A Critical Review on Ultrasonic-Assisted Fouling Control and Cleaning of Fouled Membranes

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Abstract

Fouling is one of the most challenging problems impacting the performance of membrane-based separation technology. In recent years, ultrasounds have been widely applied as an unconventional method to control membrane fouling, as well as to enhance membrane cleaning. The aim of the present work is to review the current literature and the recent developments related to the use of ultrasounds as an innovative and alternative approach to improve the fouling behavior of membrane separation processes. The theory underlying ultrasonic-assisted phenomena is reviewed, together with operational factors that influence the effectiveness of the ultrasound treatment, such as frequency, power intensity, pressure, temperature, pH, and operation mode. Ultrasound irradiation effectively aids the cleaning of contaminated surfaces and enhances the permeate flux, owing to cavitation phenomena and powerful convective currents, associated with secondary phenomena, such as microstreamers, shock waves, and heating. However, the lifetime of the membranes should be carefully evaluated when applying ultrasonication as a technique of cleaning or controlling membrane fouling. Indeed, the integrity of membranes after sonication and the control of erosion produced by high ultrasonic intensities are key issues hindering the scale-up of this approach in the membrane industry. This reviews highlights the topics requiring more investigations, specifically to evaluate the economic aspects of ultrasonic assisted fouling control and cleaning in membrane processes.

Keywords: Ultrasonic; Membrane filtration; Cavitation; Fouling; Cleaning

Contents

1. Introduction

Membrane filtration is widely applied in different fields, such as water and wastewater treatment [1-6], dairy [7] and food processing [8, 9], chemical [10, 11], biotechnological [12] and pharmaceutical [13] industries, and it is rapidly being deployed in an extensive range of other fields [14-19]. The feasibility, versatility, substantial effectiveness, and lower construction costs of membrane filtration in comparison with other separation techniques are major factors favoring the expansion of this technology [20-22].

One of the significant impediments to the efficient application of membrane processes is the decline in the performance caused by membrane fouling [23-26]. Fouling is due to the reversible and irreversible accumulation of contaminants, such as particles, colloids, macromolecules, microorganisms, and salt crystals on the surface of a membrane or within its structure producing dense cake or gel layers, and in many cases causing pore clogging [27-31]. In addition, accumulation of rejected solutes inside a thin boundary layer adjacent to the membrane results in the concentration polarization, which contributes to increased deposition of foulants onto the membrane [32, 33]. These phenomena are usually associated with a reduction in permeation and negative changes in the membrane selectivity and hence, greater operating costs [34, 35]. Feed properties (contaminant concentration and characteristics, feed ionic strength and pH), membrane surface features (charge, hydrophobicity, roughness, pore size), and operation conditions (hydrodynamics, applied pressure) are main factors influencing fouling [36, 37]. Consequently, the negative effects of membrane fouling can be reduced by appropriately pre-treating the feed stream, by applying methods for cleaning membranes physically and/or chemically, by optimizing the operating and process conditions, and of course by choosing suitable membrane materials and module layouts [38, 39].

Substantial research has been carried out to tackle membrane fouling and much of these studies have focused on membrane cleaning procedures. The most common cleaning techniques include mechanical, hydraulic, chemical, and electrical methods [40-42]. Each of these methods has benefits and limitations, and should be applied under specifically suitable circumstances [43]. Backwashing of a fouled membrane has important limitations for practical application, due to non-recoverable flux reduction between backwashes and to the associated interruptions of membrane operation [40, 44]. Chemical cleaning is considered as the most effective approach to recover membrane permeability and remove irreversible fouling [45]. However, it is a time-consuming process lowering the production time [46] and often requiring harsh chemical conditions (e.g., very high or low pH) causing secondary pollution and producing a deterioration of membrane materials at the cost of a lifetime reduction [36]. Various chemical cleaning agents are used to remove foulants from the membrane surface and to recover the membrane productivity [47-49]. Caustic soda (sodium hydroxide, NaOH) solutions increase the solubility of solutes by hydrolysis and solubilization. Oxidants, such as hydrogen peroxide (H_2O_2) and chlorine from sodium hypochlorite (NaOCl), oxidize typical functional groups found in organic macromolecules to carboxyl, ketonic, and aldehyde groups, which increase their hydrophilicity and facilitate their degradation and detachment from the membrane surface [48]. Hydrochloric acid (HCl), sulphuric acid (H_2SO_4), ethylenediamine tetraacetic acid (EDTA), and other acids are other effective cleaning agents that promote the solubilization and provide chelation capacity for scaling compounds and metals, such as calcium and barium[49, 50].

In addition to these methods, membrane filtration of industrial wastewater in the presence of electric field is used as an electrically-based cleaning technique [51, 52]. This method is affected by the complexity of the feed streams in terms of ionic constituents and is associated with high energy needs [53]. While the techniques mentioned so far are currently being improved, alternative and innovative cleaning procedures are being developed. Recently, the application of ultrasounds has been introduced and proven as a potential approach to achieve membrane fouling control [43, 54-57]. Ultrasonic cavitation is already widely applied to assist and enhance reactions (especially endothermic reactions) and chemical processes, for example oxidative desulfurization [58], extraction [59, 60], leaching [61, 62], and drying [63, 64]. The use of ultrasounds as a cleaning technique is in the early stages of development but it is increasingly used by the industry and in membrane filtration.

Ultrasonic techniques provide an alternative method for cleaning contaminated surfaces and for enhancing the permeate flux in membrane operations. This enhancement is predominantly caused by a phenomenon called cavitation, which induces strong convective currents, recognized as acoustic streaming, associated with phenomena of microstreaming, microstreamers, microjets, shock waves, and heating [46, 65, 66]. Ultrasound waves can propagate through a physical medium (gas, liquid, and solid), in a cycle of alternate and adiabatic compressions and diffractions, thus creating high and low pressure oscillating regions [20, 46, 67]. During the rarefaction phase, the medium is subject to a negative net pressure and ultrasonic cavitation is triggered [43, 44]. Cavitation bubbles collapse in the compression phase, leading to the formation of hot spots (specifically in aqueous solutions) with increased local temperatures and pressures up to 5000 K and 1000 atm, respectively [20, 68]. The amount of released energy from the collapse of the cavities is high enough to overcome the foulant–membrane interactions [69], thus removing portions of the fouling layer from the surface of the membrane, and/or prevent the accumulation of foulants [36, 70].

In 2012, Ahmad and coworkers provided a first review of the mechanism of ultrasonic cavitation and its effect on fouling reduction [71], also discussing the combination of ultrasounds with other antifouling or cleaning techniques. The use of ultrasonic technology for mitigation of membrane fouling was also reviewed by Qasim et al [72], addressing on the fouling mechanisms, ultrasound cavitation phenomena, and the effect of ultrasound parameters. This previous study focused primarily on desalination and water treatment applications. As the field of ultrasound assisted fouling control is growing quickly, some novel and attractive topics have developed in recent years, such as piezoelectric membranes, which require further evaluation within the broader perspective of membrane deployment in industrial applications. In this paper, we provide a systematic review of the current literature and of the recent developments related to the use of ultrasounds as an innovative and alternative method to control and to prevent fouling in membrane operation. A brief summary of the theory of ultrasonic-assisted effects is presented, analyzing the operational factors that influence the effectiveness of the ultrasound treatment and also the effects of ultrasonic irradiation on membranes. Other issues regarding the advantages and disadvantages of the ultrasound-based approach, its research challenges, and future research needs are also addressed. These phenomena and their implications are discussed to aid researchers and engineers in their efforts to apply ultrasonic-assisted operation in membrane processes for a wide range of applications.

2. Ultrasonic cavitation: effects and parameters

Sound waves are generated when a single or a group of displacements happen in a sound-conducting medium [73-75]. The number of pressure phase changes (periods) per unit of time is defined as frequency; when this parameter is higher than 18 KHz, we call this phenomenon ultrasounds.[76]. The propagation of ultrasonic waves in an elastic medium starts a cycle in which compression and rarefaction occur alternately and adiabatically [77]. Compression cycles increase the pressure and decrease the molecular distance, while in rarefaction cycles the molecular distance increases [78]. When the pressure amplitude exceeds the tensile strength, tiny vapor-filled cavities known as cavitation bubbles are generated [79, 80]. Afterward, microbubbles start to grow and compress in rarefaction and compression phases, respectively [27]. At a specific size, the diameter of the bubbles reaches a critical value, and the next compression causes a sudden collapse (Fig. 1). The phenomenon of formation and collapse of bubbles during the propagation of ultrasound waves through an elastic medium is called ultrasonic cavitation [81].

The total emitted irradiation energy from an ultrasonic transducer is described by the following equation discussed by Perusich and Alkire [82]:

$$E_u = \frac{I}{C}\pi a^2(2\xi) \tag{1}$$

Where ξ, a, I, and C are the acoustic particle displacement amplitude (cm), the transducer radius (cm), its intensity (W/cm^2), and the speed of sound (cm/s), respectively. By substituting the ξ and I with their definition, an expanded expression of the total energy emitted from the ultrasonic transducer can be presented as follows:

$$E_u = \frac{2}{C\omega}\left[\frac{\eta\pi^5 a^4 W_c I^4}{4\,\rho\, I_c}\right]^{1/3} \tag{2}$$

Where ω, η, W_c, ρ, and I_c are the angular wave frequency (rad/s), the transducer efficiency, calibration power (W), density (g/cm^3), and calibration intensity (W/cm^2), respectively.

2.1. Energy effects

During cavitation, once a bubble grows rapidly and cannot effectively absorb the energy, the liquid rushes in and the cavity will ultimately implode [83]. The explosion of cavities results in the release of large amounts of energy in a very short time, causing light emission (sonoluminescence) [84]), shock waves, localized high temperatures and pressures up to 5000 K and 1000 atm, respectively, as well as cooling rates as fast as 109 K/s [83, 85]. The released energy is sufficiently high to disintegrate H_2O molecules into basic constituents, i.e., hydrogen (H) and hydroxyl radicals (OH) [44]. Additionally, the occurrence of secondary phenomena, such as dispersion and coagulation, is likely during cavitation [53]. The creation of a bubble will produce two kinds of cavitation: stable and transient cavitation. In stable cavitation, the cavities grow slowly and undergo many acoustical cycles before collapsing [86], while accelerated growth of cavities induces their collapse in fewer cycles, a phenomenon described as transient cavitation.

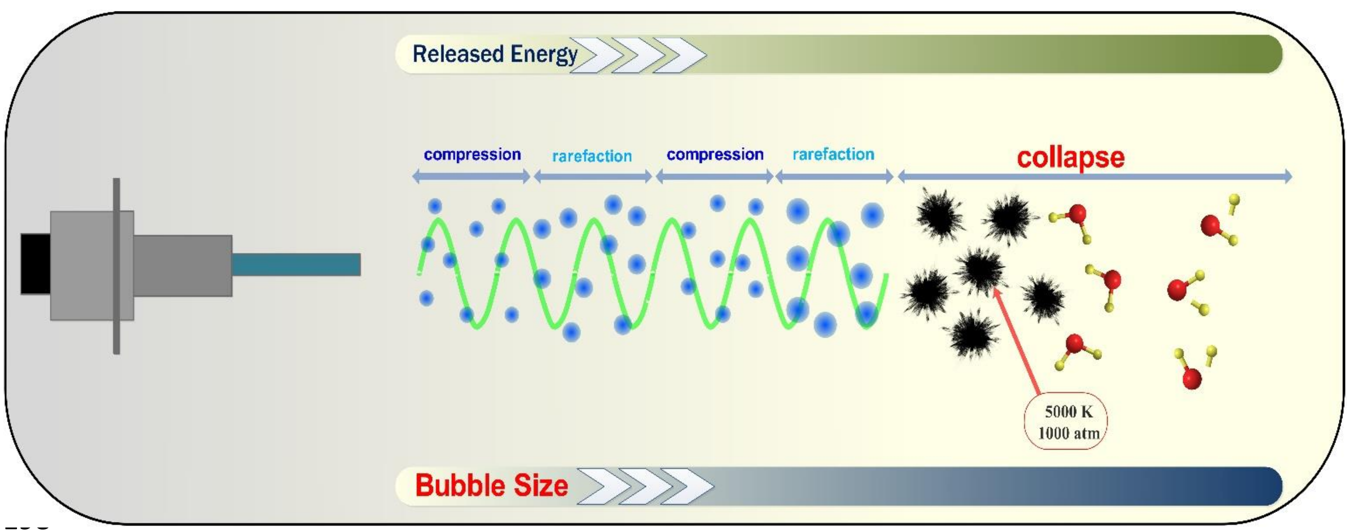

Fig. 1. Ultrasonic cavitation phenomenon: formation, growth, and collapse of bubbles.

2.2. Parameters affecting ultrasonic cavitation

The cavitation phenomenon is affected by many parameters, such as frequency and power intensity, which are considered acoustical parameters. At low ultrasound frequencies, the average size of the cavitation bubbles increases more easily and more powerful cavitational collapses can occur [87]. In contrast, increasing the frequency will weaken the sonication effects by reducing the rarefaction and compression cycles and, correspondingly, lowering the lifetime and the size of the microbubbles before collapse [88]. Increasing the intensity of sound waves will increase the acoustic pressure amplitude, resulting in more powerful cavitational effects [89]. However, there is a critical value of ultrasound intensity above which the bubbles tend to become too large and insufficient time is available for collapse during the compression cycle [87]. Additionally, high ultrasound intensities create a large number of bubbles, resulting in inhibitory effects and an overall decrease of the efficiency of the ultrasound phenomenon [90].

The external pressure is also affecting cavitation. The sonicated medium vapor pressure is decreased by high external pressure, leading to a growth in the ultrasound intensity necessary to initiate cavitation [91]. On the other hand, the number of bubbles generally increases by increasing the vapor pressure (or reducing the external pressure), while the bubbles collapse less violently because more vapor enters into the bubbles [92]. Similarly, the existence of soluble gases accelerates the nucleation of cavities and increases the number of cavitation bubbles. However, the diffusion of gas molecules into bubbles also results in higher gas pressures inside the bubble, leading to a less violent cavitational collapse [78, 93]. Larger effects of local heating are produced by gases with low thermal conductivity during bubble collapse [94]. Table 1 summarizes the influence of the various parameters on acoustic cavitation.

Table 1. Influence of parameters on acoustic cavitation.

Parameter	Influence
Frequency	The lower the ultrasound frequency, the larger will be the average size of the produced cavitation bubbles, associated with more powerful cavitational collapses.
Intensity	Increasing the intensity increases the acoustic amplitude, and the collapse pressure.
Viscosity	Must be minimized for maximum cavitation effect.
Vapor pressure	The cavitation effects greatly improve as the vapor pressure decreases. It is difficult to induce cavitation in a solvent of high vapor pressure.
Presence of gas	Gases make cavitation less powerful by diffusion of gases into the cavitation bubbles.
Surface tension	The higher the surface tension, the more intense will be the cavity collapse.
External pressure	Decreasing the external pressure will decrease the intensity of cavitational collapse, but requires lower intensity to induce cavitation.
Temperature	An increase in temperature induces an increase on the chemical activity

2.3. Hot spot theory

Presenting a theory to rationalize the cavitation phenomenon is complex. Among the existing theories around the sonochemical effects, the hot spot theory is corroborated by numerous experimental data [79]. According to this theory, microbubbles may be considered as microreactors, generating different reactive regions during their collapse [78]. Suslick and coworkers were the first scientists who successfully determined the effective temperature reached when a cavity collapses [95]. In the presented technique (comparative-rate chemical thermometry), two sonochemical reaction sites were detected: (i) the gas phase in the interior of the bubble and (ii) the initially liquid phase. The study showed that the effective temperatures of generated hot spots can exceed 5200 K around the gas phase area and 1900 K around the initially liquid area [95].

In a study by Sharma et al. [79], the results showed that three zones formed as a cavity collapse occurred: the thermolytic center (5000 K, 500 atm), the interfacial region (2000 K, 1 atm), and the bulk region (300 K, 1 atm). The released energy inside the thermolytic center is sufficiently high to achieve the pyrolysis of the liquid molecules. Despite the lower pressure levels, water pyrolysis also occurs in the interfacial region, together with the recombination of OH to form H_2O_2. The temperature and pressure in the bulk region does not change since the process progresses adiabatically. However, reactions between hydrolyzed radicals and bulk molecules occur in this region.

2.4. Physical effects

Ultrasonic irradiation brings about major physical effects [79]. Acoustic streaming generated by ultrasonic irradiations along with secondary phenomenon, such as microstreaming and microjets, causes the turbulent movement of fluid and results in a considerable velocity gradient (in the micro scale) around the generated bubbles [20]. Ultrasound waves can also rapidly melt low melting point metal particles, such as those consisting of Zn and Sn [96, 97]. Ultrasonically-induced movement of fluid enhances the mass transfer in the interface of the solid and gas phases. As a consequence, the associated sonophysical effects can enhance mixing, desorption, extraction, and cleaning processes [79]. Usually, the physical effects of ultrasonic cavitation increase by reducing the frequency of the ultrasounds, and this effect is mainly exploited in cleaning and food processing applications [98]. However, the dependence of physical effects on frequency has not been experimentally clarified and requires more in-depth investigations [79].

3. Ultrasound-assisted membrane treatment

3.1 System configurations

Ultrasonic cavitation also has significant effects on the solid-liquid interface. During the past decades, the effect of ultrasonic cavitation on the efficiency of membrane cleaning has been investigated [56, 90]. Various effects on foulants and membrane surfaces can be expected (Fig. 2), which depend on a variety of parameters, as described in the following sections. The experimental details and key observations of the main investigations are also listed in Table 2.

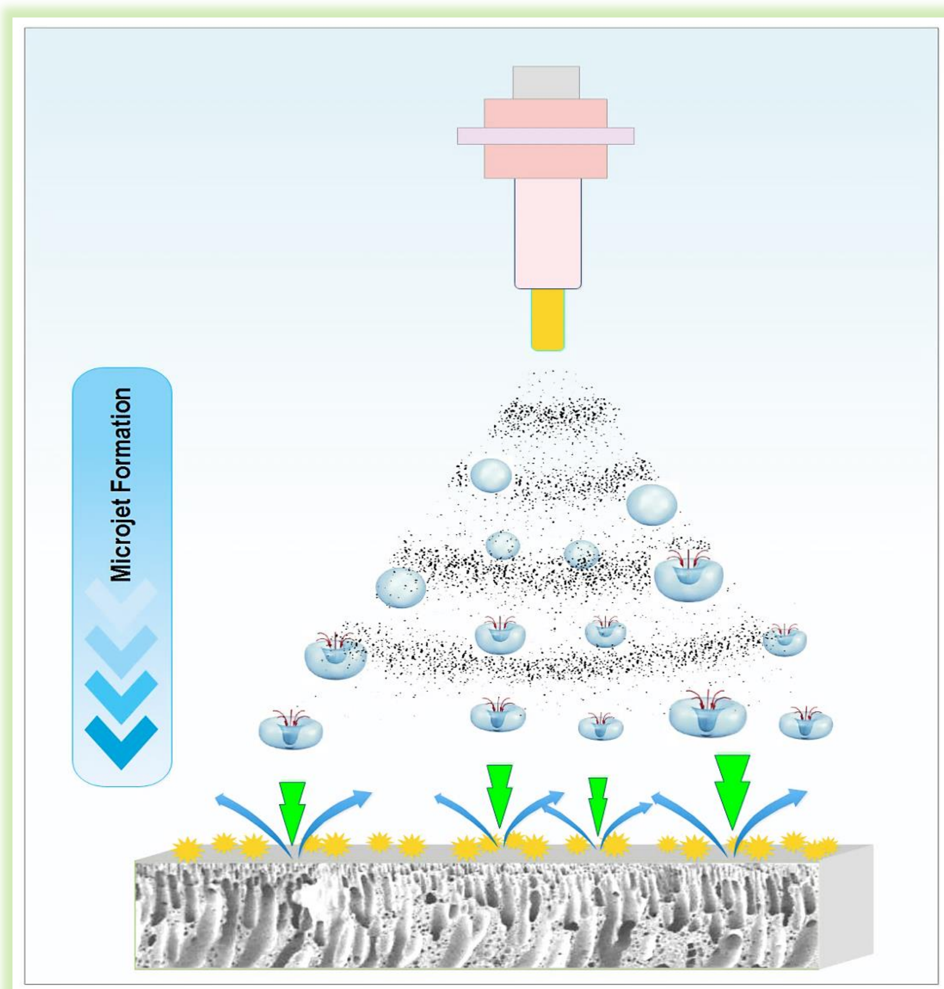

Fig. 2. Formation of microjets on the membrane surface and the corresponding cleaning effects.

Among the various ultrasonic-assisted anti-fouling systems that have been presented in the past three decades, there are two dominant ones: (i) ex-situ ultrasonic transducers, such as ultrasonic baths and (ii) in-situ ultrasonicators, also known as ultrasonic homogenizers. In ex-situ systems, the membrane module is inserted into a sonication bath, where the energy spreads evenly over a large volume via cavitation and collapse of the generated bubbles. The released energy can force surface contaminants to detach from the membrane, so this system is ideal for cleaning processes and has been used widely for membrane cleaning [99, 100]. On the contrary, the in-situ system involves inserting the ultrasonic probe directly into the liquid in contact with the membrane, with a more localized and intense release of energy near the probe. While the ex-situ systems do not require a high amount of electrical power but propagate the ultrasonic energy diffusively (Fig. 3), in-situ systems may be more effective although they are generally more energy-demanding and are not commonly used for cleaning purposes (Fig. 4).

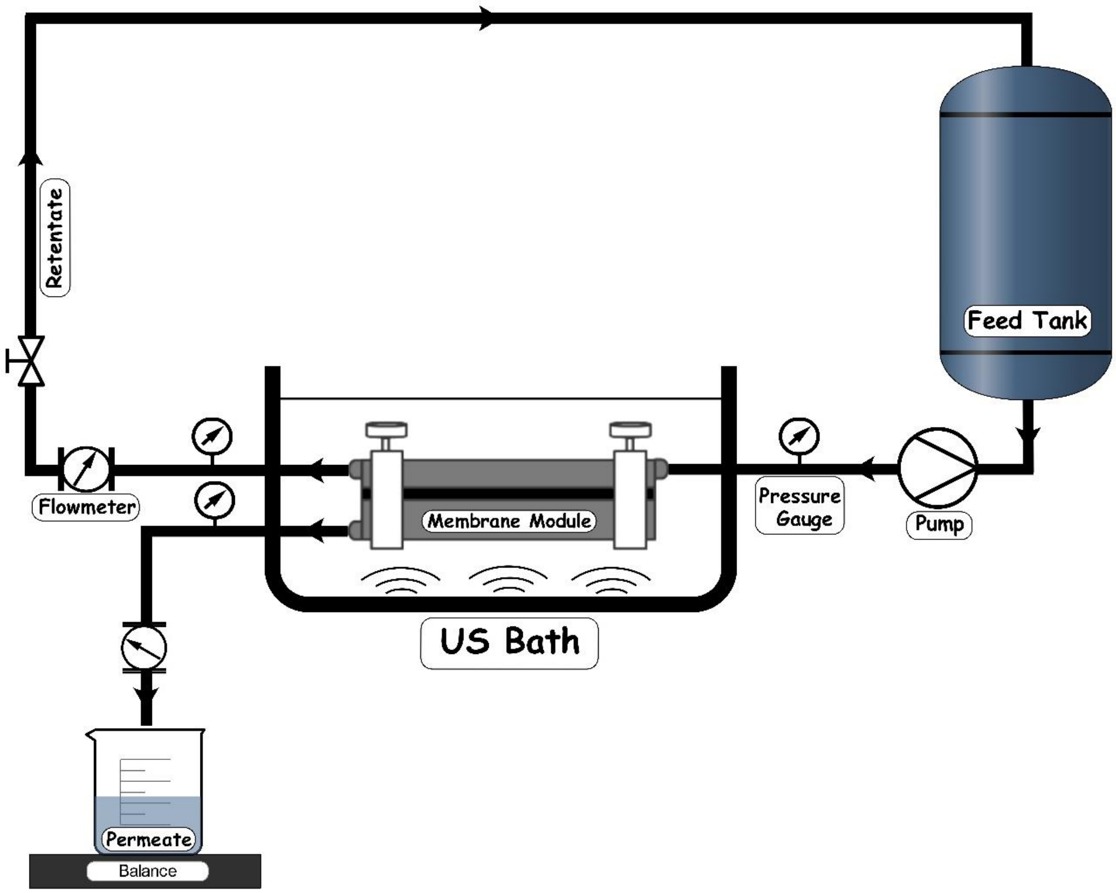

Fig. 3. Schematic of a simple ultrasonic bath used for membrane cleaning [101].

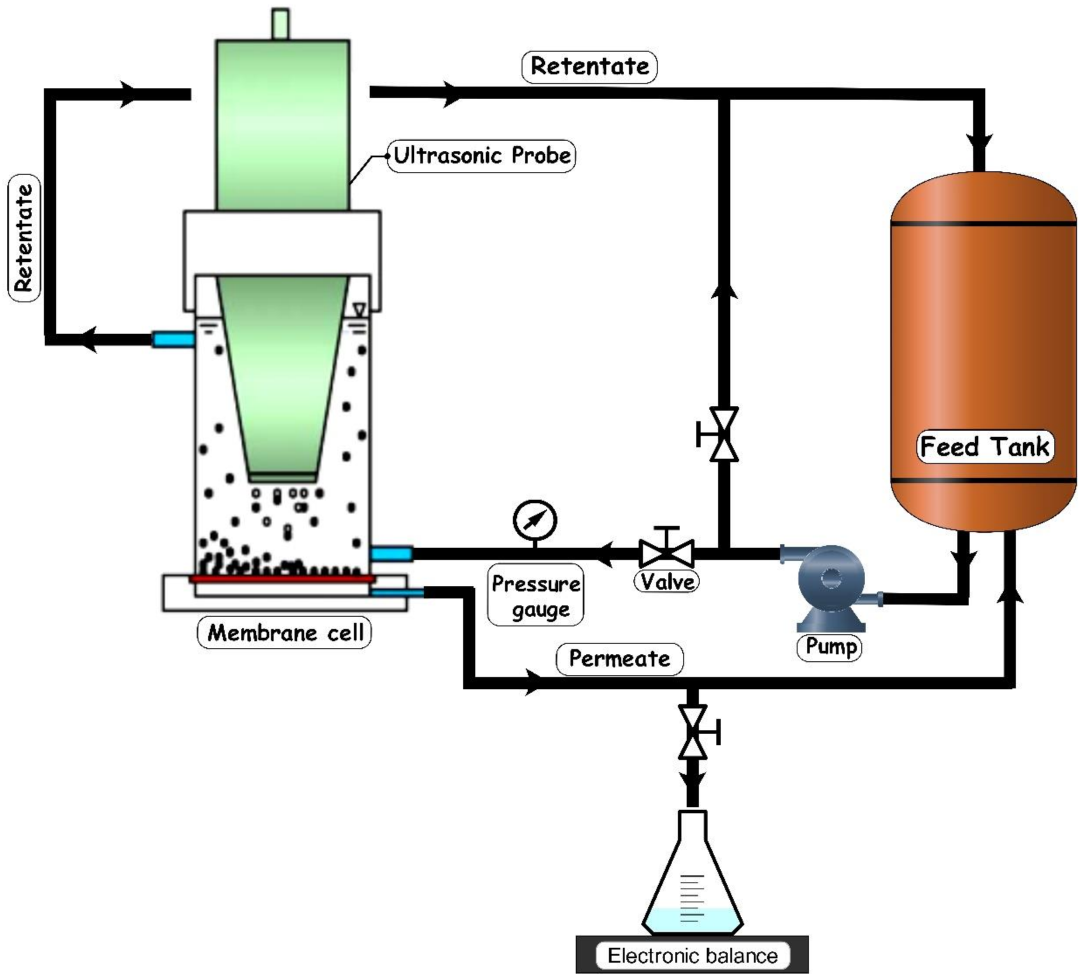

Fig. 4. Diagram of a membrane filtration system assisted with an ultrasonicator (homogenizer) [102].

3.2. Parameters affecting the ultrasonic-assisted membrane cleaning

3.2.1 Frequency

The amount of released energy from the bubble explosion and the maximum bubble size before collapse (resonance diameter) are a strong function of frequency [36, 103]. The size of the generated bubbles increases by reducing the frequency, while the number of bubbles decreases (Fig. 5). Larger bubbles release more energy, resulting in more energized but fewer hot spots, while smaller and more bubbles (higher frequencies) produce a larger density of low-energy hot spots [104, 105]. A trade-off exists between the size (released energy) and the number (cavitation rate) of bubbles, so selecting the optimal range of frequencies is critical as a function of the membrane cleaning process.

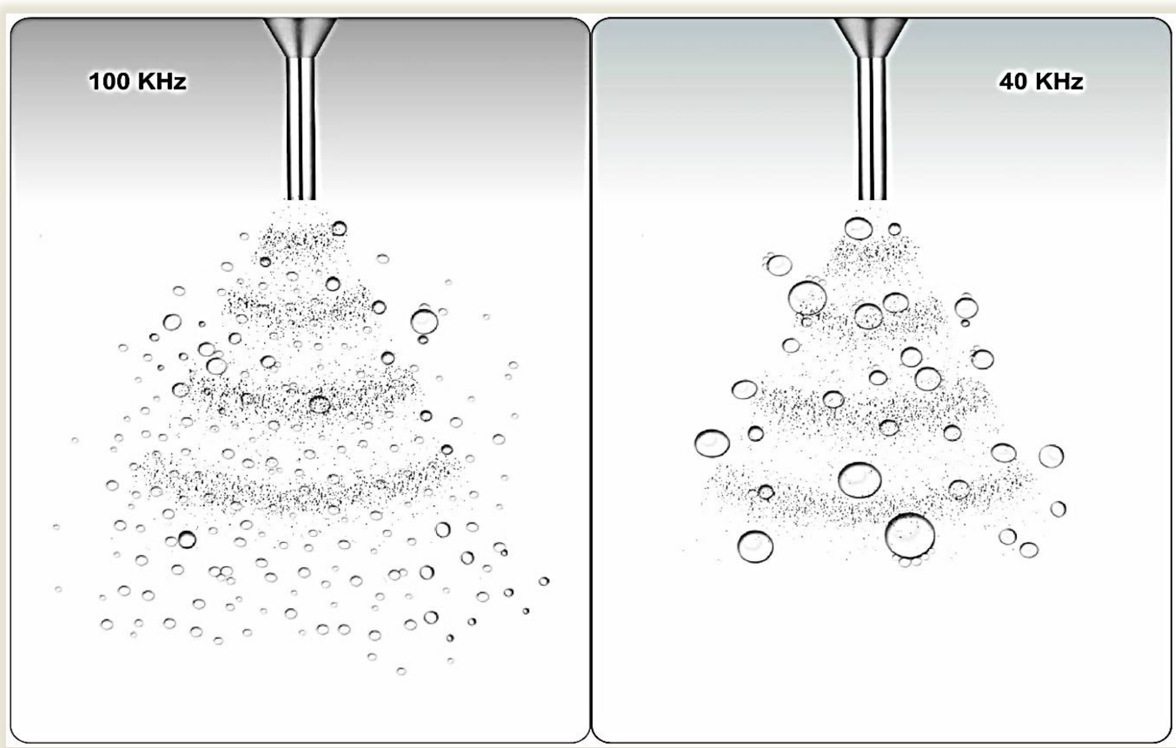

Fig. 5. The effect of implemented frequency on the size and population of the generated micro-bubbles.

Tarleton and Wakeman [81] studied the effect of frequency on the flow rate of a microfiltration process with three polymeric membranes fouled by inorganic particles in cross-flow filtration. An enhancement in filtration flux was observed by reducing the frequency from 40 to 23 KHz, which was considered the result of a different sound adsorption by the particulate matter. The effect of frequency on permeation of a dextran solution in an ultrafiltration process was presented by Kobayashi et al. [106]. The permeate flux increased by decreasing the frequency and no enhancement of permeation was observed when the frequency was set to its highest value (100 KHz). The same results were reported in UF and MF processes for the treatment of peptone and milk aqueous solutions [103]. Different frequencies, all higher than 200 KHz, were applied to treat a ceramic membrane fouled by sulfate polystyrene latex particles [44]. Results indicated that at high frequencies, full recovery in flux was achieved only by intensifying the power to a value larger than 1.05 $\mathrm{w/cm^2}$ for treatments longer than 30 s, while the same effect was observed for smaller power values at lower frequencies. The same influence of frequency was reported by Kyllonen et al. [53], and Lujan Facundo et al. [107]. Furthermore, Ohl et al. [108] showed that the combination of high and low frequencies (tandem frequency) in the membrane cleaning process resulted in better results compared to using a single high or low frequency. This technique increased the number of microbubbles on the surface of the membrane. The effect of tandem frequencies was then confirmed by Maskooki et al. [109]. Hou et al. [110] studied the effect of ultrasound waves with different frequencies (20, 30, 40, and 68 KHz) on the mass transfer rate over a PVDF membrane in a direct contact distillation system. The permeation was enhanced by reducing the ultrasonic frequency, explained with the lower production and intensity of cavitation at higher frequency. Similar results were obtained later in different membrane systems and different ranges of frequency [111, 112].

Summarizing the literature reports, it can be concluded that a lower frequency can help in the recovery of the flux by decreasing the filtration resistances of the membrane (especially the resistance associated to the cake layer) more effectively than high frequencies, which would generate more cavities but with lower energy [18]. However, as the frequency increases, the probability of particle degradation increases, lowering the probability of fouling by the same particles. The selection of a proper frequency highly depends on foulant and membrane characteristics and it should be optimized considering operational factors, such as transmembrane pressure (TMP) [46] and feed temperature [113].

3.2.2. Power intensity

The ultrasonic intensity is defined as the ultrasonic power per unit area and is an important factor influencing the efficiency of ultrasonic-assisted membrane cleaning [111]. The simplicity of power tuning compared to frequency changes, also from a practical point of view, is the main advantage of exploiting this parameter as an optimization variable. Matsumoto et al. [114] showed that the membrane permeation increased by increasing the sonication power in an ultrasonic-assisted cross-flow microfiltration process. Five types of ceramic membranes with different pore size were used to investigate the effect of pore size on permeate flux in the presence of ultrasonic waves. Observations suggested that a larger effect of increasing the ultrasonic power was obtained for smaller membrane pore sizes. Kobayashi et al. [115], Simon et al. [116], and Lamminen et al. [44] reported similar effects in ultrafiltration. The effect of ultrasonic power, horn type, and membrane-horn distance were studied by Juang and Lin [27]. At stronger ultrasonic intensities, the flux linearly increased by increasing the ultrasonic power, while at weaker intensities no significant trend was observed. In a study by Maskooki et al., the effect of ultrasound power was evaluated on membrane cleaning during and after the microfiltration of milk [109]. In accordance with the results summarized so far, the permeate flux increased linearly with ultrasonic power, rationalized with the enhanced cavitation. Muthuakumaran et al. confirmed that changing the ultrasonic power and keeping constant all the other conditions during the ultrafiltration of whey, the cleaning efficiency increased linearly with increased power [117]. The effect of the applied ultrasonic power on the permeate flux of different membranes was also studied by Wang et al. [118]. Four types of polymeric MF membranes were irradiated by ultrasonic waves of different intensities. For three of the membranes (PES, CN-CA, and N6), but not for the PVDF membrane, the permeate flux was affected by changes in the power intensity.

While larger values of power intensity clearly result in higher cleaning efficiency, they also lead to higher risks of membrane damage, especially in membranes with sensitive selective layers. The effect of power intensity on the permeation of an aromatic polyamide NF membrane fed by synthetic arsenic-rich brackish water was investigated by Wang et al. [46]. Water flux and rejection rate were monitored for different power intensities. Power intensities higher than a value of 1 w/cm^2 induced irreversible damage on the membrane surface, while power intensities lower than this threshold improved the permeation with no significant changes in rejection rate. It has also been reported that the membrane flux was reduced at very high ultrasonic powers [111]. This observation was rationalized with the very fast growth of cavitation bubbles, then connecting and forming a gaseous barrier preventing the spread of ultrasound waves. Based on these observations, an optimum value of power intensity seems to exist [111]. This effect was also observed for the separation of oil-in-water emulsions by hollow-fiber and flat-sheet polyurethane (PU) membranes [119]. Below a certain threshold, the mean size of the emulsion oil droplet increased with increased ultrasonic power. However, beyond the threshold, the droplet sizes decreased and oil rejection was impaired. Higher power can also induce emulsification of oily wastewater, negatively affecting its treatment.

To summarize, higher ultrasonic power produces on average more violent collapses of the generated bubbles. Consequently, the amount and intensity of the released energy will increase, commonly resulting in improved cleaning efficiency and higher recovered fluxes. During operation, very high power intensities may be counterproductive under certain circumstances. However, if the main objective of the operation is the ultrasonic-assisted cleaning of fouled membranes, the power may be maximized as long as the membrane structure is not damaged. The membrane performance should be evaluated and taken into account before and during changes of the ultrasonic power to ensure the feasibility of the process.

3.2.3. Pressure

Pressure has a two-fold effect: it induces considerable influences on both the number and the size of cavities and it influences the transport of foulants onto the membrane surface [20]. First, at higher filtration pressure the cavitation threshold increases and hence, fewer bubbles form [120, 121]. This phenomenon may actually enhance the intensity of bubble collapse with stronger mechanical effects that can improve the cleaning effect by ultrasounds [122, 123]. Additionally, bubble shielding may be minimized by decreasing the number of bubbles at higher pressure; this facilitates the propagation of ultrasound waves through the medium and in turn the average intensity of the ultrasound waves reaching the membrane/liquid interface increases [124]. Intensified acoustic streaming along with cavitation-induced turbulence at the membrane/liquid interface results in increased shear stress and improved cleaning efficiency [20]. Kobayashi et al. [106]] examined the effects of ultrasounds on the permeate flux when filtering dextran solutions through polyacrylonitrile ultrafiltration membranes and showed that increasing the TMP improved the effectiveness of ultrasounds on enhancing the permeate flux. Finally, Hemmati et al. [125] evaluated the effect of TMP on the efficiency of carrot juice clarification using dead-end microfiltration and reported improvements in the permeate flux at 0.5 bar compared to 0.2 bar.

Increasing the filtration pressure may be disadvantageous in some circumstances. A higher pressure increases the tendency of detached particles to be transported back near the membrane and redeposit on its surface [20]. Pressure might also force foulants into the membrane pores [126] or it may increase the density of the cake layer [127], both effects resulting in a larger resistance to permeation. Alventosa-deLara et al. [43] evaluated the effect of TMP (0.5, 1.5, 2.5 bar) on the ultrasound-assisted cleaning efficiency of an ultrafiltration ceramic membrane fouled by dye particles. The cleaning efficiency reached a peak value at the medium value of the range of investigated pressures. This result was attributed to the simultaneous effect of cavitation causing particle detachment from the membrane and the ability of acoustic streaming to overcome the drag force from the applied pressure to maintain the detached particles away from the membrane surface. Kyllonen et al. [53] studied the effect of TMP on online ultrasound-based cleaning during filtration of industrial wastewaters. They used a cross-flow membrane module inside of which the transducer was assembled, and they reported difficulties in membrane cleaning under high-pressure conditions. Chen et al. [20] investigated the effect of pressure in the cross-flow ultrafiltration of silica colloids using γ-alumina membranes. They reported a decline of the relative permeate flux from 100 ± 2% to 59 ± 5%, by increasing the pressure from 1 to 8 psi. As experimental results revealed, higher pressure had a detrimental effect on cavitational efficiency and increased the permeation drag force of the particles toward the membrane surface [126]. In this study, these mechanisms were more significant than the beneficial effects of stronger acoustic streaming or of ultrasonically-generated turbulence and therefore, the relative permeate flux was impaired. Similarly, Muthukumaran et al. [33] reported that increasing the TMP reduced the permeate flux during experiments because of the greater compressive forces applied to the cake layer in the ultrasonic-assisted cross-flow ultrafiltration of dairy whey solutions. In other works, Matsumoto et al. [114] and Duriyabunleng et al. [128] observed an optimum applied pressure at which a maximum steady-state flux was obtained. Beyond the threshold pressure, the removal of the denser cake layer by the ultrasonic was lower, and thus, the flux recovery was negatively affected.

To summarize, the removal of the cake layer by ultrasonic waves at high applied pressures is often less effective due to a stronger compaction of the cake layer. Increased filtration pressures increase the compressive forces driving cavitation collapse and leads to fewer cavitation bubbles absorbing and scattering sound waves as well as increased sound wave penetration [56]. On the other hand, permeation drag amplifies at high filtration pressure, which may lead to lower flux improvements [56, 71]. Therefore, also in this case an optimization procedure is required to achieve the most effective membrane filtration configuration [71].

3.2.4 Temperature

Temperature influences the ultrasound-assisted membrane filtration in a non-trivial way. From the filtration view point, increasing the temperature leads to enhanced diffusion, higher solubility, and reduced viscosity with correspondingly higher Reynolds numbers, all phenomena contributing to better cleaning efficiency and flux [54, 56]. In the study by Chai et al. [129] on ultrasound-assisted cleaning of polymeric UF and MF membranes fouled by peptone, the authors reported that the permeate flux was higher as the temperature increased from 20 to 40 °C. They also observed that the recovery of permeate flux during cleaning was faster at higher temperature, due to the larger solubility value of the peptone. In another study, Muthukumaran et al. [33] achieved similar results when investigating the feasibility of ultrasonic-assisted cleaning of polysulfone membranes in the cross-flow ultrafiltration of dairy whey solutions. The cleaning efficiency was improved when using a higher temperature, which was attributed to the reduction of viscosity of the solution. Additionally, in a recent study on the effect of ultrasound in membrane distillation, increasing the feed temperature from 40 to 70 ℃ led to a higher permeate flux at different ultrasonic powers (110-260 W), keeping the frequency constant [110].

Temperature also affects the cleaning efficiency of ultrasounds through its effect on ultrasonic cavitation [130]. Once the temperature of the solution increases, the vapor pressure rises and causes more rapid bubble formation, which in turn is associated with a lower shock-wave intensity of bubble collapses on the membrane surface [55, 131]. When Zhu et al. [130] investigated the effect of temperature on the permeate flux of membrane distillation, they found that the enhancement ratio was reduced with the increase of solution temperature due to a higher saturation vapor pressure. Changes in temperature also cause alteration of the cavitation threshold and consequently a change in ultrasonic effectiveness [131]. In general, decreasing the temperature leads to an increase in the threshold value, which may be due to the lower liquid vapor pressure, higher viscosity, and/or the different surface tension of the liquid [132]. Li et al. [131] applied the ultrasound technique to clean nylon microfiltration membranes fouled by pulp and paper effluents. The water permeate flux of the membrane decreased when the cleaning temperature increased from 23 to 40 °C, which was attributed to changes in the ultrasonic cavitation intensity behavior. Wang et al. [132] investigated the conditions of ultrasound-assisted cleaning of nanofiltration membranes fouled with inorganic scales: experimental results showed that there may be an ideal temperature to maximize the flux recovery rate. These authors proposed that the optimal transport and cavitation phenomena occurred at approximately 30 °C. Similarly, Muthukumaran et al. proposed that there may be an ideal temperature at which cavitation intensity is the highest and related to the colligative properties of the liquid [55].

3.2.5. pH

The role of pH on membrane filtration is complex because this parameter affects the properties of both the membrane and the feed solutes. The effect of pH on membrane fouling was investigated in several studies and conflicting reports exist [132, 133]. The net charge on both the membrane and the protein is changed by changing the solution pH because of the ionization or deionization of various acidic/basic groups on the protein and membrane surface. Accordingly, attractive or repulsive interactions can occur as a function of the equilibrium constants (pKa values) and charge density of these surfaces [134]. Usually, the permeation rate increased with increasing pH, due to lower foulant-membrane attractive interactions at more basic pH values [135, 136]. For example, the filtration of proteins, such as whey, through negatively charged membrane in acidic solutions can intensify fouling because this protein is positively charged below pH 7[137, 138]. Furthermore, the pH has strong effects on foulant-foulant interactions [139]. For example, Kilduff et al. [140] found that the highest specific cake resistance occurred at neutral pH due to the weak electrostatic repulsion between foulants. Looser deposits are usually formed in basic feed solutions [141].

Several authors [54, 57, 132, 142, 143] have studied the relationship between the feed pH and filtrate flux in the presence and absence of ultrasounds and confirmed that pH is also an important parameter controlling the efficiency of the ultrasonic field in membrane cleaning. Gao et al. [143] applied ultrasounds at a frequency of 20 kHz and a power of 16 W in a cross-flow ultrafiltration system treating natural surface water containing significant levels of natural organic matter. These authors observed that the normalized permeate flux improved from 0.61 to 0.71 as the pH increased from 4 to 8, and then decreased to 0.59 at pH 10. Wang et al. [132] studied the ultrasound-assisted cleaning of nanofiltration membranes fouled by inorganic scales in arsenic-rich brackish waters. They reported that the efficiency of cleaning was significantly improved when the pH was lower, as lower pH is more favorable for the dissolution of scales. The water flux and rejection rate of the membrane were recovered entirely within 15 min as the pH dropped to 3. Muthukumaran et al. [54] examined the effect of pH on the ultrasonic cleaning of polysulfone ultrafiltration membranes fouled by dairy whey. When the pH increased, the cleaning efficiency improved and maximum cleaning efficiency occurred at pH values between 11.5 and 13, a result also confirmed in a previous study by the same authors [57]. However, these studies mostly corroborated the trends typically observed in membrane operation at different pH values in the absence of ultrasounds. Ultrasonic-assisted filtration was applied to increase the magnitude of these well-known cleaning mechanisms but pH has not shown to have a direct specific effect on ultrasound-related mechanisms and on cavitation phenomena.

3.2.6. Operation mode

The sonication mode is directly related to energy costs and to the required duration of the sonication time, in turn associated with membrane damage. The effect of sonication mode in a dead-end ultrafiltration process was investigated by Simon et al. [144]. Both pulsed and continuous modes provided an enhancement of permeate flux. However, continuous mode was more effective than intermittent mode, while factors such as energy consumption and membrane lifetime were not compared. On the other hand, the destructive effect of continuous application of ultrasonic irradiation was reported by several authors [145]. Accordingly, through SEM analyses of the membranes, Lamminen et al. [146] reported badly damaged materials subjected to ultrasound cleaning in continues mode, whereas in pulse mode the flux improved with negligible damage to the membrane surface. The authors also applied ultrasounds in continuous or pulsing mode at different powers. The experiments were conducted with PVDF membranes fouled with 10 mg/L latex particles for 4 h, in which under pulse conditions the ultrasound was applied for cycles in which it was on for 1 s and off for 10 s. At higher applied powers, improvement in flux occurred with a concurrent evidence of damage to the membrane when using continuous ultrasound. Although pulsed ultrasound was slightly less effective than continuous ultrasound, its application during cleaning provided a flux of around 97% of the original water flux before fouling, compared to the untreated membrane which showed a value of 74%. The flux recovery without membrane damage was attributed to the fact that the fouled membrane has a thin layer of fouling material on the surface which can actually act as a barrier preserving the membrane surface from damage from short pulses of ultrasounds.

Chen et al. [20] examined pulse intervals of ultrasounds and observed that increasing the intervals between pulses decreased the flux recovery. Energy consumption was also investigated by these authors, and it was seen that using short pulse intervals can result in remarkable energy savings compared to the continuous mode. It was claimed that the ineffectiveness of some bubbles produced in continuous mode may weaken the efficiency and performance of ultrasonic waves, while the number of energized bubbles in pulsed mode was larger. More recently, the effect of sonication mode was specifically studied by Agi et al. [119]. In this study, the effect of intermittent and continues irradiation of ultrasonic waves in the ultrafiltration of an oil/water emulsion using hollow fiber and flat-sheet polyurethane membranes was investigated. The results showed that using ultrasonic waves intermittently could enhance the permeate flux in the filtration process, even better that applying ultrasounds in a continuous mode.

Table 2. Studies on Ultrasonic-Assisted de-fouling of Membranes from the literature.

Membrane type	Membrane Apparatus	Foulant	Frequency (KHz)	Power/Power intensity	Operating Pressure	Temperature (℃)	PH	Membrane erosion	Best reported condition(s)	Ultrasonic-assisted effects on fouling control and cleaning of membrane	Ref
Polyacrylonitrile	Cross-flow UF	Dextran	28, 45, 100	2.5-3.3 W/cm^2	0-140 kPa	25	Nm	Nm	Permeate flux of ~5 × 10^{-6} m^3.m^{-2} s^{-1} @45 kHz 0.5 wt% dextran	Reduction of foulant layer resistance and enhancement of mass transfer	[106]
$\boldsymbol{\gamma - Al_2O_3}$	Dead-end MF	Sulfate polystyrene latex	70, 205, 354, 620, 1062	0.21-2.1 W/cm^2	0.7 atm	15	7	Negative	Full flux recovery @ 620 kHz 0.42 W.cm^{-2}	Increase in the number of cavitation bubbles and increase in acoustic energy	[44]
Polyvinylidenefluoride	Hollow fiber UF	$CaCl_2$	Nm	0, 241, 1097, 1639, 2320 W/m^2	21, 44 , 64, 80 kPa	Nm	Nm	Positive	73.2% flux recovery @ 2 Kw.m^{-2} With 2 g.L^{-1} Citric acid soaking	Cavitation, acoustic streaming and vibration	[147]
nylon	Cross-flow MF	paper mill effluent	20	82.9 W/cm^2	0.5 atm	23, 30, 40	4.96	Negative	97.8% flux recovery @ 20kHz 375 W With forward flushing	Ultrasonic cavitation and bubble collapse on the membrane surface	[148]
Polysulfone	Cross-flow UF	Whey solution	50	300 W	55 kPa	20-22	7-10	Negative	112% flux recovery @ 50 kHz 300 W With adding surfactant pH=7 Sonication time=20 min	Physical cleaning	[57]
Polysulfone	Cross-flow UF	whey solutions	50	300 W	55-300 kPa	25, 55	4.5-14	Nm	91% cleaning efficiency @ 50 kHz 300 W pH=12 Surfactant con.: 10 mM	Enhancement of the turbulence within the cleaning solution	[54]
polyacrylonitrile	Cross-flow UF	dextran	45	248 W	30 kPa	25	Nm	Nm	Permeate flux of 6 × 10^{-6} m^3.m^{-2} s^{-1} @45 kHz 248 W 1 wt% dextran 30 min sonication	Enhancement of bulk mass transfer in the concentration polarization layer near the membrane surface	[149]
Ceramic	Tubular UF	Ovalbumin, dextran, PVA	200-400	70-100 W	5 kg/cm^2	25	Nm	Nm	140% higher mass transfer coefficient @ Under ultrasonic irradiation	Reduction in the permeation resistance caused by a gel and boundary layers, Increase in the mass transfer coefficient	[150]

Membrane type	Membrane Apparatus	Foulant	Frequency (KHz)	Power/Power intensity	Operating Pressure	Temperature (℃)	PH	Membrane erosion	Best reported condition(s)	Ultrasonic-assisted effects on fouling control and cleaning of membrane	Ref
Microfilter (Not mentioned the type)	Cross-flow MF	Anatase, China clay	23, 40	<600 W	20 psi	Nm	6.6, 6.2	Nm	~2.6 $m^3.m^{-2}\ h^{-1}$ @23 kHz (600W) + Electric field (400V DC 10A max) 1.7% v/v China clay	Imposition of acoustic field	[151]
Polyethersulfone	Dead-end UF	Dextran	20	8-20 W	2 bar	30±1	Nm	Nm	No optimal conditions were reported (Comparative Study)	Inducing convective currents and cavitation effects, influencing hydrodynamics, reducing part of the boundary layer at the solution/membrane interface	[116]
Polytetrafluoroeth-ylene (PTFE)	Air gap membrane distillation (AGMD)	NaCl	10-65	0.5-6.5 W/cm^2	Nm	40-65	Nm	Nm	Enhancement ratio of 200% for permeate mass flux @20 kHz 5 $W.cm^{-2}$ ~ 40 ℃	Inducing membrane absorption and transmission of an acoustic energy, membrane vibration, and acoustic energy dissipation	[130]
γ -alumina	Cross-flow UF	Silica particles	20	19±0.5 W	5 psi	20	5.6±0.4	Negative	Full flux recovery @20 kHz ~ 19 W 20 ℃ Particle con.: 0.1 $g.L^{-1}$ Distance: 3.5 cm Cross flow rate: 500 $ml.min^{-1}$	Acoustic streaming and ultrasonically generated turbulence	[70]
Polysulfone	Cross-flow UF	Peptone	28	8-33 W	60 kPa	25	Nm	Negative	Permeate flux of $1.2 \times 10^{-5}\ m^3.m^{-2}\ s^{-1}$ @28 kHz 33 W 1 wt% peptone Operating pressure: 60 kPa 25 ℃	Cavitation, dislodging the foulant layer from the membrane	[152]
Polysulfone, cellulose	Cross-flow UF	Polyethylene glycol, dextran	30±10, 20	40, 10 W	50 psi	Nm	Nm	Nm	800% flow rate enhancement @30 kHz 40 W 0.45wt% dextran	Strong local turbulence, minimizing concentration polarization	[153]

Membrane type	Membrane Apparatus	Foulant	Frequency (KHz)	Power/Power intensity	Operating Pressure	Temperature (℃)	PH	Membrane erosion	Best reported condition(s)	Ultrasonic-assisted effects on fouling control and cleaning of membrane	Ref
Cellulose	Cross-flow UF	Cu^{2+}–polyethylenimine, W/O emulsions	20	0-150 W	69 kPa	25	9.5	Negative	70-80% flux recovery @20 kHz >30 W Distance: 65 mm Cu-PEI solution	Producing an alternating adiabatic compression and rarefaction, forming microbubbles and imploding during the compression	**[27]**
Polytetrafluoro-ethylene	Hollow fiber MF	Silica	20	260 W	Nm	53	7	Nm	98% flux recovery @20 kHz In the presence of calcium ions	Mechanical and thermal effects, microstreaming, shock wave, and acoustic vortex streaming.	[154]
Aromatic polyamide	Cross-flow NF	Arsenic	40	1, 1.5, 2, 2.5, 4.5 W/cm^2	0.5, 1, 1.5, 2, 2.5 MPa	15, 20, 30, 40, 50, 60	2, 3, 3.5, 4	Positive	99.99% flux recovery @40 kHz 1 $W.cm^{-2}$ Assisted with citric acid pH=3 30℃	Acoustic streaming or microstreaming, mechanical vibration of the membranes	[132]
Polyethylene	Cross-flow (hollow fiber) MF	Waste activated sludge	28	0.18, 0.24, 0.3, 0.4, 0.5 W/cm^2	Nm	35±2	7.4±0.2	Nm	Fouling can be controlled successfully with ultrasonic intensity	Greater sludge decomposition, improved digestion performance, smaller flocs of the bulk sludge in the ultrasound-assisted AnMBR.	**[155]**
Polysulfone	Cross-flow (hollow fiber) UF	Municipal wastewater	35, 130	35, 29 W/l	0-15 KPa	25±2	7.91±0.24	Nm	57.33% fouling reduction @35 kHz Flow rate = 150 $L.m^{-2}\ h^{-1}$	Cavitation, generation of microstreams and vibration, degradation of water molecules and increasing production of hydroxyl radicals	[156]
Polytetrafluoroethylene (PTFE)	Direct contact membrane distillation (DCMD)	Bovine serum albumin (BSA)	20	26 0 W	Nm	53	7	Nm	98% flux recovery @20 kHz	Mechanical and thermal effects, microstreaming, shock wave, acoustic vortex streaming	[157]
Polysulfone, cellulose	Cross-flow UF and MF	Peptone and milk aqueous solutions	28, 45, 100	23 W/cm^2	60 kPa	25	Nm	Nm	Permeate flux of $\mathbf{9\times10^{-5}}$ $m^3.m^{-2}\ s^{-1}$ @28 kHz 23 $W.cm^{-2}$ 1 wt% milk 25 ℃	Cavitation	**[103]**
Polyetersulfone, alumina-based ceramic membranes	Cross-flow, dead end	Industrial wastewaters from the paper industry	27, 40 or 200	120, 200, 400 W	0, 1, 2, 3 bar	Ambient temperature	Nm	Positive	Fully cleaned @27 kHz 1.1 $W.cm^{-2}$ Atmospheric pressure	Cavitation	[53]
γ -alumina	Cross-flow UF	Silica particles	20	3.8 ± 0.1 W/cm^2	5 psi	22.4	5.6±0.4	Positive	100% flux recovery @20 kHz 3.8 $W.cm^{-2}$ 1psi 0.2 g/L Silica Distance: 3.5 mm Continuous mode	Acoustic streaming and ultrasonically-generated turbulence	[20]

Membrane type	Membrane Apparatus	Foulant	Frequency (KHz)	Power/Power intensity	Operating Pressure	Temperature (℃)	PH	Membrane erosion	Best reported condition(s)	Ultrasonic-assisted effects on fouling control and cleaning of membrane	Ref
Polyethylene	hollow fiber MF	Aqueous milk solution	28	300 W	60 kPa	Nm	Nm	Nm	Permeate flux of 9.5×10^{-5} $m^3.m^{-2}$ s^{-1} @28 kHz 300 W Distance: 8 cm	Sonic pressure around the membrane module, generation of cavity bubbles	[158]
Polysulfone	Cross-flow UF	Dairy whey solution	50	105 W	55, 150, 225, 300 kPa	10-55	6.3	Negative	170% flux recovery @50 kHz 300 W	Acoustic streaming and mechanical vibration	[33]
Polysulfone	Dead-end UF	Dextran	20	3,4, 5, 7.5, 12, 16 W	0.5, 1, 1.5, 2 bar	30±1	Nm	Negative	89% retention rate @20 kHz 16 W	Physical effects associated with ultrasound wave propagation decreasing boundary layer resistance	[144]
Mixed ester, Cellulose nitrate, Polycarbonate	Cross-flow MF	Anatase, Calcite, China clay	23, 40	0.26, 0.8, 1.7 W/cm^2	20 psi	25	3.9, 9.4, 1.2	Nm	Permeate flux of ~3.7 $m^3.m^{-2}$ h^{-1} @23 kHz 1.7 $W.cm^{-2}$ 0.1 wt% anatase pH: 8.1 cm	Microstreaming, Cavitation	[159]
PES, CN-CA, N6, PVDF	Dead-end MF	soybean protein	40	0, 1.43, 2.13, 3.57	20, 30, 40, 50, 60, 70	20±2	Nm	Positive	Permeate flux of 86.3 $kg.m^{-2}$ h^{-1} @23 kHz 3.57 $W.cm^{-2}$ PVDF membrane	Cavitation, acoustic streaming, and ultrasound-induced vibration	[118]
PVDF, γ-Alumina	Cross-flow MF	Sulfate polystyrene latex particles	476	3.3-61.3 W	0.7 atm	Nm	6	Positive	97% flux recovery @476 kHz 60.1 W Continuous mode	Cavitation	[137]
piezoelectric ceramic microfiltration	Cross-flow MF	Sulfate latex particles	70-80	2.2-22 kW/m^2	10 psi	20.2±0.2	Nm	Nm	Full flux recovery @72.6 kHz 2.2 $W.cm^{-2}$ Poled PZT membrane	Surface vibration, cavitation	[160]
Polyethersulfone/ polyolefin	Cross-flow UF	Whey protein concentrate	20	101 W	2 bar	5±2	Nm	Nm	103% flux recovery @20 kHz 5 wt% whey solution	Disruption of hydrophobic interactions of whey protein aggregates, delays in gelling of the protein, inducing shear forces, reducing this cake growth factor.	[161]
Polytetrafluoroethylene	Dead-end MF	Humic acid/bentonite mixture	Nm	0, 5, 15 W	50 kPa	Nm	7±0.1	Nm	45% flux recovery @15 W Distance= 2 cm 25 min sonication 15 $mg.L^{-1}$ coagulant dose	Growing of microbubbles and imploding within a specific region in the aqueous media	[162]

Membrane type	Membrane Apparatus	Foulant	Frequency (KHz)	Power/Power intensity	Operating Pressure	Temperature (℃)	PH	Membrane erosion	Best reported condition(s)	Ultrasonic-assisted effects on fouling control and cleaning of membrane	Ref
Polyethersulfone, hydrophilic polyethersulfone	Cross-flow UF	Bovine serum albumin (BSA)	21, 25, 30, 38	300 W	1 bar	25-45	9-11	Negative	~ 84% flux recovery @21 kHz Membrane type: UP005	Enhancement of the cake layer removal and reduction of the pore plugging	[142]
Al_2O_3 ceramic membrane	Cross-flow UF	NOM (surface-water)	20	16 W	5 psi	18	4, 8, 10	Negative	88% flux recovery @ pH=8 40 mg.L^{-1} calcium concentration	Limiting the fouling potential by creating nano-sized particles and enhancing the solubility of fine particles in solution	[143]
Polyamide	Cross-flow RO	CaSO4,Fe^{3+}, carboxyl cellulose	20	2.8 W/cm^2	100 kPa	20±1	4.5	Negative	264% flux recovery @ pH=8 500 mg.L^{-1} CMC 2 h sonication	Acoustic vortex microstreaming within the pores of the membrane, mechanical cleaning due to high-speed microstreams.	**[163]**
Polyvinylidene- fluoride	Dead-end MF	Carrot juice	20	400, 600, 800, 1000 W	0.2, 0.5 bar	Nm	Nm	Nm	Best condition @ 1000 W 30 min sonication 100 ml solution	Reduction of cake layer as the main fouling mechanism, decreasing kinetics of cake formation	[125]
Polytetrafluoroethylene (PTFE)	Direct contact membrane distillation (DCMD)	Humic acid	20	260 W	Nm	53	7	Nm	94% flux recovery @ concentration factor of 4	Mechanical and thermal effects, microstreaming, shock wave, acoustic vortex streaming	[164]
Polyvinylidene-fluoride	Hollow-fiber UF	Domestic sewage effluent, Suwannee River Humic Acid, and local tap water	38	85 W	1-10.4 kPa	20±2	7.64±0.04	Nm	No optimal conditions were reported (Comparative Study)	Removing hydrophilic organic matter and polysaccharide materials, reducing foulant-membrane bonding strength, removing large MW organic matter	[165]
Single-fiber of polysulfone	Cross-flow UF	Domestic wastewater	35, 130	0.3-1.1 W/cm^2	0-4 psi	Nm	7.19±0.08	Nm	Higher fouling reduction @ 35 kHz	Cavitation	[36]

4. Membrane erosion

In spite of the proven efficiency of ultrasounds in cleaning fouled membranes and preventing flux decline due to fouling, this technique may impact the lifetime of the membrane. Higher ultrasound power intensity and longer ultrasound irradiation time improve the effectiveness of the method while possibly producing destructive effects on the membrane selective layer [166]. Specifically, when a cavitation bubble is captured at the membrane interface, it may lead to physical erosion of the surface by repeated oscillations within the stagnant interface [57]. The nature of the polymeric material of the membrane and the parameters of the ultrasound operation are the main factors affecting the extent and magnitude of the membrane damage. It is important to note that investigations around the effects of ultrasound irradiation on the membrane performance have led to inconclusive and often contradicting results. Some studies revealed that the ultrasound treatment brought damage the membrane surface [27, 55, 167], while a number of different studies, partly from the same authors, observed no significant changes in membrane performance in spite of the continuous use of ultrasounds [33, 54, 168]. Table 3 summarizes the published literature around the topic of membrane erosion in ultrasonic-assisted operation.

Wang et al. [46] studied the impact of ultrasonic power intensity on the integrity of membrane selective layers in the ultrasonic-assisted cleaning of nanofiltration membranes that were fouled with inorganic scales. They reported membrane damage when the applied power intensity exceeded 1.5 W/cm^2, with the consequent impairment of rejection performance [167]. These results are in accordance with their previous report [118], suggesting that polymeric microfiltration membranes exposed to ultrasounds at a frequency of 40 kHz and an intensity of 1.43-3.57 W/cm^2 were negatively affected over their entire surfaces, resulting in the interconnection of neighboring pores forming large cracks. In the study of Kyllönen et al. [53], ultrasound irradiation inside the membrane module was used to achieve an effective cleaning of the membranes previously fouled by real wastewaters from the paper industry. The authors observed that membranes exposed to ultrasounds at frequencies of 27 or 40 kHz showed damaged at some locations on the surface while the presence of cross-flow (e.g., more than 0.6 m/s, Re 12000) during irradiation may have reduced the erosion on the membrane surface. According to Masselin et al. [167], significant variations occurred in membranes after exposure to ultrasonic waves. In their work, the influence of 47 kHz ultrasonic waves on polyethersulfone (PES), polyvinylidenefluoride (PVDF), and polyacrylonitrile (PAN) membranes with various molecular weight cut-off values was investigated. Over the three tested membranes, polyethersulfone (PES) was highly affected by the ultrasonic irradiation, while the two other materials showed fewer damages, with the exception of specific PAN and PVDF membranes with 50 and 40 kDa molecular weight cutoffs, respectively. Results revealed that the damage of the membrane surface under ultrasonic irradiation led to the formation of large cracks particularly at the edges of the membrane samples, as shown in Fig. 6. In another study, Chen et al. [20] investigated the mechanism of membrane damage when 20 kHz ultrasounds were applied in a cross-flow ultrafiltration system with γ-alumina membranes fouled by colloidal silica particles. They reported that when the membrane was located outside but near the cavitation zone, the integrity of the membrane was maintained. However, consequences such as damage, pitting, and membrane cracking may be unavoidable when the membrane module is placed inside the ultrasonic cavitation zone (1.3 cm or shorter distances from the ultrasonic horn). According to their findings, the surface damage caused by ultrasound waves was probably mechanical (e.g., cracking) due to microjets and/or shock waves during cavitation collapse of high energy densities [169-171]. Similarly, Wei et al. [166] found damage to the surface of

PVDF hollow fiber ultrafiltration membranes when the intensity of ultrasounds generated by a flat plate transducer was above 3 kW/m^2. Hou et al. [110] applied ultrasonic technology in direct contact membrane distillation, revealing that the skin layer of PVDF membranes was eroded, some portions of the material removed, and mechanical strength impaired by ultrasounds of 20 kHz frequency at a power of 260 W. Although the polypropylene (PP) membrane morphology was not changed significantly after three hours of ultrasonic irradiation, the pore size distribution analysis indicated that the membrane pores were enlarged and that the stretching strain of the PP fibers was reduced by almost 15%.

Although the abovementioned studies have revealed damage on the membrane surface upon sonication, other studies [54, 70, 149, 168] have observed little or negligible damage even following repeated use of ultrasound waves. When Muthukumaran et al. [57] investigated the effect of ultrasounds on the cleaning of whey-fouled membranes, they found that the ultrasound treatment did not affect the intrinsic permeability of the membrane and observed no significant erosion of the membrane. Similarly, Chen et al. [168] confirmed that no damage to the membranes occurred as a result of ultrasonic-based control of membrane fouling induced by natural organic matter and silica particles. In the study by Chai et al. [149], the application of 45-kHz ultrasounds to a cross-flow ultrafiltration cell immersed in an ultrasonic bath did not produce evident impairment of the membrane integrity. Agi et al. [119] investigated the effect of ultrasounds on membrane integrity in the application of synthesized polyurethane membranes for oil-in-water emulsion separation. No significant change in the permeate flux was observed, suggesting that ultrasounds did not damage the membrane structure. These authors attributed the lack of damage to the cross-flow filtration mode and also to the membrane mechanical strength provided by the finger-like structure and the dense skin layer. These results are consistent with the findings of Naddeo et al. [145], who found that their polysulfone membranes remained intact during the filtration of distilled water in the presence of continuous ultrasounds throughout a 4-h test. The SEM micrographs collected by Ruiz et al. [112], suggested that hollow fiber ultrafiltration membranes made of PVDF were not compromised after continuous irradiation with ultrasounds with power of 15 W and at different frequencies (20, 25, 30, and 40 kHz) over a period of four months. The complexity of these mechanisms is also attested by the fact that the same authors reported the integrity failure and the significant enlargement of pores of flat-sheet microfiltration membranes made of PES irradiated continuously with ultrasounds with powers ranging from 100 to 400 W and over a period of six months [172]. It appears that the impact on hollow fiber membranes is less important compared to that on flat-sheet membranes. This observation may be rationalized with the fact that hollow fiber membranes are fixed only at their ends and they can vibrate and dissipate energy when sonication is applied. In this way, the irradiated energy is less focused on localized spots of the membrane [112].

According to these results and based on the current knowledge of this topic, it is presently challenging to make reliable predictions on whether a membrane will or will not be damaged upon exposure to ultrasounds. Several parameters seem to concur in a complex and interrelated way to this effect. Selecting the appropriate membrane materials, structure, ultrasound frequencies, power, exposure times, membrane-module configuration, or most likely a combination of the above is clearly crucial to maximize the benefits of the ultrasonic technique while preventing significant membrane impairment [112]. The current and future research should focus on disentangling the various confounders and proposing guidelines to design the best possible modus operandi.

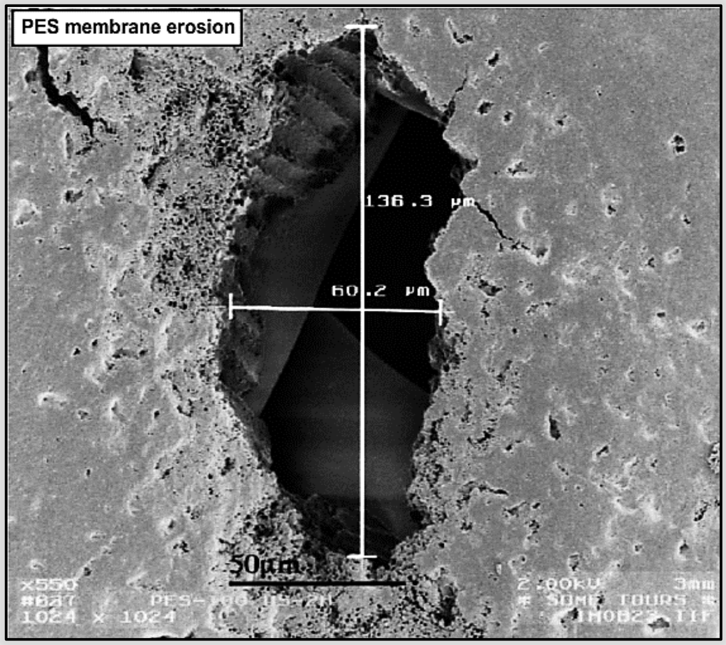

Fig. 6. Microscopic images of the highly damaged area of the PES membrane irradiated by ultrasonic waves; at a magnification of 550 [167].

Table 3. Studies on membrane erosion in ultrasonic-assisted membranes.

Power/ Power intensity	Frequency (KHz)	Probe situation	Membrane type	Exposure time (min)	Sonication mode	Probe-membrane distance (cm)	Damage report	Ref
18 W	20	In situ	Polysulfone	120	continues	1.4	None observed	[144]
Nm	47	Out situ	PES, PVDF, PAN	120	Periodic	Nm	All the membranes affected by ultrasonic waves but PES membranes affected more.	[167]
300 W	50	Out situ	Polysulfone	1 month	Periodic	Nm	None observed	[173]
160-400 W	40	Out situ	Polyethersulfone, N6, CN-CA, PVDF	30, 50, 90	continues	Nm	All the membranes were affected by ultrasonic irradiation, but PVDF membrane more resilient	[118]
3.8 ± 0.1 W/cm^2	20	In situ	γ-alumina	Nm	Nm	3.5-2.6-1.7-1.3	Pitting on the surface of the membrane was observed, possibly close to the main cavitation regions	[20]
120, 200, 400 W	27, 40, 200	Out situ	Alumina based ceramic membrane, Polymeric membrane	Nm	continues	1	Both polymeric and ceramic membranes affected by ultrasonic cavitation but ceramic membranes seemed to be more resilient. The larger the operating pressure and the higher the ultrasonic power, the likelier the membrane damage	[53]
2-8.68 kW/m^2	-	In situ	PVDF hollow fiber (UF)	360	continues	1	Likelier damage at higher ultrasonic power and sonication time	[166]
1, 1.5, 2, 2.5, 4.5 W/cm^2	40	Out situ	Aromatic polyamide (NF)	30	Nm	Nm	1 W/cm^2 found to be the optimum value of sonication power intensity which prevented damage	[46]

5. Hybrid cleaning

In some studies, ultrasonic irradiation was implemented in combination and to improve a conventional method of membrane cleaning, and/or for comparison. The general observations from the studies on hybrid cleaning are listed in Table 4. For example, the effect of electric and ultrasonic techniques on the flux recovery of a microfilter fouled by anatase and china clay was studied by Talerton [174]. The application of either ultrasonic or electric field provided some recovery of the previously lost flux, but different results were observed by implementing a combination of the two methods. In another study, Muthukumaran et al. [57] compared the cleaning efficiency of ultrasound irradiation alone, addition of sodium dodecyl sulfate (SDS), and of using SDS under ultrasound irradiation for polysulfone (PS) flat sheet ultrafiltration membranes. The surfactant was more effective than ultrasonic cleaning. More importantly, when the surfactant was used in combination with ultrasounds, the flux improvement was higher than when either methods were applied individually Popovic et al. [175] found that synergistic effect of chemical cleaning (NaOH) and a mixture of commercial detergents (P3-Ultrasil 67 and 69) and ultrasound-assisted cleaning removed the fouling layer from the membrane surface and allowed recovery up to 86.5 ± 4.9% of the initial flux after 30 minutes of sonication; this value increased to 96.3 ± 0.4% after a second sonicationThe cleaning efficiency was thus improved significantly by applying an ultrasound field in batch mode, combined with chemical agents [71]. In a study by Jin et al. [147], it was suggested that in cleaning the polluted PVDF ultrafiltration membrane, combination of flat plate transducer and 2 g/L citric acid aqueous solution resulted in the highest flux recovery of 81%.Water washing along with ultrasonic-assisted cleaning of polymeric membranes fouled by peptone particles was evaluated by Chai et al. [106], suggesting it as a very effective method to provide complete recovery of the flux in many cases. Because of the complexity of the synergetic effect of the two methods, a definitive hypothesis has not been presented. While the superiority of the simultaneous application of ultrasounds and another cleaning method is likely in most scenarios, the unexpected observations of the opposite effect in several circumstances suggest that some disturbance may occur. Clearly, this topic requires further optimization and case-specific cost analyses should be performed to determine the optimal combinations and designs.

Table 4. Summary of hybrid cleaning from the literature.

Combined method	Outcomes	Reference
Sonication+ electric field	Depending on the feed type, different results observed: Anatase suspension: Sonication improved the electric field effect on permeate flux. China clay suspension: Sonication weakened the electric field effect on permeate flux.	[81, 174]
Sonication+ water washing	The interaction of the two methods led to an excellent synergetic effect and cleaning efficiency increased significantly.	[106]
Sonication + surfactant injection in feed solution	Synergetic effect was observed and the permeate flux increased compared to the case of either cleaning method alone.	[173]

Sonication + surfactant injection in the feed solution	Combination of the two methods led to higher cleaning efficiency compared to the use surfactant alone.	[117]
Sonication + acid cleaning	Foulants were removed more effectively by ultrasound-assisted acid cleaning in comparison to simple acid cleaning. Additionally, by combining acid cleaning with ultrasounds, cleaning duration reduced by up to 2/3 and recovery of water flux reached 99.99%.	[46], [166]

6. Self-Cleaning piezoelectric membranes

Self-vibrating membranes with piezoelectric characteristics could also help solving the fouling problem. In the past few years, this technique has attracted more and more attention for its considerable benefits. Using this technique can significantly decrease the energy cost, increase the permeate productivity, and extend membrane life compared to other cleaning and antifouling methods [176]. In previous research studies on ultrasonic-assisted cleaning, the ultrasonic source (ultrasonic bath or probe) and the membrane were separate. As a consequence, the irradiated energy could not completely reach the fouled membrane surface and a significant amount of energy is thus wasted. Moreover, membrane damage was reported in many studies, which was generally a result of non-equivalent energy distribution on membrane surface, especially when ultrasonic probe/horn is utilized. Piezoelectric membrane technology may reduce the main and long-lasting challenges of the traditional ultrasonic technique in which the waves meet the membrane surface from an external source. A series of self-vibrating membranes with piezoelectric characteristics were introduced by Mao and his co-workers during the past two years [177-179]. Generally, the self-cleaning piezoelectric membranes were mounted between two flat electrodes (mostly copper) followed by application of a strong direct electric field that changes the ferro-electric domains from random to aligned (Fig. 7). The results indicated that the in-situ ultrasound emission can be very effective in avoiding fouling and the membrane flux can be maintained close to its initial value. However, more research is needed for optimizing the thickness of the layers, stability, to eliminate hazardous elements (e.g., use of lead-free materials), and to achieve application on other geometries and cost effective designs.

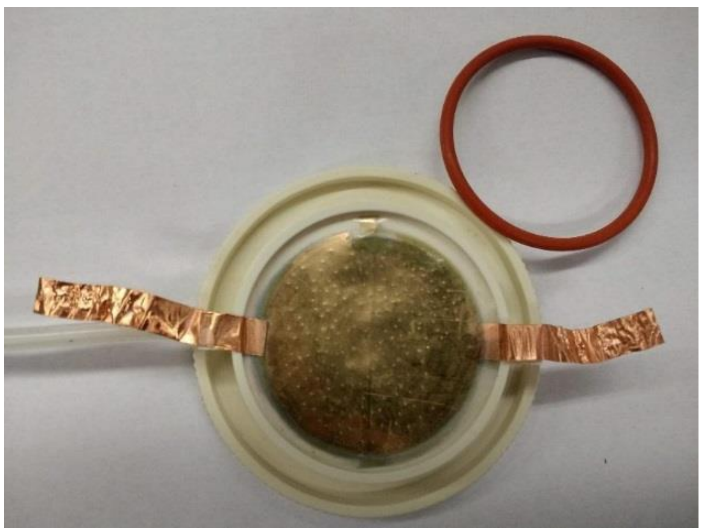

Fig. 7. Piezoelectric PVDF membrane sitting in the membrane cell. The membrane was sandwiched between two Au plates and the coating of each side was connected to a conductive copper tape respectively, [176].

7. General evaluation

7.1. Technology restrictions and utilization

Although the use of ultrasonic waves is a generally effective procedure for membrane antifouling applications, practical considerations should be made when it comes to the issue of its applicability. Membrane damage is the first and foremost major concern in the sono-treatment of fouled membranes. Extreme conditions, especially for low frequencies and high power designs could seriously damage the membrane surface, producing cracks, changes in structural integrity, and deformation of pores [102, 112, 180]. Moving from laboratory to industrial-scale application is another key challenge. Most of the research in this field has been conducted using small-scale setups under controlled conditions. The membrane dimensions and corresponding applied flow characteristics of some relevant research on ultrasound-assisted membrane fouling control are shown in Table 5. From careful evaluation of the literature, one of the conclusions is that small membrane samples were usually exposed to ultrasonic waves in an ultrasonic bath or using ultrasonic horns; on the other hand, at larger scale where the membranes, membrane modules, or vessels are larger, the majority of the membrane surface will not be placed near the source of the ultrasounds or inside a sonication cell. As a consequence, the upscaling of this technique needs technological improvements especially concerning the system design and the development of new and customized ultrasound transducers.

Table 5. Membrane area and input flow properties of ultrasonically cleaned membranes.

Membrane area (cm^2)	Input flow properties	Reference
20^{FS}	TMP: 1 MPa Cross velocity: 0.5 $m.s^{-1}$	[181]

198.4^{HF}	Feed velocity: 0.25 $m.s^{-1}$	[67]
120^{HF}	Feed velocity: 1.0 $m.s^{-1}$	[155]
6.5^{HF}	Membrane flux: 75&150 $L.m^{-2}.h^{-1}$	[156]
8.48^{SF}	Membrane flux: 75 $L.m^{-2}.h^{-1}$	[36]
96^{FS}	Feed flow rate: 325 $ml.min^{-1}$	[139]
39.6^{FS}	Nm	[116]
96^{FS}	Nm	[106]
69.39^{FS}	TMP: 34.5 kPa Cross flow rate: 500 $mL.min^{-1}$	[70]
30^{FS}	TMP: 55 kPa Cross flow rate: 600 $mL.min^{-1}$	[57]

FS: Flat Sheet, HF: Hallow Fiber, SF: Single Fiber

Despite the promising results of ultrasound-assisted cleaning of membranes at laboratory scale, the industrial feasibility of this method is not still addressed properly. Strictly considering the required power, the ultrasonic fouling control of membranes is not an energy-effective method. Further studies are necessary to estimate the energy balance at industrial scale. Furthermore, most of the lab-scale studies focused on a small range of membranes, generally flat sheet membranes, while the effectiveness of ultrasounds on other configurations have not been appropriately evaluated. Bridging the current gap between the laboratory and the industrial scale of this method still requires extensive studies on different systems and membranes, including pilot scale testing.

7.2. Process economics

As shown by several studies, the sonication of the feed solution before or during membrane filtration can effectively reduce fouling, or virtually eliminate it. However, the objective of reducing fouling is ultimately related to the minimization of process costs; if the cost of the ultrasound-based technique is too high, this will defeat the main purpose of its application. The economic assessment of ultrasonic cleaning of membranes has been performed only in few cases. The power consumption of a field-assisted crossflow microfiltration process was investigated by Tarleton and Wakeman [81]. Three types of polymeric membrane with different characteristics were applied to treat three different suspensions containing powders with different particle sizes (calcite, anatase, china clay). Observations indicated that both electric and ultrasonic fields, either used alone or simultaneously, mitigated fouling. Moreover, factors including applied field strengths, acoustic frequency, suspension concentration, liquid viscosity, particle size, and particle surface charge affected the fouling control process. Also, large flux increases were obtained when electric and/or ultrasonic fields were applied, but the energy consumed in achieving such flux values was equally significant. Consumed power by pumping requirements, as well as application of electric and ultrasonic field were measured to calculate the total power consumption. The results demonstrated that the energy required to produce a unit volume of permeate could decrease significantly for filtration of both anatase and china clay suspensions under some conditions. Specifically, the consumed energy could be reduced by 25-30% and 50-60 % in case the electric and ultrasonic field, respectively. Furthermore, the filtration time taken to extract a unit volume of filtrate reduced considerably as well. All and all, the results showed that applying a lower cross-flow rate and correspondingly achieving lower pumping costs was a necessary conditions to achieve a cost-effective filtration process when simultaneously implementing an ultrasonic and/or an electric field [81].

The feasibility study performed by Ahner et al. [153] suggested that their ultrasound-assisted ultrafiltration was cost-effective and economically-competitive compared to non-membrane processes for solvent recovery in de-asphalting operations (See Fig. 8). In particular, the ultrasonic process had the lowest capital costs, while the cost of energy was nearly equivalent to that of more conventional antifouling methods. Calculations suggested that ultrasound-assisted cleaning could reduce the total cost of the process by simplifying waste management and reducing additional pumping costs. Furthermore, the cost of maintenance, especially in the ultrasonic bath configuration, is potentially lower than that associated with other methods. Similar to any electric device, consumed energy in an ultrasonic apparatus directly depends on the supplied power and sonication time, so by decreasing these parameters the cost reduces almost linearly [182]. Accordingly, automation of the filtration process and other monitoring and feedback methods, such as pulsed mode sonication, could significantly reduce the cost [57].

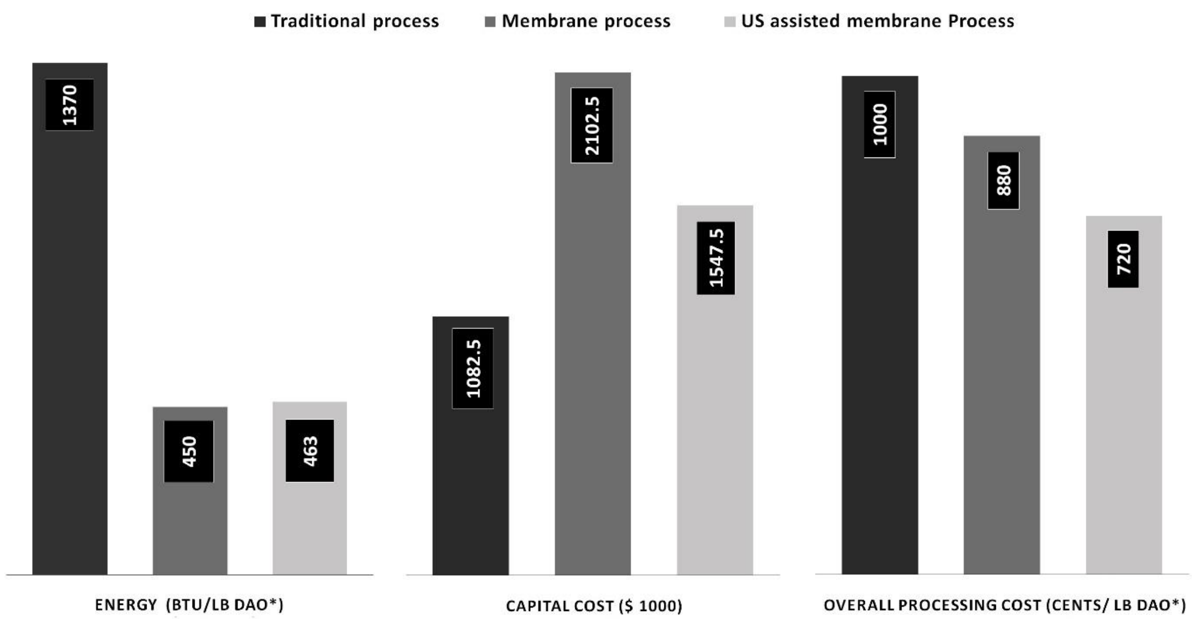

Fig. 8. Comparison of economic and energy advantages of piezoelectric membrane technology for solvent recovery in deasphalting operations [153].

8. Concluding remarks and future perspectives

The ultrasonic technique is an alternative method of membrane cleaning and applicable to enhance the permeate flux. These benefits stem from the phenomenon called ultrasonic cavitation and from secondary phenomena known as acoustic convective currents. The collapse of cavitation bubbles promote the formation of localized hot spots in a liquid medium with average bubble temperatures up to 5000 K and bubble core pressures of as much as 1000 atm. Cavitational mechanisms, such as microstreaming and shock waves, disrupt the interactions between foulants and the membrane, detach the fouling film from the surface of the membrane, and/or prevent the accumulation of microscopic particles at the membrane/solution interface.

Experimentally, the effectiveness of the ultrasound treatment is affected by several operational factors. By reducing the frequency, the size of the generated bubbles increases. Larger bubbles release more energy resulting in highly energized hot spots, while smaller bubbles (higher frequencies) produce less localized amount of energy with a larger number of hots spots but associated with smaller power. Therefore, there usually is an optimum frequency of operation. Large intensity values of the released energy are accessible by applying higher ultrasonic powers, thus leading to an increase in the effectiveness of the technique, provided that the cavitation intensity is not too high to cause problems of self-limitation. Also, risks of membrane damage and performance impairment should be considered before increasing the ultrasonic power.

The effect of hydraulic pressure during filtration is two-fold: higher pressures increase the cavitation threshold and hence, fewer bubbles form with higher localized energy. Moreover, at higher pressure, acoustic streaming intensifies and the consequent turbulence near the membrane surface can lead to increased shear stress at the membrane surface. On the other hand, an increase in the filtration pressure forces foulants closer to the membrane surface and possibly into its pores; it also causes a larger compressive force applied to the cake layer, producing a more densely packed fouling films, hence larger permeation resistances and difficulties in fouling layer destruction.

The effect of temperature is also two-fold. Firstly, increasing the temperature of the feed solution leads to improvements of the cleaning efficiency and flux due to enhanced diffusion, higher solubility, and higher Reynolds numbers. However, increasing the temperature decreases the cleaning efficiency of ultrasounds through its effect on ultrasonic cavitation. There is a relationship between the feed pH and the filtrate flux in the presence and in the absence of ultrasound, and hence, while not strictly related to the ultrasonic phenomena themselves, pH is also an essential factor related to the efficiency of sonication in membrane cleaning. The sonication mode, i.e., continuous mode or intermittent mode, has clearly significant effects on the cost of energy and on the enhancement of the permeate flux or recovery.

When considering the application of ultrasounds as a cleaning technique, one needs to ascertain that this method does not affect the integrity and the performance of the membrane. Some research reports highlighted that ultrasound treatment damaged the membrane surface, whereas other studies observed no significant erosion of the membrane. The issue of membrane damage is still not entirely understood, but clearly ultrasound irradiation time, frequency and intensity, as well as the nature and structure of the membrane material are all interrelated factors in this issue. Ultrasonic irradiation has been successfully coupled with some other treating methods to achieve a synergetic effect. Even in this topic, a definitive hypothesis of the effectiveness and optimization of the combined anti-fouling techniques has not been presented yet.

In this work, recent studies on the application of ultrasounds in membrane fouling control were reviewed and summarized, together with an evaluation of the advantages and disadvantages of this technique. The ultrasonically-induced effects consist of cavitation, microstreaming, microstreamers, microjets, shock waves, and heating. The effectiveness of this technique does not depend solely on the ultrasound parameters but closely on the nature of the membrane and foulants. Although most of the results indicate that applying ultrasound individually or in combination with other methods could enhance the overall permeation, these studies are almost always originated from laboratory scale investigations. Important obstacles are related to the efficient and cost-effective scale-up of this technology under real, and often complicated, industrial conditions. It follows that research investigations are required to evaluate the effectiveness of ultrasounds in full-scale membrane systems. Most studies have be conducted with the use of ultrasonic probes, baths, or horns which are likely to be ineffective in large-scale applications. Accordingly, the source of ultrasound is considered as a key challenge in the successful application of ultrasounds to control the fouling of full-scale membrane modules and hence, it is important to investigate novel ultrasound transducer technologies [72]. Since most studies in ultrasound-assisted cleaning of membranes have focused only on flat sheet membranes, investigations on different types of membrane modules present another key challenge for the successful application of ultrasounds in membrane fouling control [72].

While issues of membrane damage and cavitation control have not been yet overcome, innovative recent technologies, such as self-cleaning piezoelectric membranes, could overcome long-lasting challenges in this field. That being said, the ultrasonic technology has been developing rapidly in the recent decades, and new applications of this approach are constantly being proposed. Sustained efforts are necessary from scientists in different fields, from acousticians to biomedical engineers, which will aid the better design of proper ultrasonic apparatuses to overcome current limitations.

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