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1 **Ultrasound-assisted membrane technologies for fouling control** 2 **and performance improvement: A review**

3
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16 17 **Abstract**

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

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

38

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

41

42 **1. Introduction**

43 Population increase and rapid industrial development imposed additional demand on
44 freshwater resources [1, 2]. Although developed countries enjoy good quality water provided
45 by centralized municipal water supply systems, safe drinking water remains scarce in
46 developing countries. Contaminants in drinking water are among the most significant issues,
47 and millions of people suffer from their hazardous effects. Different filtration processes and
48 adsorption processes were applied for water cleaning and contaminants removal [3].
49 Membrane-based processes are increasingly applied to overcome water shortage and produce
50 high-quality drinking water by separating water molecules from contaminants. Different types
51 of pressure-driven membrane processes are commercially available for water treatment,
52 including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis
53 (RO) [4-6]. Recently, membrane distillation (MD) is introduced as an emerging technique that
54 combines thermal and membrane separation [7-19]. Membrane distillation (MD) relies on a
55 partial vapour pressure gradient generally caused by a temperature difference across the
56 membrane [20, 21]. Although MD was suggested decades ago, it is still in the developmental

57 stages. One of the major reasons behind its late commercialization is the low recovery rate,
58 severe temperature polarization, and, to a lesser extent, fouling of the membrane, especially
59 when treating concentrated feed solutions [22, 23].

60 A range of cleaning techniques was used to control membrane fouling, including physical [24]
61 and chemical [25, 26] cleanings. The advantages of these techniques include removing fouling
62 materials from the membrane surface and increasing water flux by reducing concentration
63 polarization. On the other hand, disadvantages are mainly i) reduced membrane lifetime [16,
64 27], ii) generation of contaminated wastewater [27], and iii) changes in membrane
65 hydrophobicity and surface morphology [28, 29].

66 Recently, ultrasound (US) was proposed among other innovative cleaning techniques for water
67 treatment processes, such as CO₂ nucleation, which was tested for ultrafiltration [30] and
68 reverse osmosis (RO) [31]. In water treatment context, US can be defined as the application of
69 sound waves in frequency range higher than the human hearing limits. The detailed definition
70 of terminologies used in ultrasound field and the parameters affecting its throughput will be
71 discussed in the following section. US was integrated successfully with pressure-driven
72 membrane separation [32-36] and emerging technology such as MD [37-42] to remove foulants
73 from the membrane surface. The US-assisted membrane processes can significantly improve
74 membrane performance. For example, water flux increase of up to 600% can be achieved with
75 US help [5, 7]. In addition, US technology was applicable for fouling mitigation for various
76 feed solutions such as surface water [4], milk solution [5], soybean [7] and oil wastewater [12].
77 The advantages of this cleaning technique are no chemical usage [43], no system shutdown and
78 no need for membrane removal from the system for *ex situ* cleaning so that possible membrane
79 contact with the air is minimized. Ultrasound removes deposited particles from the membrane
80 surface as a result of it shaking. As a result, permeate flux through the membrane is increased.
81 Ultrasound can also increase a membrane's operation time by reducing the occurrence of

82 fouling events. Several concerns are associated with applying US for mitigating membrane
83 fouling, such as high energy requirements [44], adverse effects on membrane integrity, and the
84 selection of best system configuration that suits large-scale applications [45]. On the other
85 hand, the advantages of US application for fouling mitigation are immense. They include
86 reduced or no chemical usage [46], no system shutdown [39, 40], minimal effects on the
87 environment and human health [39, 40, 47, 48] and high potential of scaling and biofouling
88 removal [49, 50].

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

107 data and recent development in systems configurations. Recommendations and future research
108 directions are also proposed based on literature research findings and authors own views of the
109 process.

110 **2. Ultrasound effects on membrane processes**

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

130 To maximise the benefits of ultrasound application with membrane-based technologies, it is
131 imperative to understand the enhancement mechanisms of ultrasound and how the operating

132 parameters and process environment influence these mechanisms. Ultrasound impacts
133 membrane filtration through three pathways: detaching deposited foulants and driving particles
134 and molecules away from the membrane interface (i.e. reducing concentration polarization)
135 (cleaning effects), improving water transport across the membrane (mass transfer effects) and
136 boosting heat transfer of water for thermally-driven membrane processes [45].

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

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

155 Contrary to the power, increasing frequency reduces the intensity of acoustic events except for
156 acoustic streaming. A study conducted by Costalonga et al. [60] demonstrated that acoustic

157 streaming velocity increases with frequency. The fluid pattern changes with frequency, and
 158 rotational flow diminishes as the frequency increases. A linear motion occurs, especially in the
 159 middle of the irradiating surface, as shown in Figure 1. When it comes to the cleaning effects
 160 of ultrasound, the linear motion can be problematic as it may push the fouling particles deeper
 161 into the membrane pores instead of pushing them away, as observed in the circular motion.
 162

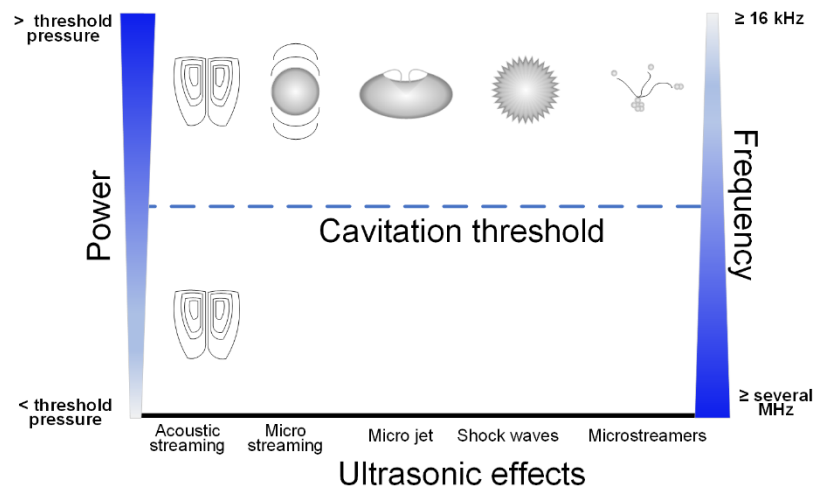


Figure 1: a) Influence of power and frequency on ultrasound effects.

163
 164 Temperature and pressure of the medium can influence ultrasonic events through their effects
 165 on medium properties and bubble dynamics. For instance, higher power is required for the
 166 ultrasonic wave to propagate and generate bubbles in a pressurized medium. The opposite is
 167 also true in a medium under high temperature [61]. Increasing the temperature reduces the
 168 medium viscosity and surface tension facilitating the generation of cavitating bubbles.
 169 However, such action can also generate bubbles with less violent collapse [62].
 170 Fluid properties may also impact the nature of its interaction with ultrasonic waves. For
 171 example, the type of dissolved gas affects the thermal product of the collapse. Gases with a
 172 high adiabatic ratio result in bubble collapse with high temperature [63]. Heavy gases can
 173 produce high collapse temperatures, but they have low thermal conductivity and convey the
 174 heat from collapse sites to the bulk slower than light gases [63]. The fluid content of dissolved

175 and suspended solids can also influence ultrasound effects. It was found that both the number
176 of bubbles and their size decreases with an increasing salt concentration in water [64].

177 The operation details of ultrasonic devices can also play an important role in controlling
178 ultrasonic effects. The effect of such details on ultrasound performance in assisting membrane
179 filtration is scarcely investigated in the literature. These details include the operation mode
180 (continuous or pulsed), vibration generation techniques (piezoelectric or magnetostrictive) and
181 the excitation wave (sinusoidal, square, triangle etc.). Applying pulsed mode was more
182 effective in utilizing energy and producing more cavitation effects [65]. In terms of the
183 operation mode on non-cavitation effects (i.e. acoustic streaming), it was reported that
184 applying this mode reduces the acoustic streaming velocity [66]. Therefore, depending on how
185 vigorous the acoustic streaming needs to achieve treatment performance, such as removing a
186 fouling layer or improving mass/heat transfer phenomena, one can decide whether to apply
187 continuous or pulsed mode. Some studies found continuous mode more beneficial for
188 improving membrane filtration flux [67], while others found that pulsed mode is more effective
189 [68]. The techniques used to generate mechanical vibrations in the transducer impact both the
190 efficiency and durability of ultrasonic devices. Magneto-strictive transducers are reported to
191 be more resistant to mechanical impact, more tolerant to high temperatures and have longer
192 working life compared to piezoelectric transducers [69]. The latter type of transducers is
193 commonly used in membrane filtration studies due to its availability as an off-the-shelf product
194 in the market. This may be one reason that makes the ultrasound technique perceived to be
195 costly. Kyllönen et al., [35] concluded that the main reason that hinders the commercialisation
196 of ultrasonic-assisted membrane technology is the absence of active efforts for developing
197 transducers that cater for this application. The effect of the excitation wave on the transducer's
198 electrical output and the cavitation chemical yield (measured by OH^\cdot and H_2O_2 production)
199 was evaluated by Al-juboori et al. [69]. The results showed that among the tested waveforms,

200 square wave resulted in the best transducer displacement and the highest concentration of OH[·]
 201 and H₂O₂. A numerical study by another team Kerboua, and Hamdaoui [70], on bubble
 202 dynamics under different excitation waveform showed that a square wave produces the highest
 203 pressure and temperature inside the bubble compared to triangle and sinusoidal waves.

204 **2.1. Effects of ultrasound on fouling**

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

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

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

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

220

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

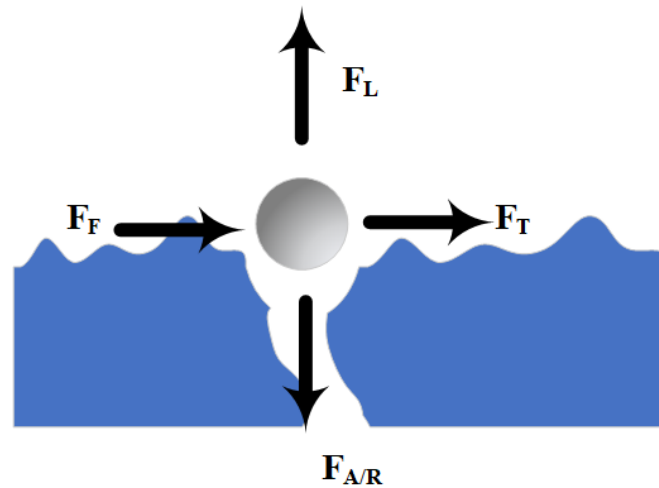


Figure 2: Forces acting on a particle being deposited onto a membrane pore.

225
226 As for the effective range of ultrasonic events, an illustrative representation is provided in
227 Figure 3 [24, 57, 75]. It is clear that except for acoustic streaming, other ultrasonic effects need
228 to occur close to the membrane-water interface to remove particles from the membrane surface.
229 Several studies [68-70] reported these effects are more intense than acoustic streaming, raising
230 concern of possible membrane damage. Strong forces such as those generated by the
231 cavitation effects are only needed when the fouling layer is already established. This also
232 depends on the fouling type: cake layer or pore blocking. The US was found to be less effective
233 in removing pore-blocking fouling as opposed to the cake layer fouling [76]. Given the fact
234 that the US is not effective in removing all forms of developed fouling on the membrane and
235 the potential damage cavitation effects may cause, one can deduce that the most efficient way
236 to apply the US for alleviating the fouling problem is by utilizing low power non-cavitation
237 effects to prevent/reduce fouling formation at early stages of filtration.

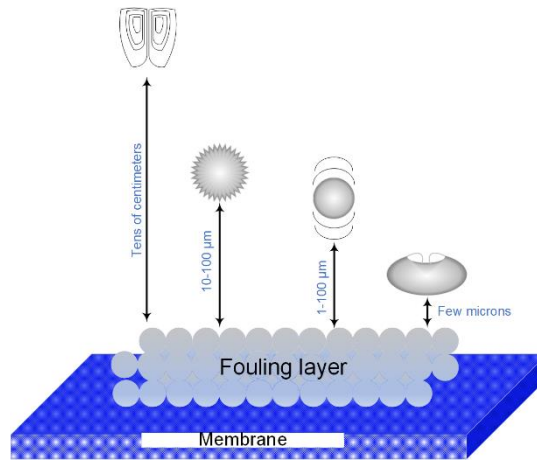


Figure 3: Active ultrasonic effects' distance.

238

239

240 2.2. Mass and heat transfer enhancement

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

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

251

252 The effect of the US on heat transfer is mainly related to its impact on the convective heat
 253 transfer coefficient on the feed side. The US increases the convective heat transfer coefficient
 254 by a component (h_{as} , W/k·m²) presented in eq. 4, where C_p is the specific heat of water

255 (kJ/kg·K). Knowing the velocity of the acoustic streaming and the feed water properties, one
256 can estimate the extent of enhancement expected with a chosen set of operating conditions.

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

257

258 **3. Integration of US with pressure-driven membrane technology**

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

271 **3.1. MF-US.**

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

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

284 A range of synergistic techniques has been reported to improve membrane throughput when
285 combined with the US. Sanderson et al. [80] found that combining forward washing with
286 ultrasound for offline cleaning of MF membrane fouled with paper mill wastewater improved
287 permeate flux by 750% compared to only 300% with ultrasound alone. Another study reported
288 that adding ethylenediaminetetraacetic acid (EDTA) to feed solution while applying ultrasound
289 on the fouled membrane with milk solution enhanced the flux further. A mixed frequency of
290 28, 45 and 100 kHz was the least affected by EDTA addition than individual frequencies [81].
291 There is other possible synergistic processes that incorporate ultrasound with membrane
292 technology and adsorption in a hybrid system as a combination of filtration and adsorption has
293 proven to be effective for treating wastewater [82]. As an example for such hybrid system is
294 the combined UF, US and activated carbon processes tested by Mona et al. [83] for removing
295 industrial dyes. The outcome of these studies is summaries in Table 1. Another synergy that
296 can benefit from ultrasound application is the hybrid electro-chemical and adsorption system
297 such as the one reported by Kadhum et al. [84] if combined with membrane technology assisted
298 by electro-chemical techniques [85]. Although such combination has not been reported in the
299 literature, one can postulate the potential benefit of ultrasound. For instance, ultrasound can
300 improve the adsorption capacity of adsorbents [83] and alleviate the impact concentration
301 polarization on membrane and electrodes [86]. However, ultrasound physical and chemical
302 effects can lead to the destruction of electrodes just as it is the case with possible membrane

303 surface deterioration [87]. The other possible risk with such combined systems is that if the
304 adsorbents are immobilised on membrane surface, ultrasound effects could detach them
305 rendering the membrane structure weak and more prone to serious damage.

306 The compiled information in Table 1 is useful to gain an in-depth understanding of the effects
307 of membrane and ultrasound operating conditions on the overall performance of the US-
308 assisted membrane process. There are three ways for pressure-driven membrane processes
309 through which the US is applied: online flux enhancement, pretreatment and offline cleaning.

310 Online flux enhancement appears to be the most effective form of US application. Examples
311 of common US-membrane design systems are illustrated in Figure 4. In addition to the 3
312 designs mentioned above, there are self-cleaning US-vibrated piezoceramic membranes that
313 have recently been developed and found to increase the flux by about 30% when the vibration
314 is in operation [88]. It appears that increasing the input US energy either through increasing
315 the applied power or the irradiation time affects the permeate flux negatively. This is likely to
316 occur due to high power density, resulting in the breakdown of particles leading to severe pore-
317 blocking fouling. For the case of MF, two studies [67, 89] showed the adverse effect of
318 ultrasonic energy on membrane flux used a high power density of 200 W/l - 300 W/l. Similarly,
319 the increasing frequency seems to result in lower permeate flux enhancement. This has been
320 attributed to the negative effect of frequency on cavitation threshold and bubble growth [90].

321 Evaluating the effect of frequency of flux with non-cavitation effect has not been addressed
322 in the literature. In this case, a higher frequency may be useful as more wave cycles are
323 generated. A mix of low and high frequency was more powerful than the low frequency alone
324 [81]. It appears that the pressure has an inverse correlation with permeate flux enhancement.

325 The latter is expected since pressure increase raises the resistance against the propagation of
326 the sound wave.

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

334 Some researchers have investigated other parameters, such as the distance between the emitting
335 surface of the ultrasound and the membrane surface. Mirzaie and Mohammadi [67] observed a
336 drop in flux enhancement of MF-US from 228% to 145% when the distance between the US
337 horn and the membrane surface was increased from 2.6 cm to 4.4 cm. However, increasing the
338 distance between the ultrasound source and membrane surface does not always have a negative
339 impact on flux enhancement. Thus, [92] showed that increasing the distance between ultrasonic
340 transducer and membrane from 4 cm to 8 cm increased the flux from $5.8 \times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$ to 7.5
341 $\times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$. However, when the distance was further raised to 12 cm, permeate flux declined
342 to $7.1 \times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$. The observed effect was attributed to the uniformity and intensity of the
343 ultrasonic field governed by the applied power, the reactor design and the nature of the
344 irradiated fluid. As stated earlier, the content of the water being irradiated could influence
345 ultrasound performance. It was reported that increasing particles concentration in water from
346 0.1 g/L to 1.8 g/L resulted in a decrease in permeate recovery of US-assisted membrane
347 technology by ~ 60% [93].

348

349

350

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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	Flux enhancement: 300 %.	Not reported.	[94]
	Polyvinylidene fluoride (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: <i>Ex-situ</i> .	Flux enhancement: 400 %, reduced resistance: 77 %.	Not reported.	[81]
	Polyvinylidene fluoride (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: <i>Ex-situ</i> .	Flux enhancement: 280 %, reduced resistance: 71.6 %.	Not reported.	[81]
	Polyvinylidene fluoride (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: <i>Ex-situ</i> .	Flux enhancement: 160 %, reduced resistance: 60.6 %.	Not reported.	[81]
	Polyvinylidene fluoride (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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	Flux enhancement: 36 %.	Not reported.	[90]

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: <i>Ex-situ</i> .	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: <i>In-situ</i> .	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: <i>Ex-situ</i> .	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: <i>In-situ</i> .	Flux enhancement: 400 % reduced resistance: 78 %.	Not reported.	[97]
	Mixed 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: <i>In-situ</i> .	Flux enhancement: 228 %.	None (confirmed by SEM analysis).	[67]
	Mixed 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: <i>In-situ</i> .	Flux enhancement: 184 %.	None (confirmed by SEM analysis).	[67]
	Mixed 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: <i>In-situ</i> .	Flux enhancement: 27 %.	None (confirmed by SEM analysis).	[67]
	Mixed 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: <i>In-situ</i> .	Flux enhancement: 490 %.	None (confirmed by SEM analysis).	[67]
	Mixed 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: <i>In-situ</i> .	Flux enhancement: 274 %.	None (confirmed by SEM analysis).	[67]

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: <i>In-situ</i> .	Flux enhancement: 317 %.	None (confirmed by SEM analysis).	[92]
	γ -alumina membranes	Feed: 0.5 g/L of 1.56 μ m 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: <i>In-situ</i> .	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: <i>In-situ</i> .	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: <i>In-situ</i> .	Flux enhancement: 634 %.	Not reported.	[91]
	Cellulose nitrate 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: <i>In-situ</i> .	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: <i>In-situ</i> .	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: <i>In-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>In-situ</i> .	Flux enhancement: 340 %.	None (estimated 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: <i>Ex-situ</i> .	Reduced resistance: 32.2 %.	None (estimated permeability and selectivity).	[100]
	Polysulfone	Feed: Real wastewater, configuration: hollow fiber, Flowrate: based on fixed flux of 75 L/m ² 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: <i>Ex-situ</i> .	Reduction in fouling rate (kPa/h): 36.54 % increased 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 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: <i>Ex-situ</i> .	Reduction in fouling rate (kPa/h): 15.38 % increased turbidity removal: ~14%, increased UV ₂₅₄ removal: ~60%.	Not reported.	[101]
	Polyethersulfone	Feed: <i>Radix astragalus</i> 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: <i>Ex-situ</i> .	Flux enhancement: 12-15 %, reduced resistance: 40%.	Increased irreversible fouling.	[102]
	Polyethersulfone (PES)	Feed: <i>Radix astragalus</i> 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: <i>In-situ</i> .	Flux enhancement: 68 %.	Rapid temperature increase of up to 55°C.	[102]
	Polysulfone	Feed: <i>Radix astragalus</i> 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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	Increased cleaning efficiency: 17 %.	Not reported.	[106]
	Polysulfone	Feed: real wastewater, configuration: hollow fibre, Flowrate: 127mL/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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>In-situ</i> .	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: <i>Ex-situ</i> , Powder activated carbon (PAC) dose of 0.75 g/m ³ 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: <i>Ex-situ</i> , Powder activated carbon(PAC) dose of 0.75 g/m ³ 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: <i>Ex-situ</i> , Powder 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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> , 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: <i>Ex-situ</i> , 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: <i>Ex-situ</i> .	Flux recovery: 78.51 %.	None (confirmed by re-testing the treated membrane), however, cracks and cavities were observed at frequency of 40 kHz.	[79]
RO	Polyamide	Feed: CaSO ₄ solution (500 mg/L), Configuration: flat sheet, Flowrate: 5 mL/min, Pressure: 100 kPa, Temperature: 20°C, effective area: 56 cm ² .	Power: 2016 W, Frequency: 20 kHz, Time: 3 h, Mode: continuous, Connection: <i>Ex-situ</i> .	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/min, Pressure: 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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>Ex-situ</i> .	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: <i>E.coli</i> 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: <i>Ex-situ</i> , 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: <i>Ex-situ</i> , US was used with NaOH 2% w/v at 25°C.	Increased rejection by 6.4%.	Trivial reduction in flux by 0.6%.	[117]

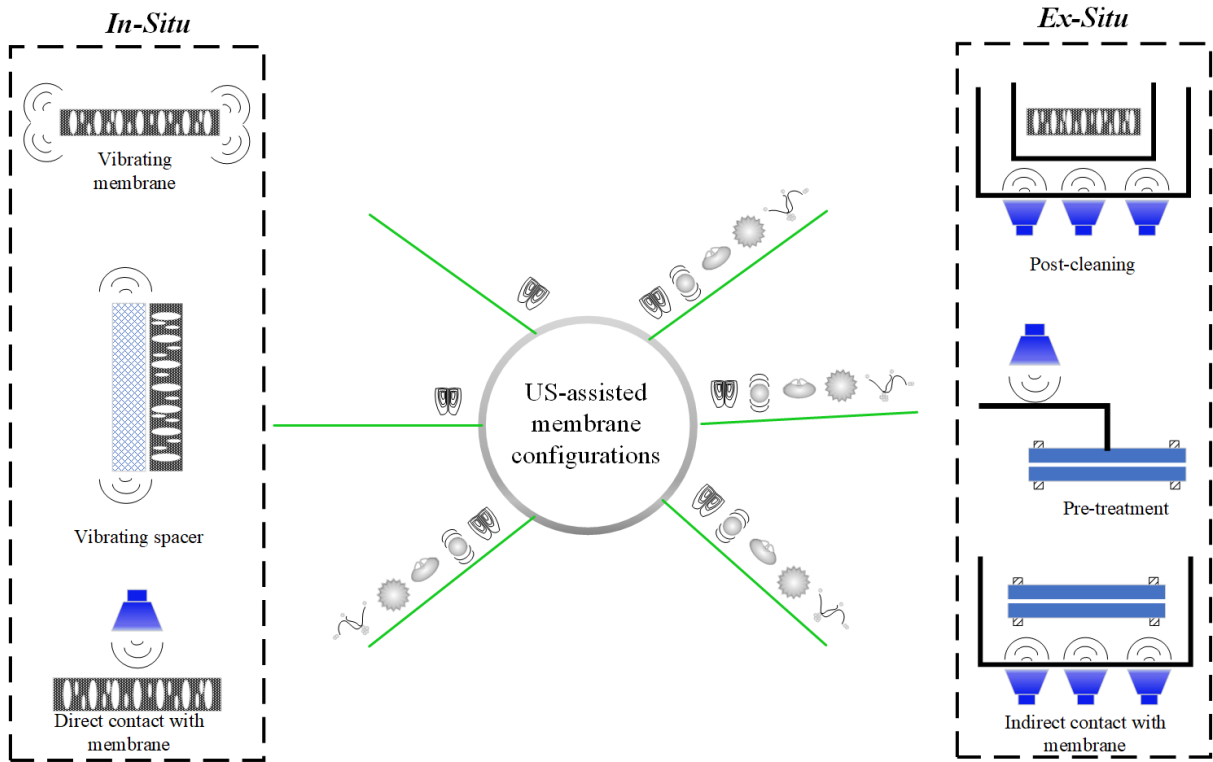


Figure 4: US-assisted membrane configurations with their effective mechanisms for membrane performance improvement.

358

359 3.2. UF-US

360 Ultrafiltration (UF) is a promising separation process that covers a wide range of industrial
 361 processes, including concentration, fractionation, water treatment and macromolecular species
 362 elimination or macro-solutes elimination from various industrial effluents [118, 119].

363 Membrane fouling is a serious issue with UF membranes, causing a decrease in permeate flux
 364 and increasing process and maintenance costs. Different chemical and physical methods have
 365 been used for UF membrane cleaning [100, 120]. Physical cleaning might change the

366 membrane's hydrodynamics, while chemical cleaning can be expensive [121]. US technology
 367 is considered for cleaning the UF membrane using a bath configuration, as illustrated in Figure
 368 5.

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

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

383

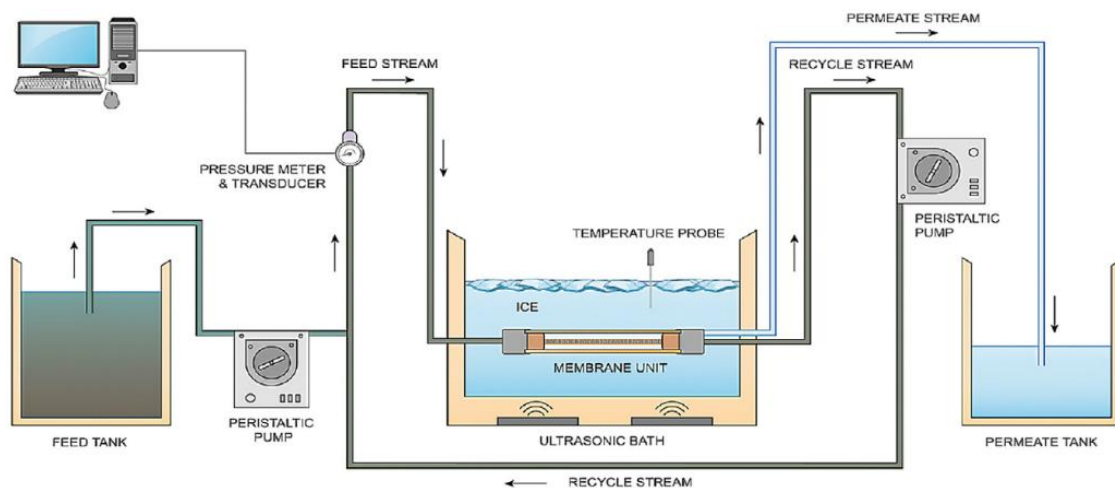


Figure 5: Schematic of UF-US system [83].

384 Even though US technology can remove the fouled layer from the membrane surface and
 385 increase the permeate flux Latt and Kobayashi [92], Matsumoto et al., [97] Muthukumaran et
 386 al., [124], its effect also depends on the feed solution matrix. For example, Chai et al. [104]
 387 used the US for cleaning polyacrylonitrile membrane after UF of dextran solution and found
 388 that the US technology could not remove the fouled layer from the membrane surface. This

389 might be due to membrane vibration caused by the US waves, which increased bulk mass
390 transfer [104]. Yu et al. [125] pointed out that applying the US for 10 mins could separate more
391 organic matter from the membrane surface. The US removed predominantly hydrophilic, high
392 molecular weight organic matter from the UF membrane. Concurrently, the US process had a
393 low effect on the accumulation of smaller molecular weight organic matter. Chen et al. [126]
394 studied the impact of particle characteristics on the ultrasonic control of membrane fouling.
395 The authors found that US cleaning was affected by the particle size and higher permeate flux
396 recovery was observed when feed water contained large particles. Interestingly, the authors
397 reported membrane damage when the US was applied at a short distance from the membrane
398 surface under high pressure.

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

414 The US power is considered an important parameter due to the high effect on fouling removal
415 and water flux recovery [92, 97, 124]. , In general, permeate flux increases linearly with US
416 power up to a certain limit, after which no significant permeate flux improvement is observed.
417 Furthermore, operating at elevated US power could also damage membrane material [109].
418 Researchers also evaluated the coupling of US with backwash techniques. Chai et al. [34]
419 utilized an ultrasonic bath at 45 kHz and 2.73 W/cm² to clean UF and MF membranes fouled
420 by peptones using a cross-flow filtration cell. The study revealed that cleaning fouled
421 membrane by combining US and backwash was better than the US alone. Furthermore,
422 Secondes et al. [83] and Naddeo et al. [129] reported removing emerging contaminants from
423 wastewater by activated carbon adsorption was about 90%, but decreased over time. However,
424 the removal increased to almost 100% by applying the US, especially with a low frequency of
425 35 kHz.

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

433

434 **3.3. NF-US**

435 NF membrane fouling is a critical issue, as it is responsible for the deterioration of the
436 membrane performance [130, 131]. It was mentioned that the cost of fouling control is almost
437 30% of the total operating cost [132]. US technology was proposed by many researchers as an
438 alternative cleaning technique to control NF membrane fouling [79, 112, 113]. US frequency

439 is one of the main parameters that significantly affect the cleaning process. Tejal and Kaushik
440 [111] studied low-frequency US effects with two different modes (continuous and intermittent)
441 to remove the fouling accumulated on the membrane surface. They found that permeate flux
442 increased by 3% - 4% when the US was applied continuously or intermittently for 160 min.
443 Continuous ultrasonic irradiation mode was more effective than the intermittent mode, but the
444 intermittent mode is still a better option when energy efficiency is considered [133, 134].
445 The second main parameter is the US's power, which could have a massive impact on the
446 treatment process. Some researchers investigate the effect of high power on cleaning efficiency
447 and permeate flux enhancement. In a study by Renata et al. [135], high ultrasonic power of 240
448 W was applied to clean the NF membrane used in treating artichoke's solid wastes no
449 significant effect on the fouling layer was observed. Still, the highest chlorogenic acid recovery
450 was achieved when the US power was at 240 W. Thombre et al. [79] used US technology for
451 cleaning fouled NF membranes. An ultrasonic power of 135 W achieved the best cleaning
452 process, while with a higher power of 150 W, pitting and corrosion was detected on the
453 membrane surface. These results agree with a study by Muthukumaran et al. [106], who used
454 300 W of US power. The authors also mentioned that permeate flux recovery of the NF
455 membrane increased by 90% in only 4 min of US.

456 It should be noted that applying a high power US increases the energy required for UF process
457 and the major parts of US waves would be wasted [136]. Many researchers used the US to
458 assist other cleaning techniques such as chemical and physical to avoid more energy waste. Liu
459 et al. [113] used the US-assisted chemical cleaning at a frequency of 25 kHz and a power of 70
460 W. They found that US technology is a more effective way to improve chemical cleaning. They
461 reported that the recovery rate reached up to 95.6% by applying US-assisted chemical cleaning.
462 Also, Jian et al. [112] used US-assisted chemical cleaning for fouling removal caused by
463 inorganic scales in arsenic-rich brackish water. Despite the increase in permeate flux, which

464 reached 80% when the membrane was cleaned only by chemicals, the NF membrane water flux
465 reached 99.99% when the US power intensity of 1 W/cm^2 was applied.

466 **3.4. RO-US**

467 Reverse osmosis (RO) is a well-established conventional desalination and water purification
468 technology that uses a semipermeable membrane. RO technology is successfully used for the
469 treatment of seawater and groundwater. Despite the advantages of RO technology in water
470 purification [87, 88], it presents some disadvantages, such as sensitivity to pH and ionic
471 strength, high energy consumption, and requirements for pre-treatment and membrane fouling
472 [137]. The RO process requires high pressure (usually 0.2-1.7 MPa) for fresh and brackish
473 water and 4-8.2 MPa for seawater treatment [138, 139]. The high-pressure demand translates
474 into a higher pressure drop inside the module and reduced membrane permeability, which
475 increases the pumping cost and alters rejection [140]. By applying high pressure, the
476 membranes also become susceptible to fouling which clogs their pores [141] and reduces the
477 permeate flux.

478 Researchers tested different chemical, physical, and US technology techniques to improve the
479 permeate flux and reduce membrane fouling [39, 91, 109]; the latter technique is the subject of
480 interest. Most US applications for alleviating RO fouling and improving permeate flux have
481 been implemented in ultrasonic bath configuration on a lab-scale, as shown in Figure 5. Rarely,
482 the US could cause damage to the RO membrane during the treatment process, which required
483 more attention when US parameters were selected. Yong et al. [115] compared US application
484 with acid and alkali agents for RO membrane cleaning while treating pharmaceuticals
485 wastewater loaded by organic compounds. It was found that 50 kHz frequency and 0.64 Wcm^{-2}
486 power were the most effective US cleaning parameters. However, membrane damage occurred
487 when the US power was 0.636 Wcm^{-2} and applied for 60 min. The study highlighted two
488 observations from the membrane damage test, including lengthy treatment time and dry storage

489 of membrane make it more susceptible to structural damage. Feng et al. [114] tested a combined
490 RO-US system to reduce the fouling layer on the membrane surface and increase the permeate
491 flux without causing any damage to the membrane. Permeate flux improvement of the RO
492 process was attributed to the US cleaning. However, the increase in permeate flux of the RO
493 process by the US technology was not high enough. The authors explained this by the
494 deposition of CaSO_4 due to hot spots created by US cavitation leading to a reduction in the
495 CaSO_4 solubility [142]. A slight improvement is likely due to the advanced crystallisation stage
496 such that complete dislodgement was not possible, especially that US effects are contactless
497 with the membrane.

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

513 where some cells were killed, while others survived but remained injured, which in turn caused
514 starvation.

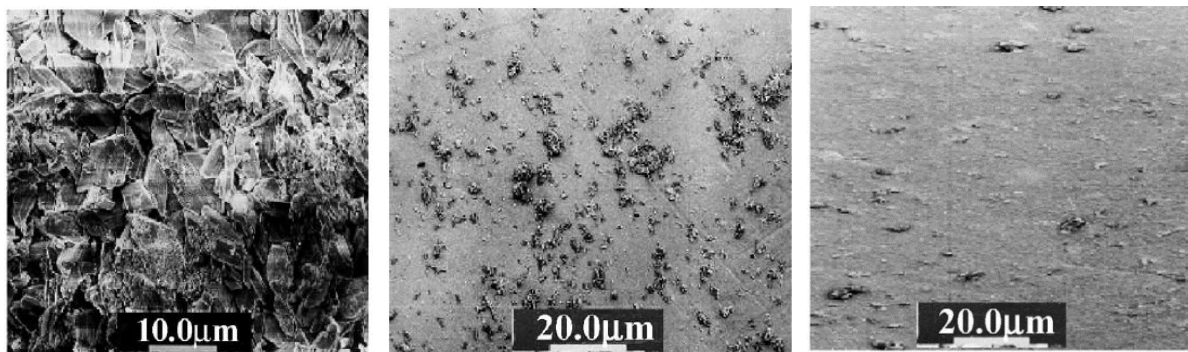


Figure 6: SEM images of a membrane surface: (a) after 7 hrs of operation; (b) after 3 hrs of cleaning with water; (c) after 0.5 h US treatment with dilute HCl [80].

515

516 **4. US application with membrane bioreactor systems (MBR)**

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

529 In a study carried out by Xu et al. [153], an integrated AnMBR-US system (Figure 7) was
530 applied for the digestion of sludge under high volatile solids (VS) at a loading rate of (3.7
531 gVS/L d) for 54 days. Although the US process could successfully control the cake layer

532 formation on the membrane surface, it had only a slight effect on the gel layer removal formed
 533 by the adsorption of proteins and humic compounds. Sui et al. [154] tested intermittent US
 534 applications with an MBR system to reduce fouling development on the membrane surface.
 535 This study found that increasing the sludge concentration in the reactor increased the need for
 536 longer ultrasonic irradiation. The study pointed out that the US irradiation had a small negative
 537 impact on the anaerobic bacteria activity; however, this did not affect chemical oxygen demand
 538 (COD) removal. Ruiz et al. [155] studied the effect of ultrasonic frequencies in a range of 20
 539 kHz - 40 kHz on membrane integrity, process performance and effluent quality using four
 540 different UF modules. The fouled membrane received two different cleanings: the US for 3 s
 541 every 3 min with the power of 150 W and various frequencies or backwash for 1 min with 5 s
 542 of aeration. The highest cleaning effect was observed with a frequency of 20 kHz with no sign
 543 of damage to the membrane surface.

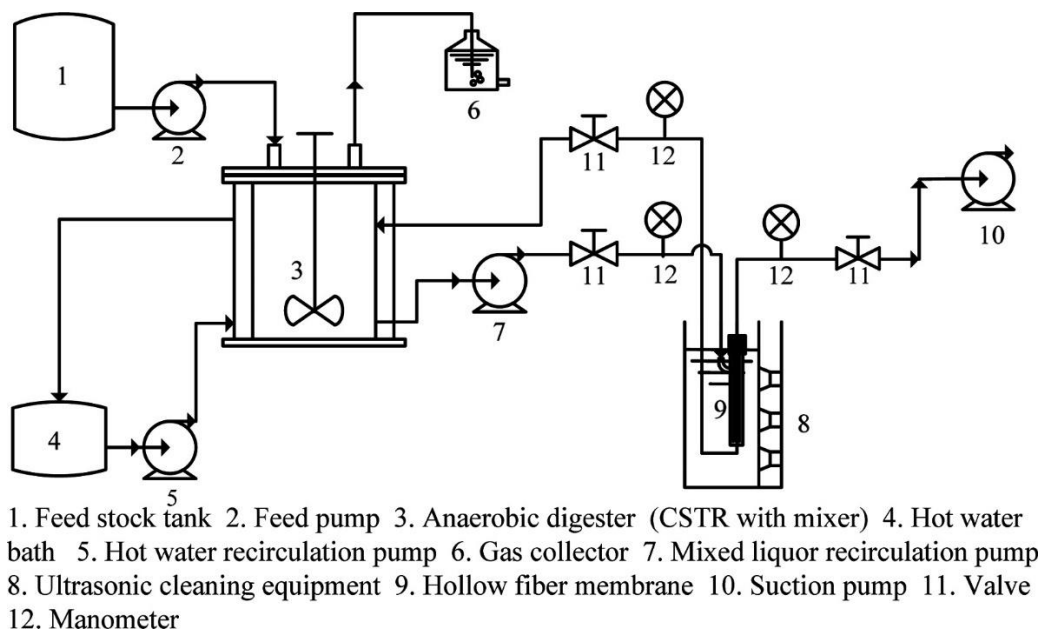


Figure 7: The flow diagram of the US-AnMBR [153].

544
 545 Another study by Ruiz et al. [156] found that even though the high US power of 300 W and
 546 400 W increased the turbidity of the effluent from 2 NTU to 20 NTU, other parameters like
 547 viscosity, colour, effluent COD and total suspended solids concentration did not change. This

548 could be due to the deflocculation of the sludge under ultrasonic irradiation [157]. Li et al.
549 [158] used US for cleaning the fouled membrane in a submerged membrane bioreactor under
550 different US frequencies of 25 kHz - 90 kHz, and applied power of 200 W – 300 W. The results
551 of this study showed that the US could reduce the quantity of the sludge produced with the
552 MBR system. The higher the transmembrane pressure, the higher the fouling layer on the
553 membrane surface. Hence, fouling removal by the US would reduce the transmembrane
554 pressure in the filtration process.

555 Sui et al. [159] applied US technology to clean the membrane used in an anaerobic membrane
556 bioreactor. It was noted that the US effect on fouling diminished when the crossflow velocity
557 was greater than 1 m/s as the fouled membrane could be cleaned by hydrodynamic forces. On
558 the other hand, the total filtration resistance was drastically improved when applying the US
559 with a crossflow velocity of less than 1 m/s, and the membrane fouling rate was $8.33 \times 10^6 \text{ m}^{-1}$
560 s^{-1} and $3 \times 10^7 \text{ m}^{-1} \text{ s}^{-1}$. The study reported a stable low total filtration resistance for one week
561 with ultrasonic power of 150 W. Abdurahman and Azhari [160] tested US-AnMBRs to treat
562 oil mill effluent with multi-frequency ultrasonic transducers. The study found that this system
563 could achieve COD removal of 98.7% with a hydraulic retention time of 4 days and maximum
564 methane production of 0.47 L/g COD·day. Similar findings were also reported by Shafie et al.
565 [161]. The authors indicated that the violent mechanical effects of the US are responsible for
566 membrane damage and the interaction of the strong oxidants produced due to bubbles collapse
567 with membrane materials. Liu et al. [162] investigated the effect of the online US-MBR system
568 on removing organic pollutants from the membrane surface. The study found that the activity
569 of the biological process was increased when the US was applied with the power of 10 W. This
570 increment in the activity of biological was due to the turbulences accompanying propagation
571 of US waves and the cavitation effects, which can increase the mass transfer by moving the
572 particles in a fluid and increase the production of the extracellular enzyme.

573 Yoon et al. [163] studied the effect of *ex-situ* US on removing sludge production in the MBR
574 system using submerged hollow fibre membrane with the power of 600 W and a frequency of
575 20 kHz. This study found that the mixed liquor suspended solids (MLSS) were kept constant
576 in the range of 7000 mg/L - 8000 mg/L when the US was applied, while the range of the MLSS
577 increased from 7000 mg/L to 13700 mg/L without US application. This is attributed to the
578 virtue of the US in preventing excess sludge production. Joshi and Parker [164] used the US as
579 a pretreatment with hydrogen peroxide to treat waste stream before digesting in a submerged
580 anaerobic membrane bioreactor. The study showed that COD solubilization increased by about
581 40% when the hydrogen peroxide dose was 50 g/kg TS and sonication operated for 60 min.
582 Wu et al. [165] investigated the US irradiation effect on the liquor properties of activated sludge
583 using a power range of 40 W - 300 W, volatile suspended solids concentration of 6 g/L, and a
584 concentration range of mixed liquor suspended solids of the activated sludge of 10 g/L -12 g/L.
585 The results showed that US treatment with an intensity of up to 2 kJ/mL could increase the
586 width of particle size distribution and the biodegradability of the activated sludge. Pardo et al.
587 [166] used US combined with ozonation (O₃-US) to treat wastewater prior to the submerged
588 MBR. The study found that the decomposition of the organic compounds was increased due to
589 the O₃-US treatment, resulting in a decrease in the microorganism's growth. Hence, the
590 concentration of extracellular polymeric substances was reduced by around 50%. Overall, US
591 technology could improve MBR performance by increasing nutrients degradability. Improving
592 mass transfer across the membrane and reducing membrane fouling. However, the positive
593 effects can only be achieved if the proper US parameters are selected.

594 **5. Integrating US with emerging membrane technologies**

595 **5.1. Ultrasound- Forward Osmosis (US-FO)**

596 The FO process uses natural osmotic pressure difference of feed solutions of different
597 concentrations to transfer water through a semipermeable membrane from the higher solute

598 concentration side to the lower solute concentration side. FO is far more energy-efficient and
599 lower membrane fouling than the RO process [167]. However, the FO process also suffers from
600 membrane fouling, especially when treating a low-quality feed solution. Integrating US
601 technology with FO could be an attractive solution to this problem. Heikkinen et al. [168] tested
602 the US-assisted FO process and found that permeate flux of FO system was increased after
603 applying US technology. The US irradiation improves the FO process performance by reducing
604 both internal (ICP) and external (ECP) concentration polarization effects (Table 2).

605 Choi et al. [169] found that US combined with FO decreased the concentration polarization
606 occurrence and membrane fouling and improved membrane efficiency. However, the US effect
607 on the membrane's durability is not obvious, and the outcome of previous studies on this issue
608 has been contradictory. Chanukya and Rastogi [170] investigated the US effect on FO
609 membrane concentration polarization while treating fruit juice and natural colorant. The
610 authors found that US applications can increase permeate flux due to ECP mitigation on the
611 feed side and ICP in the support layer. Despite the permeate flux of the FO membrane enhanced
612 by US technology when the concentration of sucrose was up to 5%, the authors found that US
613 was not able to mitigate the ECP and prevent fouling layer formation when pectin was present
614 in the feed solution which resulted in a reduction in the permeate flux. Chio et al. [171] also
615 studied the effect of US on ICP during the FO process with flat sheet membrane by utilizing
616 different US frequencies of 25 kHz, 45 kHz and 72 kHz and power of 10 W 70 W. The authors
617 found that US technology could significantly mitigate the ICP by improving the diffusion rate
618 of a draw solution. The authors also reported that membrane damage was observed at the US
619 frequency of 25 kHz and 50 W of the applied power, leading to a 3000% increase in permeate
620 flux. This damage is likely caused by changes in membrane properties which were significantly
621 affected by the US. The low-frequency US irradiation was proposed by Wang et al.[172] to
622 improve the permeate flux of the FO process with TFC PES-based polymeric membranes. The

623 authors found that the US significantly improved the FO process permeate flux via mitigating
624 ECP effect. Lee et al. [173] studied the effect of US cleaning on the FO membrane fouled by
625 activated sludge was investigated. An effective fouling removal was also observed when the
626 US was combined with flushing. Thus, 40 % of permeate flux was recovered when the US was
627 used for 60 s, while with flushing only, the permeate flux of the FO was recovered only by
628 29% [153]. Nguyen et al. [174] used the US to control the fouling on the FO membrane. The
629 study found that the sludge concentration reached 20,400 mg/L and 28,400 mg/L from the
630 initial sludge concentration of 3,000 mg/L and 8,000 mg/L with 40 kHz after 22 hours.
631 However, from an energy requirements perspective, this method is not an energy-efficient
632 technique.

633 **5.2. US-MD**

634 *5.3. Ultrasound- Air Gap Membrane Distillation (US-AGMD)*

635 AGMD has many advantages, including cost efficiency [1], lower chemical demand [175], no
636 feedwater pretreatment [40] and low membrane damage [176, 177]. Moreover, AGMD is
637 capable of separating all non-volatile matter under moderate operating conditions that ensure
638 system reliability and durability with no requirement for additional complex condensers [178,
639 179], which are needed in vacuum membrane distillation (VMD) and sweeping gas membrane
640 distillation (SGMD) [180]. Although AGMD has witnessed many physical developments, the
641 permeate flux of the AGMD is still low compared to some other membrane separation
642 processes. Another limitation of the AGMD system is that the fouling layer is quickly built on
643 the membrane surface due to the relatively big pore size of the membrane (0.2 μm – 1.0 μm)
644 [181]. This layer can prevent water vapour from crossing the membrane, resulting in low
645 permeate flux. The AGMD process can be integrated with US technology to overcome fouling
646 and improve water flux, as seen in Figure 8 [35, 59, 180]. Technically, the US energy could be
647 converted to heat which can reduce the heat transfer loss across the membrane and therefore

648 increase the permeate flux of the AGMD). Table 2 shows a summary of studies which utilized
 649 US for mitigating fouling problem in MD processes, including AGMD. Although the US has
 650 a benign environmental effect, it can increase the water treatment energy consumption. Also,
 651 applying high power of US, waves may damage the membrane surface [39].

652

653 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]

654

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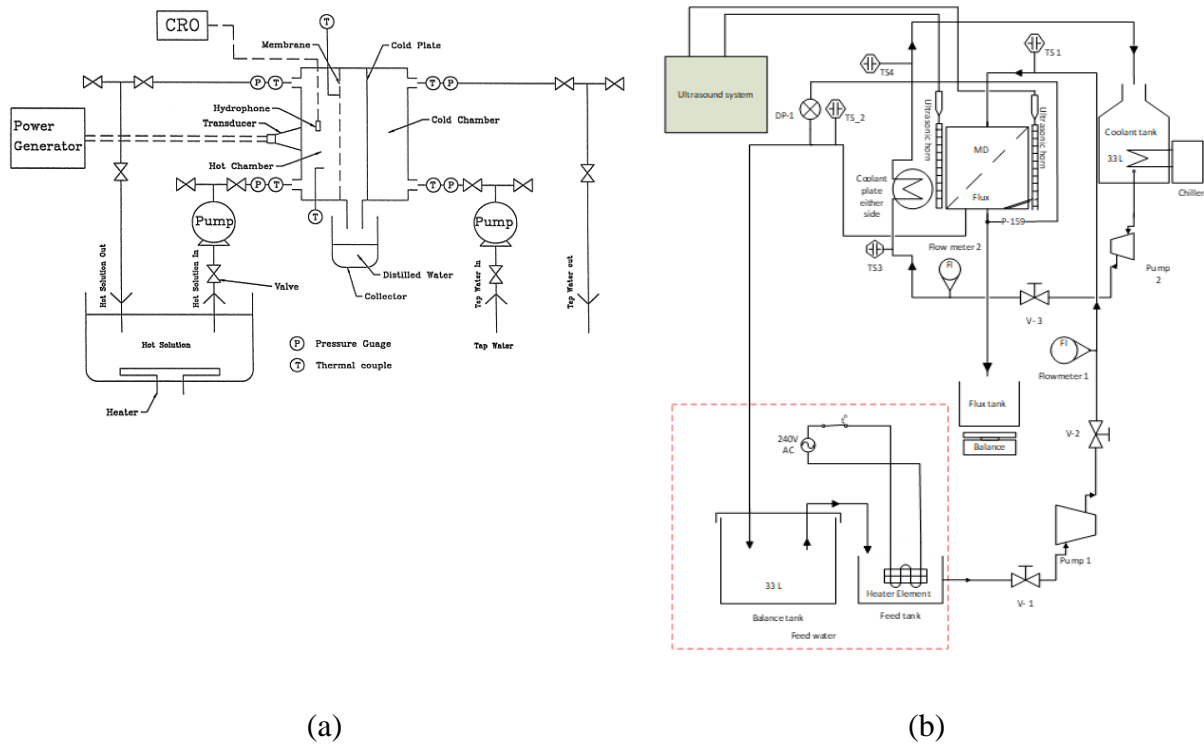


Figure 8: Shows the AGMD-US system, (a) *ex-situ* [180] and (b) *in-situ* [39].

656

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

670 5.4. US-DCMD

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

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

696 DCMD-US process was insignificant and comprised ~97% of the virgin membrane permeate
 697 flux. The SEM images (Figures 10a-c) confirmed an amorphous silica-scaling layer formed on
 698 the membrane surface after the stand-alone DCMD process. In comparison, SEM images of
 699 the PTFE membrane surfaces used in DCMD-US experiments had no silica layer on the
 700 membrane surface. Figures 10d-e demonstrate the effectiveness of US technology to remove
 701 fouling materials from the membrane surface even when Ca^{2+} ions exacerbated membrane
 702 fouling. Furthermore, permeate flux of the combined US-DCMD system was 2 kg/m^2 , 34%
 703 higher than the permeate flux observed with stand-alone DCMD. Another study by Hou et al.
 704 [183] utilized PTFE membrane to treat synthetic feed, which incorporated 50 mg/L of humic
 705 acids (HA) and CaCl_2 in a range of 2 mM – 20 mM. The authors found that US irradiation
 706 enhanced permeate flux by more than 30% without affecting HA rejection. In addition,
 707 permeate flux enhancement increased with a concentration factor
 708

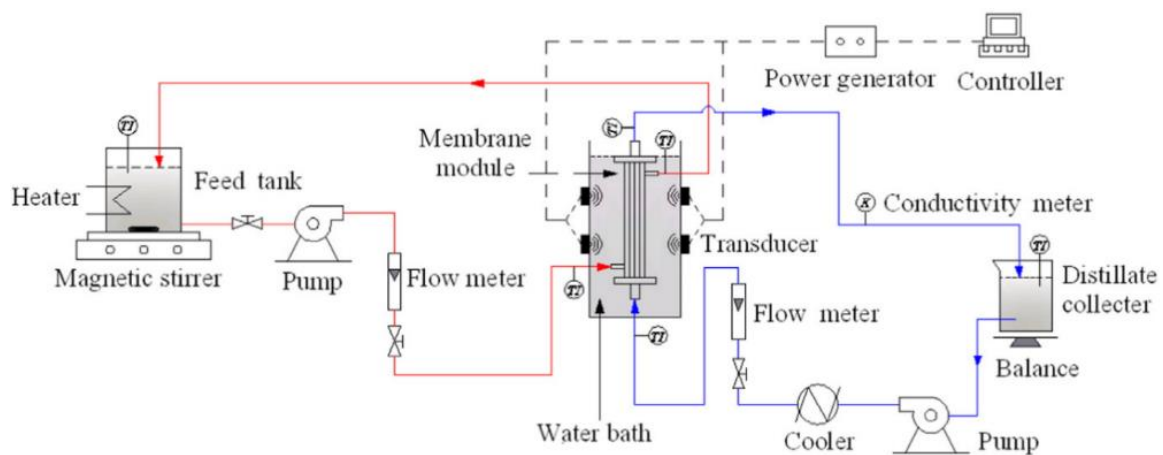


Figure9: DCMD integrated with US [38].

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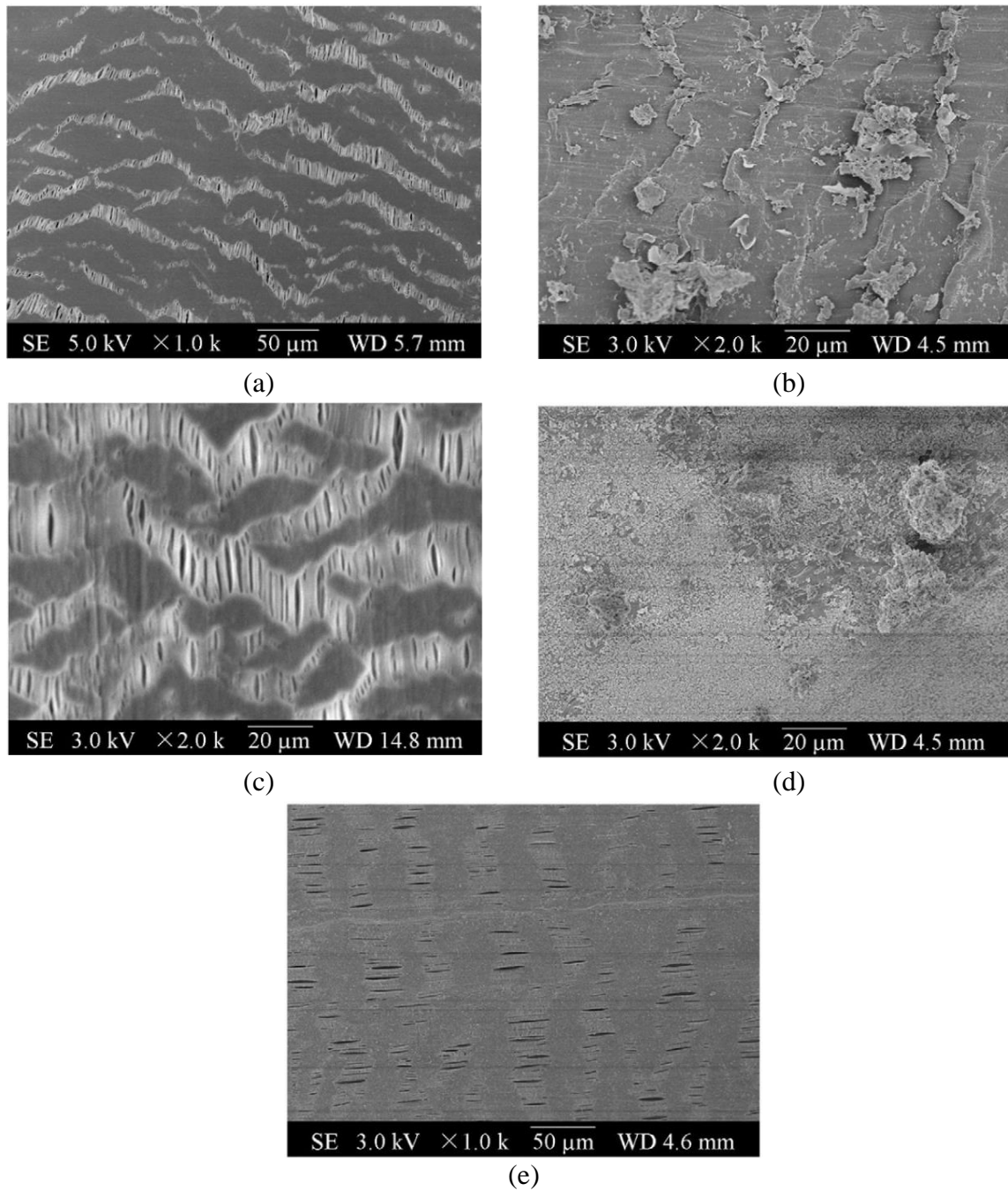


Figure 10: SEM images of the PTFE membrane, (a) virgin PTFE membrane, (b) PTFE membrane after silica fouling, (c) PTFE membrane after silica solution concentration with US irradiation, (d) after silica solution concentration experiment running 30 min in the presence of Ca^{2+} ions, (e) after silica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

710

711 **6. Overview of ultrasound effect on membrane properties**

712

713 The discussion regarding the change in membrane properties upon exposure to ultrasound

714 effect has been mainly focused on membrane physical structure as discussed in previous

715 sections. It is important to ponder about this point beyond optical or microscopic examination.

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

730 The impact of the physical effects of ultrasound on membrane properties have adequately been
731 studied, however the impact of the chemical effects is rarely discussed in the literature. It is
732 important to remember here that high frequency ultrasound produces more chemical effects
733 (i.e. generation of free radicals) compared to low frequency. Hence, high frequency ultrasound
734 is expected to cause change in membrane surface chemistry. It was reported that the production
735 of radicals such as $\text{OH}\cdot$ $\text{O}\cdot$ and oxidant agents such as H_2O_2 may cause chemical bonds scission
736 of membrane materials [51]. The quantity and the aggressiveness of produced radicals and
737 oxidants depends on many factors such as power intensity and presence or absence of radicals
738 scavenging and promoting agents. For example, the presence of Fe^{+2} facilitated the degradation
739 of ionomer membrane, Nafion® 117 through the hydroperoxyl radical attack on main and side
740 chain of the polymer [188]. The produced free radicals with ultrasound can also interact with

741 the membrane surface altering its properties. Free radicals can interact with the dissolved
742 oxygen and the carbonous structure of organic membrane producing carboxyl and carbonyl
743 groups that makes the membrane more hydrophilic [189]. In order to accurately capture the
744 changes that occur in membrane properties, long term tests and advanced analytical chemical
745 examinations are recommended to be applied as such changes can be subtle and hard to detect
746 in short-term tests and crude analyses.

747 **7. Membrane-assisted ultrasound technology: recommendations for** 748 **future research directions** 749

750 There is a plethora of successful US applications to improve membrane separation
751 technologies. However, studies in this field seem to linger at the lab testing phase. This is likely
752 due to the limited knowledge available on the intimately linked interactions between the US
753 effects and the operation parameters of different membrane processes. The majority of the
754 research in this field utilizes off-shelf US systems not designed for this particular purpose.
755 Failing to tailor US reactor design and operating conditions to suit process requirements may
756 mislead the evaluation of its true value and capacity. Since some aspects of the US-assisted
757 membrane technology were investigated more extensively than others, we believe it is worth
758 conducting a stocktaking exercise of the research maturity in these aspects as presented in
759 Table 4. The content of Table 4 was formulated based on the up-to-date literature survey carried
760 out in this study. The level of research maturity of each process aspect was categorized based
761 on the number of studies available into comprehensive, reasonable and insufficient. It appears
762 that among all the identified research aspects, only US power and the use of piezoelectric
763 transducers in *ex-situ* configuration were studied in an adequate depth. Other aspects such as
764 US frequency, type of feed water and operation mode were explored only in a few studies.
765 Furthermore, most of these studies were focused on treating synthetic feed waters by applying
766 continuous US with frequencies below 100 kHz. Unlike low frequency, high frequency is

767 expected to produce less vigorous US effects, reducing the risk of membrane damage. High
768 frequency may bring about chemical changes in the membrane surface if it is applied at power
769 level higher than the cavitation threshold. At low power level, high frequency is expected to
770 produce larger number of vibration cycles compared to low frequency, and this might be
771 beneficial especially for the dislodgment of foulants from membrane surface. As such, there is
772 a need to test US-assisted membrane technology with different natural waters (e.g., seawater,
773 groundwater, industrial and municipal wastewaters, etc.) in a high-frequency range of 200 kHz
774 - 1 MHz in different operational modes. Water samples with high ionic strength such as
775 seawater and groundwater were found to enhance ultrasonic effects [190]. They also contain
776 ions such as chloride that could scavenge hydroxyl radicals reducing its possible negative effect
777 on membrane structure.

778 The use of magneto-strictive transducers, various waveforms, *in-situ* configuration, large and
779 long-term trials, as well as cost analyses are also hardly investigated. As explained in Section
780 2, magneto-strictive transducers are more robust and suitable for industrial applications than
781 piezoelectric transducers. Therefore, assessing US application for improving membrane
782 separation performance could also be explored by utilizing this type of transducers. Most of
783 the available US equipment is driven by a sinusoidal wave, while there is a wide range of other
784 forms such as square, triangle and sawtooth that may offer a better choice for US-assisted
785 membrane technology.

786 Incorporating US technology with membrane filtration processes may increase the capital and
787 operational cost. To justify US applications in membrane filtration processes, there should be
788 a remarkable improvement in the filtration processes, especially in treating low-quality
789 wastewaters laden with contaminants that cause irreversible membrane fouling. Such
790 wastewaters require intensive pre-treatment and frequent membrane cleaning, leading to
791 significant operating costs increase. US application could also be justified to reduce the process

792 downtime by providing a constant filtration process without interruptions. The cost of US
793 application in water treatment would be justifiable in membrane processes for resources
794 recovery to achieve an elevated recovery rate without membrane fouling or damage.

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

809 Regarding the scale-up opportunities for US-assisted membrane technology, the authors can
810 offer adjudication informed by literature knowledge and experience in the subject matter. The
811 opportunities for scaling up US-assisted pressure driven membranes lie in pre-treatment and
812 post-cleaning applications. Other configurations of the process (see Figure 4) require high
813 energy to overcome the pressurised environment in the first instance prior to producing any
814 useful effects. This portion of energy can be considered waste as it does not return any benefits
815 to the overall process. The scale up opportunities for other membrane types (e.g. thermally and
816 osmotically driven) are wider. Technically, all systems configurations shown in Figure 4 can

817 be applied in thermally and osmotically driven membranes as sound waves do not need high
 818 energy to breakdown the cohesive forces of the medium. Some of these membrane processes
 819 are in the development phase in the present time and this offers a great opportunity for
 820 considering the integration of US at early stage of system design.

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

825

826 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	<i>In situ</i>	✓		
	<i>Ex situ</i>			✓
Large scale trials		✓		
Long term trials		✓		
Proper analysis for capital and operational cost		✓		

827

828 8. Conclusions

829 US coupling with membrane separation technologies has been proposed to reduce fouling and
 830 permeate flux increase. The present study reviewed the theoretical and experimental aspects of
 831 US technology and links between the US design and membrane system operating parameters
 832 and its impact on fouling mitigation and mass and heat transfer enhancements. The efficient

833 application of the US requires prior knowledge of the US design and application method and a
834 deep understanding of the nature of the treated solution and its conditions. Overall, US-assisted
835 membrane processes can maintain the filtration processes without interruption and improve the
836 permeate flux substantially. However, technology is still under investigation, and it is energy-
837 intensive with the potential of negatively affecting membrane integrity if the operating
838 conditions are not properly selected.

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

850

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