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PII: S0260-8774(21)00036-4

DOI: https://doi.org/10.1016/j.jfoodeng.2021.110511

Reference: JFOE 110511

To appear in: *Journal of Food Engineering*

Received Date: 17 August 2020

Revised Date: 23 December 2020

Accepted Date: 26 January 2021

Credit Author Statement

Please check the following as appropriate:

- All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

| Author's name | Affiliation |
|---|---|
| John Tobin | Teagasc |
| Herehau Blais | Teagasc |
| Eoin Murphy | Teagasc |
| Quang Tri Ho | Teagasc |
| Karin Schroen | WUR |
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# A cascade microfiltration and reverse osmosis approach for energy efficient concentration of skim milk

Herehau Blais[1,3], Quang Tri Ho[1,2], Eoin G Murphy[1,2], Karin Schroën[3], John T Tobin[1,2] (corresponding author)

[1]Food Chemistry and Technology Department, Teagasc Food Research Centre, Moorepark, Fermoy, Co.Cork Ireland.
[2]Dairy Processing Technology Centre, Teagasc Food Research Centre, Moorepark, Fermoy, Co. Cork, Ireland
[3]Dairy Science and Technology department, Wageningen University, P.O. Box 8129, 6700 EV Wageningen, The Netherlands

herehau.blais@teagasc.ie, quangtri.hqt@gmail.com, eoin.murphy@teagasc.ie karin.schroen@wur.nl, john.tobin@teagasc.ie

**ABSTRACT**

To improve the efficiency of water removal from skim milk, a cascade membrane process of microfiltration and reverse osmosis (RO) was developed whereby skim was concentrated to 18 % dry matter (DM) by RO at either 15 or 50°C. The average flux of the RO process at 50 °C was 89 % higher than that observed at 15°C, linked to altered membrane surface fouling behaviour due to lower viscosity, higher cross-flow velocity and increased diffusivity of the solvent phase. In corollary, a ~57 % energy reduction per unit volume of water removed was observed when the RO process was operated at 50°C. Evaluation of the physicochemical properties of control (9 % DM content skim milk) and RO retentates post-heating (at 80, 90 and120°C) and post-evaporation (to 42 % DM) demonstrated a clear relationship between heating at  elevated DM contents and solution viscosity, an effect that was compounded at higher heating temperatures.

**Keywords**: membrane cascade, microfiltration, reverse osmosis, evaporation, milk concentration, energy efficiency

# 1. Introduction

With a global production estimated at 4–4.5 million tonnes in 2014 (Schuck, 2014), skim milk powder is one of the most widely produced dairy commodities, used as an ingredient in various food products such as yogurt, dairy desserts, baby food or animal feed. To produce skim milk powder, whole milk is pasteurised at 71–74°C for 15 s, prior or after skimming using a centrifugal separator. Before evaporation, the skim milk is normally exposed to an additional heat treatment ranging from 75–125°C for 5–15 s depending on product requirements relative to either microbiological safety or heat classification i.e. low, medium or high-heat (ADPI, volume IV, issue 5). Commercially milk is typically concentrated using falling-film evaporators that operate under vacuum removing ~ 90 % of the intrinsic water by indirect heat transfer. However, evaporation is an energy-intensive process, limited by product characteristics including viscosity and stability of heat labile components (Hasanoğlu and Gül, 2016). To reduce energy consumption, skim milk can be pre-concentrated using reverse osmosis (RO), followed by evaporation to reach dry matter (DM) contents suitable for efficient stabilization through spray-drying (Cheryan et al., 1990; Ramirez et al., 2006).

RO membranes have a pore-equivalent diameter <0.1 nm and therefore retain all ions and larger components while allowing water to permeate. As the process is driven by pressure as opposed to heat transfer, RO preserves the native physicochemical properties of the resulting concentrates, while altering their residence time during subsequent evaporative concentration steps (Cheryan et al., 1990; Kulozik and Kessler, 1990; Syrios et al., 2011). However, RO pre-concentration remains limited to relatively low volume concentration factors (VCF) due to performance limitations linked to increasing osmotic pressure and viscosity of the retentate/concentrate stream. To overcome osmotic resistance, it is necessary to apply high transmembrane pressures (TMP) which negatively impact permeate fluxes due to the higher compaction of fouling materials on the membrane surface (Meyer and Kulozik, 2016). Thus, Meyer and Kulozik (2016) found that subjecting skim milk to an ultrafiltration (UF) step before RO enhanced the processing efficiency of the latter. Indeed, owing to a larger membrane pore size facilitating the permeation of small components (e.g. lactose and minerals), the UF step yielded a protein-free serum, negating the effects of both protein-induced fouling and viscosity development during subsequent RO concentration. Consequently, these authors achieved a final VCF of 5.8 during concentration of a UF permeate by RO, which appeared advantageous compared to the maximum VCF of 3.8 observed during concentration of skim milk by RO. Meyer and Kulozik (2016) considered the RO of UF permeate to be economically favourable when directly compared to the RO of skim milk, relative to both maximum achievable VCF and flux performance. However, the study did not elaborate on the total mass and energy balances of the cascade UF/RO process compared to a conventional RO process, which are key determinants of the industrial feasibility.

In this study, RO alone or a cascade of microfiltration (MF) and RO were assessed for pre-concentration of skim milk to a VCF of 2 before evaporation. MF was chosen to i) retain vegetative microorganisms and spores (Elwell and Barbano, 2006), which would allow the subsequent RO process to be performed at higher temperatures, resulting in an increased flux and a reduced energy consumption per unit permeation and ii) retain residual fat globules and somatic cells (Saboya and Maubois, 2000) to alter the fouling behaviour and by proxy flux, in the subsequent RO process. The impact of heat treatment (low, medium or high) of pre-concentrated skim milk (18 % w/w DM) on the physicochemical properties of the resultant evaporated concentrate (42 % w/w DM) was also assessed to reflect the implications of pre-concentration relative to viscosity and whey protein nitrogen indexes post-evaporation.

## 2. **Materials and Methods**

### 2.1. Materials

Pasteurized skim milk (73.8°C x 15s) was obtained from a local dairy processor and was stored at 5°C for 2 days maximum before use. Its composition was 0.5 $g \cdot kg^{-1}$ fat, 36.7 $g \cdot kg^{-1}$ total protein and 46.9 $g \cdot kg^{-1}$ lactose as measured using a MilkoScan™ FT2 (Foss Electric, Denmark), and 92.1 $g \cdot kg^{-1}$ DM as measured according to the ISO 5537-IDF26 method. Somatic cell content was measured using a Fossomatic 300 (Foss Allé, Denmark).

### 2.2. Preparation of skim milk concentrate

Concentration of pasteurized skim milk was performed according to four case scenarios (performed in duplicate) as described in Fig.1. In the first scenario (control), skim milk was subjected to a heat treatment, followed by evaporation to 42 % (w/w) DM content. In the second scenario, skim milk was pre-concentrated to 18 % (w/w) DM content by RO operated at 15°C, followed by heat treatment and evaporation to 42 % (w/w) DM content. The third (‘MF/RO’) and fourth (‘MF/RO hot’) scenarios comprised an MF step at 50°C followed by RO concentration to 18 % (w/w) DM content at 15 or 50°C, followed by heat treatment and evaporation to 42 % (w/w) DM content.

#### 2.2.1. *Membrane filtration*

Both MF and RO processes were performed using a pilot-scale membrane plant (GEA Process Engineering A/S, Denmark) operated in continuous mode, with the retentate and permeate collected in separate tanks (Fig.1.B). The processing parameters are reported in Table 2. The feed and recirculation (retentate pressure in and retentate pressure out) pressures were maintained constant over the filtration run, yielding a constant TMP. No permeate back pressure was applied during MF nor RO. The plant and membranes were cleaned according to the standard clean-in-place procedure (Appendix (A)).

Three tubular ceramic MF membranes with a nominal size cut-off of 1.4 µm (Isoflux$^{TM}$, Tami Industries, France) were used in parallel, with a total surface area of 1.05 $m^2$. MF process was performed at 50°C, at a VCF of 11, and for at least 10 h starting with ~ 4800 kg of feed to ensure enough permeate was generated to feed the subsequent RO processes.

RO processing was performed using two spiral-wound composite polyamide RO membranes (Dairy AF3838C30, General Electrics) connected in series, with a total surface area of 14.0 $m^2$. The RO processes ran for 8 h at a VCF of 2; ~1300 kg of skim milk or MF permeate was fed to the RO or MF/RO processes, respectively, and ~2100 kg of MF permeate was fed to the MF/RO hot process. The RO and MF/RO processes were performed at 15°C, using a shell-and-tube heat exchanger within the recirculation loop. In the hot RO process, a plate-and-frame heat exchanger was employed upstream of the feed inlet in order to heat the feed entering the membrane plant to ~42°C. The heat generated from the high pressure pump brought the overall operational temperature to 50°C, which was maintained throughout processing.

Parameters of membrane filtration such as recirculation, retentate and permeate flow rates, as well as temperature, pressure and energy consumption of the pumps (i.e. feed, booster and recirculation pumps) and the heat exchanger were recorded using a data logger (Endress+ Hauser AG, Switzerland). The average energy consumed per unit volume of permeate produced (or water removed for RO processes) was calculated for all filtration processes and compared to that of a conventional thermal evaporation. All equations used in modelling of filtration performance are outlined in Appendix (B).

### 2.2.2. *Heat treatment*

Heat treatment was performed using a MicroThermics tubular heat exchanger (MicroThermics, UHT/HTSTLab-25HVHE, USA), operated at a flow rate of 2 $L \cdot min^{-1}$. Briefly, 20 kg of skim milk (9 % (w/w) DM) and 10 kg of skim concentrate (18 % (w/w)

DM, obtained from RO processing) were heated at 80, 90 or 120°C for 30 s. Samples were cooled to 45°C before evaporation.

### 2.2.3. *Evaporation*

Evaporation was performed using a pilot-scale single-effect falling-film evaporator (Anhydro F1 Lab, Denmark), operated at 66°C (under vacuum) in recirculation mode, at a flow rate of 50 L·h$^{-1}$, until a DM content of 42 % (w/w) was achieved. The approximate evaporation time was 5 minutes. The DM content was chosen as the highest level achievable whereby the properties of the evaporated samples would remain stable before analysis.

## 2.3. Physicochemical properties of the concentrates

### 2.3.1. *Viscosity*

Viscosity measurements of the evaporated samples were performed at 50°C, using a controlled stress rheometer (AR2000ex Rheometer, TA Instruments, UK), equipped with a concentric cylinder geometry and controlled peltier heating system. A shear rate ramp from 0 to 300 s$^{-1}$, followed by a holding step at a shear rate of 300 s$^{-1}$ for 5 min, was applied to each sample.

### 2.3.2. *Particle size*

Particle size was measured by static light scattering using a laser-light diffraction unit (Hydro MV, Mastersizer 3000, Malvern Instruments Ltd, UK). The maximum diameter under which 90 % of particles reside, $D_{90}$, is reported. Measurements were performed in triplicate, at 20°C, using a dispersant refractive index of 1.330, a particle refractive index of 1.380, a particle adsorption index of 0.001 and an obscuration range of 3.5 – 12 %. Size distributions were recorded using polydisperse analysis.

### 2.3.3. *Whey protein nitrogen index*

Whey protein nitrogen index (WPNI) was measured according to the GEA Niro method (GEA Niro method A21, 2009). Results are presented as mg native protein per g of DM (mg·g$^{-1}$). A WPNI (mg·g$^{-1}$) value higher than 6 corresponds to a low heat treatment, 1.5–6 corresponds to a medium heat treatment and below 1.5 corresponds to a high heat treatment.

### 2.3.4. *DM content, density, osmolality and osmotic pressure*

DM content was measured according to the "ISO5537-IDF26" method (ISO, 2004). Density of skim control and RO concentrates was measured with a portable densitometer (DMA35, Anton Paar GmbH, Austria) at 25°C.

Osmolality of skim control and RO concentrates was measured with a cryoscopic osmometer (Osmomat auto, GONOTEC, Germany) at 25°C. Samples (50 µL) were placed in an Eppendorf tube and positioned on the machine. The freezing point depression of samples was measured and compared to that of pure water. The osmolality indicating the concentration of all osmotically active dissolved parts in the solvent was calculated by the instrument according to equation (1) (Gonotec 2009):

$$C_{osl} = \frac{\Delta T}{K} \tag{1}$$

where $C_{osl}$ is the osmolality ($osmol \cdot kg^{-1}$), $\Delta T$ is the temperature difference between the sample temperature and the freezing point depression (K) and $K$ is the freezing point constant ($1.858°C\ kg \cdot osmol^{-1} \cdot K^{-1}$). Osmolality values of the samples were used to calculate the osmotic pressure $\pi$ (Pa) according to equation (2) (Janácek and Sigler, 2000):

$$\pi = C_{osm} \cdot \rho \cdot R \cdot T \tag{2}$$

where $C_{osm}$ is the osmolarity ($osmol \cdot m^{-3}$), $R$ is the universal gas constant ($8.31441\ N \cdot m \cdot mol^{-1} \cdot K^{-1}$), $T$ is the solution temperature (K) and $\rho$ is the density ($kg \cdot m^{-3}$).

## 2.4. Statistical analysis

Physicochemical properties including viscosity, WPNI values, and particle size were analysed using one-way analysis of variance (ANOVA), with post-hoc Tukey method using the SPSS statistics software (SPSS V.18, IBM, US).

# 3. **Results and discussion**

## 3.1. MF performance

The permeate flux was recorded as soon as the VCF had been adjusted to 11 and that the DM content of the retentate had reached 9 % (w/w) to ensure minimal inclusion of water during transition to product. To maintain the TMP at 210 kPa, both feed and recirculation pressures were kept constant at 310 and 110 kPa, respectively, throughout processing. The initial flux of ~ 400 $L\cdot m^{-2}\cdot h^{-1}$ gradually decreased to ~ 200 $L\cdot m^{-2}\cdot h^{-1}$ yielding an averaged flux of 319.05 $L\cdot m^{-2}\cdot h^{-1}$ (Fig.2). While an initial flux increase was observed, most likely related to plant stabilisation effects, the overarching process behaviour was a progressive decrease in flux, as expected since various components (e.g. somatic cells, residual fat globules, protein aggregates) were retained, leading to a higher fouling resistance, limiting flow through the membrane. Compositional analysis (Table 1) showed that most of the residual fat globules and somatic cells from the skim milk were retained, which may improve the efficiency of the subsequent RO step, as somatic cells and residual fat globules may affect fouling accumulation.

These results aligned well with the study of Elwell and Barbano (2006) in which somatic cell content was reduced from $129\cdot10^3$ $cells\cdot mL^{-1}$ in raw skim milk to less than $3\cdot10^3$ $cells\cdot mL^{-1}$ in the permeate obtained from a 1.4 μm MF process. As expected at this membrane cut-off, smaller components such as minerals and lactose were found in relatively similar proportions as in skim milk. Particle size analysis (Table 1) indicated that significantly larger particles were retained in the retentate compared to those present in the permeate and the size thereof suggested that these were mainly fat globules and protein aggregates.

No microbial analysis was performed in this study as the main focus was on process efficiency; however, it can be inferred that more than 99.9% of bacteria present in raw skim milk are retained by a 1.4 µm MF treatment Elwell and Barbano (2006). It was thus assumed that a subsequent RO concentration of the MF permeate could be performed at 50°C without compromising the subsequent microbiological quality of either the membrane plant itself or the subsequent concentrated product. It should be noted that the utilization of the MF retentate was not described in this study as the main focus was on assessing potential efficiency gains during RO at 50°C versus 15°C, the MF process being employed simply to remove microbes and other foulants. The VCF of 11 applied during MF was based on the limitations of the pilot filtration plant and the challenges surrounding accurate control of the retentate flow rate during  continuous operation.

Similar observations relative to the filtration performance of skim milk using large pores size MF membranes have been made in the literature. Tan, Wang et al. (2014) observed a similar flux evolution to this study when investigating a cold 1.4 μm MF treatment of skim milk under continuous operational conditions. These authors hypothesized a physicochemical effect whereby whey proteins tended to adsorb onto the ceramic membrane surface, while casein micelles contributed to the fouling layer proportionally to the pressure applied. Similar to the present study, Gosch, Apprich et al. (2013) obtained an average permeate flux of 205 $L \cdot m^{-2} \cdot h^{-1}$ when subjecting skim milk to 1.4 μm MF at 30°C (VCF 2.4) in batch mode, with the lower averaged flux likely an artefact of the lower processing temperature. When using a ceramic 1.4 μm MF membrane filled with glass beads to ensure a uniform TMP of 100 kPa, Pafylias, Cheryan et al. (1996) obtained a flux of 400 $L \cdot m^{-2} \cdot h^{-1}$ during filtration at 50°C (VCF 10) in batch mode, most likely imputable to a higher cross-flow velocity and altered fouling behaviour compared to this study.

Table 1. Composition of the different fractions obtained by membrane filtration.

| Composition | Skim milk | MF permeate | MF retentate | RO retentate | MF/RO retentate | MF/RO hot retentate |
|---|---|---|---|---|---|---|
| Total solids (%) | 9.21±0.07$^{a}$ | 9.05±0.30$^{a}$ | 9.28±0.16$^{a}$ | 17.71±0.35$^{b}$ | 17.26±0.30$^{b}$ | 17.48±0.22$^{b}$ |
| Fat (%) | 0.17±0.03 | 0.06±0.06 | 0.19±0.07 | 0.09±0.03 | 0.05±0.05 | 0.06±0.05 |
| Total protein (%) | 3.79±0.12$^{a}$ | 3.68±0.34$^{a}$ | 3.79±0.01$^{a}$ | 6.87±0.22$^{b}$ | 6.70±0.11$^{b}$ | 6.77±0.13$^{b}$ |
| Lactose (%) | 4.69±0.13$^{a}$ | 4.54±0.18$^{a}$ | 4.73±0.03$^{a}$ | 9.55±0.34$^{b}$ | 9.35±0.15$^{b}$ | 9.47±0.19$^{b}$ |
| Casein (%) | 2.82±0.11$^{a}$ | 2.73±0.37$^{a}$ | 2.82±0.01$^{a}$ | 5.36±0.15$^{b}$ | 5.17±0.05$^{b}$ | 5.25±0.07$^{b}$ |
| Somatic cells (cell·mL$^{-1}$) | 105× 10$^{3a}$ | 5 × 10$^{3b}$ | 789 × 10$^{3c}$ | - | - | - |
| Particle size $D_{90}$ (µm) | 0.369±0.007$^{a}$ | 0.357±0.008$^{a}$ | 0.59±0.001$^{b}$ | - | - | - |

± standard deviation. Values within a row not sharing a common superscript differ significantly ($P$ < 0.

## 3.2. RO performance

Flux evolution during RO, MF/RO and MF/RO hot processes is shown in Fig. 3. The permeate flux was recorded as soon as the DM content of the retentate reached 17 % (w/w) (approximately 15 min after the introduction of skim or MF permeate into the plant). In all three processes, the flux rapidly declined during the first hour of filtration, followed by a gradual decrease throughout the remainder of the filtration process. The strong initial decline can be associated with the increasing viscosity and DM content in the retentate during plant stabilization. Once steady-state conditions relative to VCF and DM were achieved all processing parameters were kept constant thereafter, with the gradual flux decline likely attributable to the accumulation of additional fouling materials at the membrane surface leading to a concomitant increase in fouling resistance. Drawn by convective forces towards the membrane surface, solutes (protein, lactose and minerals) slowly accumulate to form a fouling layer (Skudder et al., 1977), which increases in thickness and compaction relative to the duration of the filtration cycle (Hiddink et al., 1980).

The averaged flux values of RO, MF/RO and MF/RO hot processes were 5.3 ± 0.1, 5.9 ± 1.0 and 10.5 ± 2.0 $L \cdot m^{-2} \cdot h^{-1}$, respectively. Surprisingly there was little difference in the performance characteristics of RO and MF/RO processes carried out at 15°C, as it was initially hypothesized that the removal of foulants by the MF step may alter subsequent fouling behaviour during RO leading to improved performance. Thus it may be inferred that larger foulants such as residual fat globules, somatic cells and microorganisms may play a lesser role in the fouling behaviour of skim milk during concentration by RO. A flux value ~ 89 % higher at 50°C was observed during RO, compared to either cold processes. Despite a slightly higher osmotic pressure for the MF/RO hot process, the improved performance is likely linked to the 35 % lower viscosity of the retentate coupled with a higher cross flow velocity (Table 2).

Table 2. Processing performance parameters.

| | MF | RO | MF/RO | MF/RO hot |
|---|---|---|---|---|
| Recirculation flow rate ($kL \cdot h^{-1}$) | 14.5 | 6.8 | 7.0 | 8.4 |
| Feed pressure (kPa) | 308 | | 3005 | |
| Recirculation pressure (kPa) | 113 | | 2830 | |
| Permeate flux ($L \cdot m^{-2} \cdot h^{-1}$) | 319.05 | 5.28 | 5.86 | 10.50 |
| TMP (kPa) | 210±10 | 2920±10 | 2920±10 | 2920±10 |
| Viscosity of the retentate at trial temperature (mPa.s) | - | 5.32±0.18 | 5.33±0.07 | 3.46±0.01 |
| Osmotic pressure of the retentate (MPa) | - | 1.59±0.04 | 1.60±0.06 | 1.78±0.05 |
| VCF | 11 | 2 | 2 | 2 |
| Trial temperature (°C) | 50±2 | 15±2 | 15±2 | 50±2 |

In corollary, as all RO retentates had similar compositions and DM, the improved performance for the MF/RO hot process could be due to increased diffusivity of the solvent phase and altered fouling accumulation. These observations were consistent with those of Ibrahim and Mohammad (2001) regarding the positive effect of temperature on RO performance, although the order of magnitude change found was not consistent with this study, whereby temperature was the most significant parameter influencing RO performance, with an increase of 1°C resulting in 3 % higher flux.

An accurate comparison with studies focusing on skim milk concentration through RO is difficult due to the prevalence of batch concentration processes in the literature compared to the continuous concentration process investigated in this study, with the latter being a closer approximation of commercial plant operation (Cheryan et al., 1990; Meyer and Kulozik, 2016). Indeed, while a fixed quantity of fouling materials is recirculating in a plant operated in batch mode, a continuous mode implies an increasing quantity of fouling materials being fed to the plant, potentially altering fouling accumulation dynamics, whereby an increasing fouling resistance causes an altered flux decline in studies operated in continuous as opposed to batch modes.

The most relevant study describing a cascade membrane approach to improve the efficiency of RO concentration of milk components is that of Meyer and Kulozik (2016) who assessed the efficiency of a cascade of UF and RO compared to that of RO alone for concentration of UF permeate and skim milk respectively. Logically these authors observed improved performance in the absence of proteinaceous material during RO of UF permeate, compared to RO of skim milk, with volume reduction ratios (VRR) of 5.8 and 3.8 achieved respectively. Evaluating the VRR applied by these authors using either UF/RO or RO and considering an arbitrary skim milk volume such as 1000 kg of skim milk as initial feed, then the following observations can be made:

- If the conventional RO is carried out until a VRR of 3.8 then ~737 kg of RO permeate is produced.
- In the cascade UF/RO process, to produce ~737 kg of RO permeate from a UF permeate of 5.6 % DM at a VRR of 5.8 necessitates a UF permeate feed of ~890 kg.
- To produce ~890 kg of UF permeate from 1000 kg of skim milk necessitates that the UF process be performed at a VCF of 9.1 i.e. with the remaining 110 kg being the UF retentate.

- To produce a UF retentate at a VCF of 9.1 means that the ~ 110 kg of UF retentate would contain 34 % (w/w) protein (based on a skim milk protein content of 3.71% (w/w) and not accounting for NPN loss to the UF permeate). This concentration would not be possible in the absence of substantial DF volumes, which would necessitate additional water removal by RO.
- The production of skim milk concentrate, through recombination of the proposed cascade UF/RO retentates, necessitates a UF plant designed to produce at minimum a composition reflecting MPC70 in the UF retentate stream.
- Several authors have described the maximum concentration factors achievable during UF of skim milk relative to VCF (1.7 – 7), retentate total protein concentration (17-21 %) and the necessity for DF water (Gesan-Guiziou, 2013; Klarenbeek, 1994; Mistry and Maubois, 2017).

It is possible that the combination of UF and RO presented by the authors as more economically efficient than RO alone for concentration of total milk solids would in fact be limited by the efficiency of the UF step in terms of achievable VCF, the implications of high protein (casein) contents and high viscosity limiting UF performance at higher VCF's and the requirement for DF water addition which would have to be removed during subsequent RO processing.

In contrast a cascade MF/RO hot process as presented in this study, has a number of advantages over either a UF/RO or RO alone approach for the following reasons:

- 1.4 µm MF step can easily achieve a VCF of >50, allowing most milk components to cross the membrane.
- The large pore size MF membrane used in this study achieved an average flux value of 319 $L \cdot m^{-2} \cdot h^{-1}$ essentially limiting the need for a very large MF plant and by proxy capital and operational costs.
- Removal of >99.9 % of microorganisms (Elwell and Barbano, 2006) allows the subsequent RO process to be performed at 50°C without compromising the microbiological quality of either the RO plant or the subsequent skim concentrate.
- Operation of the RO plant at 50°C greatly enhances flux performance compared to cold operation and thus provides a realistic approach to skim milk concentration whereby capital and operational costs are minimized.

### 3.3. Fouling resistance

Throughout RO processing, fouling was expected to occur under two forms: i) organic caused by proteins, lactose or organic acids and ii) inorganic mostly related to calcium phosphate precipitation, especially at higher protein concentrations (Hiddink et al., 1980). The third common fouling form, namely biofouling associated with growth of biomass, was excluded as i) two out of the three RO processes were performed at low temperatures, ii) for the MF/RO hot process, most microorganisms originally present in the milk were expected to be retained during the MF pre-treatment, and the RO was run for a relatively short duration thereby limiting microbial growth overtime. The fouling resistance $R_f$ was empirically correlated to cumulative permeate volume $F_c$ for the three RO processes, as shown in Fig.4. As expected due to more particles accumulating onto the membrane surface in continuous mode, fouling resistance increased with increasing $F_c$ in all cases. However, while the rapid initial increase in fouling resistance during RO and MF/RO processes was similar, following a trend akin to a Langmuir adsorption model (Tong et al., 2020), it was much lower in the MF/RO hot process with a linear relationship observed. That difference can be related to the lower viscosity of the MF/RO hot retentate (3.46 mPa·s) compared to that of RO (5.32 mPa·s) and MF/RO (5.33 mPa·s) retentates, thereby reducing concentration polarization on the retentate side via a higher turbulence at the membrane surface. Indeed, at a constant TMP of 2.9 MPa, the higher temperature was found to increase the recirculation flow rate $Q_R$ during the MF/RO hot process (8207–8583 $L \cdot h^{-1}$ compared to 6554–7375 $L \cdot h^{-1}$ for the cold processes) which resulted in a higher cross-flow velocity, increasing shear at the membrane surface and reducing fouling propensity (Hiddink et al., 1980; Skudder et al., 1977).

Based on these experimental results, the rate of change of fouling resistance, $R_c$, was plotted against the cumulative permeate volume $F_c$ according to equation (B8), as illustrated in Fig.5, with the estimated parameters reported in Table 3. Compared to the MF/RO hot process, $R_c$ was found to be three times as high for both cold processes at the start of the filtration, indicating a much more rapid fouling build-up at 15°C. With increasing $F_c$, the rate of fouling build-up of both cold processes decreased until it was approximately four-fold as low when $F_c \sim 50\ L \cdot m^{-2}$ was reached. On the other hand, $R_c$ remained almost constant with increasing $F_c$ during the MF/RO hot process, indicating a linear build-up of fouling resistance over the entire trial duration.

Table 3. Parameters of fouling resistance $R_f$ in function of cumulative permeate volume for RO, MF/RO and MF/RO hot processes.

| | Coefficient $c_1$ ($m^{-1}$) | Coefficient $c_2$ ($L \cdot m^{-2}$) |
|---|---|---|
| RO | $28.2\times10^{13}$ | 37.0 |
| MF/RO | $20.6\times10^{13}$ | 26.3 |
| MF/RO hot | $120.1\times10^{13}$ | 504.0 |

## 3.4. Physicochemical properties of the concentrates

Skim control samples 9 % (w/w) DM, as well as RO, MF/RO and MF/RO hot 18 % (w/w) DM concentrates were heat-treated (80-120°C) to ascertain the impact of pre-concentration on the physicochemical characteristics of the concentrated system post-heat treatment/evaporation using conditions commonly applied in commercial processes. The viscosity of the control and concentrates was measured directly after evaporation to 42 % (w/w) DM (Fig.6). Heat treatment of RO, MF/RO and MF/RO hot concentrates (~18 % (w/w) DM) at 80 and 120°C did not significantly increase solution viscosity compared to control samples. In contrast heat treatment at the intermediate temperature of 90°C yielded significantly ($P<0.05$) higher post-evaporation viscosity for RO, MF/RO and MF/RO hot concentrates relative to the control sample, which demonstrated a similar viscosity to that observed at 80°C. WPNI values as presented in Fig. 7, showed no significant ($P>0.05$) difference in heat classification between control and concentrated samples post-heat treatment at each individual treatment condition (80, 90 or 120°C).

It is well-established that casein micelle structure is relatively heat stable (Vasbinder and de Kruif, 2003), with viscosity increases post-heat treatment likely related to whey protein denaturation/aggregation. Additionally, some of the unfolded whey proteins (primarily β-lactoglobulin) may interact with the hairy brush of casein micelles through covalent bonds between thiol groups and disulfide residues of κ- and $\alpha_{s2}$-casein, increasing the volume fraction of the whey-casein micelle complexes and promoting their interactions, an effect likely exacerbated at higher DM contents (Vasbinder and de Kruif, 2003). While there was limited differences in sample properties within a given temperature treatment in this study, this may relate to slight compositional (protein/dry matter) differences between replicate samples post-evaporation which may mask true in-process behaviours.

Heat treatment, before evaporation, remains necessary to inactivate pathogenic bacteria or prevent spoilage, thus ensuring the production of microbiologically-safe concentrates; however, the impact on physicochemical properties may have far reaching consequences relative to process efficiency and heat classification at higher DM contents and by proxy high protein contents. Processing implications surrounding increased solution DM/viscosity may include reduced heat transfer coefficients, a higher propensity for fouling in heat exchangers/pipework, which may negatively impact equipment run times, CIP intervals and discharge of milk solids to effluent treatment (Wijayanti et al., 2014). If concentration of skim milk by RO before both heat treatment and evaporation was to be implemented at commercial scales, the addition of a MF step prior to RO could facilitate the use of lower heating temperatures before evaporation. This could limit any potential deleterious effect on both solution viscosity and WPNI values post-evaporation, while ensuring the microbiological stability and safety of the final product.

### 3.5. Energy consumption

The energy consumption of all filtration processes (MF, RO, MF/RO and MF/RO hot) was calculated based on the power consumption of the feed, recirculation and high pressure pumps, as well as that of the heat exchanger (employed to maintain the RO plant at 15°C). The total energy consumptions of the RO and cascade MF/RO and MF/RO hot processes were 396.49±8.76, 421.21±21.19 and 178.46±25.42 $kJ \cdot L^{-1}$ of water removed, respectively (Table 4). While the energy, utilities and chemical consumption of the cleaning cycles were not considered in this manuscript, they would be relevant when evaluating operational cost at an industrial scale.

427 Table 4. Total energy consumption during performance of MF, RO, MF/RO and MF/RO hot processes.

| | Feed pump (kW) | Recirculation pump (kW) | Booster pump (kW) | Heat exchanger (kW) | Total energy (kW) | Energy consumption ($kJ\cdot L^{-1}$ permeate) |
|---|---|---|---|---|---|---|
| RO | 0.50±0.02 | 1.14±0.03 | 3.67±0.05 | 2.84±0.04 | 8.15±0.04 | 396±9 |
| MF | 0.17±0.11 | 1.85±0.05 | - | - | 2.03±0.03 | 21±2 |
| MF/RO | 0.52±0.01 | 1.23±0.15 | 3.87±0.00 | 2.94±0.31 | 8.56±0.15 | 380±69 |
| Combined MF and MF/RO | - | - | - | - | - | 421±21 |
| MF | 0.17±0.02 | 1.85±0.05 | - | - | 2.03±0.03 | 21±2 |
| MF/RO hot | 0.56±0.04 | 1.27±0.06 | 3.70±0.09 | - | 5.53±0.19 | 137±22 |
| Combined MF and MF/RO hot | - | - | - | - | - | 178±25 |

428 ± standard deviation

In order to compare the energy consumed per unit volume of water removed by the three RO processes (RO, MF/RO and MF/RO hot), the cascade MF/RO processes must also account for the energy consumed by the MF plant to produce a given volume of MF permeate to feed the subsequent RO process. In this study under a VCF of 2 for the RO process, 2 kg of MF permeate (RO feed) were required to produce 1 kg of RO permeate.

Due to the relatively large pore size, low operational pressures and high temperatures employed during MF, this process consumed relatively little energy (20.63 $kJ \cdot L^{-1}$ of permeate). On the other hand, the RO processes required a high hydrostatic pressure to be generated in order to overcome the osmotic resistance on the retentate side (Fell, 1995), primarily exerted by a multistage centrifugal high-pressure pump which consumed between 3.67-3.87 kW, with a large proportion of that energy converted directly into heat. Therefore, RO processes performed under cold conditions (RO and MF/RO) consumed more energy per unit of water removed than the MF/RO hot process, due to a combined effect of lower permeate flux and hence feed flow, coupled with the need to remove the heat generated by the high pressure pump to maintain the filtration process at 15°C. Although the feed temperature was ~5°C throughout both RO and MF/RO processes, a tubular heat exchanger within the membrane plant recirculation loop, equipped with a heat-meter, consumed between 2.84-2.94 kW to maintain the plant temperature at 15°C. This provides a good insight into the actual energy being utilised for separation as opposed to direct conversion into heat. The cascade MF/RO process consumed ~6 % more energy per unit volume of water removed compared to the RO process due to the additional filtration step in the former, as the flux characteristics for both RO and MF/RO were similar throughout processing. Conversely, with an energy consumption of 178 $kJ \cdot L^{-1}$ of water removed, the MF/RO hot process consumed 58 and 55 % less energy per unit volume of water removed compared to the MF/RO and RO processes, with 421.21 and 396 $kJ \cdot L^{-1}$ respectively. This lower energy consumption is primarily related to the absence of cooling of the RO plant during processing at 50°C. Essentially, the feed entering the plant at ~ 42°C coupled with the heat generated by the high pressure pump yielded an overall process temperature of 50°C. In this study, the MF permeate feeding the MF/RO hot process was pre-heated from 5 to 42°C using a plate heat exchanger; however, the energy consumed in this step has not been considered in the energy calculations as it was only included due to the logistics surrounding milk holding and quality implications thereof which were artefacts of the scheduling of the pilot-scale filtration trials. In the commercially envisaged process the cascade hot RO step would occur immediately after MF (50°C), likely with some storage buffering, thus only requiring a heat exchanger to compensate for frictional heating but without a need for intermediate cooling or reheating prior to concentration. A logical process configuration incorporating the MF/RO hot process would include pasteurisation (i.e., 73°C × 15 s), regenerative cooling to 50°C before cream separation, with the skim milk thereof directly feeding the MF and subsequent RO steps, before either cooling and storage or further processing of the concentrated skim.

In commercial dairy plants, multiple-stage evaporators equipped with either thermal or mechanical vapour recompression (MVR/TVR) are typically employed to reduce the energy consumption associated with water removal (Ramírez, Patel, and Blok 2006). These authors reported that the typical energy demand for a 7-stage falling film evaporator equipped with TVR is ~ 300 $kJ \cdot L^{-1}$ of water removed. This energy demand is almost two-fold that observed for the MF/RO hot process in this study, albeit the concentration range was significantly lower under a VCF of 2. On the other hand, considering that a MVR evaporator consumes ~ 55 $kJ \cdot L^{-1}$ of water removed with a commercial RO plant consuming 20-40 $kJ \cdot L^{-1}$ (Fox et al., 2010), a number of conclusions can be drawn. Firstly it is likely that the energy figures generated at pilot-scale greatly underestimate the efficiency of a multi-loop commercial installation. Secondly while there are clear advantages for RO pre-concentration relative to TVR evaporators the similarities in energy consumption between RO and MVR evaporators per unit water removed seem to rule out the latters combined use. However, the installation of a RO pre-concentration step to limit the size of the subsequent MVR evaporator could still be advantageous from a capital cost perspective. Finally careful consideration should be given to any retrofitting of an evaporator with a RO pre-concentration step as product flow rates, tube wetting and temperature conditions within the evaporator will all likely be affected with potentially unpredictable outcomes relative to product and process performance.

## 4. Conclusion

Reverse osmosis is an attractive low cost solution for water removal from skim milk. The addition of an MF pre-treatment as part of a cascade filtration approach did not significantly alter the subsequent RO performance at 15°C compared to RO alone. However, the introduction of an MF step, as a significant microbiological hurdle, allowed the subsequent RO step to be operated at 50°C which greatly improved flux performance, limiting the accumulation of foulants at the membrane surface throughout processing. Under the concentration factors applied (VCF2), > 50% of the innate water in skim milk was removed, with >55% reduction in the energy usage for RO operated at 50 compared to 15°C. Assessment of the physicochemical characteristics of heat-treated and evaporated skim milk and RO concentrates determined no implications relative to WPNI values and by proxy heat classifications when heating RO concentrates compared to a skim milk control. However, heating RO concentrates at temperatures ⩾90°C yielded a higher post-evaporation viscosity, which suggests that altered heating conditions pre-evaporation may be necessary to ensure subsequent drying performance is not compromised.

Further work is required to determine the longevity of polymeric RO membranes subjected to operational use at 50 °C, in addition to careful monitoring of the microbiological quality of the MF permeate feeding the RO plant and the implications of high temperature processing on the growth of microorganisms within the RO plant itself during commercially representative production cycles.

## 5. Acknowledgments

H. Blais was funded under the Teagasc Walsh Scholarship. This research was supported by Enterprise Ireland under the Dairy Processing Technology Centre, project number TC 2014 0016.

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## 7. **Figure captions**

Figure 1: (A) Process scenarios investigated in this study. Scenario 1 refers to the conventional concentration process while scenarios 2, 3 and 4 describe the combination of RO, MF/RO and MF/RO hot with evaporation. (B) Schematic of the filtration plant.

Figure 2: MF permeate flux (blue) and temperature (red) as a function of time.

Figure 3: Typical evolution of RO permeate fluxes as a function of time.

Figure 4: Fouling resistance $R_f$ of RO, MF/RO and MF/RO hot processes as a function of cumulative permeate volume.

Figure 5: Rate of change of fouling resistance $R_c$ as a function of cumulative permeate volume.

Figure 6: Apparent viscosity (300 $s^{-1}$, 50 °C) of skim control and RO concentrates at 42% DM, subjected to heat treatments (80-120 °C). Samples not sharing a common superscript differ significantly ($P < 0.05$). Analysis of variance was performed within discrete treatment temperatures.

Figure 7: WPNI values of skim control and RO concentrates subjected to heat treatments (80-120 °C). Analysis of variance was performed within discrete treatment temperatures.

## 8. Appendix

### (A) CIP procedure

Before each filtration, 2 % aqueous solution of P3-Ultrasil-115 (caustic) was recirculated for 15 min at 45-50°C and flushed with RO water. Post-filtration, three discrete cleaning steps were applied: i) a solution of 1 % enzyme/caustic Ultrasil-69:67 in a 1:2 ratio (Eco lab, USA), ii) a 1 % aqueous solution of Ultrasil-78 (nitric acid) (Eco lab, USA) and iii) a 2 % aqueous solution of P3-Ultrasil-115. Each cleaning solution was recirculated for 15 minutes at 45-50°C, followed by flushing with RO water for 15 minutes. Clean water flux was measured gravimetrically before and after the filtration, as well as after CIP, using reverse osmosis water under operational conditions for both MF and RO processes.

### (B) Modelling of filtration performance

The transmembrane pressure $\Delta P_{TMP}(t)$ was calculated as follows:

$$\Delta P_{TMP}(t) = \frac{P_f(t)+P_r(t)}{2} - P_p(t) \qquad \text{(B1)}$$

where $P_f(t)$ is the feed inlet pressure (Pa), $P_r(t)$ is the outlet pressure of the retentate (Pa) and $P_p(t)$ is the permeate pressure (Pa) at time $t$.

The initial RO membrane resistance at $t_o$, $R_o$, was calculated as follows:

$$R_o = \frac{A\left(\frac{P_f(t_0)+P_r(t_0)}{2} - P_p(t_0) - \Delta\pi(t_0)\right)}{Q_p(t_0).\eta(t_0)} \qquad \text{(B2)}$$

where $A$ is the membrane surface area ($m^2$), $Q_p(t_o)$ is the permeate flow rate across the membrane ($m^{-3}\cdot s^{-1}$) at $t_o$ and $\eta$ is the viscosity of the RO retentate (Pa·s).

The fouling resistance $R_f$ was expressed as follows (Persson and Nilsson, 1991):

$$R_f(t) = \frac{A\left(\frac{P_f(t)+P_r(t)}{2} - P_p(t) - \Delta\pi(t)\right)}{Q_p(t).\eta(t)} - R_o \qquad \text{(B3)}$$

The total resistance $R_{tot}(t)$ is considered to be the sum of the initial membrane resistance at $t_o$, $R_o$, and the fouling resistance $R_f(t)$.

$$R_{tot}(t) = R_o + R_f(t) \qquad \text{(B4)}$$

Fick's law for permeate flow rate $Q_p$ ($m^3\cdot s^{-1}$) across the RO membrane is related to the hydraulic pressure and osmotic pressure across the membrane as follows (Shirazi et al., 2010):

$$Q_p(t) = A \cdot K_p(t) \frac{\Delta P_{TMP}(t) - \Delta\pi(t)}{\eta} = A \frac{\Delta P_{TMP}(t) - \Delta\pi(t)}{\eta \cdot R_{tot}(t)} \quad \text{(B5)}$$

where $K_p(t)$ is the membrane permeability (m).

During RO performance, $R_f$ was empirically expressed against cumulative permeate volume $F_c$ in a non-linear relationship, similarly to a Langmuir model (Tong et al., 2020):

$$R_f = \frac{c_1 \cdot F_c}{c_2 + F_c} \quad \text{(B6)}$$

where $F_c$ is the cumulative permeate volume across the membrane ($L \cdot m^{-2}$), $c_1$ ($m^{-1}$) and $c_2$ (m) are the coefficients of the model. Note that if $c_2 >> F_c$, the correlation between $R_f$ and $F_c$ would become linear as follows:

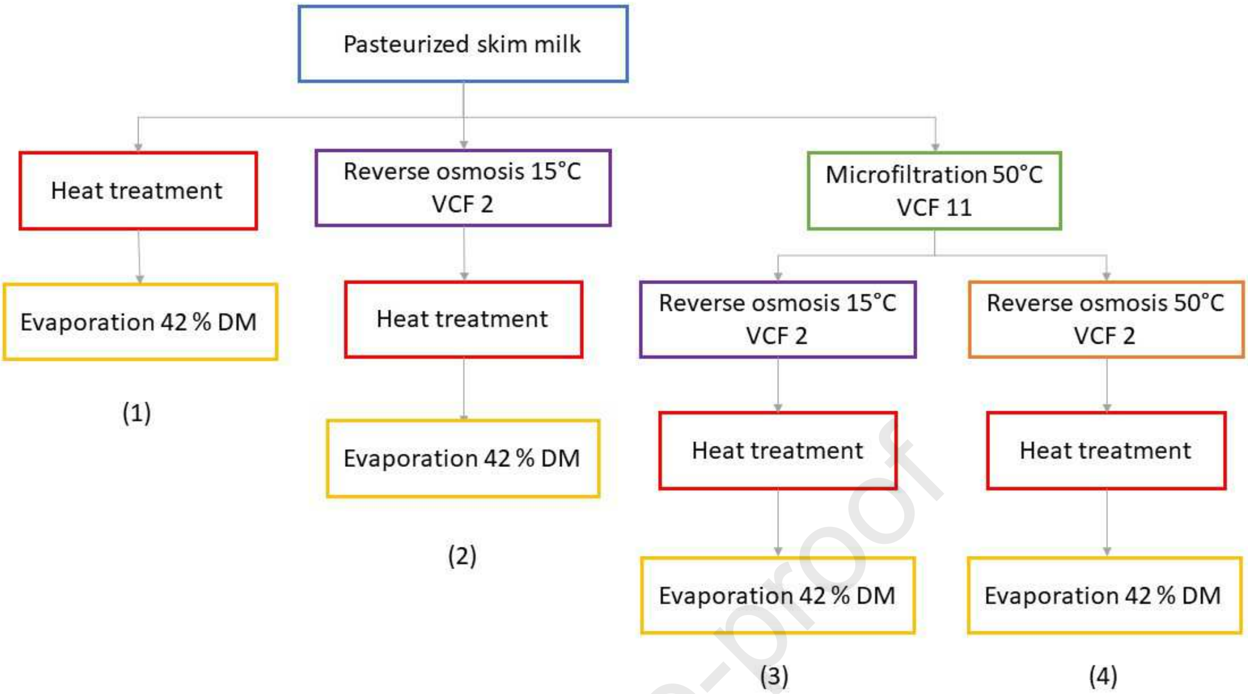
$$R_f = \frac{c_1 \cdot F_c}{c_2} \quad \text{(B7)}$$

For each replicate trial, parameters $c_1$ and $c_2$ of this resistance model were estimated by minimising the sum square difference between the resistance values predicted by the model and the experimental ones using a non-linear estimation programme written in Matlab (The Mathworks, Inc., Natick, USA). The averaged coefficient values of both replicate trials were eventually used to model the fouling resistance $R_f$.

The rate of accumulation of fouling resistance $R_c$ ($m^{-2}$) relative to cumulative permeate volume $F_c$ was expressed as follows:

$$R_c = dR_f/dF_c = \frac{c_1 * c_2}{(c_2 + F_c)^2} \quad \text{(B8)}$$

**A**


Pasteurized skim milk
Heat treatment
Reverse osmosis 15°C VCF 2
Microfiltration 50°C VCF 11
Evaporation 42 % DM
Heat treatment
Reverse osmosis 15°C VCF 2
Reverse osmosis 50°C VCF 2
(1)
Evaporation 42 % DM
Heat treatment
Heat treatment
(2)
Evaporation 42 % DM
Evaporation 42 % DM
(3)
(4)

**B**

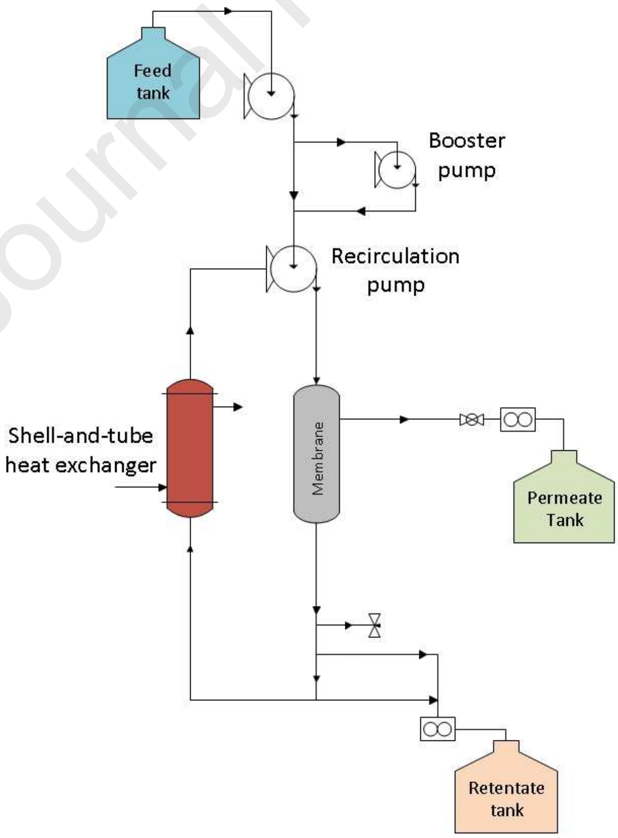

Feed tank
Booster pump
Recirculation pump
Shell-and-tube heat exchanger
Membrane
Permeate Tank
Retentate tank

Fig.1. (A) Process scenarios investigated in this study. Scenario 1 refers to the conventional concentration process while scenarios 2, 3 and 4 describe the combination of RO, MF/RO and MF/RO hot with evaporation. (B) Schematic of the filtration plant.

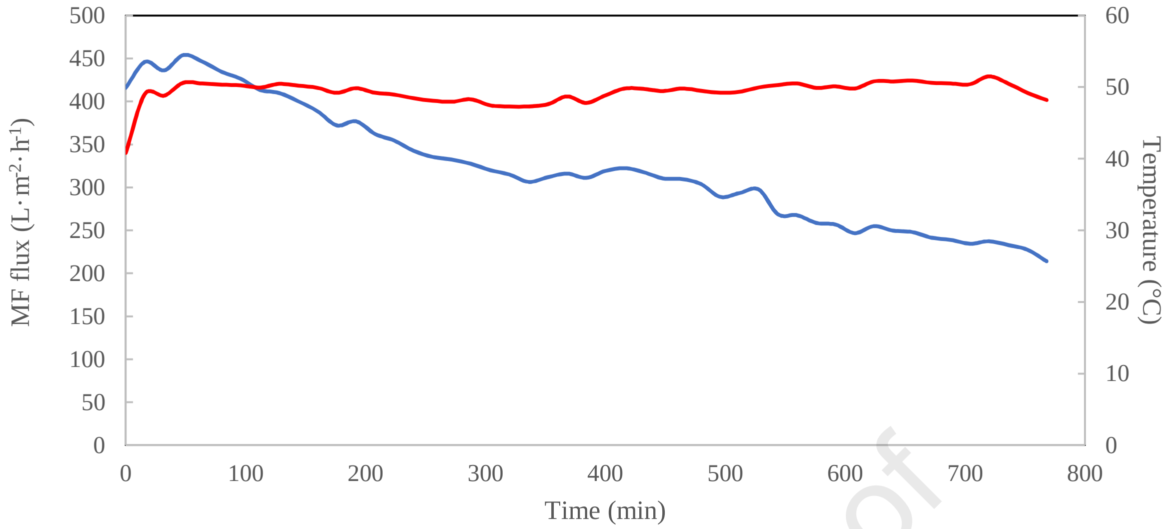



Fig.2. Permeate flux (blue) and temperature (red) as a function of time during MF.

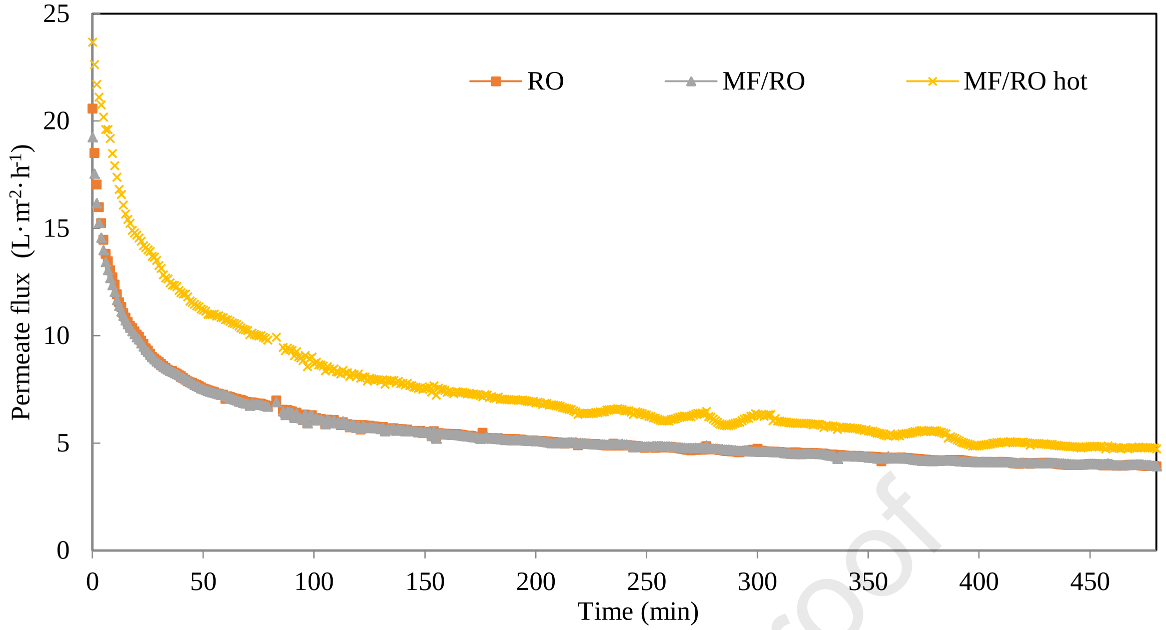



Fig.3. Typical evolution of RO permeate flux as a function of time.

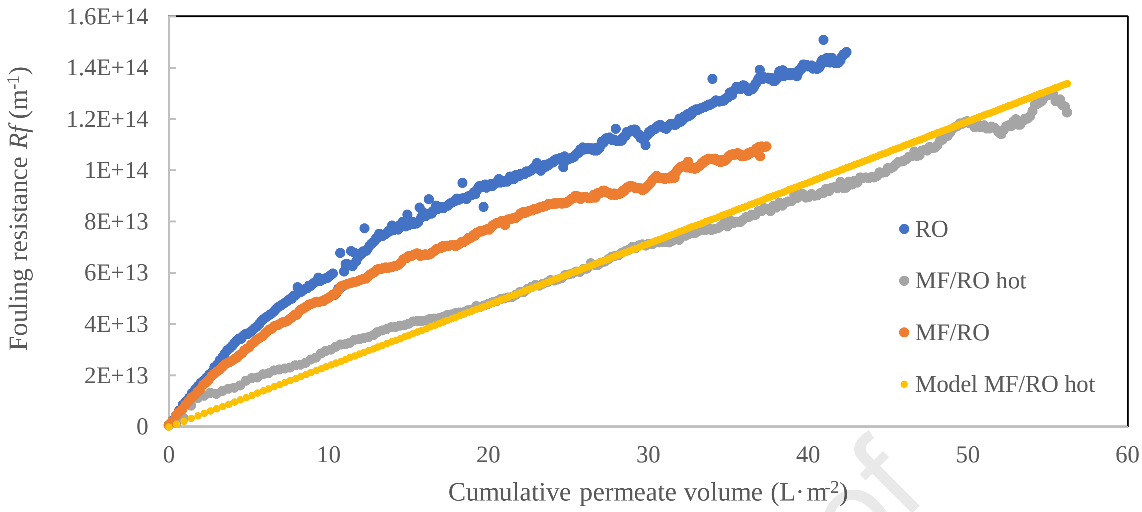



Fig.4. Fouling resistances $R_f$ of RO, MF/RO and MF/RO hot processes as a function of cumulative permeate volume.

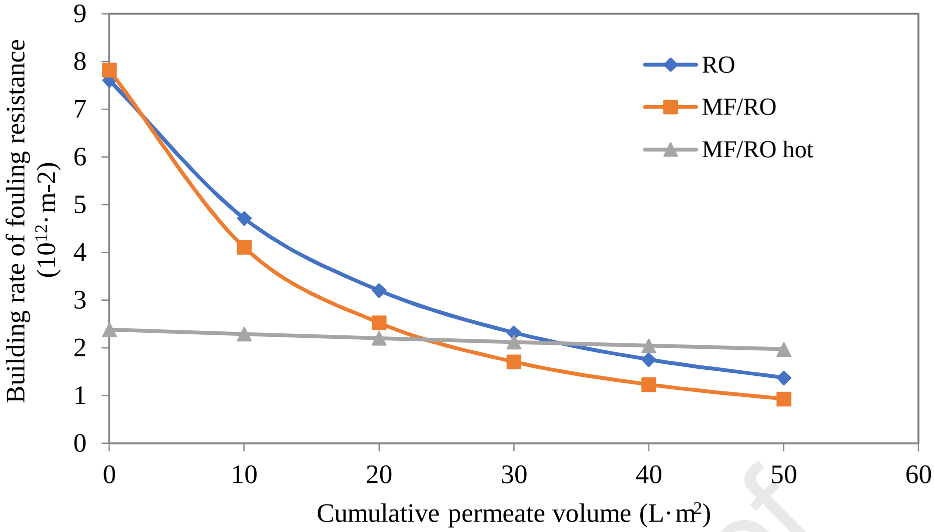



Fig.5. Rate of change of fouling resistance $R_c$ as a function of cumulative permeate volume.

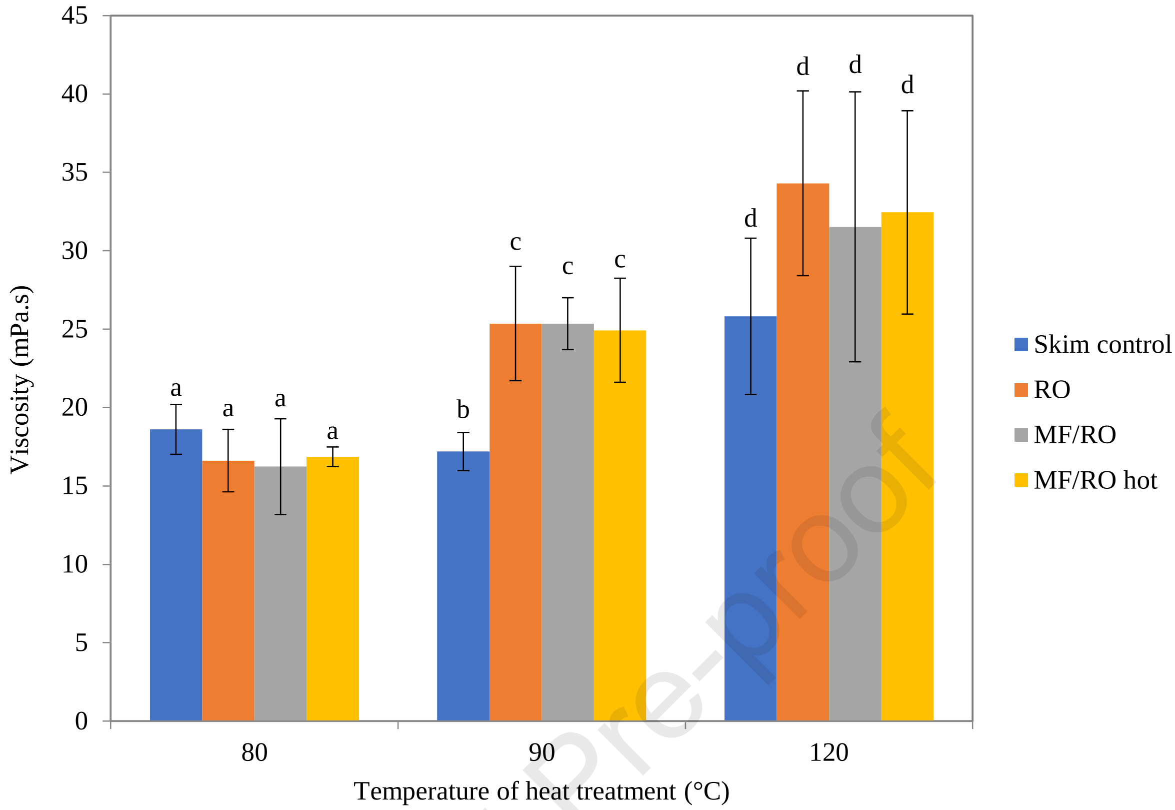



Fig.6. Apparent viscosity (300 $s^{-1}$, 50 °C) of skim control and RO concentrates at 42% DM, subjected to heat treatments (80-120 °C). Samples not sharing a common superscript differ significantly ($P$ < 0.05). Analysis of variance was performed within discrete treatment temperatures.

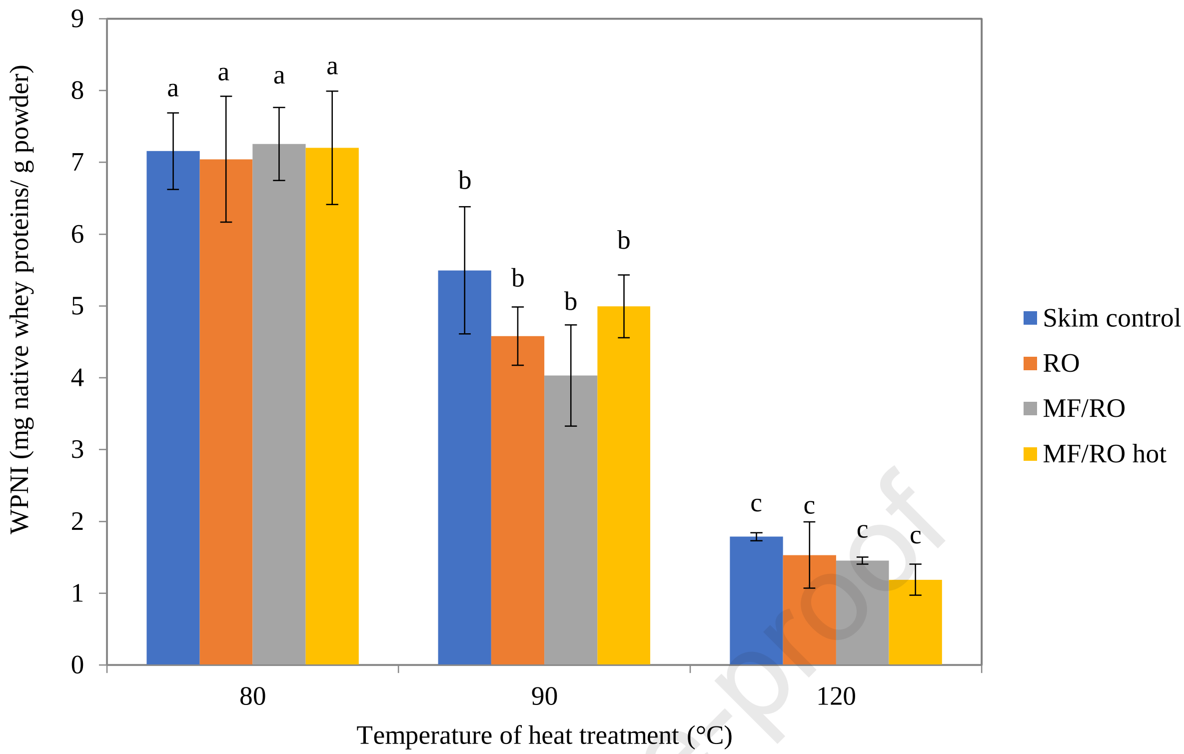



Fig.7. WPNI values of skim control and RO concentrates subjected to heat treatments (80-120 °C). Analysis of variance was performed within discrete treatment temperatures.

Highlights

- A cascade filtration process for efficient removal of water from skim milk
- Fouling resistance is reduced when reverse osmosis is performed at 50°C
- Flux is increased when reverse osmosis is performed at 50 versus 15 °C
- Heat classification of skim milk is not affected by heat treatment at 18 % dry matter

Conflict of Interest and Authorship Conformation Form

Please check the following as appropriate:

- All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

- This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue.

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| Author's name | Affiliation |
|---|---|
| John Tobin | Teagasc |
| Herehau Blais | Teagasc |
| Eoin Murphy | Teagasc |
| Quang Tri Ho | Teagasc |
| Karin Schroen | WUR |
| | |
| | |
| | |