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Flux behavior and energy consumption of Ultrafiltration (UF) Process of milk

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Abstract

Flux decline and recovery of UF process used in concentrating milk at different operational pressures were studied. Filter medium resistance during filtration and cleaning was estimated from the experimental data. Energy consumption during filtration and cleaning processes were estimated. Regarding the flux behavior, retentate and permeate fluxes declined with time at different operational pressures during filtration, on contrary, flux during cleaning increased with time at all pressures studied. Filter medium resistance increased linearly with time of filtration at different operational pressures during concentration. It increased during the first 40 min of cleaning process and decreased with time to reach the minimum resistance at the full flux recovery. The total energy consumed for filtration ranged from 50.08 to 62.54 kJ/L of retentate, while it ranged from 18.18 to 21.65 kJ/L of permeate. The energy consumed for cleaning ranged from 87.08 to 107 kJ at different operational pressures.

Keywords: Ultrafiltration, Permeate, Retentate, Flux decline, Fouling, Flux recovery.

Abbreviations

A_m_Membrane area (m²); D_Effective diffusivity (m²/s); k_s_Mass transfer coefficient (m/s); t_Filtration time (s) V_Filtration volume (m³); v_Cross-flow velocity (m/s); ΔP _Transmembrane pressure (Pa); r_The molecular radius (m); R_m_Membrane medium resistance (m⁻¹); μ _Filtrate viscosity (Pa.s); ρ _Density (kg/m³); α _Specific cake resistance (m⁻¹); σ _Boundary layer thickness (m); d_h_Hydraulic radius (m); T_Temperature (K); *J*_The permeate flux (L/min.m²); *P*_Pressure at the inlet of the module (Pa); *P*_Pressure at module outlet (Pa); *Q*_{circ}_Average circulation flow rate (m³ s⁻¹); η _Pumping efficiency (%).

Introduction

Membrane technology constitutes an efficient and ecological process for the extraction (concentration, purification and fractionation) of valuable molecules from wastes or by-products in agro-food industry. Dairy applications probably account for the largest share of studies, particularly for the understanding under different processing conditions (i.e. pH, temperature, ionic strength, etc.) of the interactions between individual components of milk or between these components and membranes (Gourley et al., 1995, and Daufin et al., 1998). Membrane separation process for liquid systems is conventionally classified in terms of the size ranges of materials separated (microfiltration, 10 μ m–0.1 μ m; ultrafiltration, 0.1 μ m–5 nm; nanofiltration, 5 nm–0.5 nm; reverse osmosis 0.5 nm). The ultrafiltration process has become particularly important for concentrating proteinaceous solutions. Ultrafiltration performance is limited, however, due to the build up of the solutes at the membrane surface. This is the so-called concentration polarization effect (Bowen and Williams, 2007). The major problem in membrane separation process is decline in flux over time of operation. This flux decline is attributed to the fouling of membrane. Membrane fouling is affected by three major factors, namely, the membrane material properties, the feed characteristics and the operating parameters (Platt and. Nyström, 2007, Matzinos and Álvarez, 2002, Zhang, and Liu, 2003, Kazemimoghadam and Mohammadi, 2007, Juang and Lin, 2004, Rai et al., 2007).

Tong et al. (1988) studied the characteristics of proteinaceous foultants and flux decline during the early stages of whole milk ultrafiltraion, and they found that in early stages of milk UF, adsorption fouling is probably the primary mechanism of flux decline.

A pilot study was undertaken to evaluate the UF process in dead-end filtration compared to cross-flow filtration mode in terms of water loss, energy consumption (flux rate achievable). The study performed on surface water, i.e., high variable water quality, has shown that the membrane specific flux in cross-flow filtration seems to be more stable at high flux, i.e., 100 L/h.m². However, results have shown that in dead-end filtration, a gain of about 0.3 kWh/m³ is realized on energetic balance. In this case, the energy consumption is around 0.2 kWh/m³ and represents only a small percentage of the operation and maintenance costs. However, maximum flux achievable needs to be tested for both operating modes in order to evaluate the capital cost in terms of membrane surface requirement (Glucina et al., 1998).

An energy analysis has demonstrated that the major energy consumption takes place in the thermal process and not in the mechanical pumping of the fluid. In addition, higher increments in permeate volume can be achieved by increasing transmembrane pressure, not temperature. The mathematical analysis evaluated of optimum values of the engineering parameters necessary to design and operate skim milk ultrafiltration units (Rinaldoni, et al., 2009).

Performances and energy consumption of pilotscale spiral wound and hollow fiber modules were compared (Cheryan and Kuo, 1983). Exponential flux decay behavior was typical for both units. Flux was affected by pressure in the first 60 min of operation; for the spiral wound unit, flow rate had a beneficial effect only at pressures above 135 kPa. After 3 to 5 h of operation, however, flux became independent of pressure for both modules; flow rate affected the flux of the hollow fiber module but not of the spiral wound module. The average flux of the hollow fiber unit during concentration of whey was double that of the spiral unit, but energy required for recirculation within the hollow fiber unit was higher. Compared to tubular and plate-and-frame units, the spiral and hollow fiber units consumed 10 to 100 times less energy per unit volume of permeate removed.

Chabeaud et al. (2009) studied the performances of ultrafiltration membranes for fractionating a fish protein hydrolysate. They concluded that the cleaning efficiency of the membrane regeneration, at 10 bars, two cleaning procedures are necessary to recover 90% of the initial water permeation flux. The pressure increase makes the cleaning more difficult (only 76% of flux is recovered). Specific energy requirements are practically constant and equal to 0.9 MJ/kg of peptide recovered until a volume reduction factor (VRF) of 2.3 but they increase then significantly to reach 1.20 MJ/kg at VRF = 4.8.

In the present study, milk concentration using UF under different operational pressures as well as the flux behavior during filtration and cleaning processes were investigated. Also, Filter medium resistance and energy consumption during filtration and cleaning processes at different pressures were estimated.

Materials and methods

Milk supply

Fresh Cow's and Buffalo's milks (1:1) were obtained from the herds of the Faculty of Agriculture, Moshtohor, Benha University, Egypt. The total solids and fat contents of the mixed milk were 13.0 and 5.3%, respectively.

UF equipment description

The UF unit consists of 200L stainless steel storage tank, two screw pumps (feeding and circulation pumps), 2 pressure gages, Carbosep M2 membrane (91.Sc.37.5206.1.08 model, 01703 Miribel, Codex, France) it has the following specifications: 37 tube/module, total area of 0.84 m², membrane length is 120 cm, has an external and internal diameters of 1.0 and 0.6 cm, respectively and empty weight is 8.8 kg, The details of schematic diagram of the experimental set up are presented in fig. 1. Membranes were cleaned and sanitized at different pressures and the cleaning efficiency was evaluated and the procedure of Bird and Bartlett, (2002) and Madaeni et al. (2001) was followed.

Determination of fouling

For fouling and performance studies, three batches, 160L each were used. A total recycled mode was used, retentate was returned to the feed tank and permeate was removed continuously to reach the desired milk concentration. Solid content was measured in the retentate and the process was stopped when



Fig 1. Schematic of the experimental setup.

the desired concentration was achieved (32 to 33%). Retentate and permeate flow rates were measured directly with a flow-meter (fixed in the UF equipment) placed in the flow stream. Readings were taken every 15 min. Solid not fat (SNF) in retentate were measured using a hand-held Brix refractometer (Agato, Brix 32, made in Japan). The operational pressures were selected as 3, 4, 5 and 6 bars. Pressure drop was kept on 2 bars for all processes under different conditions and time of process. The averages of the three batches were taken.

Filter Medium Resistance Determination

In UF of milk concentration, permeate flux declines with time due to membrane fouling, which very com-

plicated phenomenon is caused by many chemical and physical properties interactions. It is believed that membrane fouling is a dynamic process starting with pore blocking followed by continuous cake formation on the membrane surface. Pore blocking is a fast process observed at the beginning of UF for a clean membrane due to its high initial permeates flux. As UF process goes on, accumulation and deposition of particles on the membrane surface begin and gel layer is formed. The layer resistance becomes dominant after the initial stage. Ignoring the pore blocking resistance, the whole resistance can be considered as membrane resistance, deposited solute resistance and boundary layer resistance (Marshall et al., 1993). Therefore, for UF of milk concentration

Operational	Co	Coefficient of \mathbf{p}^2								
pressure, bar	а	b	_ determination, R ⁻							
For retentate										
3	8.88	- 0.67	0.979							
4	13.41	- 0.71	0.929							
5	13.33	- 0.65	0.803							
6	8.90	- 0.49	0.885							
For permeate										
3	22.52	- 0.887	0.814							
4	22.90	- 0.844	0.958							
5	23.99	- 0.837	0.968							
6	16.24	- 0.688	0.990							

Table 1. Equation constants of the relationship between retentate flux and process time at different operational pressures.

using Darcy's law, the following equation can be written:

$$J = \frac{1}{A_m} \frac{dV}{dt} = \frac{\Delta P}{\mu (R_m + R_d + R_b)}$$
(1)

Equation 1 could be rewritten as:

$$\frac{1}{A_m}\frac{dV}{dt} = \frac{\Delta P}{\mu(R_m + R_p)}$$
(2)

Where, R_p is polarized solute resistance, using conventional filtration theory, the following equation can be derived:

$$R_p = \alpha_p \left(\frac{VC_p}{A_m} \right) \tag{3}$$

Combining equations 2 and 3 and integrating gives the following filtration equation:

$$\frac{t}{V} = \frac{\mu \alpha C_p}{2A_m^2 \Delta P} V + \frac{\mu R_m}{A_m \Delta P}$$
(4)

The Sperry equation has been modified to account for changing resistance with increasing time. De la Gaza and Boulton (1984) assumed Alpha to be constant and modified the Sperry equation such that the filtration rate is a power function of filtrate volume.

$$\frac{dt}{dV} = \frac{\mu}{A_m \Delta P} \left[\alpha c \left(\frac{V}{A_m} \right)^n + R_m \right]$$
(5)

Integration of equation 5 gives:

$$t = \frac{\mu}{\Delta P A_m} \left[\frac{\alpha c (V / A_m)^{n+1}}{n+1} + R_m V \right]$$
(6)

Bayindirli et al. (1989) tested equation 5 and discovered that a common n in equation 6 could not be found to describe all data at different body feed concentration. Consequently, as alternative equation was proposed which combines the specific cake resistance and solids concentration into a parameter, k.

$$\frac{dt}{dV} = \frac{\mu}{A_m \Delta P} R_m [e]^{kV/A_m}$$
(7)

Integration of equation 7 gives:

$$t = \frac{\mu}{\Delta Pk} R_m [e]^{kV/A_m} \tag{8}$$

Taking a natural logarithm of equation 8:

$$\ln(t) = \ln\left[\frac{\mu R_m}{\Delta P k}\right] + \frac{k}{A_m}V \tag{9}$$

Energy consumption

The energy required to concentrate milk by ultrafiltration is mainly attributed to the pumping (Ghidossi et al., 2006). These pumps are feeding and circulation pumps. The feed pump power, W_{feed} (W) can be expressed as $W_{\text{feed}}=JA_{\text{m}}(P_2)$ (11) The circulation power, W_{circ} (W) can be expressed as: $W_{\text{circ}}=Q_{\text{circ}}(P_1-P_2)$ (12)

Then a total power W_{tot} is calculated as the sum: $W_{\text{tot}} = W_{\text{feed}} + W_{\text{circ}}$ (13) The energy consumed (E_c) per m³ of permeate

produced (kJ m⁻³) is given by:

$$E_c = \frac{W_{tot}}{JA_m \eta} \tag{14}$$

Results and discussion

Flux Behavior

The flux of retentate at different operational pressures (3, 4, 5 and 6 bars) is shown in Figure 2. The results showed that the flux declined rapidly during the first 40 min, slowed down gradually during the period of 40 to 120 min, and after this period, flux was constant until it reached the end of concentration process. In

Operation	Time of Milk concentration process				Cleaning process		
Pressure,	Concentration,	W _{feed} , kJ/L	Weire kJ/L	Wtot, kJ/L of	kJ/L of	Time to full	Energy
bar	min	of retentate	of retentate	etentate	permeate	recovery, min	consumed, kJ
3	175	1.34	50.23	51.57	19.22	100	102.63
4	105	1.84	48.20	50.04	18.18	80	107.00
5	95	2.32	56.95	59.27	21.25	55	87.08
6	80	2.89	59.65	62.54	21.65	50	103.00

Table 2. Energy consumption for batch concentration of milk using UF and cleaning in place.

W_{feed}, W_{circ} and W_{tot}, refer to the power required for feeding, recirculation, and total, kJ



Figure (3) Permeate flux at different operational pressures of UF process.

In general, retentate flux increased with increasing the operational pressure. After 15 min, the flux was as high as 2.17 L/s at 6 bars, while it was as low of 1.35 L/s at 3 bars. By the end of process, flux decreased to reach 0.27 and 0.40 L/s at 3 and 4 bars,

respectively. While at 5 and 6 bars, the fluxes at the end were 0.51 and 0.9 L/s, respectively. These results are in agreement with the results of Tong et al. (1988). In early stage, of milk concentrating using UF, adsorption fouling is probably the primary mechanism of flux decline (Matthiasson, 1985). In the second stage, flux decline is probably due to the concentration polari- zation. But the majority of declination was due to the adsorption fouling. Regression analysis was carried out to obtain a relationship between retentate flux (RF, L/s) and process time (t, s) at different operational pressures. The best relationship between the flux and process time was exponential as follows: $RF = a(t)^{b}$ (15)

The equation constants (a and b) at different operational pressures are listed in table (1).

The flux of permeate at different operational pressures (3, 4, 5 and 6 bars) is shown in Figure 3. The results revealed that the permeate flux increased with increasing the operational pressure. The flux declined rapidly during the first 40 min, slowed down gradually during the period of 40 to 120 min, afterward, flux was constant until it reached the end of concentration process. At 3 bars, operational pressure, the flux decreased from 1.72 L/m^2 .min after 15 min to 0.143 L/m^2 . min by the end of process. At 3 bars, the flux decreased from 1.72 L/m². min after 15 min to 0.143 L/m^2 min by the end of process. At 4 bars, the flux decreased from 2.29 L/m². min after 15 min to 0.191 L/m^2 . min by the end of process. At 5 bars, the flux decreased from 2.87 L/m².min after 15 min to 0.239 L/m^2 . min. by the end of process. At 6 bars, the flux decreased from 3.44 L/m². min after 15 min to 0.287 L/m^2 . min by the end of process. In early stages of milk UF, adsorption fouling is probably the primary mechanism of flux decline and concentration polarization is the second phase of flux decline (Matthhiasson, 1985). At high pressures, a high flow rate decreased the rate of fouling, improving average permeate flux, but increasing energy consumption (De Bruijin et al., 2003).

Regression analysis was carried out to obtain a relationship between permeate flux (PF, L/m^2 . min) and process time (t, min) at different operational pressures. The best fit for the obtained data was in the

250 200 🔹 at 3 bar 150 at 4 bat 100 ▲ at 5 bar 50 ×at6bar Û 20 40 60 80 0 100 120 Time (min) Figure (4) Flux recovery during cleaning at different operational pressures. 35 30 ◆ at 3 bat Total Solids (%) 25 at 4 bas 20 ▲ at 5 bat 15 × at 6 bas 10 0 50 100 150 200 Time (min)

Figure (5) :Milk concentration at different operational pressures

Flux recovery during cleaning process

The flux recovery of the cleaning solution at different UF operational pressures is shown in Figure 4. The full flux recovery of this system was at 280 L/m^2 .h. The results showed the solution flux increased rapidly during the first 40 min of cleaning process, where it reaches 250 L/m².h, and then it was approximately constant until the end of the cleaning cycle. In this process, initially, small cleaning flux increases to a maximum, this occurs due to the removal of surface deposits happened gradually with the cleaning time

until it reaches the complete cleaning, flux restores as a pure water. After 10 min, flux recovery percentages were 5.2, 17.2, 31.0 and 40.4 % at 3, 4, 5 and 6 bars operational pressures, while, by 40 min, flux recovery percentages were the same (86.2%) at different pressures under study. At both 3 and 4 bars, flux recovery percentages were the same by the end of cleaning process (96.55%), while it reach 100% flux recovery after 90 and 80 min when it works at 5 and 6 bars.

Milk Concentration at Different UF Operational Pressures

The total solids (TS) as indicator for milk concentration with time at different UF operational pressures are shown in figure 5. TS were 13% as an average for the whole milk at the beginning of UF process. It increased gradually until it reaches around 30% during the first 50 min., which represents more than 80% of the desired concentration that is suitable of Feta cheese making. As the process goes on, TS increment was slowing down until it reaches around 32 % and then the TS of milk increases very slightly with time at different UF operational pressures under study (3, 4, 5 and 6 bars).

Filter Medium Resistance

Filter medium resistance during concentration process

The effect of operational pressure of UF during the concentration of milk on the predicted medium resistance (R_m) with the process time is shown in Figure 6. It indicated that the R_m increased linearly with the time at different operational pressures. Rm increases with increasing the pressure. Also, it is worthy to notice that at the higher pressures (5 and 6 bars), Rm seems to have no big difference as affected by those two pressures, while there were big differences between R_m values when it works at 3 and 4 bars. The trend of these results is in agreement with those obtained by Rai et al. (2005a).

Filter medium resistance during cleaning process

Medium resistance (R_m) during cleaning is shown in Figure 7 at different operational pressures. It indicated that the R_m increased linearly with the time at different operational pressures during the first 40 min and then decreased. This maybe due to removing the surface deposits and loose particles tend to block off the membrane which in turn increase the resistance, after flushing this solution with these loose particles, R_m decreases (Bird and Bartlett, 2002). The results revealed that R_m increased with increasing the pressu-



exponential for (equation 15) and its constants are

listed in table (1).



Fig. (6): Effect of UF opertaional pressure on the filter medium resistance (Rm) as a function of time during milk concentration.



re. R_m reached the maximum $(1.2x10^{10} \text{ 1/m})$ at the higher operational pressure (6 bars) after 40 min. Meanwhile, it was $6x10^{10}$ 1/m at the lower pressure (3 bars) at the same time. This could be attributed that the quality of cleaning water has a great importance to membrane fouling because of the presence of suspended particles which deposit on the membrane surface and as the pressure increases; these deposited particles became compacted and increase the filter resistance (Renner and Abd El-Salam, 1991). Rm decreased gradually after 40 min to reach 4.5x10⁹, 7.06x10⁹, 9.73x10⁹, and 1.16x10¹⁰ 1/m, by the end of cleaning process at 3, 4, 5, and 6 bars.

Energy Consumption

Energy consumption in UF is divided into parts, one for retentate recirculation and the other for feeding pump.

For retentate recirculation

The accumulative energy consumption at different operational pressures during UF process of recirculation of milk retentate is shown in Figure 8. The results indicate that the accumulative energy consumption increased gradually with time of concentration. where it was 405, 495, 549 and 651 kJ after the first15 min. at 3, 4, 5, and 6 bars. After one hour, energy consumed values were 1080, 1428, 1767 and 2026 kJ for the same pervious order, which means by increasing the process time to 4 folds, energy increased 2.67, 2.89, 3.22, and 3.11 times for the same pervious order. By the end of process, total energy consumed were 2009 kJ after 175 min at 3 bar pressure, 1928 kJ at 4 bar for 105 min, 2278 kJ at 5 bar for 95 min and 2386 kJ at 6 bars for 80 min operation time. It could be concluded that UF consumed more energy at the higher operational pressures but in a shorter time.

For feeding pump (permeation flow)

The accumulative energy consumption at different operational pressures during UF process for permeate is shown in Figure 9. The results indicated that the accumulative energy consumption increased gradually with time of concentration, where it was 15.31, 21.76, 28.77 and 36.45 kJ after the first15 min. at 3, 4, 5, and 6 bars. After one hour, energy consumed values were 33.81, 55.52, 74.00 and 90.50 kJ for the same pervious order, which means by increasing the process time to 4 fold, energy increased by 2.21, 2.55, 2.57, and 2.46 times for the same pervious order. By the end of process, total energy consumed were 53.66 kJ after 175 min at 3 bars, 73.39 kJ at 4 bars for 105 min, 92.98 kJ at 5 bar for 95 min and 115.54 kJ at 6 bars for 80 min operation time.

During cleaning process

Figure 10 shows the energy consumption during cleaning process at different UF operational pressures (3, 4, 5 and 6 bars). UF flux recover for this equipment was at 280 L/h.m2. the energy consumed to reach the full recovery of flux were 88.63 kJ for 90 min at 3 bars, 107 kJ for 80 min at 4 bar operational pressure, 87.71 kJ for 55 min at 5 bar and 103 kJ for 50 min at 6 bars.

Specific energy consumption

The energy consumption in milk concentration and cleaning at different operational pressures in UF process is shown in Table (2). The results indicated that the time required for concentration decreased



different operational pressures during UF milk concentration.



different operational pressuresUF milk concentration.



with increasing the pressure, where it decreased to one third when pressure increased from 3 bars to 6 bars. The total energy consumed for milk concentration ranged from 50.08 to 62.54 kJ/L of retentate, while it ranged from 18.18 to 21.65 kJ/L of permeate. The time required to reach full recovery of flux during UF cleaning decreased with increasing pressure where it decreased from 100 min at 3 bar pressure to 50 min at 6 bars. The energy consumed for cleaning ranged from 87.08 kJ at 5 bars to 107 kJ at 4 bars.

Conclusions

Milk was concentrated from 13% to 32% TS using ultrafiltration (UF) at different operational pressures. Retentate and permeate fluxes decline during concentration of milk was measured with time of process, as well as, flux recovery during cleaning. Filter medium resistance during filtration and cleaning were determined from the experimental data, and finally energy consumption for feeding, recirculation and cleaning were estimated as well. The study has shown that the operational pressure has a great effect on the UF performance. The results indicated that to concentrate milk from 13 to 32% at 3 bar operational pressure needed 175 min and consumed a total energy of 51.57 kJ/L while it needed 80 min only at 6 bars and consumed total energy of 62.54 kJ/L with a difference of energy consumption of 10.97 kJ/L (3.05 kWh/ m³) of retentate. To reach the full flux recovery during cleaning process at 3 bars needed 100 min and consumed an energy of 102.63 kJ while it needed half of this time to reach full recovery at 6 bar and consumed 103 kJ, which gave us very useful indicator for UF operation during the cleaning process, it is recommended to be done on higher pressures.

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