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Potential uses of ultrasound in the dairy ultrafiltration processes

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There has been a growing interest in the industrial application of ultrasound, especially in the food industry. Power ultrasound can have a number of physical effects; it can increase turbulence through both the introduction of vibrational energy and through acoustic streaming, it can cause both particle agglomeration and particle dispersion and clean surfaces with a scouring action. Our work in this area has focused on the use of ultrasound to enhance membrane processing. Low frequency ultrasound has been used to facilitate cross flow ultrafiltration of dairy whey solutions for both during the ultrafiltration production cycle and the cleaning cycle. During the production cycle, the use of ultrasound reduces both pore blockage and the specific resistance of the fouling cake layer. This leads to higher flux rates and the potential for longer production cycles. During the cleaning cycle, ultrasound systematically increases cleaning efficiency, thus has the potential to reduce both total chemical consumption and system downtime. There was no deterioration in cleaning effectiveness or membrane condition, which implies that sonication, has not damaged the membrane itself. Similarly, there was no change in the chemical nature of soluble proteins following sonication.
Key words: Ultrasound; Spacers, Whey proteins; Ultrafiltration; Flux; Fouling; Cleaning

1 Introduction

Membrane ultrafiltration (UF) provides an extremely attractive technique for whey processing which can fractionate the whey components, thus enhancing their utilization and reducing the pollution problem. One of the critical issues in the development of effective whey ultrafiltration processes is the decline in system performance due to both concentration polarization and membrane fouling. Frequent fouling and subsequent cleaning of dairy whey ultrafiltration membrane significantly affects the economics of such processes. Fouling results in a significant reduction in the separation efficiency and increase the costs of membrane replacement by decreasing the life span of the membrane. Similarly, cleaning of the fouled membrane requires expensive chemicals and significant downtime and physical cleaning methods interrupt the continuous filtration process leading to a longer processing time. The application of ultrasound (US) has been studied as an alternate technology for enhancing permeation in membrane separation processes. Ultrasound can be used either during the operational cycle to enhance the permeation or as a cleaning technique to improve the cleaning efficiency.

The use of ultrasound to assist membrane filtration and reduce fouling has been studied in both cross-flow systems [1,2,3,4,5,6] as well as in dead end filtration [7,8], often in combination with chemical and or water cleaning [9]. Our studies [10,11,12,13,14,15] revealed that that the use of low frequency (50 kHz) ultrasound at low power densities enhances whey ultrafiltration and the cleaning of whey fouled membranes. This paper is an extension of our previous work and provides insight into ultrasonic mechanisms that has the potential to enhance the economics of whey ultrafiltration processes.

2 Experimental Setup and Procedure

The same experimental set up as described in our previous reports [11,12,13] has been used for this study. A single 30 cm^2 polymeric UF membranes membrane sheet of 30000 MWCO was sandwiched with a spacer of 1.3 mm thickness on the feed side of a cross flow Minitan S unit (Millipore Inc). The unit was completely immersed in a 50 kHz ultrasonic bath, which was switched on as required. All experiments used re-constituted spray-dried whey powder to foul the membrane.

Different stages of filtration experiments are shown in Fig.1(a). Initially the pure water permeate flux J_{wi} was measured and this value was used to obtain the clean membrane resistance R_m using the well known equation:

$$J = \frac{\Delta P}{\mu R} \tag{1}$$

where ΔP is the transmembrane pressure, μ is the viscosity of the permeate solution and R is the resistance to solvent permeation. Subsequently, the membrane was fouled for 4 hours with freshly prepared 6% w/w whey solution under different conditions. The steady state permeate flux J_f was determined by averaging the last 10 recorded values of permeate mass. The total fouling resistance R_{total} was calculated from this steady state value, again using Eq.(1). Milli-Q water/distilled water was then fed through the ultrafiltration unit and the final water flux J_{wr} was measured. This rinsing is intended to remove the reversible fouling resistance R_b, that results from both concentration polarization and labile surface deposits, leaving the more tenacious deposits. The membrane was finally cleaned using conventional alkali cleaning cycles and Milli-Q water/distilled water was fed into the unit to flush out the cleaning solution. Final permeate flux J_{fc} recorded and compared to the initial water flux J_{wi}. The individual resistances were determined using the resistance in series model,

$$R_{total} = R_m + R_b + R_f \tag{2}$$

Also Ho & Zydney [16] model was used to describe the flux decay as a function of time when both pore blockage and cake filtration mechanisms are active. This model requires the simultaneous solution of two equations:

$$\frac{J}{J_0} = \exp\left(-\frac{\alpha \Delta P C_b}{\mu R_m} t\right) + \frac{R_m}{R_m + R_p}\left[1 - \exp\left(\frac{\alpha \Delta P C_b}{\mu R_m} t\right)\right] \tag{3}$$

$$R_p = \left(R_m + R_{po}\right)\sqrt{1 + \frac{2 f' R' \Delta P C_b t}{\mu \left(R_m + R_{po}\right)^2}} - R_m \tag{4}$$

The model contains three adjustable parameters i.e., the pore blockage factor α which describes the extent of blocked pores, the initial resistance of the cake deposit, R_{po} and the cake growth factor ($f' R'$) which is the product of the fractional amount of protein present that contributes

to deposit growth and the specific protein layer resistance. The best-fit values of these parameters were determined by minimizing the sum of the squared residuals between the experimental filtrate flux data and the model calculations.

Different stages of cleaning cycle experiments are shown in Fig.1(b). After fouling, the membrane was rinsed with water for ten minutes to remove labile surface deposits. The permeate rate of water J_{wr} was recorded and used to calculate irreversible fouling hydraulic resistance R_r. The rinsed membrane was cleaned for ten minutes using enzyme (Ultrasil 56), but respectively with and without the use of ultrasound. The membrane resistance after cleaning R_c was calculated at the end of this step by recording permeate rate of water J_{wc} again. The membrane was finally cleaned using conventional alkali cleaning cycles in order to return the initial cleaning resistance R_m. The cleaning efficiency (CE) has been used as the criterion to assess the cleaning process, which is defined as [17],

$$CE = \frac{R_r - R_c}{R_r - R_m} \times 100 \qquad (5)$$

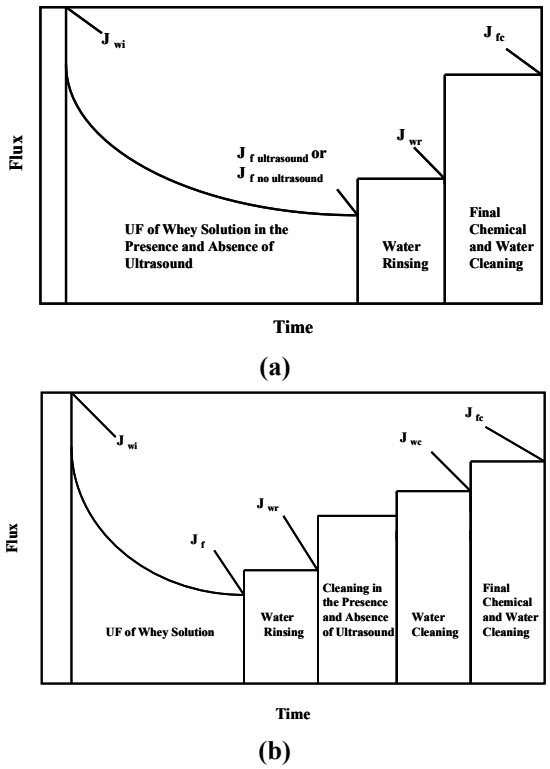

Fig.1 A schematic diagram showing different stages of filtration cycle (a) and cleaning cycle (b).

3 Results and discussions

3.1 Influence of TMP and feed spacers

Permeate flux declines at high transmembrane pressure (TMP) and this trend is attributed to the higher convective mass of particles toward the membrane surface at higher pressures (Fig.2(a)). As the TMP increases, the compressive force exerted on the cake layer favour a thicker and more densely packed cake layer which leads to lower permeate fluxes. The result shows that both spacers and ultrasound can improve the permeate flux. The spacer is ineffective at high TMP but results in a consistently higher permeate flux when this TMP is decreased. Both the particle deposition behaviour on the membrane surface and cavitational properties of ultrasound are affected by the TMP. An ultrasonic enhancement factor is defined as the ratio of ultrafiltration fluxes in the presence and absence of ultrasound shows that ultrasound is effective under all TMPs with an enhancement factor increases slightly as the TMP increases (Fig.2(b)). This suggests that the ultrasound operates through increasing acoustic streaming and mechanical vibration rather than through bubble cavitation.

The irreversible fouling resistance provided in the presence and absence of ultrasound are comparable (Fig.3(a)), indicating that the ultrasound is relatively ineffective in reducing the extent of both the tightly bound deposits within the fouling cake and any pore blockage.

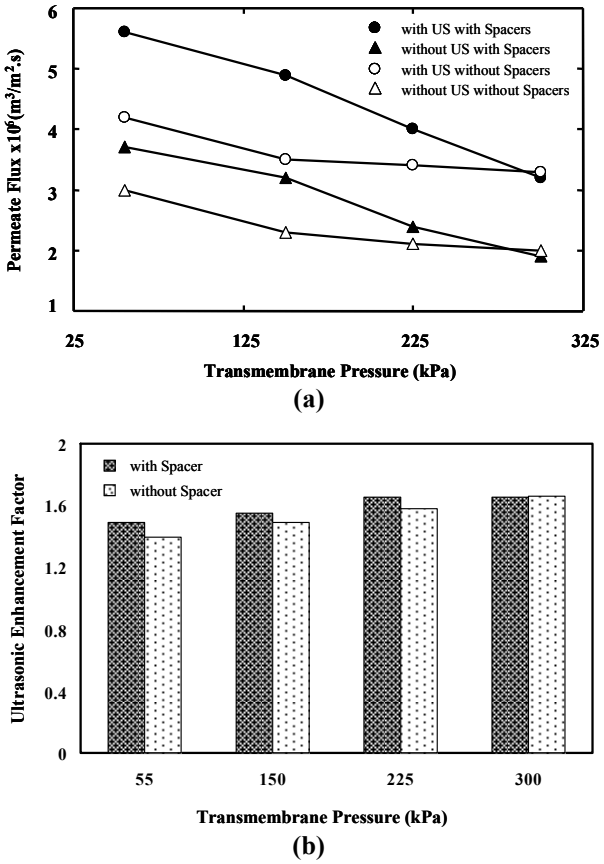

Fig.2 Permeate flux of whey solution after 4 hr of run at variable TMP (a), ultrasonic enhancement factor values as a function of TMP (b) [CFR 550 ml/min, C_{whey} = 6 wt % T = 20^0 C, and nominal ultrasonic power = 300 W].

The reversible fouling resistance increases with TMP and is clearly lower when ultrasound is applied, and this is more significant at higher TMPs (Fig.3(b)). This result again tends to indicate that ultrasonic enhancement does not occur through cavitation. It also suggests that ultrasound works mainly by either reducing the resistance in the looser and more labile cake and/or reducing concentration polarization by increasing turbulence.

Similarly, the results were analyzed using the theoretical model developed by Ho and Zydney [16] (Fig.4). The pore

blockage parameter α is essentially independent of TMP. Pore blockage is reduced slightly when ultrasound is employed. The best fit values of both $f'R'$ and R_{po} increase with increasing TMP, which is due to the compressibility of the whey protein deposit. However these fouling parameters are always lower when ultrasound is used. The effects are most pronounced in the cake growth factor where the ultrasound causes this parameter to fall by a factor of 5.

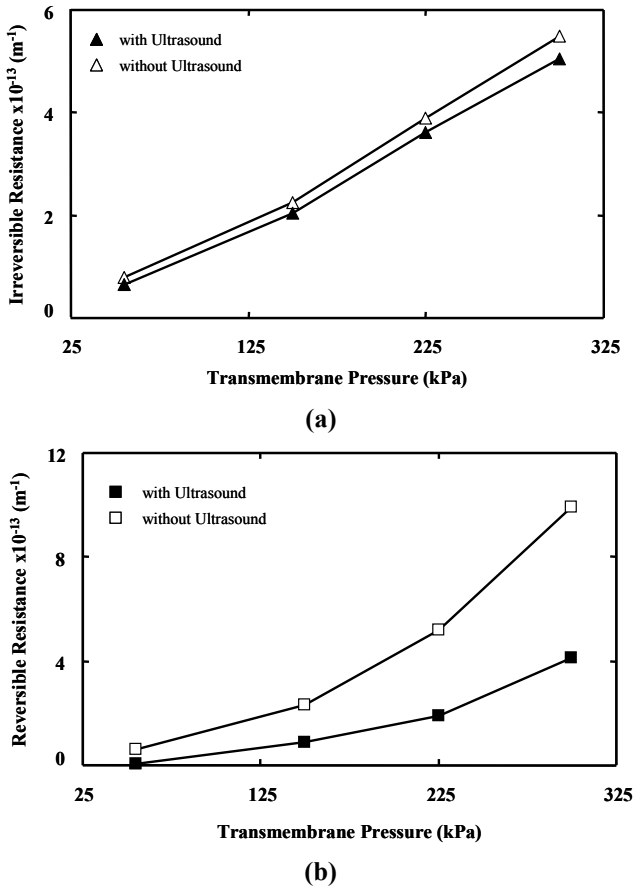

Fig.3 Irreversible resistance (a) and reversible resistance (b) as a function of TMP in the presence of spacers.

This result suggests that the ultrasound acts by 'loosening' (less compressed) the cake and reducing its compressibility. This is because sonication causes agglomeration of fine particles thus reducing cake compaction and the turbulence associated with ultrasound which is used to separate physical aggregates of protein molecules by disrupting the intermolecular forces.

3.2 Influence of ultrasound on enzyme cleaning

Our earlier investigations have shown that the concurrent use of ultrasound enhances the cleaning of whey-fouled UF membranes under all experimental conditions [11,13]. The optimal experimental parameters for effective ultrasonic cleaning are high temperatures, low transmembrane pressures, a solution pH of 12 and a surfactant concentration close to that of the critical micelle concentration.

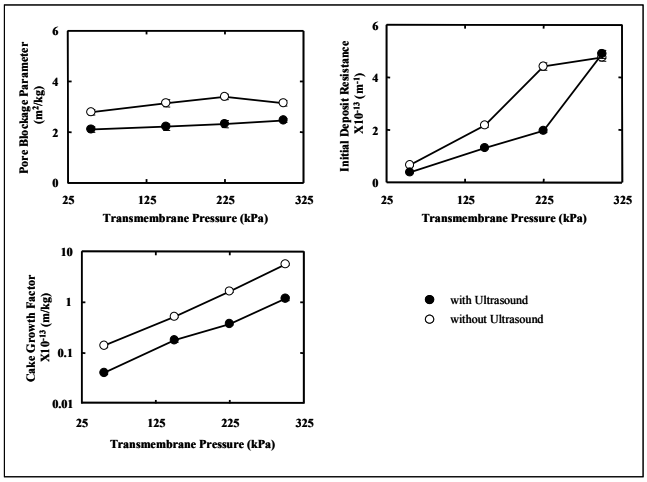

Fig.4 Best-fit values of the fouling parameters as a function of TMP in the presence of spacers.

Experiments were carried out with a range of enzyme (Ultrasil 56) concentrations, which is combination of detergent and enzyme to determine the effect of ultrasound on the cleaning efficiency. Parkin [18] shown that the effect of temperature on the detergent containing enzyme and found a temperature of greater than 50^0 C is required for maximum activity. Hence, the cleaning experiments alone were carried out at 50^0 C. All water fluxes were measured at 55 kPa and 550 ml/min and at 20^0 C as per normal experimental procedure.

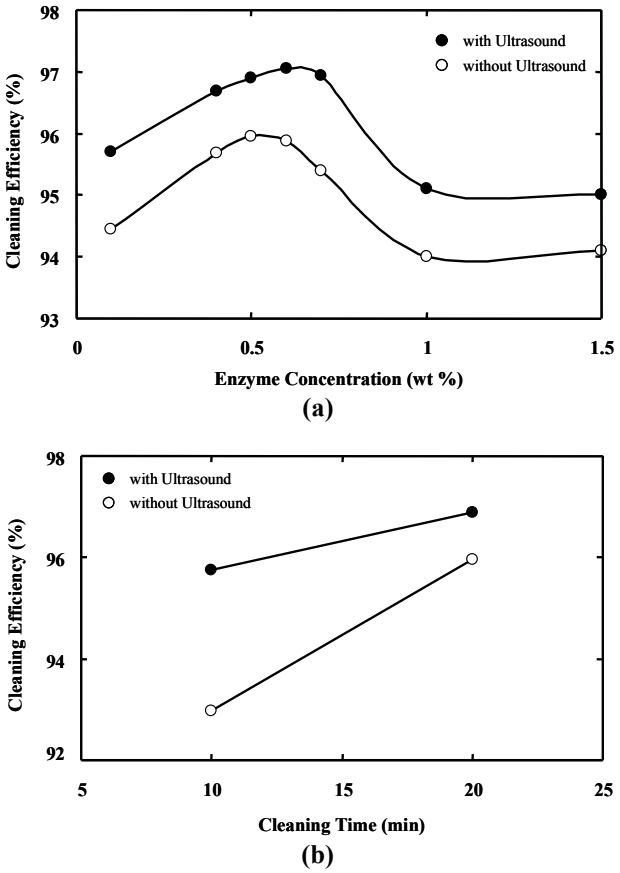

Fig.5 (a) Effect of enzyme concentration and ultrasound (b) effect of cleaning time on the cleaning efficiency.

From the economic point of view the determination of the optimum concentration of enzyme required to clean the

membrane is very important. Fig.5(a) shows that the cleaning efficiency increases with increasing enzyme concentration up to 0.6 wt%. Beyond this concentration, the excess enzyme could possibly contribute to fouling of the membrane, thus decreasing cleaning efficiency consistent with other studies [19,20].

The cleaning efficiency is higher when enzymatic formulations were used in combination with ultrasound. In order to find whether shorter cleaning time with ultrasound favors the cleaning efficiency, selected experiments were carried out at same enzyme concentration but with the cleaning time reduced to 10 min. The result in Fig.5(b) again suggests that the ultrasonic influence is significant when cleaning is carried out with 10 minutes. The cleaning efficiency with 10 min of ultrasound is equivalent to that of 20 min without ultrasound. Thus use of ultrasound could reduce the enzyme cleaning time required.

3.3 Effect of ultrasound on membrane life and on dairy solutions

No damage was observed to the experimental membranes used in this work following many hours of ultrasonic exposure. This is qualitatively supported by consistent values of the water flux through cleaned membranes over many weeks of experimentation. Field emission scanning electron microscope (FESEM) images of membrane surfaces further support the fact that the membrane was not damaged during sonication (Fig. 6).

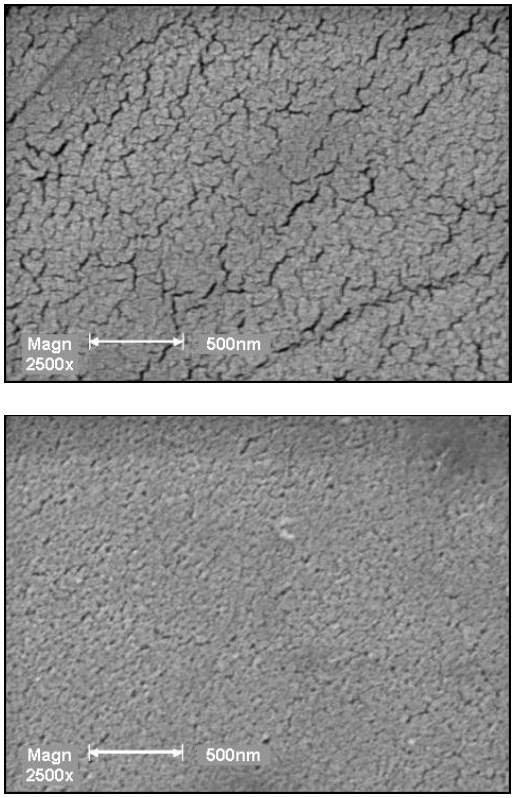

Fig.6 Surface view of unused (top) and sonicated (bottom) PS30000 MWCO membrane (x 2500 magnification)

The size of the surface pores appears slightly smaller in the sonicated membrane, which may be due to either irreversible pore blockage or swelling of the membrane material itself. However, there is no structural difference between the membranes indicating that the intrinsic permeation properties of the membranes are not modified by the exposure to low frequency ultrasound. This is consistent with other studies [6,8].

In an alternate series of experiments, the membrane holder was removed from the bath and a beaker containing 6 wt% of whey solution was added in its place. This beaker was exposed to ultrasound for up to 4 hours. Analysis of the soluble protein content of the resulting solutions using high performance liquid chromatography (HPLC) showed identical concentration profiles in all samples (Fig.7). This is consistent with the findings of other workers [21,22].

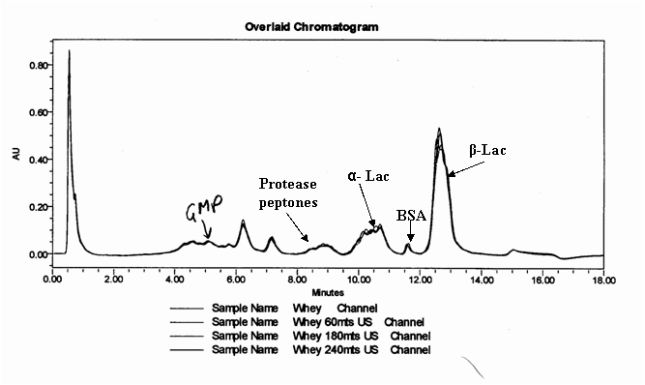

Fig.7 HPLC elution profile from whey protein in the presence and absence of ultrasound [C_{whey} = 6 wt%, T = 20 ± 2^{o}C, ultrasonic frequency = 50 kHz and nominal ultrasonic power = 300 W].

4 Conclusion

Experimental results reveal that the combined effect of spacers and ultrasound can thus lead to a doubling of permeate flux. The main mechanisms involved in flux enhancement are thought to arise from increased acoustic streaming and mechanical vibration. However, the influence of acoustic cavitation can not be completely excluded. The ultrasonic irradiation acts to reduce the resistance of both the initial protein deposit and the growing cake, reducing the compressibility of these deposits. Further, the use of enzymes in combination with the ultrasound had a synergistic effect, leading to a substantial improvement in the flux recovery. Electron microscopy results showed no evidence that the ultrasonic irradiation altered the membrane integrity. HPLC analysis of the whey proteins in the feed solution before and after sonication showed that the sonication process did also not affect the concentration profile of the whey proteins.

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