Improved ultrasonic cleaning of membranes with tandem frequency excitation

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A R T I C L E   I N F O
Article history:
Received 13 February 2012
Received in revised form 29 May 2012
Accepted 29 May 2012
Available online 5 June 2012
Keywords:
Ultrasound-cleaning
Membrane-fouling
High-speed video imaging

A B S T R A C T
In water purification membrane fouling is the most common problem that decreases efficiency in the purification process. In this work we propose a simple method to remove the particles deposited on the membrane surface by applying two different ultrasound frequencies in tandem. First a high ultrasound frequency, here 220 kHz, is used to create microscopic bubbles that are immediately after excited with a lower ultrasound frequency, e.g., 28 kHz. High speed video of the bare membrane shows the higher number of bubbles after the application of the high ultrasound frequency, as opposed to applying only a low ultrasound frequency. The method is then evaluated by recording the flux delivered and the transmembrane pressure (TMP) in this small scale experiment. The results show that after the application of the tandem frequency the transmembrane pressure is restored to the value of the non fouled membrane.

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1. Introduction

Membrane separation process is a widely used technology in areas such as food and dairy products processing, and water purification. Membrane fouling, one of the major problems encountered in membrane filtration, occurs by irreversible deposition of retained particles, colloids, macromolecules, salts, etc. Consequently, fouling causes significant decrease in permeate flux [1]. Currently, the most common methods used to clean membranes include backflushing/backwashing and chemical cleaning of membranes, but these methods have some drawbacks and limitations [2]. Though backflushing is used in both hollow fiber [3] as well as flat sheet membranes[4,5], it results in degradation of maximum flux after repeated backflushes. Chemical cleaning may damage the membrane and cause secondary pollution [6].

Ultrasound (US) is used for the cleaning of contaminated surfaces [7,8]. Various studies report the enhancement of permeate flux using US [9–11]. This enhancement is attributed to phenomena related to bubble oscillations, acoustic streaming, and heating [12–14] which increase flux by affecting the concentration polarization at the membrane’s surface [15].

The frequency of the US driving is an important parameter. For example the effect of permeate flux with US irradiation at three different frequencies of 28 kHz, 45 kHz and 100 kHz had been investigated by Kobayashi et al. [16]. They found that lower frequencies (here 28 kHz) was more effective in cleaning fouled membranes. Similar results were obtained by Lamminen et al. [14]. Maskooki et al. [17] tested three ultrasound frequencies as well as a combination of these both with and without a chemical cleaning agent (ethylenediaminetetraaceticacid, EDTA). Interestingly, they found the best cleaning performance when the frequency was alternated between 28 kHz, to 45 kHz, and 100 kHz. Kim et al. [18] have demonstrated that high frequency can be applied to remove particles from silicon wafers. By means of high speed images they show that the bubble oscillating close to their resonant size induce fluid motion that causes the detachment of the solid particles attached to the silicon wafer. More recently, Thiemann et al. [19] studied the bubble structure present in a 230 kHz ultrasonic field; their work is relevant because their studied ultrasound frequency is similar to the high frequency of this work. They captured high speed video and performed sololuminescence and surface cleaning tests; two groups of bubbles were investigated, bubbles larger than the linear resonant size and bubble near or slightly below the linear resonant size. They concluded that both bubble populations showed cleaning potential, although they acknowledged that the mechanisms associated with particle removal may be different.

It is commonly accepted that the large amplitude bubble oscillations from so-called cavitation bubbles are responsible for
the cleaning [14,18,19]. Even though the detailed mechanisms are not fully understood, experimental evidence points to the importance of the wall shear stress [20,21]. The wall normal velocity gradient is caused by radial and surface oscillations as well as fast jet-like flows.

Generally, cavitation bubbles grow from stabilized micron and sub-micron sized bubbles which may be stabilized by some shells or are trapped in crevices. These crevices are found on surfaces of the container or on particles suspended in the liquids. Also microbubbles are shed from strongly oscillating bubbles which serve as nuclei for later cavitation bubble growth. The complex flow pattern and acoustic field in ultrasonic cleaning devices makes it to impossible to predict the location of these "natural" cavitation bubble nuclei, nor to control them. Only nuclei close to the membrane contribute to the cleaning. Thus two strategies can be applied to improve the cleaning results, either increase the US power to create and drive more bubbles, or increase the number of nuclei close to the surface. Increasing the power not only increases the energy cost, but also may cause side-effects such as membrane damage [11,12] and a lesser control of the sound field due to nonlinearities. The second strategy of actively generating bubble nuclei near the membrane is more promising. In this work we report on a technique to populate the membrane with bubbles which allows to keep the US driving at low amplitudes. The cost evaluation of these two options could be compared in a similar way to that shown by Kang and Choo [22].

Membranes surfaces possess pores which may act as gas pockets and become the place of bubble nucleation. Bubbles in the liquid are attracted by an attractive force to the membrane boundary. Once on the surface these seed bubbles may grow in size by coalescence or by rectified gas diffusion [23]. Bubbles of similar size and driven in phase experience an attractive interaction force, the so-called secondary Bjerknes force [24]. This is valid for sufficiently mild oscillations; some of the details are discussed here [25]. The secondary Bjerknes force is also responsible for the attraction of bubbles from the bulk towards the membrane. This field force can be simply explained using potential flow theory. In potential flow the oscillation of a bubble at a distance s from a boundary can be constructed from the oscillation of two bubbles oscillating in phase at distance 2s using image theory [24]. Hence, bubbles become attracted towards the boundary. Thus, our novel method accumulate bubbles directly on the membrane before they are activated into cavitation bubbles; the accumulation and coalescence is induced with a high frequency ($f_h=220$ kHz) ultrasound. Then the driving sound field is quickly changed to a lower frequency ($f_l=28$ kHz). At this lower frequency these bubbles undergo much larger radial oscillations which then lead to cleaning of the membrane surface. We termed this technique tandem frequency excitation.

The objective of this study is to propose and test this method to clean and/or prevent fouling of microfiltration membranes.

### 2. Experimental setup

To evaluate the tandem frequency method we compare the bubble dynamics and resulting cleaning with that from a single frequency excitation. The water filtration membrane (hollow fiber membrane, 1.9 mm in diameter, 30 mm effective length, 0.035 μm average pore size, GE Corp.) was placed inside a basin filled with water. A focused piezoelectric transducer (80 mm diameter, 8.5 mm height) with a brass backing was placed below. The distance between the membrane and the focused transducer is 33 mm which is the distance where the largest acoustic pressure was measured with the PVDF hydrophones (RP Acoustics, Germany). The transducer was driven with a sine signal from a function generator (Model 33220A, Agilent, USA) connected to an amplifier (AG Series 1006, T&C Power Conversion Inc., USA). The transducer is operated at up to 136 V in the high frequency and at up to 370 V in the low frequency mode. For the tandem frequency driving a digital pulse/delay generator (Model 575, Berkeley Nucleonics, USA) was used to set the desired timing of the two frequencies.

The bubble activity near the membrane was recorded using a high speed camera (Photon SA1.1, Japan) at frame rates of up to 112,500 frames per second. Care was taken for a good illumination of the scene. The membrane was illuminated from the front by a metal halide light source (LS-M250, Sumita, Japan) coupled to a dual light guide; a mercury lamp (Olympus ILP-2) was used to illuminate the membrane from the back. Fig. 1A and B are a sketch of the experimental setup. The center of the membrane is the origin of the coordinate system, see Fig. 1B.

The filter cake of bentonite was deposited on the membrane in the following way: First, a clean membrane was immersed into a bentonite suspension; one end of the membrane was sealed while the other end was connected to a peristaltic pump operating at a flow rate of 6 ml per minute. A thick bentonite cake deposits onto the membrane surface within approximately 2 h. Next, the wetted membrane still connected to the operating pump is now submerged into a basin shown in Fig. 1A and filled with fresh tap water. The pump continues to run at the set pump rate of 6 ml/min.

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**Fig. 1.** Sketch of the experimental setup (A) and the geometry of the basin as seen from the camera position (B). The coordinate system is used in mapping of the pressure field, see Fig. 2. The plot in (C) sketches the timing of the two frequencies used in the tandem frequency driving.
Membrane cleaning typically utilizes ultrasound excitation at a single frequency, most common at the acoustic resonance of the specific transducer. The focused transducer has two resonances, one at 28 kHz and a second at 220 kHz. For the tandem frequency (TF) method, the membrane was sonicated with a high frequency ($f_h = 220$ kHz) for 8 s and then immediately the driving frequency was changed to the low frequency ($f_l = 28$ kHz) for 2 s, see Fig. 1C. The TF method is compared to a single frequency driving by sonication of the membrane for 2 s of the low frequency ($f_l = 28$ kHz) only.

To document the acoustic parameters of the sound field, we mapped the pressure in the area around the membrane with PVDF hydrophones. For the low frequency a hydrophone with 3.8 mm tip diameter and for the high frequency a hydrophone with a thinner tip of 1.4 mm in diameter is used (PVDF hydrophones, RP Acoustics, Germany, type p and l, respectively). The pressure measurement was done in the absence of the membrane. The region sampled has the same size as the region captured by the camera’s field of view in the tests of low frame rate, the tests.

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**Fig. 2.** (A) and (B): Typical waveforms at the origin showing at maximum driving for the high and low frequency driving, respectively. (C) and (D): Pressure map showing the amplitude distribution in the region of the membrane for the driving frequency of 220 kHz and 28 kHz, respectively. The ‘+’ mark on (C) and (D) denotes the origin and center of the membrane, see Fig. 1(B). Note that (C) and (D) are recorded at a reduced driving voltage, therefore the pressure values are lower than in (A) and (B).

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**Fig. 3.** Upper row: frames taken from high speed recording of bubbles grown on the bare membrane. Time zero (t = 0) is set as the time when the acoustic driving is switched from high to low frequency. Hence, frames in the first column are in the high frequency regime while frames in the second and third columns are in the low frequency regime. Original frames from high speed recording. Lower row: frames subtracted with a background frame to enhance the contrast.
with the largest viewing area. The typical pressure values and waveforms for the actual maximum amplitude used are depicted in Fig. 2A and B. Here, only a few cycles of the strong ultrasound is applied to the hydrophone. For obtaining a map of the sound field (see Fig. 2C and D) the transducer was run at a lower voltage. Here, a purely sinusoidal pressure was measured. It is evident that the pressure map shows distinct differences for the two frequencies used. For the high frequency driving the pressure field has a strong maximum near the origin (Fig. 2C), while for low frequency a more homogenous pressure distribution is found. This difference may be caused by the focusing property of the transducer operated at high frequencies while for low frequencies a standing field is generated. The structures in the sound field should be of the order of half the wavelength, \( \lambda/2 = 27 \text{ mm} \). The high frequency driving will create a standing wave, too, which is overlaid onto the focused field. Fig. 2A and B: Typical waveforms at the origin showing at maximum driving for the high and low frequency driving, respectively. C and D: Pressure map showing the amplitude distribution in the region of the membrane for the driving frequency of 220 kHz and 28 kHz, respectively. The “+” mark on (C) and (D) denotes the origin and center of the membrane, see Fig. 1B. Note that (C) and (D) are recorded at a reduced driving voltage, therefore the pressure values are lower than in (A) and (B).

3. Results

3.1. Bare membrane

At first we demonstrate that the high frequency driving is able to nucleate bubbles on the surface of a bare membrane. Unfortunately, the resulting size of most of the gaseous bubbles are below the spatial resolution of the camera, which is about 13 \( \mu \text{m/pixel} \). They only become visible once the low frequency driving is switched on, and they expand multiple times their rest radii. Fig. 3 depicts in the top row the bare membrane in the first frame after approx. 8 s at low frequency driving, just before the low frequency is switched on at \( t=0 \). Two round bubbles are visible on the upper part of the left frame (indicated with circles in Fig. 3). At time \( t=0 \) the low frequency excitation is started (the high frequency is switched off). We see within less than 100 \( \mu \text{s} \) large bubbles in the recording. The number of visible bubbles is counted manually as a function of time. For the bubble detection the image contrast is enhanced by subtracting a background image from each image. The background subtracted frames are depicted in the lower row of Fig. 3. The relative high framing rate of 112,500 frames/s makes it possible to visualize the bubble oscillation within a period at \( \text{fl} = 28 \text{ kHz} \). Fig. 4 presents the number of bubbles as a function of time for different driving voltage amplitudes of the high frequency. For the highest voltage of 136 V \( \text{p-p} \) we find already after 2 cycles of the ultrasound (period of oscillation \( \text{T} = 36 \mu \text{s} \)) many bubbles. The number of bubbles oscillates with time and increases. For voltages down to 48 V \( \text{p-p} \) (which...
corresponds to 1.4 bars) of the high frequency we observe similar behavior, yet the bubble appearance is retarded and their number is less. For 44 \(V_{p-p}\) and below no bubbles can be found. The oscillation of the number of bubbles is due to the limited image resolution. Thus when bubbles shrink below approx 13 \(\mu m\) in size they cannot be detected. Therefore, Fig. 4 relates indirectly to the bubble size as a function of time.

Fig. 4 demonstrates that for lower acoustic pressures (lower voltage amplitudes) it takes more time for bubbles to appear on the membrane surface; also the number of bubbles detected is lower. If the acoustic pressure falls below 1.4 bars no more bubbles are created on the membrane that can be used as seeds for cleaning during the application of the low frequency. On the other hand, increasing the ultrasound power too much may lead to membrane damage [26]. Yet, we have not observed ultrasound induced membrane damage in all measurements, e.g., we find a recovery of the trans membrane pressure (see below) and by visual inspection no bentonite particles in the filtrate.

### 3.2. Membrane with filter cake

Now we show and compare the membrane cleaning using a single frequency (SF) ultrasound with the tandem frequency (TF) method, Fig. 5. Fig. 5A (SF) and B (TF) show each a freshly prepared membrane with a bentonite filter cake. Fig. 5A depicts the cleaning for the first second of acoustic exposure showing very little bubble activity, hardly any activity is noticeable in the direct picture. Yet in the lower row of Fig. 5A the background subtracted frames reveal some bubbles, circled in Fig. 5A. Also fragments of the filter cake are ejected as visible in the upper left corner of the last frame.

We contrast this finding with the tandem method where the membrane is first exposed to 8 s of high frequency (driving voltage 136 \(V_{p-p}\)). The first frame of Fig. 5A \(t=240 \text{ ms}\) depicts a very small bubble weakly visible in the background subtracted image. This bubble is probably only mildly oscillating as no filter cake ejection is observed till then. However, as the frequency is reduced at \(t=0\), a cluster of bubbles emanates. Also pronounced ejection of the filter cake from the membrane surface is found. This originates from the bubble indicated with the arrow \((t=32 \text{ ms})\). Again, this bubble develops into a cluster of bubble populating the upper side of the membrane and leading to the ejection of the filter cake.

We quantify the number of bubbles found for the TF method vs. the single frequency excitation. Fig. 6 demonstrates that within 100 \(\mu m\) more than 40 bubbles appear whereas it takes more than 0.5 ms for the first bubbles to appear using the low frequency only. The total number of bubbles is in order of magnitude larger for the TF method. As already visible in Fig. 5, the bubble dynamics leads to ejection of filter cake material into the bulk liquid. We now study the progress of membrane cleaning using the tandem and the single frequency excitation.

We now apply the protocol for the US multiple times and follow the removal of the filter cake. Fig. 7 depicts in the left column the SF and in the right column the TF method. For the SF excitation 2 s is waited between runs while for the TF method the high/low frequency is applied consecutively. For the TF method after nine runs of the protocol large bright patches become visible indicating removal of the filter cake. In contrast, the SF treated membrane (left column in Fig. 7) is still fully covered with the filter cake. From frame 6 (seventh frame from the top, right column), pronounced ejection of debris into the bulk becomes visible.

We find much more bubble activity if we apply a high-frequency field prior to the cleaning with a low frequency US field. High-speed video reveal that the formation of bubbles close to the membrane surface is enhanced. Some more detail of this process is visible in Fig. 8. This recordings show the bubble dynamics on the membrane together with the filter cake removal. In the rightmost frame of Fig. 8 bentonite particles are ejected in a round plume originating from a single position. Likely a single bubble or dense cluster of bubbles grows between the membrane and the cake, once the low frequency is switched on particles become ejected from this position, visible after 4.5 ms (see arrow in the last frame of Fig. 8). We notice also that the bubble location is slightly moving to the right, it is translating as well as oscillating.

Besides the usual bubble random translation for the cleaning of membrane surface, we also noticed that bubble may translate along a straight path on top of the membrane, as can be seen in Fig. 9. While translating, the bubble ejects materials from the cake. The low frame rate used (250 fps) does not allow unambiguous measurement of the velocity in this measurement. However, we found that the lower bound of the velocity of the ejected materials is approx. 0.125 m/s. The frames of Fig. 9 have been subtracted with a background frame to enhance the bubble’s motion and the particles removed by it.

### 3.3. Tandem frequency validation experiments

Of the two methods tested, only the tandem frequency is able to remove the accumulated bentonite particles on the membrane, the single frequency method does not. We remove some of the cake but a large portion remains attached to the surface of the membrane. Besides this visual verification of the proposed membrane cleaning technique we tested the membrane cleaning performance by registering the trans membrane pressure (TMP) and the flux through the membrane.

Fig. 7. Comparison of the membrane surface covered initially with a bentonite filter cake before (#0) and after 1–9 cycles (#01–09) of single frequency (SF) and tandem frequency method (TF) on the left and right column, respectively. Both videos were captured at 250 fps.

Our small scale experiment was setup and conducted as follows: First the membrane is immersed in a small beaker that contains a water and bentonite solution (12 g/l) to form a cake for 25 min, while the pressure at the exit of the membrane is recorded with a Wilka pressure sensor (−100 kPa to 100 kPa range).
and the weight of the permeate is measured in an Ohaus scale (model PA413C, read out with a computer interface). When the cake is formed the peristaltic pump is turned off and the membrane is transferred to the container previously described that houses the focused transducer. Because of the rather small focal zone of the transducer we placed the membrane in clean water to make sure it is located at the right position. The time it takes to transfer the membrane from the small container with the water bentonite mixture to the container housing the focused ultrasound is less than 60 s; we do not expect a softening of the integrity of the cake from the time the pump is turned off to the time when the ultrasound is applied.

In the test shown in Fig. 10a about 35 cycles of TF excitation were necessary to clean the membrane. Once the membrane is cleaned it is placed again in the small beaker with bentonite and the pump is turned on again.

Fig. 10 shows the results of tests conducted in a hollow pressure membrane at constant flux. Fig. 10a depicts the change of TMP as a function of time. The “pure water” curve is a control for the TMP: no change of the TMP is observed. The “No US applied” curve represent the pressure obtained when the membrane is kept in the water-bentonite solution and no ultrasound is applied; this curve shows that the TMP increases with time. These two curves are used as a benchmark against which the tandem frequency is compared. The “Tandem Freq applied” curve depicts the TMP obtained with the application of ultrasound to remove the filter cake. When the test begins the TMP increases just as it does in the “No US applied” curve, then when the pump is turned off the TMP drops to zero. After the membrane is cleaned and the pump is turned on again the TMP reduces to approximately the same value it had at the beginning of the test, i.e., the value for the pure water case. Even after repeatedly fouling and cleaning of the membrane the minimum TMP returns to the control value, i.e., no residual fouling is observed [3].

We now analyze the mass flux. Fig. 10a depicts four regions labeled a–d and we compare the flux for the three cases, pure water, water–bentonite, and TF excitation. Fig. 10b depicts the weight delivered during the time the tests are conducted. The flux delivered in the pure water case is practically constant. The “No US applied” curve follows the pure water curve closely at the beginning of the test but its slope decreases and after 90 min it is already noticeable that the flux is lower than that of the pure water test. Finally, the slopes of the TF curve in regions a–d are very similar to the slope of the “pure water” curve, the three flat sections shown in the “tandem frequency curve” correspond to the time when the pump was turned off thus no liquid was delivered.

Fig. 10. The transmembrane pressure TMP and the weight of liquid delivered for the three conditions tested. (A) The TMP as a function of time. Regions a–d correspond to the pump-on time during the tandem frequency tests. and (B) The weight delivered by the low pressure system for the three conditions tested, from the weight information the average flux obtained in regions a–d was obtained.
The TF method was tested on a hollow membrane yet we find that the use of two ultrasound frequencies is more efficient in removing particular matter deposited on a circular membrane. We attribute the better cleaning performance of the TF method to a larger number of bubbles created on the surface of the membrane prior to the application of a lower frequency which induces cavitation. These low frequency driven cavitation bubbles acts as scrubbers that perform the cleaning of the membrane. The technique may be generally applied to seed the surface to be cleaned before the high amplitude/low frequency US is applied. This splitting of the seeding of the surface from cleaning could lead to less damaging protocols in US cleaning. Still, the details of the enhanced bubble formation at high frequency need to be elucidated. The proposed technique was applied in a small scale experiment that showed that the flux obtained can be constant thus it can be implemented as a mechanism to de-foul membranes in low pressure ultrafiltration applications.

Other issues such as the quality/type of the filter cake that can be removed with the present technique, long term effects of ultrasound on the membranes, and the energy balance need to be addressed. Nevertheless, we hope this work stimulates others to have a fresh look at US cleaning through the deeper knowledge of bubble dynamics and its correlation to the cleaning process.

Acknowledgments

We greatly acknowledge the help we received from the Singapore Membrane Technology Center (SMTC). In particular we thank Filicia Wicaksana and Anthony Fane. This research grant (MEWR C651/06/176) is supported by the Singapore National Research Foundation under its Environmental & Water Technologies Strategic Research Programme and administered by the Environment & Water Industry Programme Office (EWI) of the PUB.

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