Separation of casein micelles from whey proteins by high shear microfiltration of skim milk using rotating ceramic membranes and organic membranes in a rotating disk module

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A R T I C L E   I N F O

Article history:
Received 2 May 2008
Received in revised form 28 August 2008
Accepted 6 September 2008
Available online 17 September 2008

Keywords:
Milk microfiltration
Casein concentration
α-Lactalbumin and β-lactoglobulin extraction
Dynamic filtration

A B S T R A C T

This paper investigates the microfiltration of skim milk in order to separate casein micelles from two whey proteins, α-lactalbumin (α-La) and β-lactoglobulin (β-Lg), using a modified dynamic filtration pilot (MSD) consisting in 6 ceramic 9-cm diameter membrane disks of 0.2 μm pores, rotating around a shaft inside cylindrical housing. A comparison was made with another dynamic filtration module consisting in a disk rotating near a fixed PVDF 15.5 cm diameter membrane with 0.15 μm pores. Maximum permeate fluxes were 120 L h⁻¹ m⁻² with the MSD module at 1930 rpm and at 40°C, and 210 L h⁻¹ m⁻² at 2500 rpm and 45°C, with the rotating disk module. Casein rejection was around 99% at high speed for both membranes. α-La transmission decreased with increasing transmembrane pressure (TMP) from 75% to 60% for ceramic membranes and from 25% to 10% for the PVDF one. β-Lg transmissions were lower, ranging from 23% to 15% for ceramic membranes and from 20% to 5% for the PVDF one. In a concentration test with the PVDF membrane at 2000 rpm, the flux decayed from 200 L h⁻¹ m⁻² at initial concentration to 80 L h⁻¹ m⁻² at VRR = 3.2 and 22.1% of the initial α-La mass was recovered in the permeate, against 8.1% for β-Lg. Permeate fluxes in the mass transfer limited regime (J₂lim) of the MSD and rotating disk module operated at various speeds were well correlated by the equation J₂lim = 17.13 Vavo, where Vavo denoted the disk azimuthal velocity averaged over the membrane area. Measurements of J₂lim, taken from Ref. [G. Samuelsson, P. Dejlmek, G. Tragardh, M. Paulsson, Minimizing whey protein retention in crossflow microfiltration of skim milk. Int. Dairy J. 7 (1997) 237–242] during MF of skim milk using tubular ceramic membranes at velocities from 1.5 to 8 m s⁻¹ with permeate co-current recirculation were found to obey the same correlation.

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1. Introduction

The separation of casein micelles from whey proteins is important in several applications, as a concentrated casein solution can be mixed with cream for production of cheese, or for standardisation of milk and production of dried native casein used as food additives [1,2]. Whey proteins, which can be recovered from milk, have valuable physicochemical properties. For instance, α-lactalbumin (α-La) has pharmaceutical applications while β-lactoglobulin (β-Lg) can be used for emulsification, foaming and gelling. Lactoferrins are found in infant formulas and as meat preservative [2]. Unlike traditional methods such as acidification or coagulation, casein extraction by membrane microfiltration (MF) does not modify the casein structure. But it is well known that MF of milk induces severe fouling and requires generally combining very high membrane shear rates with low transmembrane pressure, in order to preserve whey protein transmission, as noted by several authors [3–6]. Daufin et al. [3] separated casein from whey proteins using 0.1 μm pore ceramic membranes (Exekia, 65460 Bazet, France) in a filtration bench equipped with co-current recirculation of the permeate in order to obtain a low uniform transmembrane pressure (UTP mode of operation) as in the Bactocatch system of Alfa Laval [7] proposed for bacteria removal from milk. They conducted tests at constant flux with a pump on permeate and obtained a whey protein transmission of about 70–80%. Géron-Guiziou et al. [6] used a similar ceramic membrane (Kerasep 0.1 μm, TechSep Miribel, France) and the same filtration bench in UTP mode as [3] and obtained fluxes at 50°C of about 80 L h⁻¹ m⁻² and an α-La transmission of 50–80%, but permeate turbidity was relatively high, at 100–200 NTU, corresponding to a 80% casein rejection. Casein concentration tests were also made by Vadi and Rizvi [8], using a 0.2 μm ceramic Membralox multi-channel membrane from Exekia.
with 3.7 mm inner diameter, both in UTP and non-UTP modes. They reported that the non-UTP mode gave higher flux up to a concentration factor (CF) of 4, while the UTP mode performed better at higher CF. They also observed that the cake formed during MF in non-UTP mode was more difficult to erode than the cake produced under UTP conditions. They estimated the optimal crossflow velocity to be 7.1 m s\(^{-1}\) for their membrane. Samuelson et al. [9] used a 0.14 μm ceramic tubular membrane (TechSep) for casein concentration in skim milk while minimizing whey protein retention by using crossflow velocities up to 8 m s\(^{-1}\). They obtained a maximum flux of 145 L h\(^{-1}\) m\(^{-2}\) at a speed of 8 m s\(^{-1}\) and 55°C which fell to 80 L h\(^{-1}\) m\(^{-2}\) at 4 m s\(^{-1}\). Whey protein transmission was 88% at 8 m s\(^{-1}\) and 74% at 6 m s\(^{-1}\), but casein retention was only 90%. A recent investigation of casein concentration by MF using polymeric membranes was made by Lawrence et al. [10] who used 0.3 and 0.5 μm pore PVDF membranes, both in a flat sheet laboratory module and in a spiral wound industrial pilot in non-UTP mode. They observed a casein rejection which increased from 96% at a TMP of 50 kPa to 98% at 150 kPa and 100% at 258 kPa. The β-Lg transmission decreased from 22% at 50 kPa to 8% at 150 kPa and 1% at 258 kPa. In the flat sheet module at 50°C and a velocity of 0.44 m s\(^{-1}\), the permeate flux decayed from 60 to 52 L h\(^{-1}\) m\(^{-2}\) over a period of 2 h. In the spiral module at same velocity and 40°C, the flux remained steady with time, at near 32 L h\(^{-1}\) m\(^{-2}\).

This literature survey has stressed the importance of using high shear rate and low uniform TMP in milk MF, in order to obtain both a good casein rejection and acceptable whey protein transmission. These requirements could only be met in crossflow filtration by a co-current recirculation of the permeate together with a high crossflow velocity of 7 to 8 m s\(^{-1}\). Our laboratory has previously shown [11–13] that dynamic filtration, using a vibrating VSEP system, or metal disk rotating at high speed near a fixed circular membrane, could be well suited to milk MF and UF, as it yielded very high shear rates, in excess of 10\(^5\) s\(^{-1}\) with low feed flow rates and relatively uniform TMP.

Since most tests in MF of milk have been performed with ceramic membranes, which seem to be preferred to organic membranes for this application, we have used in this work a new multishaft disks dynamic filtration pilot (MSD) with ceramic membrane disks rotating inside a housing (Westfalia Separator, Aalen, Germany). For comparison, we have also used a rotating disk system designed in our laboratory equipped with a fixed organic MF membrane. The goal was to find the best compromise between high permeate flux, good α-La and β-Lg transmissions and high casein rejection.

### 2. Material and methods

#### 2.1. MSD pilot

The MSD pilot, described in [14], consists of two parallel hollow shafts rotating at the same speed, each one normally bearing six ceramic membrane disks and enclosed in a stainless steel housing (Fig. 1). However, only six membranes on the same shaft were used in these tests. The permeate was collected inside each disk by flat hollow channels. Actual TMP could not be measured in the MSD, but was calculated from measurements of operating pressure \(p_c\) at a pressure tap in the housing, close to disk periphery, using a Validyne DP 15 pressure transducer (Validyne Corp., Northridge, CA, USA) as described in [14], as

\[
\text{TMP} = p_c - \frac{\rho \omega^2 (R_1^2 + R_2^2)}{4}
\]

where \(R_1 = 4.5\) cm and \(R_2 = 1.02\) cm are, respectively, the outer and inner radius of membrane disks.

#### 2.2. Rotating disk module

The rotating disk system module, shown in Fig. 2, has been described previously in [15]. It is equipped with a single-organic membrane, of 188 cm\(^2\) area (outer radius \(R_1\): 7.75 cm, inner radius \(R_2\): 1.75 cm) was fixed on the cover of the cylindrical housing in front of the disk. The disk can rotate at adjustable speeds, ranging from 500 to 2500 rpm. It can be smooth or equipped with eight 6 mm-high vanes in order to increase the core fluid angular velocity \(ka\) between the membrane and disk where \(\omega\) is the disk angular velocity. The maximum membrane shear rate at disk rim

![Fig. 1. Schematic of MSD pilot and data acquisition system.](image-url)
(R_d = 7.25 cm) has been calculated in [15] to be

\[ \gamma_{\text{max}} = 0.0296 \rho_d^{8/5} k_w^{9/5} \nu^{-4/5} \tag{2} \]

The peripheral pressure (p_c) was measured at the top of the cylindrical housing by a Validyne DP15 pressure transducer and inlet pressure by a Bourdon Haenni manometer. Values of velocity factor k were obtained from measurements of p_c at different speeds and found to be 0.89 for a disk equipped with 6 mm vanes [16]. The pressure is adjusted by acting a valve on outlet tubing. The transmembrane pressure (TMP) was then determined in [15] as

\[ \text{TMP} = p_c - \frac{1}{4} \rho k^2 \omega^2 R^2 \tag{3} \]

where R is the inner housing radius.

2.3. Membranes and cleaning procedure

Ceramic membranes used in the MSD were made by Westfalia Separator and had a nominal pore size of 0.2 μm according to the manufacturer. Their hydraulic permeability was determined by measuring the permeate flux of demineralized water at various TMP and found to be 186 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) at 20 °C, corresponding to a membrane resistance (R_m) of 6.0 × 10\(^5\) m\(^{-1}\). The membrane in the rotating disk system was made of PVDF with 0.15 μm pores and obtained from Alfa Laval Nakskov, Stavangervej, Denmark. Its initial hydraulic permeability, measured with demineralized water, was 92 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) at 20 °C (R_m = 3.0 × 10\(^5\) m\(^{-1}\)). According to supplier recommendation, this membrane was dipped in an ethanol solution for 1 h and washed with demineralized water before use.

After a test, ceramic and organic membranes were rinsed with demineralized water and cleaned with a P3 Ultrasil 10 (Ecolab) solution at 0.5% and 40 °C for 1 h in closed circuit and 5 min in open circuit in order to empty the system. Then the circuit was rinsed with demineralized water until the initial pH of 7.0 was recovered. Hydraulic permeabilities of membrane were measured after successive cleaning. They varied from 273 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) after 1st cleaning to 330 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) after 3rd cleaning for ceramic membranes and for PVDF membranes, from 129 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) after the 1st cleaning to 206 L h\(^{-1}\) m\(^{-2}\) bar\(^{-1}\) after the 3rd one. This was probably due to adsorption of surfactant into the membrane which leads to a less hydrophobic surface than the new membrane [17].

2.4. Test fluid

Since fresh skim milk was not locally available, the test fluid was a commercial UHT skim milk (Carrefour, France) with the following composition: casein 25.6 g L\(^{-1}\), whey proteins 6.4 g L\(^{-1}\), including 0.40 ± 0.05 g L\(^{-1}\) of α-La and 0.16 ± 0.02 g L\(^{-1}\) of β-Lg, lactose 46 g L\(^{-1}\) and calcium 1.2 g L\(^{-1}\). Although UHT milk has a smaller whey protein concentration than fresh milk, Akoum et al. [18] found that its filtration characteristics were identical to those of low-heat milk with normal whey protein contents and whey proteins transmissions of UHT milk should be representative of those with fresh milk.

2.5. Experimental protocol

Both modules were fed from a thermostated and stirred tank containing 5 L of milk by a volumetric diaphragm pump, and the permeate was collected in a beaker placed on an electronic scale (Sartorius B3100 P, Gottingen, Germany) connected to a computer in order to measure the permeate flux. Tests were conducted at 40 °C for the MSD and 45 °C for the rotating disk module with complete recycling, except for a concentration test with the rotating disk module. The feed flow rate Q_i was set to 3 L min\(^{-1}\) in all tests. Most tests were performed with permeate and retentate recycling in order to investigate the effect of TMP, rotation speed and time at constant protein concentrations. The rotation speed was first set to its maximum value while TMP was increased in steps of 40 kPa lasting 25 min, from 20 kPa to a maximum varying from 180 to 300 kPa. Then, the TMP was lowered back to 20 kPa and the rotation speed set to the next lower value. Samples were collected in permeate 20 min after the beginning of each TMP increment in order to have stabilized flux and transmission conditions. Casein rejection (R) and whey proteins transmission (Tr) were calculated, respectively, from

\[ R = 1 - \frac{C_p}{C_i} \tag{4a} \]
\[ Tr = \frac{C_p}{C_i} \tag{4b} \]

where C_p denotes the permeate protein concentration and C_i the feed one.

2.6. Analysis

Retentate and permeate turbidities were measured with a Hach turbidimeter (Colorado, USA). A calibration of casein concentration (C) versus turbidity (T) in NTU was made using suspensions of caseins from bovine milk (Sigma–Aldrich, Germany) in a...
1 M Na₂HPO₄ buffer and gave the following relation, valid until $T = 100$ NTU

$$C = 0.0534 \cdot T \quad R^2 = 0.997$$  \hspace{1cm} (5)

Eq. (5) was used to calculate the casein rejection from Eq. (4a). In fact, as other elements of permeate, such as whey proteins, contribute to the turbidity, the permeate casein concentration using Eq. (5) is overestimated and its rejection underestimated. By precipitating the caseins at a pH of 4.6 with 1 M acetic acid in a 30 ml sample and filtrating the supernatant at 0.45 μm, we have estimated the turbidity due to caseins to be 70% of that measured. Since rejections were very high, above 98%, this correction was small and was not used in the graphs.

α-La and β-Lg concentrations were measured in 5 mL samples collected at the permeate outlet of the module by HPLC, using a Waters 510 chromatograph, a UV detector at 280 nm and a Vydac-C4 column thermostated at 40 °C, according to the method of Jaubert and Martin [19]. A calibration curve was made using pure α-La and β-Lg samples from bovine milk (Sigma–Aldrich, Germany) of known concentrations measurements. Two solvents were used for HPLC analysis, Trifluoroacetic acid (TFA) at 0.1% in milli-Q water (Sigma–Aldrich), and acetonitrile at 80% with 0.096% TFA and 20% milli-Q water (Prolabo, France). Before injecting a sample into the column, it was pretreated in order to precipitate caseins. Its pH was lowered to 4.3 with acetic acid (10%, W/V) and restored to 4.6 after 5 min with a 1 M sodium acetate solution. After heating the sample for 20 min, the supernatant was filtered using a 0.45 μm, Minisart RC 45 (Sartorius) syringe filter, before injection into the HPLC. A standard chromatogram of α-La and β-Lg protein is shown in Fig. 3. The two protein peaks can be clearly seen on the graph, the first peak corresponds to α-La and the 2nd and 3rd to β-Lg. Proteins concentrations in g L⁻¹ were calculated from

$$C_{\alpha-La} = 6 \times 10^{-7}A$$  \hspace{1cm} (6a)

$$C_{\beta-Lg} = 10^{-6}A$$  \hspace{1cm} (6b)

where $A$ denotes the area in mm² under the corresponding peaks of the chromatogram. Measurement error was estimated to be 5%.

3. Results

3.1. Tests with MSD pilot

Fig. 4 presents the variation of permeate flux with time in response to 40 kPa TMP increments from 20 to 180 kPa, at three rotation speeds of 1492, 1044 and 745 rpm. In addition, filtration was carried out at 1044 rpm and a TMP of 27 kPa for 50 min before these tests and for 20 min after, in order to check the amount of flux decline over the duration of the experiment which lasted 7 h. Since at each change in speed the TMP was lowered to zero by opening the retentate valve, the flux decreased sharply before rising again during the next 25 min. It can be seen that the permeate flux increased very little above a TMP of 60 kPa at the two higher speeds and decayed slowly with time at 745 rpm, probably due to membrane fouling. This fouling is confirmed by the observation that the flux at 27 kPa has declined after 7 h to 10 L h⁻¹ m⁻² from an initial value of 50 L h⁻¹ m⁻². It is then possible that the apparent flux stability with pressure above 60 kPa is due to a compensation of flux decline by the rise in TMP. Fig. 5 displays the values of stabilized fluxes as a function of TMP for the tests of Fig. 4, together with another test performed at 1931 rpm, without the initial 50 min filtration period at 1044 rpm of the other tests. It can be seen that the maximum flux increased proportionally to rotation speed $N$, as will be confirmed in Section 4.

Fig. 5. Variation of stabilized permeate flux versus TMP with the MSD at different rotation speeds for tests of Fig 4 and one at 1930 rpm.
The variation of permeate turbidity due to casein with TMP during tests of Fig. 5 is presented in Fig. 6. These turbidities are very small in comparison of milk turbidity and correspond to a casein rejection larger than 97.5%. For the first test performed at 1492 rpm, this turbidity increased with increasing TMP, as more casein micelles are forced into the largest pores by pressure. In the 2nd test at 1044 rpm, the turbidity, on the contrary, decreased at large pressure, due to the growth of casein layer with time which decreases the casein transmission. Permeate turbidity was smallest for the last test which had the highest fouling and thickest casein layer.

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The transmission of α-La is depicted in Fig. 7. It is highest (85%) at the beginning of the experiment at 1492 rpm and for a TMP less than 60 kPa, when membrane fouling was probably still small. Above 60 kPa, the transmission decayed slowly with increasing TMP from 70% towards 60%, 50% and 45%, respectively, for 1492, 1044 and 745 rpm. β-Lg transmission, also shown in Fig. 7, was lower than that of α-La which has a lower molecular weight. At 1492 rpm, it decayed with increasing TMP from 22% to 13% and fluctuated between 10% and 25% at 1044 rpm.

In order to improve whey protein transmission and operate closer to industrial conditions, another test was carried out with full recycling at a rotation speed of 1044 rpm and a TMP of 60 ± 2 kPa, corresponding to the beginning of limiting flux, when it becomes pressure independent. After a small rise from 53 L h⁻¹ m⁻² corresponding to the rise in temperature from 33 to 40 °C during the first hour as the milk was heated by the thermostatted tank until it reached an equilibrium temperature, the flux remained constant, at about 58 L h⁻¹ m⁻² until the end of the test which lasted 5 h, confirming that membrane fouling was small at low TMP. Corresponding variations of permeate turbidity and casein rejection with time are shown in Fig. 8. The initial rise in turbidity is due to the rise in temperature and its decay after 90 min may be explained by the formation and the packing under pressure of a casein layer of the membrane which increases its rejection. α-La transmissions, also given in Fig. 8, fluctuate around 60%, like in the tests of Fig. 7, while β-Lg transmissions are higher, between 50% and 40%, than those of Fig. 7, due probably to a lower rate of internal fouling. These results show that no protein denaturation occurred, as protein concentrations were stable with time.

The total final mass of α-La and β-Lg proteins in permeate and retentate was measured in two of these tests and was found to be 3–4% less than their initial mass, due probably to loss in collected samples and by adsorption on the membranes.

3.2. Tests with rotating disk module and PVDF membranes

3.2.1. Tests with full recycling (VRR = 1)

The variation of permeate fluxes versus TMP obtained with a 0.15 μm pores PVDF membrane and a disk equipped with vanes rotating a 2000 rpm is shown in Fig. 9 for an initial test with a new membrane and successive tests performed later after membrane cleaning, denoted as 1st, 2nd and 3rd reuse, respectively. The limiting permeate flux, when it becomes pressure independent, increased significantly with successive reuses from a maximum of 150 L h⁻¹ m⁻² for the new membrane to about 200 L h⁻¹ m⁻² at the 2nd reuse. This may be due to the surfactant adsorption into the membrane discussed earlier. However, no further gain in flux was observed after the 2nd reuse. These fluxes were higher than with the ceramic membranes, due to the higher disk peripheral velocity, αR₁, 16.2 m s⁻¹ against 9.1 m s⁻¹ for the MSD at 1930 rpm, according to Table 1. Similar increases in permeate flux after membrane cleaning and reuse were observed at speeds of 2500 and 1500 rpm, and maximum permeate fluxes are listed in Table 1. Here again, no further increase in flux was observed after the 2nd reuse. Corresponding α-La transmissions are shown in Fig. 10 at 2000 rpm after 1–3 reuses. This transmission was largest at the 2nd reuse and decayed with increasing TMP from 35% to 22%. In the 1500 rpm test, not shown here, it decayed from 32% to 15% with increasing TMP. β-Lg transmissions, also shown in Fig. 10, were also maximal during the 2nd reuse and decayed with TMP from 15% to 10% at 2000 rpm as TMP increased from 150 to 225 kPa. The variations of permeate turbidity and casein rejection are also shown at the 3rd reuse for rotation speeds of 2500, 2000 and 1500 rpm in Fig. 11. Casein rejection was minimal and increased with TMP at the highest speed (2500 rpm) and remained independent of TMP and larger than 99% for 2000 and 1500 rpm.
Table 1
Membrane-averaged velocities and limiting fluxes at various rotations speeds for the MSD pilot (corrected at 45° C) and the rotating disk module.

<table>
<thead>
<tr>
<th>Membrane</th>
<th>Rotation speed (rpm)</th>
<th>Peripheral velocity (m s⁻¹)</th>
<th>Vav (m s⁻¹)</th>
<th>Jlim (L h⁻¹ m⁻²) at 45° C</th>
</tr>
</thead>
<tbody>
<tr>
<td>MSD pilot</td>
<td>1930</td>
<td>202</td>
<td>9.1</td>
<td>6.93</td>
</tr>
<tr>
<td></td>
<td>1492</td>
<td>156</td>
<td>7.0</td>
<td>5.33</td>
</tr>
<tr>
<td></td>
<td>1044</td>
<td>109.3</td>
<td>4.9</td>
<td>3.75</td>
</tr>
<tr>
<td></td>
<td>745</td>
<td>78</td>
<td>3.5</td>
<td>2.67</td>
</tr>
<tr>
<td>Rotating disk</td>
<td>2500</td>
<td>262</td>
<td>20.3</td>
<td>14.10</td>
</tr>
<tr>
<td>module</td>
<td>2000</td>
<td>209.4</td>
<td>16.23</td>
<td>11.26</td>
</tr>
<tr>
<td></td>
<td>1500</td>
<td>157</td>
<td>12.17</td>
<td>8.45</td>
</tr>
</tbody>
</table>

Fig. 10. Variation of α-La and β-lg transmissions with TMP for the tests of Fig. 9.

3.2.2. Concentration test
A concentration test was carried out with a cleaned membrane without permeate recycling on an initial volume of 5 L of milk at a rotation speed of 2000 rpm and a TMP of 166 kPa with the 0.15 μm PVDF membrane until a volume reduction ratio (VRR) of 3.2. The variation of permeate flux with VRR is displayed in Fig. 12. This permeate flux decayed approximately linearly with increasing VRR, as it was not completely in the mass transfer limited regime, from a maximum of 200 L h⁻¹ m⁻² to a minimum of 80 L h⁻¹ m⁻² at VRR = 3.2. Final α-La and β-lg concentrations in the cumulated permeate of 3.49 L were, respectively, 0.127 and 0.019 g L⁻¹ corresponding to recovered whey protein masses of 0.443 g or 22.1% of initial mass for α-La, and 0.0663 g or 8.1% of initial mass for β-lg. These percentages would have been larger with a higher final VRR.

4. Discussion
Our data showed that the 0.2 μm ceramic membranes gave an excellent casein rejection, and a better α-La transmission than the 0.15 μm PVDF membrane. β-Lg transmission was similar for both membranes, but fluxes were higher with the rotating disk module, because of its higher peripheral velocity. Since, in the mass transfer limited regime, the permeate flux is governed by the membrane shear rate, itself function of the mean fluid velocity relative to the membrane, we have plotted in Fig. 13 the values of maximum stabilized permeate fluxes (Jlim), corresponding to the 2nd reuse for the rotating disk, and those obtained with the MSD, as function of azimuthal disk velocity averaged over the membrane area (Vav). Since both ceramic and organic membranes were annular with an outer diameter R₁ and an inner one R₂, values of Vav are given by

$$V_{av} = \frac{2\pi}{\pi(R_1^2 - R_2^2)} \int_{R_2}^{R_1} r^2 \omega \, dr = \frac{2\pi(R_1^3 - R_2^3)}{3(R_1^2 - R_2^2)}$$ (7)
Using values of $R_1$ and $R_2$ given in the material and methods section, one finds for the MSD module,

$$V_{av} = 0.7615R_1 \omega,$$  \hspace{1cm} (8a)

while for the rotating disk module

$$V_{av} = 0.6943R_1 \omega,$$  \hspace{1cm} (8b)

Their values summarized in Table 1. We also have included in Fig. 13 values of permeate fluxes obtained by Samuelson et al. [9] in MF of using a 0.14 μm tubular ceramic membrane at various mean fluid velocities ($V$) in UTP mode. $V$ is uniform over the membrane area and is equivalent to $V_{av}$. Since their test were performed at 55 °C, we have multiplied their fluxes by the ratio of milk viscosities at 55 and 45 °C $\mu_{55}/\mu_{45} = 0.845$, and MSD fluxes by $\mu_{40}/\mu_{45} = 1.093$, as Frappart et al. [20] have observed that permeate fluxes in the mass transfer limited regime were inversely proportional to fluid viscosity. These permeate fluxes at various velocities are listed in Table 2. Fig. 13 shows that all fluxes in L h$^{-1}$ m$^{-2}$ from the three systems are proportional to $V_{av}$ and well correlated by

$$J_{lim} = 17.13V_{av} \quad R^2 = 0.980$$  \hspace{1cm} (9)

The MSD point corresponding to 1930 rpm is a little above the straight line in Fig. 13, but, as seen earlier, this is due to the fact that this measurement was made after only 30 min of filtration instead of 75 min at the lower speeds. It is interesting to find that permeate fluxes of three different systems obey the same linear function of the membrane averaged fluid velocity relative to the membrane over a wide range of velocities from 1.5 to 14 m s$^{-1}$. Data of [9] taken at high velocity would have probably been below the correlation, if they had not been obtained in UTP mode. If this result is general and holds for other fluids, it is important as the membrane averaged fluid velocity is much easier to calculate than the membrane shear rate, which in the case of MSD module, was not known.

While casein rejection was high, both with the MSD ceramic membranes and the PVDF one, $\alpha$-La transmission was between 60% and 75% with the MSD, but lower with the PVDF membrane. $\beta$-Lg transmission fluctuated between 40% and 50% with the MSD in the test shown in Fig. 9, carried out at a constant TMP of 60 kPa, but was lower, between 15% and 20%, in tests carried out at various TMP and rotation speeds for both membranes, which were not ideal for optimizing whey protein transmission. This confirms the importance of operating at a TMP corresponding to the onset of the limiting flux to reduce internal fouling in MF of milk. It must be noted that these transmissions were lower than total whey protein transmission, which is generally quoted in the literature, as other whey proteins have smaller molecular weights than $\alpha$-La and $\beta$-Lg. Gésan-Guiziou et al. [6] obtained higher transmissions $\alpha$-La and $\beta$-Lg in UTP mode with a 0.1 μm ceramic membrane, but with a low casein rejection (80%).

Another advantage of MSD industrial modules is that they do not need large pumps for recirculation of retentate and permeate at high speeds as in UTP systems. Thus, according to the manufacturer, their energy requirements at moderate rotation speeds can be 50% lower than those of tubular ceramic membrane systems of same area at same permeate flux, while their cost is only about 30–50% higher than such systems [21].

Since the hydraulic power cannot be calculated as product of flow rate by pressure drop in the MSD system, we have measured the power consumed by the motor with a wattmeter. Unfortunately, it is difficult to make realistic energy measurements in a small laboratory pilot such as our MSD unit, since a disproportionate part of the motor power is spent in friction of the rotating shaft and motor internal parts. For instance, at 1492 rpm the power consumed by the motor was 141 W for a permeate flow rate $Q_f$ of 0.086 L min$^{-1}$. To correct this, we have subtracted the power consumed at same speed without fluid, as done in [22], which was 85 W. So we can estimate the net power $P_{net}$ consumed by friction on membranes to be about 56 W, giving a net specific energy consumed by the membranes rotation per m$^3$ of permeate ($P_{net}/Q_f$) to be 10.7 kWh m$^{-3}$.

5. Conclusion

Our main purpose was to investigate the potential of the MSD concept in MF of skim milk for separating caseins and whey proteins, as it is one of the few dynamic filtration systems available with ceramic membranes, which are preferred to organic ones for this application, due to easier cleaning and longer life span. Its 0.2 μm pore membranes gave good $\alpha$-La and $\beta$-Lg transmissions with nearly complete casein rejection. The permeate flux at rotation speeds of 1492 and 1930 rpm were comparable to values reported in the literature [6,9] with UTP systems at velocities of 8 m s$^{-1}$, but with higher casein rejection. A comparison of energy consumption per m$^3$ of permeate between our MSD pilot and tubular membranes could not be made as we did not find corresponding data for UTP systems or isoflux membranes which do not require co-current permeate recirculation and can be substituted to UTP systems.

An interesting finding was the linear relation (Eq. (9)) of limiting permeate fluxes with membrane averaged fluid velocity which was valid for three different filtration systems, a disk rotating near a fixed organic membrane, rotating ceramic disks and tubular ceramic membranes in UTP mode. This finding needs to be further investigated, as it would be a convenient tool for comparing performances of very different systems and for scaling up, as the averaged fluid velocity is easier to calculate than the shear rate in a system like the MSD.

Acknowledgments

V. Espina has been supported by a scholarship No. E06D101610CL of the Alban European Union Program for Latin America. The authors thank Westfalia Separator for the loan of a MSD pilot, and Alfa Laval Nakskov for supplying membranes.

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C$</td>
<td>protein concentration (kg m$^{-3}$)</td>
</tr>
<tr>
<td>$J$</td>
<td>permeate flux (L h$^{-1}$ m$^{-2}$)</td>
</tr>
<tr>
<td>$k$</td>
<td>velocity factor</td>
</tr>
<tr>
<td>MF</td>
<td>microfiltration</td>
</tr>
<tr>
<td>$Q_f$</td>
<td>$Q_g$</td>
</tr>
<tr>
<td>$r$</td>
<td>radial coordinate (m)</td>
</tr>
<tr>
<td>$R_d$</td>
<td>housing, disk radius (m)</td>
</tr>
<tr>
<td>$R_1$</td>
<td>outer (inner) membrane radius (m)</td>
</tr>
<tr>
<td>$T$</td>
<td>turbidity (NTU)</td>
</tr>
<tr>
<td>TMP</td>
<td>transmembrane pressure (kPa)</td>
</tr>
<tr>
<td>UF</td>
<td>ultrafiltration</td>
</tr>
<tr>
<td>$V$</td>
<td>mean fluid velocity in tubular membrane (m s$^{-1}$)</td>
</tr>
<tr>
<td>$V_{av}$</td>
<td>membrane averaged fluid velocity (m s$^{-1}$)</td>
</tr>
<tr>
<td>VRR</td>
<td>volume reduction ratio</td>
</tr>
</tbody>
</table>
Greek letters

\( \dot{\gamma} (\dot{\gamma}_{\text{max}}) \) maximum membrane shear rate at periphery (s\(^{-1}\))

\( \mu \) dynamic viscosity (Pa s)

\( \nu \) fluid kinematic viscosity (m\(^2\) s\(^{-1}\))

\( \rho \) density (kg m\(^{-3}\))

\( \omega \) angular velocity (rad s\(^{-1}\))

References


