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The use of ultrasonic feed pre-treatment to reduce membrane fouling in whey ultrafiltration

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ABSTRACT

The production of whey protein concentrate powders is often limited by the fouling of the ultrafiltration membranes and the low heat stability of the whey protein solutions. Ultrasonic treatment of whey solutions has previously been shown to break down protein aggregates and improve heat stability. This study investigates the use of ultrasound as a pre-treatment step to improve downstream ultrafiltration performance. Results show that sonication alone alleviated membrane fouling to a small extent. However, the use of ultrasound following heat exposure reduced membrane pore blockage and growth of the foulant cake greatly, relative to heat exposure in the absence of ultrasound. The extent of changes to pore blockage and cake growth was greater at higher solids concentration. In all cases, the protein concentration in the permeate remained unchanged. This work has the potential to reduce energy requirements in the ultrafiltration of whey as feed pre-treatment by both ultrasound and the combination of heat and ultrasound produced a lower viscosity feed solution.

1. Introduction

The dairy industry relies heavily on membrane ultrafiltration (UF) for the concentration of whey, a by-product of cheese-making. Downstream, this concentrated whey is usually evaporated and spray dried to produce whey protein concentrate (WPC) powders of varying protein content ranging from 35% to 80%. However, membrane fouling, which is a build-up of particles on the membrane surface and within its pores, reduces ultrafiltration performance, resulting in a sharp reduction in a permeate flux and an increase in pressure drop across the membrane during filtration. Costly cleaning cycles and, in some cases, replacement of the membrane modules are required to restore the original flux, limiting the economic efficiency of the ultrafiltration operation.

Several modifications have been proposed to enhance membrane performance and reduce membrane fouling. Feed pre-treatment, installation of turbulence promoters and ultrasonic enhancements are examples of such modifications. In feed pre-treatment, the feed solution is treated using heat, pH adjustment, addition of complexing agents, precipitation or pre-microfiltration. This alteration in feed properties stabilises or removes foulants upstream of filtration. Hickey and co-workers [1] observed an increase in the permeate flux when the feed temperature and pH were increased prior to filtration. This was the result of the removal of calcium phosphate crystals, which would otherwise precipitate in the membrane pores, upon heating at higher pH. However, a decrease in protein retention made this method infeasible [2].

The addition of turbulence promoters, such as vibratory shear-enhanced filtration and rotating disk modules, in a filtration unit enhances turbulence and back-transport and subsequently increases the shear rate near the membrane surface. Particle deposition is hence prevented and fouling is subsequently reduced. Akoum and co-workers [3,4] have observed an increase in the permeate flux in these systems when compared to a standard spiral wound membrane in the microfiltration (MF) and UF of skim milk. However, the vibrating equipment is expensive and this limits membrane area and the potential for scale-up [2,4].

The use of an ultrasonic field has been studied widely in membrane filtration systems for both flux enhancement during fouling [5–13] and to improve cleaning efficiency [14–17]. When an acoustic field is applied to a liquid, acoustic cavitation, a phenomenon in which bubbles present in the liquid medium grow and collapse due to pressure fluctuations caused by ultrasound waves, is generated. Acoustic cavitation generates shear

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forces, turbulence and micro-streaming, which can enhance membrane performance [18]. Muthukumaran et al. [19–21] found that during the ultrafiltration of whey, the permeate flux is enhanced significantly when a low-frequency, low power ultrasonic field is present. They speculated that at the membrane surface, the mass transfer coefficient within the concentration polarisation layer increases due to localised flow disturbances through bubble collapse and acoustic streaming. Microjets, which are also generated during cavitation, scour the surface, enhance turbulence and promote the back-transport of deposits to the bulk solution. The cake layer was also found to be less compressible and looser. More recently, Mirzaie et al. [22] obtained similar results in the microfiltration of milk where flux was enhanced by a factor of 490% with the use of ultrasound at 20 kHz. However, higher acoustic power levels and improper installation of the ultrasonic unit to the membrane can impact the structural integrity of the membrane [19,20]. Large-scale filtration with ultrasonic enhancement may also be expensive due to high energy consumption [2].

Our recent work has shown that ultrasonic treatment of concentrated whey solutions independent of the filtration operation can reduce solution viscosity and protein aggregate size significantly [23]. The first aim of the present work is to determine whether such ultrasonic application upstream of ultrafiltration may be as effective as sonication during the UF process, as this would be considerably easier to implement.

Further, during evaporation of the whey solution downstream of filtration and in the use of the whey powders in downstream ingredient manufacture, the aqueous whey solution is often exposed to heat. Exposure to temperatures above 70 °C results in denaturation and aggregation of the whey proteins, often resulting in excessive thickening or gelling of the protein solution during processing and upon storage [24,25]. This thickening limits the solids concentration that can be achieved upstream of spray drying and may also limit the application of the whey powders as dairy ingredients. Pre-treatment procedures, such as fore-warming and pH adjustment, have been developed to improve this heat stability. Of most relevance to this work is fore-warming: Deysher and co-workers [26] described the production of a heat stable condensed milk stream by the application of fore-warming as early as 1929 [24,27–29]. However, this approach was found to be ineffective for spray-dried products as the resulting increases in viscosity that occur after heating restrict the ability to generate a dryer feed stream of high solids content. Furthermore, the aggregation that occurs during heating can result in later phase separation and protein precipitation.

Alternatively, we have used a combination of heat and ultrasound to improve heat stability [18]. When ultrasonic treatment is applied to a heated solution of denatured and aggregated proteins, there is a dramatic decrease in protein aggregate size and viscosity. It is speculated that these reductions are due to the disruption of hydrophobic interactions by shear forces that are generated during acoustic cavitation [30]. Upon further heating, the low viscosity is maintained, overcoming the problems of pre-treatment by fore-warming and poor heat stability in the reconstituted powder [18]. With such a reduction in viscosity, it may be possible to process whey solutions to higher solids content in downstream evaporator units. The combination of heat and ultrasonic pre-treatment may be a promising approach in alleviating membrane fouling and enhancing spray dryer productivity while producing heat stable powders. This represents the second aim of this paper.

2. Theory

During UF at constant feed concentration, the flux decline curves can be analysed using a combined pore blockage and cake filtration model developed by Ho and Zydney [31]. These authors assume that the initial flux decline arises from the deposition of large aggregates, which block the membrane pores (Eq. (1)) [31].

$$J_{blocked} = \frac{\Delta P}{\mu_f (R_m + R_p)} \tag{1}$$

where $J_{blocked}$ is the flux through blocked pores (m^3/m^2 s), ΔP is the transmembrane pressure (Pa), μ_f is the viscosity of the feed (Pa s), R_m is the clean membrane resistance and R_p is the resistance of the protein deposit (m^{-1}). With time, there is increasing resistance for fluid to flow through the blocked regions as more particles settle on the membrane surface and contribute to the growth of a cake layer (Eq. (2)).

$$\frac{dR_p}{dt} = f' R' J_{blocked} C_b \tag{2}$$

where f' is the fractional amount of protein that contributes to deposit growth (dimensionless), R' is the specific resistance of the protein layer (m/kg) and C_b is the bulk protein concentration (g/L). The volumetric flow rate through the open and blocked pores, Q (m^3/s), is finally determined as

$$Q = Q_w \left[\exp\left(-\frac{\propto \Delta P C_b}{\mu_f R_m} t \right) + \left(\frac{R_m}{R_m + R_p} \right) \left(1 - \exp\left(-\frac{\propto \Delta P C_b}{\mu_f R_m} t \right) \right) \right] \tag{3}$$

$$R_p = (R_m + R_{p0}) \sqrt{1 + \frac{2 f' R' \Delta P C_b}{\mu_f (R_m + R_{p0})^2} t} - R_m \tag{4}$$

where Q_w is the volumetric flux of pure water, α is the pore blockage parameter (m^2/kg), t is the filtration time (s) and R_{p0} is the resistance of a single protein aggregate (m^{-1}). The first term within the brackets in Eq. (3) represents the flow through the open pores and corresponds to the classical pore blockage model while the second term describes flow through the blocked pores. The permeate flux through the membrane is thus dependent on the pore blockage parameter (α), the initial resistance of the deposit (R_{p0}) and the cake growth rate (described by $f'R'$). The pore blockage parameter, also known as the rate of pore blockage, is equal to the membrane area blocked per unit mass of protein convected to the membrane surface and is an indication of the protein aggregate size [19]. The initial resistance of the protein deposit is a ratio of the initial resistance of the protein deposit to the membrane resistance. The cake growth factor represents the rate of increase of the protein layer resistance with time due to the growth of the protein cake layer.

This model was developed based on the microfiltration of bovine serum albumin. In further work, the authors have found the model to be in good agreement for the filtration of lysozyme, pepsin, immunoglobulin G and myoglobin [32] and the microfiltration of BSA-lysozyme and BSA-pepsin mixtures [33]. More recently, Muthukumaran et al. [19] obtained a good fit between the model and the experimental data in the ultrafiltration of WPC80 solution with and without in-situ ultrasound.

When the membrane operation is run at increasing feed concentration within a steady state, pressure independent filtration regime, a gel polarisation model can be used. This approach assumes that a concentration-polarised boundary layer exists above the precipitated cake or gel layer. Eq. (5) is the classical equation used for the model.

$$JC - JC_p = -D\left(\frac{dC}{dx}\right) \tag{5}$$

where J is the permeate flux (m^3/m^2.s), C is the protein concentration within the concentration polarisation layer (kg/m^3), C_p is the protein concentration in the permeate (kg/m^3) and D is the