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Crossflow Microfiltration in the Dairy Industry

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1.1 Introduction

1.1.1 Membrane Types

Since their introduction in the 1960s, pressure driven, crossflow or tangential filtration membrane technologies have become important in the food processing industries. The dairy industry currently uses crossflow membrane technologies for applications such as fractionation of the casein and whey proteins, whey protein concentration, demineralization of whey, removal of somatic cells and bacteria from milk, and milk concentration to save transport costs (Pouliot, 2008; Gésan-Guiziou, 2010). Membranes are also used alone or with the evaporation step in the manufacture of milk powders, and are increasingly being used in the development of new dairy-based beverages, fermented milk beverages and yogurt products. They are also finding a place in clean-in-place (CIP) processes to recover cleaning agents or to recover water used in processing (Alvarez *et al*., 2007; Luo *et al*., 2012).

Four types of membranes are used by the dairy industry: reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF). The operating parameters for crossflow filtration membranes are shown in Figure 1.1. The pressure-driven feed, with flow rate, Q_F, flows through the membrane channel parallel to the surface of the membrane. The applied pressure, P_F, must overcome the osmotic pressure, π_F, of the feed solution (Cheryan, 1998). The crossflow velocity (CFV), the velocity of the feed as it flows parallel to the membrane through the channel, has a sweeping effect that minimizes build-up of the feed particles on the membrane surface.

Some of the feed stream containing the smaller molecules flows through the walls of the membrane leaving as the permeate, with flow rate, Q_P, and pressure P_P. Q_P is often reported as the permeate flux, J, defined as the volume of permeate per unit membrane surface area per time. P_P has a gauge pressure reading of 0.0 if the stream

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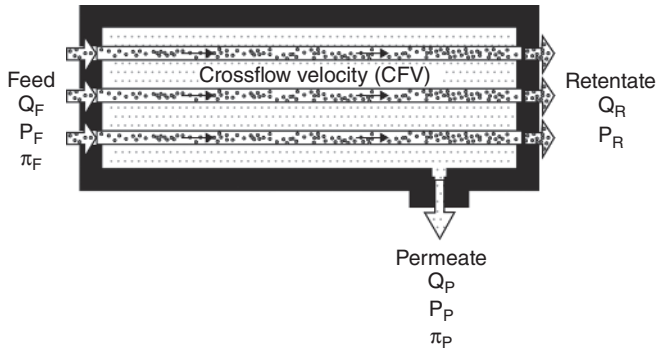

Figure 1.1 Parameters for crossflow filtration. Cross-section of a crossflow microfiltration housing for multiple membrane tubes shown.

is open to the atmosphere. The remainder of the stream, called the retentate, with flow rate, Q_R, and pressure, P_R, flows out the end of the membrane. This stream may be entirely or partially recycled back to the feed. The size distribution of the particles in the permeate and the retentate depend on the pore size distribution of the membrane. The pressure-driving force is reported in terms of the transmembrane pressure (TMP) and is given by:

$$\text{TMP} = (P_F - P_R)/2 - P_P \tag{1.1}$$

Table 1.1 shows the sizes of the milk nutrients, somatic cells and species that may populate milk, such as bacteria, spores, yeasts and moulds, and the corresponding types of membranes that would be used to separate them from smaller milk components. The wide ranges in sizes for bacteria and spores reported in Table 1.1 account for their possible lengths and widths (Garcia *et al*., 2013). The operating pressure ranges and the separation technologies that compete with the particular membrane type are also listed. Particles smaller than the rating or pore or cut-off size leave in the permeate stream, particles larger than the pore size remain in the retentate. For RO and NF, the membranes are rated by salt rejection standards defined by the manufacturer. UF membranes are rated by a molecular weight cut-off size (MWCO) and MF membranes are rated by pore size.

RO, which may be used for milk or cheese whey concentration and to concentrate milk to save on transport costs, mostly retains the milk solutes, allowing only water to pass through the membrane. NF, which is also known as leaky RO, since it allows monovalent ions to pass through the membrane along with water, can also be used for concentration and, for example, in whey demineralization to purify lactose from cheese whey by removing salt, or to reduce water hardness in dairy plants (Cheryan, 1998; Pouliot, 2008; Gésan-Guiziou, 2010). The driving force for RO and NF is osmotic pressure. Depending on the cut-off size, UF, the most commonly used membrane process in the dairy industry, produces a retentate of proteins and fat, with the permeate containing minerals, nonprotein nitrogen and lactose. UF is used for protein standardization of cheese milk, to concentrate whey, for lactose-reduced milk and to fractionate the whey proteins. MF, depending on the membrane pore size, has been used to pretreat whey to remove fat, casein fines and bacteria prior to manufacture of

Table 1.1 Membrane pore size and operating pressure ranges, milk component sizes, size range and alternative processing methods. The corresponding MW range is in parentheses

Membrane Type/ Range	Pressure Range (KPa)	Milk Component	Size Range	Alternative Process
Microfiltration 0.1 μm – 10 μm (~100 – 1000 kDa)	10 – 350	Somatic Cells	8 – 10 μm	Centrifugation
		Fat	0.1 – 15 μm; 3.4 μm average	
		Bacteria/Spores	0.2 – 10 μm	
		Yeasts, moulds	1 – 10 μm	
Microfiltration/ Ultrafiltration		Casein micelles	0.110 μm; average 0.02 – 0.3 μm	
		Immunoglobulins	150 – 900 kDa	
Ultrafiltration 0.001 – 0.1 (1 – 500 kDa)	30 – 1050	Whey proteins	0.03 – 0.06 μm	Centrifugation
		α-lactalbumin	14 kDa	
		β-lactoglobulin	18 kDa	
		BSA	66 kDa	
		lactoferrin	86 kDa	
		GMP	8 – 30 kDa	
		Enzymes	13 – 100 kDa	
Nanofiltration 0.2 – 2 kDa	1000 – 4000	Lactose	0.35 kDa	Evaporation, Distillation
		Salts		
		Vitamins		
Reverse Osmosis	1300 – 8000	Water		Distillation, Evaporation, Dialysis
		Ions		

Data from Brans *et al.*, 2004, and Garcia *et al.*, 2013.

whey protein concentrates by UF (Cheryan, 1998), to remove bacteria from milk and for production of micellar casein and whey protein from milk.

Currently, MF has limited use in the dairy industry, with an installed membrane area of 15 000 m^2 compared to that of UF with an installed area of 350 000 m^2 (Garcia *et al.*, 2013). This chapter reviews the theory and experimental techniques used in research on MF and then focuses on the current status of MF for removal of bacteria from milk to create extended shelf life (ESL) milk, processes which use MF to separate milk into value-added enriched fractions and newer developments in MF applications. The greenhouse gas emissions, energy use and estimated costs for a fluid milk processing plant are compared to those for the same plant with an MF installation.

1.1.2 MF Membranes

Membranes used in the dairy industry are semipermeable and are manufactured to achieve various pore sizes and pore size distributions tailored for a particular

application. MF membranes for dairy applications have a well defined pore size distribution and are manufactured from ceramic materials or polymeric materials. Milk MF is usually performed with membranes in tubular form (ceramic membranes) or, in limited applications, a spiral-wound (SW) design (polymeric) to fit laboratory, pilot plant and commercial scale equipment.

Ceramic membranes have an asymmetric structure consisting of two layers. The top layer, also known as the skin layer or active membrane layer, is very thin and, depending on the pore size and pore size distribution, is a factor in determining the performance of the membrane in terms of fouling. Fouling lowers the permeate flux, J, and may also prevent or alter the transmission of the feed components to the permeate. The bottom layer is a macroporous support structure for the membrane (Figure 1.2). Ceramic membranes are made from metal oxides such as zirconia, titania, or alumina and silica and formed into tubes. MF membranes for dairy applications are usually made from alpha-alumina. Polymeric SW membranes for MF are manufactured mainly from poly(vinylidene fluoride) (PVDF). Their manufacture is not discussed here but details can be found elsewhere (Cheryan, 1998).

Regardless of membrane type, membranes for the dairy industry must be able to withstand the rigorous cycling of chemicals and high temperatures during cleaning. The PVDF SW membranes can withstand temperatures up to about 60°C but are susceptible to chemical cleaning, which limits their use to about one year (Cheryan, 1998). Ceramic MF membranes are more expensive than the SW membranes and can withstand liquid temperatures of up to approximately 95°C, but the actual temperature limits are set by the tolerances of the gaskets and o-rings to the higher temperatures and chemical cleaning. These membranes can last for up to 10 years (Cheryan, 1998).

Hydrophilic membranes are chosen for milk processing applications because they minimize protein binding by the hydrophobic proteins that contribute to fouling and affect permeability (Bowen, 1993). Their high surface tension attracts water molecules

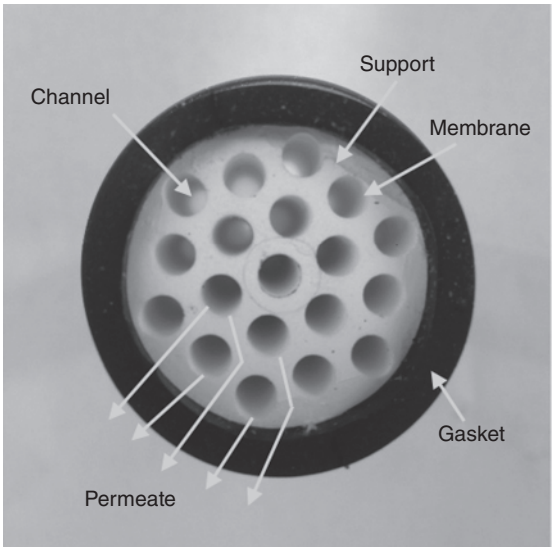

Figure 1.2 Cross-section of a ceramic membrane tube.

to the surface; this helps to prevent protein fouling. Ceramic membranes are naturally hydrophilic, since they are derived from the hydrophilic metal oxides. PVDF membranes are hydrophobic but are available in modified form to reduce hydrophobicity (Liu *et al*., 2011).

In addition to hydrophilicity, the charge, surface roughness and morphological properties of the membrane and the sizes and tortuosity of the membrane pores have also been shown to affect the extent of fouling by proteins (Bowen, 1993; Cheryan, 1998). For example, milk has a pH of 6.6, with many of its proteins negatively charged. It would be expected that a negatively charged membrane would be more preferable for milk processing than a positively charged membrane. However, many of the ionic species in milk, particularly calcium, would bind to the membrane and, in turn facilitate, binding of the negative proteins and phosphates (Bowen, 1993).

The selectivity is also an important consideration when choosing a membrane. Selectivity may be adversely affected by the pore size distribution, uneven TMP across the membrane and fouling (Brans *et al*., 2004). A large pore size distribution may result in undesired transmission or retention of milk components adversely affecting permeate composition. Uneven TMP across ceramic membranes, typically caused by the high CFV required for high permeate flux, J, causes variations in permeate flux that may lead to fouling on and in the pores of the membrane and undesired transmission and retention of milk components.

In the late 1980s, the uniform transmembrane pressure (UTP) process was developed by Alfa-Laval to address uneven TMP in milk MF (Sandblom, 1978; Malmberg and Holm, 1988; van der Horst and Hanemaaijer, 1990). The process is known as 'Bactocatch'. The Bactocatch process operates by addition of a pump to recirculate the permeate through the permeate side of the membrane cocurrently with the retentate. Plastic balls added to the permeate side decrease the amount of permeate required and, thus, the pump size. This modification was shown to result in a constant pressure drop on both sides of the membrane so that J on the order of 500 l/m^2/h with almost complete transmission of the milk proteins, and total bacteria retention could be sustained for 10 hours with low fouling (Saboya and Maubois, 2000). However, this method incurs higher operating costs due to the additional permeate recycle pump.

More recently, graded permeability (GP) membranes (Pall Corporation) and Isoflux membranes (Tami Industries) have been introduced; these do not require installation of an additional pump as in the Bactocatch process to maintain uniform J and low fouling. The GP membranes include a longitudinal permeability gradient in the support structure, which is located around the active membrane layer, that maintains TMP along the length of the membrane. The Isoflux membranes include a change in thickness in the active layer to maintain TMP, and thus J, along the length of the membrane. MF plants today use either the UTP process or the GP and Isoflux membranes.

The ceramic membranes used to process milk are multichannelled tubes, up to 1.2 m long with 3–39 channels (Figure 1.2). The channels are usually circular with inner diameters ranging from 2 to 6 mm. Isoflux membrane tubes have multichannel configurations named daisy, sunflower and dahlia, in which the channels are roughly triangular in shape. These patterns provide more surface area per tube, thus increasing J. Star-shaped channels, which provide more surface area than circular channels, are also available for membranes.

For small-scale pilot testing, a single tube is placed in a housing and installed in the supporting MF equipment. In larger pilot-scale equipment or commercial-scale operations processing several litres of milk per hour, several tubes are placed in a single

housing and then installed. Commercial plants use several housings. These configurations affect the method for calculating CFV.

For a single membrane channel

$$CFV = Q_F / 3600\, A_{xs} \tag{1.2}$$

Q_F is the flow rate of the feed to the channel and A_{xs} ($= \pi d^2/4$) is the cross-sectional area of a single channel with diameter, d. If a membrane has several channels, as shown in Figure 1.2, then $A_{xs} = n\,\pi d^2/4$, where n is the number of channels. For a membrane module in which several membrane elements are contained in a housing (Membralox, 2002), $A_{xs} = Nn\pi d^2/4$, in which N is the number of membranes in the housing.

1.1.3 Pilot Plant Testing

Milk MF is typically conducted using a 1.4 μm membrane at temperatures ranging from 40 to 55°C, with 50°C the most commonly used. CFV ranges from 5 to 9 m/s with CFV and TMP chosen so that J is optimized. TMP typically ranges from 30 to 50 KPa (Cheryan, 1998; Brans *et al*., 2004) for the UTP process but is higher for the GP and Isoflux membrane processes, with reported values ranging from 50 to 200 KPa depending on CFV (Fritsch and Moraru, 2008; Skrzypek and Burger, 2010; Tomasula *et al*., 2011). Higher values of TMP are observed for the smaller membrane pore sizes (Tomasula *et al*., 2011; Adams and Barbano, 2013). For applications involving removal of bacteria and spores from milk, only skimmed milk is filtered because of the overlap in sizes of the bacteria and spores with that of the fat globules (Table 1.1). A 1.4 μm membrane is used for skimmed milk MF, although 0.8 μm membranes may optionally be used since they are more effective for spore removal (Tomasula *et al*., 2011).

A schematic diagram of a pilot plant MF process is shown in Figure 1.3. Several companies provide the equipment necessary for pilot testing of milk MF in batch or continuous modes in skid form. This equipment is of sanitary construction and usually includes a 115 or 190 litre feed tank, single or multiple membrane modules, a recirculation pump with variable speed drive, a heat exchanger, pressure gauges and transducers to measure the inlet and outlet pressures, thermocouples, flow meters and valves for control of the permeate and retentate flows, and process control equipment. CFV is controlled using the recirculation pump and the retentate valve. Many units are equipped with an optional back-pulsing system, which is used to push foulant from the membrane to be cleared by the CFV. This is accomplished by applying pressure on the permeate side so that $P_P > P_F$. The frequency and duration of the pulses may be varied. Back-pulsing is limited to the pilot scale.

Prior to running a milk MF pilot process, the membrane is cleaned according to the manufacturer's instructions. Then, water MF is conducted to determine the clean water flux, CWF, to ensure that it is approximately the same value determined from previous experiments.

$$CWF = (Q_f * \mu/(TMP * A) \tag{1.3}$$

where Q_f is the water flow rate in l/h; μ is the water viscosity, 0.001 Pa-s at 20°C; and A is the membrane surface area for filtration, m^2. If CWF is not in agreement with previous values, the membrane should be cleaned according to the manufacturer's protocol, and tested again. Inability to clean the membrane may be indicative of irreversible fouling. After successful testing of the CWF, skimmed milk is charged to the

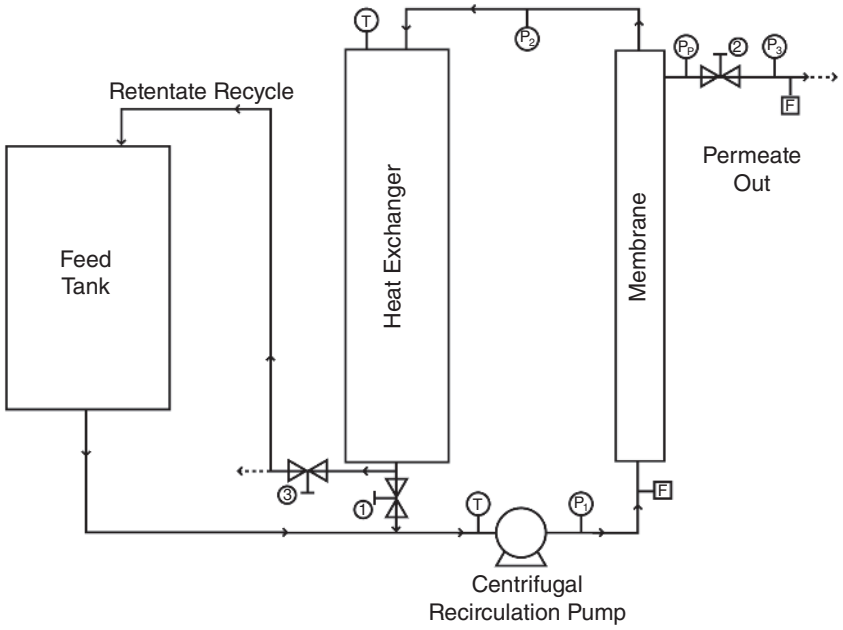

Figure 1.3 Schematic diagram of MF process skid showing batch filtration with full recycle of retentate. P_1, P_2, P_3 and P_P are pressure gauges; (1), (2) and (3) are valves; T is a temperature thermocouple.

holding vessel and then pumped to the membrane at 50°C. The weight of the permeate may be determined as a function of time to determine the experimental permeate flux, J, in l/m^2/h, often reported as LMH.

$$J = \text{weight permeate (kg)} / (A \rho t) \tag{1.4}$$

A is the surface area of the membrane, m^2; ρ is the permeate density (kg/m^3) at MF temperature; and t is the time (hours).

Experimental processes are typically conducted in batch mode with most of the retentate recycled back to the feed tank and the permeate collected. To achieve a particular volume concentration reduction (VCR), for example a VCR of 20, or 20× concentration, the following relationship is used:

$$VCR = V_F/V_R = V_F/(V_F - V_P) \tag{1.5a}$$

where V_F is the volume of feed; V_R the volume of retentate; and V_P the volume of permeate.

VCR for a continuous process is defined as:

$$VCR = (Q_P + Q_R)/Q_R \tag{1.5b}$$

where Q_P is the flow rate of the permeate and Q_R is the flow rate of the retentate. VCR may also be referred to as a volume concentration factor or concentration factor.

Research to develop models for correlation and prediction of J for various milk species–membrane interactions is carried out continuously (Kromkamp *et al*., 2007; Kuhnl *et al*., 2010). Selection of the appropriate membrane for a milk MF application should include pilot plant testing with a particular membrane to verify that the desired production rate, permeate composition, quality attributes, and process economics will be met. Consultations with vendors are also recommended.

1.2 MF Principles and Models

For the ideal case of flow of a fluid through a microporous membrane, the flow rate of the permeate stream, J, is given by Darcy's law (Cheryan, 1998):

$$J = TMP/\mu R_m \tag{1.6}$$

where μ is the solution viscosity and R_m is the intrinsic resistance of the membrane for pure water. However, J for a fluid such as milk is often less than the ideal value because of boundary layer formation, concentration polarization and fouling effects that can act as a secondary membrane along the surface of the membrane (Schulz and Ripperger, 1989; Merin and Daufin, 1990; Bowen, 1993; James *et al*., 2003). In addition to fouling that occurs due to the protein–membrane interactions discussed previously, other causes of fouling are: pore blockage, resulting in partial or total closure of pores; deposits of particles, or cake layer formation, which grow in layers at the surface of the membrane and over time act as an additional resistance to permeate flow; and gel formation of macromolecules, arising from concentration polarization (Belfort *et al*., 1994; Bacchin *et al*., 2006).

The temperature effects of milk MF on J are included in the viscosity term of Equation 1.6. As temperature increases, the viscosity of milk decreases, which leads to increased J (Whitaker *et al*., 1927; Alcântara *et al*., 2012). The viscosity of milk may also vary with pH and age (McCarthy and Singh, 1993). Permeate flow, J, for milk MF at 53°C was shown to be approximately 85% greater than that at 6°C due to the decreased viscosity (Fritsch and Moraru, 2008).

When milk MF is conducted in the range from 40 to 55°C, the viscosity of milk varies from 1.04 cP (0.00104 Pa s) to 0.77 cP (0.00077 Pa s) (Whitaker *et al*., 1927), which indicates a difference in viscosity over the MF temperature range of approximately 25%. However, temperature increases also lead to an increase in protein diffusivity, which may reduce concentration polarization and fouling, but there may be an increase in internal fouling of the membrane (Marshall *et al*., 1993).

The flow of milk at high CFV across a MF membrane helps prevent the build-up of particles along its surface but is associated with boundary layer formation due to shear stress at the membrane wall (Cheryan, 1998). The boundary layer includes the velocity profile along the wall at which the velocity is a minimum to the point where it is approximately the velocity of the bulk stream. Because the membrane is porous, the amount of water in the milk stream is decreased at the wall due to the permeate flow and is accompanied by an increased concentration of milk proteins. This is concentration polarization, which is also referred to as a gel or cake layer; it forms due to the dynamic and reversible layer of milk proteins (Figure 1.4). This layer can be removed through the proper selection of operating conditions such as CFV and TMP.

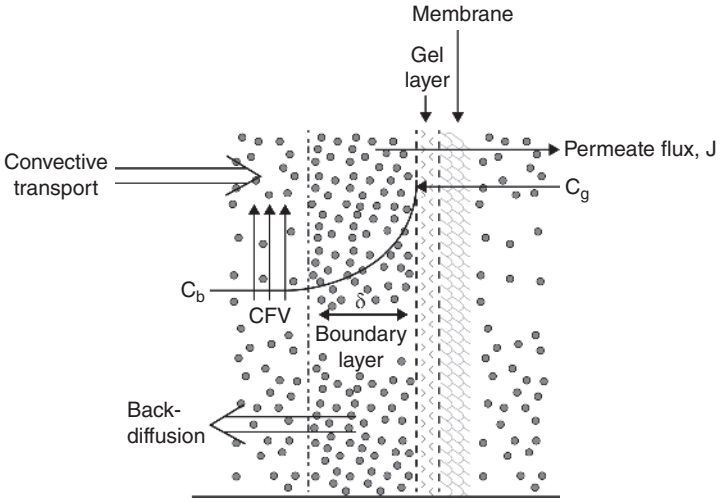

Figure 1.4 Concentration polarization.

Fouling of the membrane may be reversible or irreversible. A reversible fouling layer is indicated by a slow decline in J with MF run time and is difficult to reverse with changes in CFV or TMP alone. It may be reversed using back-pulsing, as discussed previously, although this technique is not always successful for sustained MF operations.

For MF, it was hypothesized (Field *et al*., 1995; Howell, 1995) that at a particular CFV, J is a linear function of TMP until a critical flux, J_{crit}, is reached at TMP_{crit}. Increasing TMP above TMP_{crit} initiates membrane fouling and increases J further. Strong and weak forms of J_{crit} were proposed. The strong form is characterized by a linear plot of J as a function of TMP, similar to that observed for water MF. The weak form is also linear but various membrane interactions decrease J relative to that of water.

The performance of J as a function of TMP in three distinct regions has been described for MF (Field *et al*., 1995; Howell, 1995; Brans *et al*., 2004). Milk MF in the subcritical region ($TMP < TMP_{crit}$) (Region 1 or the pressure-controlled region) is desirable for optimal selectivity of the membrane with minimal fouling (Figure 1.5). Although selectivity is optimal, a larger membrane surface area is needed because of low J (Brans *et al*., 2004). Increasing CFV or temperature at a fixed TMP also increases J. In Region 2, when $TMP > TMP_{crit}$ and $J > J_{crit}$, J is optimal and less membrane surface is required, but selectivity is not optimal. With further increases in $TMP > TMP_{crit}$, J approaches a limiting value, J_{lim}, independent of TMP as the fouling or gel layer increases in thickness. The capacity of the membrane is then saturated by fouling (Belfort *et al*., 1994; Bacchin *et al*., 2006) and J becomes independent of membrane pore size. Milk MF for bacteria reduction and for casein micelle concentration from milk is conducted from the boundary of Regions 1 and 2 and into Region 2 (Brans *et al*., 2004). In Region 3, as the fouling layer builds, compaction of the layer may occur, decreasing J further because of membrane pore blockage (Chen *et al*., 1997). Back-pulsing would be required to control the fouling. There may also be an abrupt decline in J, as shown in Figure 1.5. Upon reduction in TMP, hysteresis

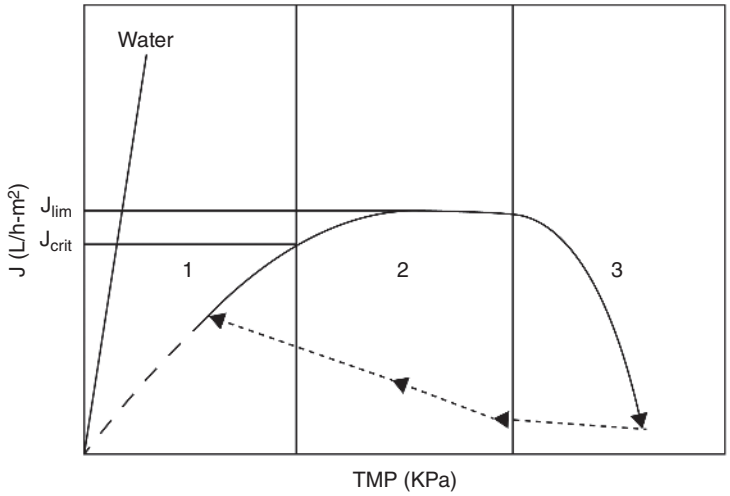

Figure 1.5 Dependence of flux, J, on transmembrane pressure (TMP). The pressure-controlled region is approximated by Region 1 and the mass-transfer-controlled region by Region 2. Increasing CFV or temperature at a fixed TMP also increases J.

in the curve is noted and J is not restored to its initial value (Chen *et al*., 1997; Guerra *et al*., 1997; Tomasula *et al*., 2011).

Semi-empirical and empirical models have been developed to obtain an understanding of the dependence of J on TMP in the subcritical region and the characteristic shift from the pressure-controlled region to the pressure-independent, mass-transfer-controlled region, as J_{crit} and J_{lim} are approached (Figure 1.5). The models are of three general types: film or gel-polarization theory models, osmotic pressure models to determine J_{lim}, and resistance models. Variations of these models have also been described but are not discussed here (Bowen and Jenner, 1995).

1.2.1 Gel Polarization Models

The film theory model was first applied to ultrafiltration (Bowen and Jenner, 1995) and assumes that the solute particles of milk migrate from the bulk stream toward the membrane surface by convective transport and return to the bulk stream by back-diffusion (Figure 1.4).

At steady-state, a boundary layer forms with thickness, δ. The mass transfer coefficient is given by:

$$k = D/\delta \tag{1.7}$$

where D is the diffusion coefficient. If a critical concentration of a solute particle, for example the milk proteins or casein, is reached at the membrane surface (Guerra *et al*., 1997; Tomasula *et al*., 2011), a gel layer may form restricting permeate flow. J is then given by (Cheryan, 1998):

$$J = k \ln (C_g/C_b) \tag{1.8}$$

where C_g is the concentration of the solute at the membrane surface or the gel concentration and C_b is the concentration of the solute in the bulk of solution (milk).

Cg is termed C_M, concentration at the membrane, if a gel layer is not formed. J for this model predicts J_{lim}, the limiting flux.

The Leveque equation for laminar flow or the Dittus–Boelter equation for turbulent flow may be used to determine k, which is related to the Sherwood number, Sh, (Bowen, 1993) but does not give an exact representation for CFV and other empirical representations may be found more suitable. For turbulent flow, defined by Reynolds number, $N_{Re} > 4000$, the Dittus–Boelter expression is (Cheryan, 1998):

$$Sh = 0.023(Nre)^{0.8}(Sc)^{0.33} \tag{1.9}$$

where

N_{Re} = Reynolds number = $D_h V\rho/\mu$
Sc = Schmidt number = $\mu/\rho D$
D_h = hydraulic diameter = 4 (cross-section available for flow/wetted perimeter of the channel).

The gel polarization model is useful for estimating J_{lim} when the concentration of the gel layer at the surface of the membrane is at constant C_g. However, since it lacks a pressure term and does not account for other operating conditions, it does not describe cases in which $J < J_{crit}$ (Samuelsson *et al*., 1997; Cheryan, 1998; Ripperger and Altmann, 2002).

1.2.2 Osmotic Pressure Model

The osmotic pressure model (Jonsson 1984; Wijmans *et al*.,1984; Prádãnos *et al*., 1995) for J takes into account the operating conditions and the osmotic pressure term, π_M, in Darcy's equation (Bowen, 1993; Cheryan 1998):

$$J = (TMP - \Delta\pi_M)/R_m \tag{1.10}$$

$\Delta\pi_M$ is the osmotic pressure difference across the membrane but is approximated by π for a concentrated solute, such as milk proteins or casein at the membrane surface in the case of milk MF, and is calculated in terms of virial coefficients, A_n (Cheryan, 1998):

$$\pi_M = A_1C_M + A_2C_M^{\,2} + A_3C_M^{\,3} + \ldots \tag{1.11}$$

C_M is the concentration at the membrane surface and may be calculated from film theory (Equation 1.8). R_M is the intrinsic membrane resistance determined for pure water. Depending on the value of C_M, the higher order terms of Equation 1.11 may become important, increasing the value of π_M so that it approaches TMP, resulting in a decrease in J.

The gel polarization and osmotic pressure models are useful for understanding the dependence of J_{lim} on operating conditions. While the gel polarization model is useful for predicting the dependence of J on C_b or CFV, it requires the appropriate Sherwood correlation to estimate k and only applies to Region 2, the mass-transfer-dependent region of Figure 1.5. The osmotic pressure model does not assume deposition or

adsorption of proteins but relies on the availability of osmotic pressure data to calculate the pressure difference across the membrane given in Equation 1.10. A unified model for prediction of J across the three regions of Figure 1.5 is still not available.

1.2.3 Resistance–in-Series Model

The model most commonly used to correlate experimental data for J as a function of time is the resistance-in-series model given by Darcy's law:

$$J = (TMP - \Delta\pi)/\mu R_{total} \tag{1.12}$$

If $\Delta\pi$ is negligible:

$$R_{total} = R_m + R_d \tag{1.12a}$$

R_{total} is the total hydraulic resistance (m^{-1}) which may be estimated from the final average value of J obtained from several milk MF trials performed under the same operating conditions. R_m is the resistance of the clean membrane obtained from CWF (Equation 1.3). R_d includes other resistances attributed to reversible, $R_{f,rev}$, and irreversible, $R_{f,irrev}$, fouling with:

$$R_d = R_{f,rev} + R_{f,irrev} \tag{1.12b}$$

Some models further define $R_{f,rev}$ and $R_{f,irrev}$ to specifically account for adsorption, pore blocking, and other possible fouling mechanisms (Bowen and Jenner, 1995). $R_{f,\,rev}$ is due to adsorption of milk components by the membrane that occurs with MF operations mainly in Region 1. It may be calculated from the values of R_{total} and R_m, since $R_{f,irrev}$, fouling that cannot be removed with changes in CFV or TMP during an experiment, is negligible in Region 1. Experimentally, the contribution of $R_{f,\,irrev}$ may be obtained by rinsing the membrane with deionized water after a milk MF trial for approximately 20 minutes to remove $R_{f,rev}$ (Tomasula *et al*., 2011). $J_{f,irrev}$ is then the value of J for the rinsed membrane and $R_{f,rev}$ may then be determined from Equations 1.12a and 1.12b if there is irreversible fouling. $R_{f,irrev}$ would occur mainly in Region 3. Determination of resistances in Region 3 may require use of the osmotic pressure term, $\Delta\pi$, in Equation 1.12 which becomes important at high TMP.

Figure 1.5 shows an example of irreversible fouling at $J > J_{lim}$ in Region 3. In this case, it was hypothesized that compaction of the gel layer, consisting mainly of casein, increased the hydraulic resistance across the membrane. This would result in an increase in the osmotic pressure at the surface of the membrane approaching that of TMP and a decrease in J (Tomasula *et al*., 2011).

To demonstrate the effects of pressure of a casein deposit on a membrane, compression and relaxation of casein with changes in pressure were observed through dead-end microfiltration experiments with a polyethersulfone (PES) UF membrane. The feed was either native phosphocaseinate powder or a sodium caseinate powder dispersed in UF skimmed milk permeate. The resistance to flow through the casein deposit was found to depend on the internal porosity of the casein micelle, which was controlled by the degree of compression (Pignon *et al*., 2004; Qu *et al*., 2012). A critical osmotic pressure, π_{crit}, was also defined as the compressive pressure to achieve a critical concentration of casein micelles and the point at which phase transition for the formation of an irreversible deposit is initiated. In future experiments, the effects of CFV and TMP on the casein deposit during crossflow milk MF and the properties of π_{crit} will be examined.

1.3 Applications of MF

1.3.1 Production of Concentrated Micellar Casein and Whey Proteins

Whey protein concentrates are produced from cheese whey using UF. An alternative method to produce whey concentrates uses MF with a skimmed milk feed instead of cheese whey to produce concentrated micellar casein in the retentate and native whey proteins or serum proteins (SPs) in the permeate. The casein in the concentrate is in its native micellar form, unlike acid casein, which is denatured when precipitated from milk using acids. SPs are a potential alternative to whey protein concentrates obtained from cheese making with the added benefits of not being denatured or containing any residual products from cheesemaking and lower fat content. Micellar casein and SP protein concentrates may also be dried or blended depending on application.

The UTP concept and later development of the GP and Isoflux membranes led to improved processes to produce micellar casein and SPs directly from skimmed or whole milk. Polymeric membranes were used in early studies but were subject to fouling and low selectivity due to a wide pore-size distribution (Brans *et al*., 2004). Since polymeric SW membranes cost less than ceramic MF membranes, many recent studies have been conducted to determine the efficacy of PVDF SW membranes for milk separation.

Early studies for the removal of native casein micelles from skimmed milk used ceramic membranes with pore sizes ranging from 0.05 to 0.2 μm (Brans *et al*., 2004) but low values of J were reported. It was noted that since similar values of J were consistently reported even though TMP and pore sizes were dissimilar, MF was conducted in the pressure-independent region (Region 2) and that J was most likely equal to J_{crit}. Higher values of J were noted when a Kenics static mixer was inserted into a 0.1 μm membrane to change membrane hydrodynamics.

Zulewska *et al*. (2009) compared the efficiency of SP removal from skimmed milk using the UTP process with 0.1 μm membranes, an MF system equipped with 0.1 μm GP membranes and an MF system equipped with a 0.3 μm PVDF SW membrane. The processes were operated in a continuous bleed-and-feed 3× concentration factor mode. SP removal was 64.4%, 61.04% and 38.6% for the UTP, GP and SW processes, respectively, compared to the theoretical SP removal rate of 68.6%. The relative proportions of casein to SP for the respective processes were reported as 93.93/6.07, 93.14/6.86 and 90.03/9.97, compared to 82.93/17.08 for skimmed milk. The values of J for the UTP, GP and SW membranes were 54.1, 71.8 and 16.2 kg/m^2/h, and indicated that SP removal was most efficient for the UTP ceramic membrane but more SP would be removed for the GP membrane during an MF trial due to higher J, although the permeate was slightly cloudy compared to the clear UTP permeate. It was concluded that the SW membranes would require additional membrane surface area or several diafiltration steps to achieve the SP removal of the UTP and GP membranes.

In another study, Hurt *et al*. (2010) demonstrated SP removal for a three-stage, 3× UTP system with 0.1 μm MF ceramic membranes and with water diafiltration between the stages, to determine the amount of SP removed relative to the theoretical values. Cumulative SP removal for the first, second and third stages was 64.8, 87.8 and 98.3%, respectively, compared to the theoretical values of 68, 90 and 97% SP, respectively. In comparison, an SW system using a 0.3 μm membrane was estimated to require more than eight stages, including five water diafiltration stages, to remove 95% of SP from skimmed milk (Beckman *et al*., 2010). Reduced passage of SPs through the SW

membrane was attributed to fouling by casein, which increased the hydraulic resistance (Zulewska and Barbano, 2013).

Later experiments with a 0.14 μm Isoflux membrane (Adams and Barbano, 2013) showed SP removal efficiency (70.2%) similar to the SW membrane (70.3%) after three stages. The GP membrane showed a removal efficiency of 96.5%. It was expected that the Isoflux membrane would have a removal efficiency similar to that of the GP membrane. The authors offered several reasons for the performance of the Isoflux membranes in this application: some pore sizes were too small for passage of SPs, reverse flow conditions and the selective layer modification served to reduce the effective surface area of the membrane, and the shape of the membrane channels promoted fouling and rejection of SPs.

Karasu *et al*. (2010) noted that SW membranes provide performance similar to ceramic membranes but that at the industrial scale high hydraulic pressure drops and low TMP are difficult to achieve unless shorter membrane lengths are used. For ceramic membranes, Piry *et al*. (2012) constructed a module containing four sections to assess the effect of membrane length on fouling effects, J and beta-lactoglobulin (β-LG) transmission. Maximum β-LG transmission depended on the position along the membrane.

Quality of Micellar Casein Concentrate and Serum Proteins The stability of micellar casein concentrates (MCCs) under sterilization processing is critical for their use as ingredients in shelf-stable, high protein beverages. Sauer and Moraru (2012) found that MCC is unstable when subjected to sterilization. UHT treatment was found to induce coagulation of MCC while retorting caused an increase in particle size, possibly due to solubility loss of calcium phosphate and dissociation of the casein micelles. MCC was found to be stable though when pH was increased, temperature was decreased, or both, although the composition and size of the MCC micelles differed from the original MCC micelles.

In a comparison of 34% SP concentrate (SPC) with WP concentrate (WPC) made from the same milk (Evans *et al*., 2009), SPC had lower fat and calcium contents, higher pH and did not contain glycomacropeptide (GMP) from cheese making. Upon rehydration at 10% solids, the SPC solutions were clear and the WPC solutions were cloudy. Sensory differences were related to the differences in fat content and the compounds generated from the starter culture and were minor but distinct. Flavour differences were mild in SPC and WPC made from the same milk compared to commercial WPC.

Commercial Developments Skrzypek and Burger (2010) reported that four commercial plants using 0.14 μm Isoflux membranes were in operation in Poland and the Czech Republic to produce MCC for casein standardization of skimmed or fat standardized milk in Quark production. Quark was described as an unripened acidic white cheese, made using traditional methods, which leads to an acid whey by-product that cannot be disposed of owing to environmental regulations. Use of MCC, microfiltered using a VCR of 1.6–2, may reduce the amount of acid whey generated by the Quark process by 40–60%. The authors state that the sweet whey permeate MF product is also used for protein standardization in the manufacture of spray dried milk and other milk-based products.

Skrzypek and Burger (2010) also reported commercial production of MCC with VCR of 2–6 corresponding to a 49% and a 72% level of true proteins in the retentate,

respectively, with the ratio of casein/true protein exceeding 90%. The plant capacities ranged from 10 000 to 23 000 l/h depending on desired VCR. Four MF modules of 50 m^2 each are used in a two-stage MF plant. The SP permeate is concentrated by NF and then spray-dried or spray-dried after mixing with milk or milk derivatives.

Native WP containing 24% alpha-lactalbumin (α-LA) and 65% β-LG, obtained by MF using polymeric membranes, demonstrated improved *in vitro* functional properties compared to cheese WP (Shi *et al*., 2012). Results indicated that the anti-obesity effects of native WP obtained by MF are inferior to those of α-LA, but it protects against diet-induced obesity during weight loss due to its α-LA content.

Alternative Processes for MCC and SP Production Still in development are rotating disks, rotating membranes and vibrating systems (Jaffrin, 2008). In an example of milk MF, a laboratory rotating disk module equipped with six ceramic disks with pore size of 0.2 μm rotating around a shaft inside a cylindrical housing was used for MCC and SP production using a pasteurized skimmed milk feed at 45°C (Espina *et al*., 2010). A UF dynamic filtration followed milk MF to separate the whey proteins, α-LA and β-LG. TMP was a low of 60 KPa and was a function of the inner and outer radii and the angular velocity of the membrane disks. Permeate flux, J, was highest (90–95 LMH) at VCR of 1. Casein rejection was reported as high as 99% with α-LA and β-LG transmission through the membrane of 0.8 and 0.98, respectively.

1.3.2 Extended Shelf Life Milk

With good quality raw milk, aseptic filling and packaging in standard containers (packaged nonaseptically), and careful handling during storage and distribution to maintain temperature below 6°C, extended shelf life (ESL) milk has a shelf life ranging from a few days to up to 28 days under refrigeration (Goff and Griffiths, 2006) and as long as a reported 45 days (Rysstad and Kolstad, 2006). The shelf life for milk is established when the total bacterial count is >20 000 CFU/ml (FDA, 2011) after a certain length of time. According to Saeman *et al*. (1988), shelf life is determined by proteolysis, which generates off-flavours when the decrease in casein as a percentage of true protein (CN%TP) is greater than 4.76%. Maintenance of refrigeration temperature below 4°C will lead to the longest shelf life of milk.

ESL milk is known commercially as ultrapasteurized (UP) milk that was heated at a temperature $\geq$137.8°C for $\geq$2 s and packaged nonaseptically. Ultra high temperature (UHT) pasteurized milk is also ESL milk if filled and packaged aseptically making it shelf-stable for about six months. ESL milk may also be heat treated at a temperature of 125°C and held for four seconds or heated to 127°C with a hold time of five seconds (Goff and Griffiths, 2006), after preheating at temperatures ranging from 70 to 85°C. However, while heat treatment ensures the safety of milk, it is also associated with cooked flavours, impaired functionality and loss of cheese making ability. Inactivated bacterial cells remain in the milk with any still active enzymes leading to reduced shelf life. Unfortunately, the temperatures used for ESL milk may also activate spores of *Bacillus* spp. (Goff and Griffiths, 2006), some of which have the potential to germinate and grow under refrigeration temperatures, without competition from other organisms. Use of bactofugation (te Giffel and van der Horst, 2004) or MF would reduce the level of bacteria and spores prior to the heat treatment used in production of ESL milk, or production of UHT milk. Bactofugation is not discussed in further detail in this chapter. However, Westfalia Separator Group claims that installation of

two separators in series before milk separation will remove up to 90% of total bacteria before pasteurization and reduce *Bacillus cereus* spore counts to less than one spore in 10 ml of milk. The milk is claimed to have a shelf life of 20 days with taste judged similar to that of high temperature, short time (HTST) pasteurized milk.

ESL milk, manufactured using MF followed by HTST pasteurization, commercially microfilters only skimmed milk because of the overlap in sizes between bacteria and spores and the milk fat globules as shown in Table 1.1. The MF equipment is installed after the separator and before the pasteurization step. Membrane pore size is typically 1.4 μm but a 0.8 μm membrane may be used. The retentate, which can range in volume from approximately 0.5% of the skimmed milk feed at VCR of 200, to 5% for VCR of 20, contains somatic cells and the bulk of bacteria removed from the skimmed milk when concentrated (Elwell and Barbano, 2006; Hoffman *et al*., 2006). Typical operating conditions for milk MF using a 1.4 μm membrane in the UTP process at 50°C are TMP of about 50 KPa, CFV from 6 to 9 m/s and average J of 500 LMH for a 10 hour run (Saboya and Maubois, 2000). Few studies of operating conditions for the Isoflux or GP membranes have been reported for milk MF (Fritsch and Moraru, 2008; Tomasula *et al*., 2011); but with the exception of higher values of TMP, which may range from about 50 to 200 KPa depending on CFV, operating conditions and J are within those reported by Saboya and Maubois (2000) for the UTP process. Caplan and Barbano (2013) reported a shelf life of 90 days for skimmed milk and 2% fat milk prepared by MF using a 1.4 μm membrane followed by HTST pasteurization at 73.8°C for 15 seconds.

Several potential plant configurations incorporating the MF process in an existing HTST fluid milk process are possible. The retentate may be added to the cream stream, heat treated at 130°C for four seconds and then added to the skimmed milk permeate followed by homogenization and HTST pasteurization. Alternatively, cream may be added to the permeate stream with high-temperature-treated retentate added to the surplus cream, which undergoes heat treatment at 130°C for four seconds. The retentate may also be directed back to the separator (te Giffel and van der Horst, 2004) or may be treated by UHT heating at 143°C for 1.1 seconds and discarded or used elsewhere (Hoffman *et al*., 2006). Other variations of this process have been reported, such as addition of another MF stage with VCR of 10 to follow the first stage with VCR of 20 to reduce the volume of the retentate stream to about 0.5% of the feed. In this case then, the retentate would be 200× concentrated and would not be used. Other variations of a MF milk process have been proposed by Hoffmann *et al*. (2006) which did not use the retentate to make ESL milk.

MF using a 1.4 μm membrane produces a permeate free of somatic cells with log reductions of bacteria averaging from 2.6 to 5.6 (Trouvé *et al*., 1991; Pafylias *et al*., 1996; Elwell and Barbano, 2006). Log reductions of bacterial spores of 2–3 log were reported in the permeate (Elwell and Barbano, 2006; Hoffman *et al*., 2006). While the permeate is free of somatic cells, the retentate may contain only about 25% of their original number (Elwell and Barbano, 2006), possibly due to the exposure to shear forces on the retentate side caused by the membrane itself or by the pump as it recycles the retentate stream through the membrane.

A milk MF process using a 0.8 μm membrane was proposed by Lindquist (2002) and became known as the Tetra Pak Ultima process (Maubois, 2011). MF conducted at 50°C with J of 400 l/m^2/h resulted in a sterile permeate with a decimal reduction of 13 (calculated from cellular volume) for *Clostridium botulinum* and a decimal reduction of nine for *Bacillus pumilus*. The milk permeate was aseptically mixed with

UHT-treated cream for fat standardization. This was followed by homogenization and then heat treatment of the stream at 96°C for six seconds to inactivate endogenous milk enzymes. The milk product was packaged aseptically and had a reported shelf life of 180 days at 20°C. The process was not commercialized.

Skrzypek and Burger (2010) reported that there are more than 10 commercial milk MF lines in Austria, Germany and Switzerland, installed in standard HTST pasteurization lines, processing from 15 000 to 35 000 l/h. The shelf life of the milk is from 20 to 25 days. In these plants, the skimmed milk stream is microfiltered using Isoflux membranes with a pore size of 1.4 μm and the cream stream is heated to a maximum of 135°C after addition of the retentate. The cream/retentate stream is added to the permeate and then pasteurized to produce the ESL milk. Bacteria removal from 60 000–160 000 to fewer than 10 counts/ml, corresponding to a log reduction of 4.20–3.78 $\log_{10}$/ml, was reported. Aerobic spore counts were from 40 to 210 spores/ml milk, corresponding to 1.60–2.32 $\log_{10}$ spores/ml, prior to MF/HTST pasteurization with only 3/35 analyses showing 1 spore/ml remaining in the permeate.

Schmidt *et al*. (2012) investigated the biodiversity of ESL manufactured by MF followed by HTST pasteurization and stored at 4, 8 and 10°C to investigate changes in bacterial counts, microbial diversity and enzyme quality. Biodiversity analyses were also conducted for samples from five manufacturers of commercial ESL milk at the end of shelf life. Even though MF reduced microbial logs by about 6 $\log_{10}$ CFU/ml, bacterial counts ranged from <1 to 8 $\log_{10}$ CFU/ml, with 8% of samples showing spoilage indicated by counts >6 $\log_{10}$ CFU/ml. The spoilage groups of bacteria were identified as post-process contaminants that included *Sphingomonas*, *Psychrobacter*, *Chryseobacterium* and *Acinetobacter*, and the spore formers *Bacillus cereus* and *Paenibacillus*, which caused enzymatic spoilage and off-flavours. Only three out of 13 isolates were identified as psychrotolerant genotypes. Overall, discrepancies in microbial loads and microflora varied even in samples of the same production run. The authors attributed this to stochastic variation of initial species in the milk packages arising from the low numbers of bacterial counts after ESL treatment. Thus, different bacterial populations would be observed during cold storage as well as occasional growth of high numbers of pathogenic species.

It is often assumed that milk MF is a nonthermal process. Even though MF is conducted over the temperature range 40–55°C, it is also assumed that energy use and the associated carbon dioxide (CO_2) emissions of MF followed by HTST pasteurization at a lower pasteurization temperature are less than that for HTST pasteurization conducted at a higher temperature alone. Using a computer simulation model of the fluid milk process developed recently (Tomasula and Nutter, 2011; Tomasula *et al*., 2013, 2014), a fluid milk process with MF/HTST (Figure 1.6) was simulated for comparison of energy use, greenhouse gas (GHG) emissions and operating costs to retrofit a plant using HTST pasteurization alone. The assumed production rate was 27 000 l/h of milk. The MF section was modelled assuming two MF processing modules in series, with each containing housings for 1.4 μm membranes. The first MF module was fed by milk at 55°C leaving the separator. The retentate from the first module fed the second module. The VCR of the first module was assumed to be 20 and that of the second 10, for an overall VCR of 200. Permeate from both modules was combined with cream, homogenized and then HTST pasteurized at 72°C to produce whole milk. The retentate, which was about 0.5% of the total feed stream, was processed as waste. Results showed that electricity and natural gas use for the MF/HTST process, which extended from the milk silos to cold storage at the plant, were 0.16 and 0.13 MJ/L, respectively,

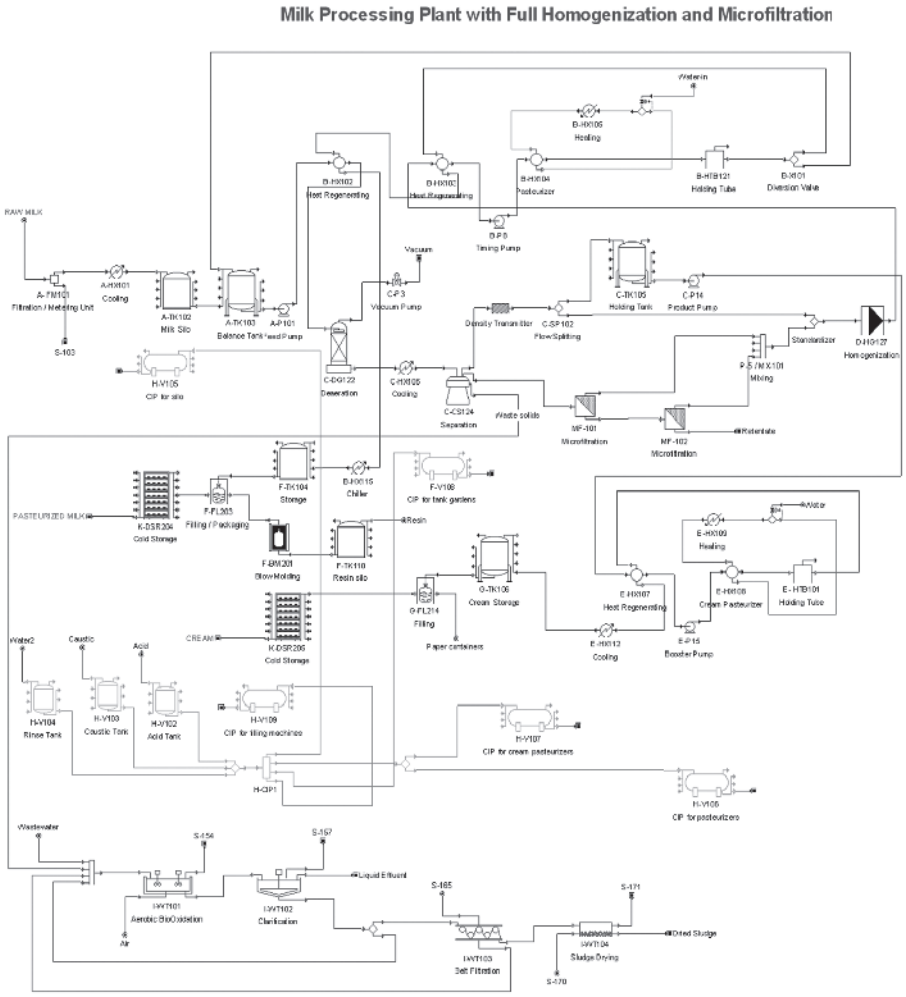

Figure 1.6 Milk Processing Plant with Full Homogenization and microfiltration.

and the carbon footprint of milk was 40.7 g CO_{2e}/kg milk. For HTST pasteurization alone conducted at 77°C, electricity and natural gas use were 0.14 and 0.13 MJ/l, and the carbon footprint of milk was 37.6 g CO_{2e}/kg milk (Tomasula *et al.*, 2014). Addition of MF prior to HTST pasteurization increased electricity use. The natural gas use increased due to the lower regeneration efficiency, which resulted from the 0.5% retentate waste, which in turn lowered the overall flow rate of milk. Less energy is thus regenerated because of the lower flow rate, in what was assumed to be an existing pasteurizer. The difference in operating costs between the two scenarios was estimated as 0.10 US cents/l. This is within the range reported by Skrzypek and Burger (2010).

The longer shelf life of the MF/HTST products may compensate for the small increase in GHG emissions of 3.1 g CO_{2e}/kg milk noted for the process over HTST

pasteurization alone. Supply chain losses and waste as well as losses at the point of consumption may account for up to approximately 0.5 kg CO_{2e}/kg milk consumed due to the 12% losses at retail and an additional 20% loss from cooking, spoilage and waste due to consumer practices (Thoma *et al*., 2013) it is conceivable that MF/HTST products would reduce retail and consumer losses due to their extended shelf life.

French regulatory authorities permit the sale of microfiltered whole milk, also known as ESL raw milk or Marguerite milk, which is not pasteurized. In this case, the cream is heat treated (95°C, 20 s), mixed with the permeate, lightly homogenized, filled aseptically and refrigerated at <6°C. The reported shelf life is about 15 days (Saboya and Maubois, 2000; Gésan-Guiziou, 2010). Using process simulation, the energy and natural gas usage for the MF process alone was 0.29 MJ/l with a hold time for cream of 15 seconds to ease comparison with the other models presented here. The GHG emissions were 41.4 CO_{2e}/kg milk. The increase in GHG emissions relative to the HTST pasteurization process was due to the increased electrical usage associated with MF. Natural gas use was 0.14 MJ/kg.

To get around the requirement of skimmed milk use for MF because of the overlap in sizes of the bacteria and spores with that of fat globules, a process was developed (Maubois, 2011; Fauquant *et al*., 2012) in which whole milk was homogenized twice prior to MF using a 0.8 μm Membralox membrane, at 50°C and J of 200 l/m^2/h in one example, to obtain fat globules smaller than 0.3 μm in the UTP process. This reduced the size of the fat globules so that they passed through the membrane with most bacteria retained. The fat standardized milk was pasteurized at 72°C with a 20 second hold time and has a shelf life of 30 days if stored at 4°C. If the fat standardized milk is heat treated at 96°C with a six second hold time, milk with a shelf life of 180 days at 20°C is obtained. Significant fouling was not observed after an eight-hour run. Milk flavour was reported identical to that of pasteurized milk. This process is not for production of a raw milk product. MF is followed by pasteurization, which is necessary to destroy the native lipase, which causes lipolysis in homogenized milk.

Quality and Safety The quality of heated milk is evaluated using the heat indicators lactulose, furosine, β-LG and lactoperoxidase (Lan *et al*., 2010). Lactulose is not present in raw milk but is formed upon heating through the isomerization of lactose. β-LG tends to decrease with heating due to denaturation while furosine, formed through the Maillard reaction, increases. Lactoperoxidase is used as an indicator to show that milk was heated over 80°C. Hoffman *et al*. (2006) measured the levels of several of these indicators for raw milk, skimmed milk, MF permeates and retentates, ESL milk treated by MF followed by HTST pasteurization and for the UHT-treated retentate. For the β-LG indicator, values of β-LG in the permeate were not affected by MF or the combination of MF and heat treatment but UHT treatment of the retentate resulted in a 90% decrease in β-LG. While furosine indicated that cream had been subjected to heat, the lactulose indicator showed values which agreed with HTST pasteurized milk.

In addition to achieving a longer shelf life than milk treated by HTST pasteurization alone, removal of somatic cells by MF prevents much of the lipolysis (the increase in free fatty acids) and proteolysis in milk associated with high or low somatic cell counts (Ma *et al*., 2000) although proteolysis, the breakdown of casein by plasmin was still observed due to native milk proteases, which cause off-flavour development (Santos *et al*., 2003; Elwell and Barbano, 2006). It was recommended that raw milk used in

MF should contain <100 000 somatic cells/ml to keep a low concentration of active plasmin, which passes through the microfilter and survives pasteurization.

MF followed by HTST pasteurization significantly reduces the microflora in milk (Trouvé *et al*., 1991; Pafylias *et al*., 1996; Elwell and Barbano, 2006; Tomasula *et al*., 2011); however, few studies have been conducted to determine the efficacy of MF in eliminating human pathogens of concern that have been occasionally reported in raw milk, such as *Listeria monocytogenes*, *Salmonella* spp. or *E. coli* 0157:H7, that are destroyed by pasteurization. Since these pathogens are rod-shaped with an approximate width of 0.5 μm and length of 1.5 μm (Garcia *et al*., 2013), MF would prevent most of these pathogens from entering the permeate, with pasteurization eliminating those in the permeate (Holsinger *et al*., 1997). Using the Bactocatch method, *Listeria monocytogenes*, *Salmonella Typhimurium*, *Brucella abortus* and *Mycobacterium tuberculosis* inoculated into skimmed milk were reduced by 3.4, 3.5, 4.0 and 3.7 $\log_{10}$, respectively (Madec *et al*., 1992; Saboya and Maubois, 2000). However, rod-shaped bacterial spores would survive pasteurization (Tomasula *et al*., 2011) and would either die off during cold storage or grow (Novak *et al*., 2005).

Few studies have determined the efficacy of MF in eliminating bacterial spores from milk using GP or Isoflux membranes. In a study examining methods to protect the milk supply from intentional addition of threat agents to milk prior to pasteurization, or to decontaminate milk, the impact of MF on removal of spores of *Bacillus anthracis* (BA) (Sterne) inoculated into raw milk was evaluated (Tomasula *et al*., 2011). The length of the spores ranged from 1.09 to 2.13 μm and diameter ranged from 0.66 to 1.09 μm (Carrera *et al*., 2007). Starting with raw milk inoculated with 6.5 $\log_{10}$ spores BA/ml, MF using a 0.8 μm membrane retained 5.91 ± 0.05 $\log_{10}$ spores BA/ml of milk and a 1.4 μm membrane retained 4.50 ± 0.35 $\log_{10}$ BA spores/ml of milk. The operating conditions for the 0.8 μm membrane were 50°C, CFV of 6.2 m/s, TMP of 127.6 KPa with an average J of 273 LMH. CFV for the 1.4 μm membrane was 7.1 m/s, TMP was 127.6, with an average J of approximately 200 LMH. Casein as a percentage of crude protein decreased 1.5% for the 1.4 μm membrane and 4.3% for the 0.8 μm membrane after 200 minutes of operation. For MF run times >10 minutes, either the 0.8 μm membrane (1.4 μm membrane not tested) or the associated pumping of the recycle stream appeared to contribute to sporulation of BA during cold storage, even though the milk was HTST pasteurized after MF. This observation would not be expected for MF of raw milk naturally containing very few spores/ml of other *Bacillus* species.

Head and Bird (2013) examined removal of psychrotropic spores from milk protein isolate (MPI) feeds ranging from 5 to 15% solids content. *Bacillus mycoides* spores were inoculated into the MPI feeds as a surrogate for spores of *B. cereus* and microfiltered using 0.8 and 1.4 μm GP membranes, 2 and 5 μm membranes without GP modification, and a 12 μm membrane comprised of a support layer only. The results showed that the 12 μm membrane at CFV of 1.4 m/s was best for the 10 and 15 wt-% high solids feeds, with spore reductions of 2.6 and 2.1 $\log_{10}$, protein transmission of 90% and 96.5% and J of 123 LMH and 27 LMH, respectively. Back-flushing was suggested as a method to improve J and protein transmission.

1.3.3 Cold Processing MF of Milk

The term 'cold pasteurization' often refers to milk MF when used to remove bacteria and spores from milk, to produce ESL milk, or when used prior to cheese making or manufacture of raw milk cheeses (Brans *et al*., 2004). It may also be associated with

MF when used as a pretreatment for skimmed milk in any dairy process to remove somatic cells and bacteria that may impact the quality and safety of the final product during prolonged storage, such as NFDM (nonfat dry milk) powder.

To attain shelf life and quality benefits beyond that of milk microfiltered at 50°C, MF processing of milk at temperatures <6.7°C to maintain the raw status of milk was studied (Fritsch and Moraru, 2008). Processing at these low temperatures may also avoid the potential problem of biofilm formation by bacteria that deposit on the large surface area of the membrane at the higher MF temperatures.

Vegetative cells, spores and somatic cells were removed at a CFV of 7 m/s, TMP of 60–80 kPa, and temperature of 6°C. J was approximately 50 LMH compared to 350 LMH for milk MF conducted at 55°C. To increase J at 6°C, a CO_2 back-pulsing technique was used; this increased J by 20% by clearing fouling in the outer membrane channels. Improvements to the back-pulsing technique, including the addition of injection ports around the membrane housing and the membrane, were suggested to improve J.

Pulsed electric fields (PEF) processing has also been combined with MF (1.4 μm membrane) for cold pasteurization of milk in two different sequences: MF prior to PEF (MF/PEF) and PEF prior to MF (PEF/MF) (Walkling-Ribiero *et al*., 2011) and is discussed in further detail in Chapter 5. Milk MF was conducted at 35°C with J of 660 LMH. MF/PEF processing of milk at a maximum temperature of 49°C resulted in a 4.8 $\log_{10}$ reduction in mesophilic aerobic counts of native microorganisms of milk. PEF/MF treatment of milk at a maximum processing temperature of 49°C resulted in a 7.1 $\log_{10}$ reduction of inoculated native microorganisms. The shelf life of PEF/MF milk and that of HTST pasteurized milk stored at 4°C was seven days. Although it would be intuitive to expect that the MF/PEF sequence would be more effective for reduction of microorganisms in milk, the authors believed that PEF may have induced agglomeration of the microorganisms resulting in a higher log count.

With the observation that low temperatures cause release of β-casein from the casein micelles, Woychik *et al*. (1992) applied microporous ultrafiltration of skimmed milk to facilitate removal of β-casein from milk. Flat plate PVDF membranes 0.1 or 0.2 μm in size were used. Casein/whey ratios of 0.7–0.9 were obtained in the permeates and ratios of 5–7 were obtained in the retentates. Higher amounts of α_{s2}-casein and lower amounts of β-casein were noted in the permeate than in the retentate. The retentate was suggested as a potential replacer for human milk. Van Hekken and Holsinger (2000) also applied this process to produce unique β-casein enriched milk gels with the potential to make simulated goats' milk cheeses. A milk MF process using SW membranes in which the permeate contains the whey proteins and β-casein was also developed (Lucey, 2012, Lucey and Smith, 2012). Suggested uses were for fortification of infant formula, cheese with improved meltability and bitterness, and using β-casein as a replacement for sodium caseinate in foaming and emulsification applications. Fractionation of whole milk was also found a possibility.

1.3.4 Separation and Fractionation of Milk Fat from Whole Milk or Buttermilk

Using MF as an alternative to centrifugation for separation of milk into skimmed milk and cream fractions may result in less damage to the fat globule membranes at low CFV and lead to cream with improved stability (Brans *et al*., 2004). The process may also be more energy efficient than centrifugation.

While processes for isolation of the native casein and whey components have been commercialized, processes for separation of the milk fat globules according to their size are still in development. The milk fat globules range in size from 0.1 to 15 μm and average 3.4 μm in size (Table 1.1). The effects of differences in compositions of the individual milk fat globules, their contribution to the functional properties of foods and their role in health and nutrition are not well understood (Singh, 2006).

Goudéranche *et al*. (2000) used whole milk MF with a 2 μm 'special' ceramic membrane to prepare two fractions containing the larger and smaller milk fat globules without damaging the milk fat globule membrane (MFGM). Michalski *et al*. (2006) optimized milk fat MF using membranes with pore sizes of 2–12 μm. The sizes of the globules were found to affect the properties of cheeses (Michalski *et al*., 2003, 2007), with smaller milk fat globules found useful in preparation of products with a finer texture. Using a similar MF technique optimized to select for small milk fat globules (about 1.6 μm) and large milk fat globules (about 6.6 μm), Lopez *et al*. (2011) determined that differences in composition varied according to size. The smaller milk fat globules contained higher amounts of the polar lipids, lower proportions of phosphatidylcholine and sphingomyelin in the MFGM and differences in the distribution of fatty acids. It was hypothesized that the sizes of the milk fat globules may play a role in delivery of biologically active compounds in the gastrointestinal tract of infants.

MF using a 0.5 μm membrane has been employed to obtain the valuable MFGM from buttermilk, which is a richer source of MFGM than whole milk (Astaire *et al*., 2003; Morin *et al*., 2007; Jiménez-Flores and Brisson, 2008). MF of buttermilk powder, whey and whey cream powders, coupled with supercritical fluid extraction (SFE) was shown to concentrate the lipids for possible new ingredients (Spence *et al*., 2009a, 2009b, 2009c). The phospholipids (PL) were concentrated fivefold. Numerous health benefits are associated with the MFGM, which also appears to inhibit rotavirus activity (Fuller *et al*., 2013).

1.3.5 Separation of Milk Bioactive Compounds

MF membranes, either alone or in combination with other types of membranes and unit operations, have also been used to extract bioactive components from milk, colostrum and whey. These bioactives, which have the potential to affect human health, may be included in food or consumer products to promote health and well-being. The isolation of growth factors, such as the insulin-like IGF-I, IGF-II, epidermal growth factor EGF, transforming growth factors TGF-β1 and TGF-β2, the basic fibroblast growth factor bFGF and the platelet-derived growth factor PDGF, using UF or MF membranes have been described (Pouliot and Gauthier, 2006, Gauthier *et al*., 2006, Akbache *et al*., 2009). Growth factors have been suggested for treatment of inflammatory gastrointestinal disorders, wound healing, bone tissue regeneration and skin diseases. Ollikainen *et al*. (2012) found that the ultimate TGF-β2 growth factor recovery was 83% if pasteurized milk was used in the initial MF step and 93% if nonpasteurized milk was used. Ben Ounis *et al*. (2010) used MF to separate TGF-β2 from WPI using a 0.8 μm membrane. Adjustments in pH and ionic strength and addition of λ-carrageenan facilitated removal of the growth factor, which was also enriched in immunoglobulins.

Manufacture of bioactive peptide-rich concentrates from whey is also a possibility using MF as a first step to reduce microbial contamination. Use of MF avoids

the potential change in bioactivity of the compounds that occurs with heat treatment (Tavares *et al*., 2012).

1.3.6 Other Applications

Skrzypek and Burger (2010) also reported the use of Isoflux MF membranes (1.4 μm) for bacteria and spore reduction of cheese brine, which is reused during the cheese making process for cheese salting. About 100% of mould, yeast and *E. coli* are removed with greater than 99.9% reduction in total bacteria reported. MF provides an environmentally-sound solution to disposal of the brine but also eliminates the more labour intensive methods used previously.

Use of MF is not limited to bovine milk. Beolchini *et al*. (2004, 2005) investigated the use of MF for reduction of bacteria in bovine and ovine milk. Extending the shelf life of milk from goats, sheep and other animals, such as camels, may be of interest, since they are usually available only in areas where they are produced. MF to produce ESL products would help satisfy consumer demand for these unique milk products and increase profits for those who produce them.

1.4 Membrane Modifications to Increase Performance

Brans *et al*. (2004) discussed the various strategies proposed in the literature to improve membrane performance through reduction of fouling. The methods include: vibrating modules, rotating disks, scouring particles and air slugs to improve shear at the membrane (Jaffrin, 2008; Ahmad *et al*., 2010; Espina *et al*., 2010); turbulence promoters (Popovic and Tekic, 2011), pulsating crossflow and use of ultrasound (Mirzaie and Mohammadi, 2012) to improve back transport near the membrane; and use of electric fields to repel charged particles from the membrane. Disadvantages to use of these approaches included high power consumption, high capital investment, difficulty cleaning, equipment wear and difficulty in scaling up.

1.5 Microsieves

Microsieve technology is a promising alternative to improving J over that of conventional milk MF using ceramic tubular or SW membranes. Microsieves are manufactured from silicon nitride or polymers. To manufacture silicon nitride microsieves, photolithographic technologies are used to produce silicon wafers with a thickness of 1 μm. According to one manufacturer (Sievecorp, Inc., http://www.sievecorp.com), microsieves come in the form of six-inch [15 cm] wafers that are assembled in stacks containing 45 sieves/stack. Each wafer can process 165 l/h of liquid and each stack processes 7500 l/h of liquid with maximum viscosity of 40 cp and particle load to be retained of less than 1 g/l. The microsieves are available in pore sizes of 0.35 or 0.35 μm slits, 0.45 and 0.45 μm slits, and 0.8 and 0.8 μm slits. Frequent back-pulsing is required to prevent fouling and to control concentration polarization effects. The advantage of microsieves over conventional membranes is their controlled pore-size and consistent morphology.

Brito-de la Fuente *et al*. (2010) determined the effects of process variables on MF of commercial UHT-treated whole milk using a pilot plant crossflow microsieve membrane system. Five-litre volumes of milk were processed at 40°C. The membranes had 0.8 μm slits and a surface area of 4 cm^2. Values of J from 5000 to 27,000 l/m^2/h, a minimum 10× greater than J reported for skimmed milk MF with ceramic membranes, were achieved with low TMP in the range from approximately 7 to 15 KPa. Run times of over two hours were possible. TMP and the frequency of back-pulsing were the most important variables to control fouling. Changes in the viscosity and particle size distribution of the components of the milk were not noted. In a comparison to HTST pasteurization, the energy demand of microsieves reported by the manufacturer was 30 kJ/kg versus a reported 220 kJ/kg for HTST pasteurization. Even though the energy demand reported for microsieves is low, this step would still need to be followed by a pasteurization step. Also, the capital and operating costs for microsieve MF were not reported.

Prior to experiments, the microsieves were pretreated to induce hydrophilicity of the hydrophobic silicon material. After a milk MF run, the microsieves were cleaned with an alkaline cleaning agent at 50°C and membrane integrity was tested before and after each experiment.

Girones i Nogue *et al*. (2006) reported the performance of polyethersulfone (PES) polymeric microsieves for skim milk MF at 7°C using a membrane with a pore size of 2 μm. PES microsieves, which were reported to have lower production costs than silicon microsieves, are manufactured using phase separation micromoulding and are available in pore sizes ranging from 0.5 to 5 μm. A crossflow module was used for experiments using an effective membrane area of 0.5×10^{-4} m^2 and channel height of 700 μm. Varying back-pulse frequencies were applied to prevent fouling. Operating pressure was reported as 2 KPa and J was reported as 1600 LMH. It was concluded that a smaller pore size membrane, which would retain bacteria, would still result in improved productivity relative to conventional MF. Back-pulsing was necessary to prevent J decline.

Milk proteins were reportedly not retained using either the silicon nitride or PES microsieves. The silicon and polymeric microsieves show impressive performance compared to crossflow milk MF. Microsieves may be the future of milk MF but research is needed on start-up/shut-down operations, sanitation and verification of quality and safety of milk after MF to validate their performance against conventional crossflow MF.

1.6 Conclusions

Although MF is a well-established technology, it has shown limited growth in the dairy industry compared to ultrafiltration. With advances in membrane manufacture, low fouling membranes are now available that ensure maintenance of the transmembrane pressure, and thus the permeate flux along the length of the membrane. Microfiltration, using membranes with pore sizes ranging from 0.1 to 10 μm, can be used for a variety of applications. Using the smallest pore sizes, concentrates of micellar casein and the whey proteins are possible, presenting the possibility of new beverages and ingredients on the market that can be used to exploit the functional properties and health benefits of these proteins. Membranes with various pore sizes are being explored for separation of the milk fat globules to manufacture dairy products

with improved texture and to produce individual fat globules and MFGM to better understand their role in health and nutrition. The biggest advantage of MF is its ability to allow for physical removal of microorganisms that lead to milk spoilage from milk. MF, used as a processing step prior to HTST or UHT heat treatment, has led to production of ESL milk and dairy-based beverages with improved organoleptic properties. The longer shelf life is an also an advantage because it allows for creation of products with targeted nutritional benefits for example, products targeted to particular segments of the population such as children, the elderly or for post-workout needs. The longer shelf life would also help further the sales and distribution of milk from other animals. In addition, production of ESL milk may help lower the greenhouse gas emissions associated with retail and consumer wastes of milk.

Increasing permeate flux would decrease operating costs attributed to electricity use. On the horizon are alternatives to tubular ceramic or SW membranes, such as microsieve technology, in which permeate fluxes exceeding 10 times that of conventional membranes have been demonstrated. Additional research is needed to develop membranes that are resistant to fouling and chemical and heat degradation, and with active and passive characteristics to improve their selectivity so that all essential nutrients in milk may be used.

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