	
Ex-situ	DCMD	Feed temp: 35°C, coolant temp: 20°C, feed flow: 0.25m/s, coolant flow: 1.0m/s, US power: 260W, US frequency: 20kHz	$CaSO_4$ 2000 mg/L	0.415	0.915	100%	100	[182]
			$CaCO_3$ 100mg/L	0.95	0.96	1%	100	
			SiO_2 150mg/L	0.8	1.0	20%	100	
Ex-situ	DCMD	Feed temp: 53°C, coolant temp: 20°C, feed flow: 0.25m/s, coolant flow: 1.0m/s, US power: 260W, US frequency: 20kHz	Humic acid (HA) 10 mg/L	1.76	NO experiment	N/A	99.97	[183]
			Humic acid (HA) 50mg/L	1.65	2.1	30%	99.97	
			HA 50mg/L + $CaCl_2$ 2 mM.	0.9	0.99	30%	99.97	
			HA 50mg/L + $CaCl_2$ 10 mM.	0.85	0.97	30%	99.97	
			HA 50mg/L + $CaCl_2$ 20 mM.	0.76	0.95	30%	99.97	
Ex-situ	DCMD	Feed temp: 53°C, coolant temp: 20°C, feed flow: 0.25m/s, coolant flow:	silica concentration 150 mg/L,	1.5	2.1	43%	100	[38]

		1.0m/s, US power: 260W, US frequency: 20kHz						
Ex-situ	FO	Feed temp: 27°C, feed flow: 150ml min^{-1}, US frequency: 30kHz	Fruit juice and natural colorant	10 $lm^{-2}h^{-1}$	12 $lm^{-2}h^{-1}$	20	N/A	[170]
Ex-situ	FO	Feed temp: 20°C, feed flow: 0.25m/s, US power: 10- 70W, US frequency: 25, 45, 72kHz	NaCl	3.7 LMH	8.4 LMH	129	N/A	[171]
Ex-situ	FO	Feed temp: 20°C, feed flow: 1L/min, pressure 5bar, US power: 30W, US frequency: 72kHz	calcium sulfate	10 LMH	16 LMH	60	N/A	[169]
Ex-situ	FO	Feed temp: 40°C, feed flow: 1.2L/min, pressure 3.1bar, US power: 50-300W, US frequency: 22kHz	sodium sulphate	11 LMH	23 LMH	110	N/A	[168]
Ex-situ	FO	Feed temp: 25°C, feed flow velocity: 3.8m/s, pressure 3.1bar, US power: 1800W, US frequency: 57kHz	activated sludge	6.5 $Lm^{-2}h^{-1}$	8.5 $Lm^{-2}h^{-1}$	40	N/A	[173]
Ex-situ	FO	Feed temp: 25°C, feed flow: 280mL/min, pressure 3.1bar, US power: 1800W, US frequency: 40kHz	waste activated sludge	N/A	N/A	N/A	98	[174]
Ex-situ	FO	Feed temp: 20°C, feed flow velocity: 0.28m/s, US power: 0.2 to 0.8W/cm^2, US frequency: 40kHz	sodium chloride solution	18 LMH	20 LMH	18	100	[172]



(a) (b)

Figure 8: Shows the AGMD-US system, (a) *ex-situ* [180] and (b) *in-situ* [39].

The possibility of integrating AGMD with US technology was proposed and tested by Zhu et al. [180] for two types of feed solutions. The study found that the higher the US irradiation power, the higher the permeate flux would be. It is also found that a higher feed temperature can improve the permeate flux of the AGMD with the same US intensity. The reason behind this increment is that water flux in the AGMD process depends on the temperature difference between both sides of the membrane. The permeate flux of the AGMD increased when the US was operated for 10 min each 30 min, demonstrating its ability to break the cake layer built on the membrane surface [40]. Another study by Naji et al. [39] designed an integrated US-AGMD system to treat natural groundwater (3,970 µS/cm), and RO rejects water (12,760 µS/cm). They found that the US technology could bring a 100% improvement in permeate flux 100% by removing the fouling cake layer and improving mass transfer across the membrane. The study used a new technique in which US transducers is directly connected to the spacers on both sides of the membrane (*in-situ*).

5.4. US-DCMD

Another MD process that has been frequently probed in water desalination is direct contact membrane distillation (DCMD). Its advantages include low working temperatures, operation at atmospheric pressure and high salt rejections [184-186]. However, DCMD performance is accompanied by membrane fouling which significantly impends the permeate flux and increases operation costs. Therefore, to improve the permeate flux of the DCMD and reduce fouling on the membrane surface, it is suggested to integrate DCMD with US technology, as shown in Figure 9. Several researchers focused on integrating DCMD with the US to overcome membrane fouling and improve the permeate flux [38, 59, 182, 183]. Hou et al. [182] designed and tested four transducers located outside the water bath to treat three different synthetic water samples containing $CaSO_4$, $CaCO_3$, and SiO_2. The study found that the permeate flux of the DCMD reduced by 55% when $CaSO_4$ concentration increased from 1 mg/L to 4 mg/L due to the precipitation of $CaSO_4$ salt on the membrane surface [182]. In another experiment, ~20% reduction of permeate flux was achieved when feed solution contained Na_2SiO_2 due to formation and deposition of colloidal polysilicic acid on the membrane surface. However, when the US was applied, no permeate flux decline was observed due to US cleaning of the membrane surface. The rejection rates in all experiments with and without US treatment were around 99.99%.

Furthermore, the US exhibited a more pronounced effect on permeate flux recovery at a higher salt concentration factor. The authors also showed that the US did not affect the membrane integrity since the *ex-situ* US connection kept the emitting surface away from the membrane surface. Another study by Hou et al. [38] used a PTFE membrane for treating synthetic water containing silica with a concentration of 150 mg/L. During DCMD experiments, the feed water was not diluted while the silica was added to the DCMD-US experiments. The study found that the permeate flux during the stand-alone DCMD process decreased by around 20% when the silica concentration factor peaked at 4. Contrarily to this, permeate flux decreased during the

DCMD-US process was insignificant and comprised ~97% of the virgin membrane permeate flux. The SEM images (Figures 10a-c) confirmed an amorphous silica-scaling layer formed on the membrane surface after the stand-alone DCMD process. In comparison, SEM images of the PTFE membrane surfaces used in DCMD-US experiments had no silica layer on the membrane surface. Figures 10d-e demonstrate the effectiveness of US technology to remove fouling materials from the membrane surface even when Ca2+ ions exacerbated membrane fouling. Furthermore, permeate flux of the combined US-DCMD system was 2 kg/m^2, 34% higher than the permeate flux observed with stand-alone DCMD. Another study by Hou et al. [183] utilized PTFE membrane to treat synthetic feed, which incorporated 50 mg/L of humic acids (HA) and $CaCl_2$ in a range of 2 mM – 20 mM. The authors found that US irradiation enhanced permeate flux by more than 30% without affecting HA rejection. In addition, permeate flux enhancement increased with a concentration factor

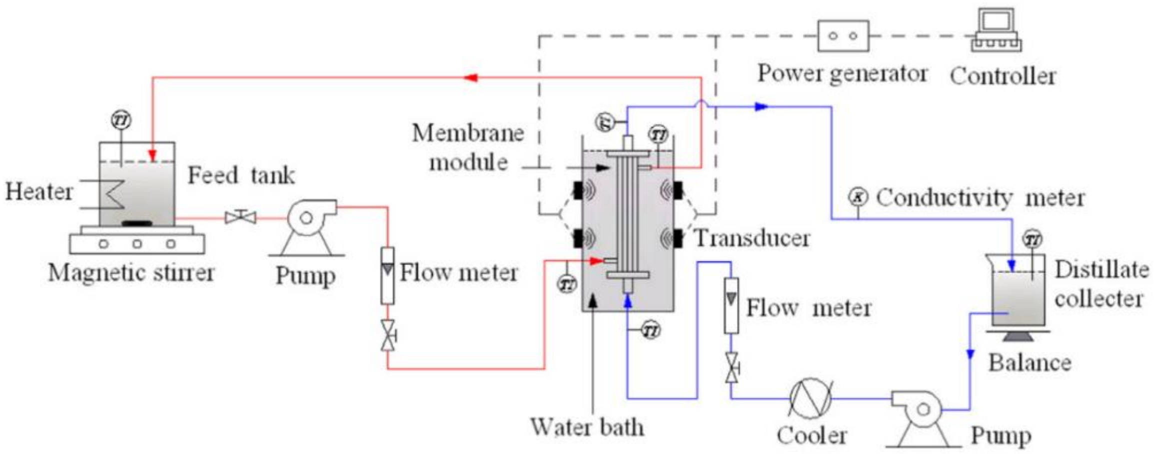


Figure9: DCMD integrated with US [38].

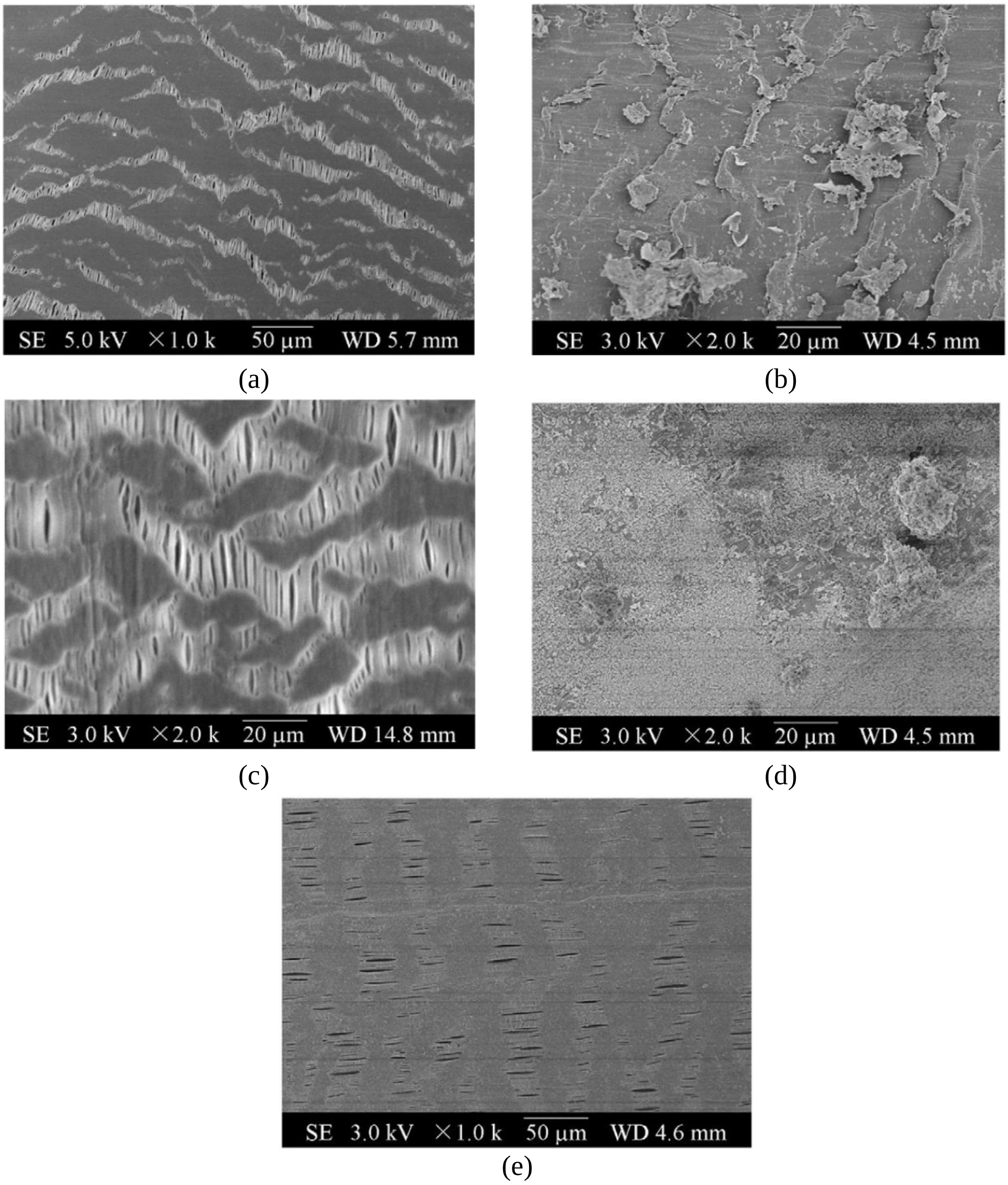


Figure 10: SEM images of the PTFE membrane, (a) virgin PTFE membrane, (b) PTFE membrane after silica fouling, (c) PTFE membrane after silica solution concentration with US irradiation, (d) after silica solution concentration experiment running 30 min in the presence of Ca^{2+} ions, (e) after silica solution concentration experiment in the presence of US irradiation [38].

6. Overview of ultrasound effect on membrane properties

The discussion regarding the change in membrane properties upon exposure to ultrasound effect has been mainly focused on membrane physical structure as discussed in previous sections. It is important to ponder about this point beyond optical or microscopic examination.

In general, membranes can be classified into organic membranes that absorb the mechanical effects of ultrasound (e.g. shock wave, streaming) and inorganic that reflect the energy produced from the mechanical effects of ultrasound. For both membrane categories, ultrasound treatment can change their roughness and porosity. Several studies reported pores enlargement and structural damage after ultrasound treatment for polymeric membrane as mentioned in Table 1. However, the extent of ultrasonic effect on polymeric membrane structure varies depending on their chemical structure. For instance, Masselin et al. [187] reported crevices in PES membrane, while PVDF and PAN membranes did not show sign of structural degradation under the same ultrasound treatment conditions. Pitting of inorganic membrane surface is a possible scenario when treated with ultrasound especially at high power and short distance between emitting surface and membrane surface. Once the roughness of membrane surface increase, the possibility of heterogeneous cavitation on membrane surface increases. This in turn can deteriorate the membrane structure through the continuous oscillation of heterogeneous cavitation bubbles [106].

The impact of the physical effects of ultrasound on membrane properties have adequately been studied, however the impact of the chemical effects is rarely discussed in the literature. It is important to remember here that high frequency ultrasound produces more chemical effects (i.e. generation of free radicals) compared to low frequency. Hence, high frequency ultrasound is expected to cause change in membrane surface chemistry. It was reported that the production of radicals such as OH• O• and oxidant agents such as H_2O_2 may cause chemical bonds scission of membrane materials [51]. The quantity and the aggressiveness of produced radicals and oxidants depends on many factors such as power intensity and presence or absence of radicals scavenging and promoting agents. For example, the presence of Fe^{+2} facilitated the degradation of ionomer membrane, Nafion® 117 through the hydroperoxyl radical attack on main and side chain of the polymer [188]. The produced free radicals with ultrasound can also interact with the membrane surface altering its properties. Free radicals can interact with the dissolved oxygen and the carbonous structure of organic membrane producing carboxyl and carbonyl groups that makes the membrane more hydrophilic [189]. In order to accurately capture the changes that occur in membrane properties, long term tests and advanced analytical chemical examinations are recommended to be applied as such changes can be subtle and hard to detect in short-term tests and crude analyses.

7. Membrane-assisted ultrasound technology: recommendations for future research directions

There is a plethora of successful US applications to improve membrane separation technologies. However, studies in this field seem to linger at the lab testing phase. This is likely due to the limited knowledge available on the intimately linked interactions between the US effects and the operation parameters of different membrane processes. The majority of the research in this field utilizes off-shelf US systems not designed for this particular purpose. Failing to tailor US reactor design and operating conditions to suit process requirements may mislead the evaluation of its true value and capacity. Since some aspects of the US-assisted membrane technology were investigated more extensively than others, we believe it is worth conducting a stocktaking exercise of the research maturity in these aspects as presented in Table 4. The content of Table 4 was formulated based on the up-to-date literature survey carried out in this study. The level of research maturity of each process aspect was categorized based on the number of studies available into comprehensive, reasonable and insufficient. It appears that among all the identified research aspects, only US power and the use of piezoelectric transducers in *ex-situ* configuration were studied in an adequate depth. Other aspects such as US frequency, type of feed water and operation mode were explored only in a few studies.

Furthermore, most of these studies were focused on treating synthetic feed waters by applying continuous US with frequencies below 100 kHz. Unlike low frequency, high frequency is expected to produce less vigorous US effects, reducing the risk of membrane damage. High frequency may bring about chemical changes in the membrane surface if it is applied at power level higher than the cavitation threshold. At low power level, high frequency is expected to produce larger number of vibration cycles compared to low frequency, and this might be beneficial especially for the dislodgment of foulants from membrane surface. As such, there is a need to test US-assisted membrane technology with different natural waters (e.g., seawater, groundwater, industrial and municipal wastewaters, etc.) in a high-frequency range of 200 kHz - 1 MHz in different operational modes. Water samples with high ionic strength such as seawater and groundwater were found to enhance ultrasonic effects [190]. They also contain ions such as chloride that could scavenge hydroxyl radicals reducing its possible negative effect on membrane structure.

The use of magneto-strictive transducers, various waveforms, *in-situ* configuration, large and long-term trials, as well as cost analyses are also hardly investigated. As explained in Section 2, magneto-strictive transducers are more robust and suitable for industrial applications than piezoelectric transducers. Therefore, assessing US application for improving membrane separation performance could also be explored by utilizing this type of transducers. Most of the available US equipment is driven by a sinusoidal wave, while there is a wide range of other forms such as square, triangle and sawtooth that may offer a better choice for US-assisted membrane technology.

Incorporating US technology with membrane filtration processes may increase the capital and operational cost. To justify US applications in membrane filtration processes, there should be a remarkable improvement in the filtration processes, especially in treating low-quality wastewaters laden with contaminants that cause irreversible membrane fouling. Such wastewaters require intensive pre-treatment and frequent membrane cleaning, leading to significant operating costs increase. US application could also be justified to reduce the process downtime by providing a constant filtration process without interruptions. The cost of US application in water treatment would be justifiable in membrane processes for resources recovery to achieve an elevated recovery rate without membrane fouling or damage.

In the future, it would be useful to test different waveforms for membrane performance improvement. Additionally, a process scale-up and proper cost analysis of the long-term experiments covering capital and maintenance expenditures and the return of investment are needed to adequately assess the viability of the US-assisted membrane technology for industrial applications. Several factors affect the US-membrane technology scale-up from laboratory to field, such as type of membrane technology, US method (direct vs indirect sonication), feed water quality, membrane configuration, and purpose of treatment. US technology would be more suitable for treating complex wastewaters containing large amounts of fouling materials that would cause membrane fouling or damage to justify the cost of US installation and use. For instance, industrial wastewaters and concentrated brine are examples of feed waters that require special fouling mitigation measures to avoid membrane fouling or damage. Future work should also investigate the impact of membrane module type on the performance of the US because studies in this field are scarce. Comparison studies will determine the best membrane module for US application, depending on its configuration, materials and packing density.

Regarding the scale-up opportunities for US-assisted membrane technology, the authors can offer adjudication informed by literature knowledge and experience in the subject matter. The opportunities for scaling up US-assisted pressure driven membranes lie in pre-treatment and post-cleaning applications. Other configurations of the process (see Figure 4) require high energy to overcome the pressurised environment in the first instance prior to producing any useful effects. This portion of energy can be considered waste as it does not return any benefits to the overall process. The scale up opportunities for other membrane types (e.g. thermally and osmotically driven) are wider. Technically, all systems configurations shown in Figure 4 can be applied in thermally and osmotically driven membranes as sound waves do not need high energy to breakdown the cohesive forces of the medium. Some of these membrane processes are in the development phase in the present time and this offers a great opportunity for considering the integration of US at early stage of system design.

In addition to the aspects mentioned above, it will be worth investigating the effect of US on other emerging processes (e.g. pressure retarded osmosis) and electrochemical processes (e.g. electrodialysis and capacitive deionization), as well as the resistance of novel membrane materials (e.g. graphene, carbon nanotubes, aquaporin, biomimetic).

Table 4: Maturity evaluation of US-assisted membrane technology research.

Process research aspects		Level of research maturity		
		Insufficiently investigated	Reasonably investigated	Comprehensively investigated
Ultrasonic power				✓
Ultrasonic frequency			✓	
Ultrasonic wave generation	Piezoelectric			✓
	Magneto-strictive	✓		
Operation mode (pulsed, continuous and sweep frequency)			✓	
Waveform		✓		
Feedwater type			✓	
Configuration	*In situ*	✓		
	Ex situ			✓
Large scale trials		✓		
Long term trials		✓		
Proper analysis for capital and operational cost		✓		

8. Conclusions

US coupling with membrane separation technologies has been proposed to reduce fouling and permeate flux increase. The present study reviewed the theoretical and experimental aspects of US technology and links between the US design and membrane system operating parameters and its impact on fouling mitigation and mass and heat transfer enhancements. The efficient application of the US requires prior knowledge of the US design and application method and a deep understanding of the nature of the treated solution and its conditions. Overall, US-assisted membrane processes can maintain the filtration processes without interruption and improve the permeate flux substantially. However, technology is still under investigation, and it is energy-intensive with the potential of negatively affecting membrane integrity if the operating conditions are not properly selected.

The efficient use of US technology to improve membrane separation seems to be limited to laboratory scale. This is likely due to the high operating cost of US technology and the lack of techno-economic studies on the applications of US technology in membrane filtration processes. A proper cost analysis for the long-term tests on a large scale, considering capital and maintenance costs and the return of investment, is needed to adequately assess the viability of applying the US in combination with membrane technology. Future studies should also focus on investigating the type of membranes' modules suitable for the US technology, type of US application method (direct vs indirect), and on-site natural samples testing. Combining the US with membrane filtration is expected to have a niche market in challenging feed such as industrial and municipal wastewater and brine to justify the technology cost. More research should be done in this field.

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