Membrane distillation for milk concentration

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ABSTRACT

Membrane distillation is an emerging technology to concentrate liquid products while producing high quality water as permeate. Application for desalination has been studied extensively the past years, but membrane distillation has also potential to produce concentrated food products like concentrated milk. Water vapour migrates from the milk feed side to the permeate side by the vapour pressure difference between the two sides of a hydrophobic membrane. Unlike pressure driven membrane filtration high solid concentrations are achievable.

Experimental results show that concentrations of 50% total solids can be achieved for milk, which makes membrane distillation a competitor to evaporation. In experiments for skimmed milk, ranging from 40 to 60°C, the flux was found to increase with higher feed temperatures. For skimmed milk with 25% total solids, at 60°C an initial flux of around 16 kg/m^2 per hour was achieved. Due to gradual fouling built the flux declined to 8 kg/m^2 per hour after 15 hours. At concentrations of 50% total solids the flux declined to 3 kg/m^2 per hour.

Confocal laser scanning microscopy (CLSM) was used to investigate the size and morphology of the fouling layer. Images indicated homogeneous fouling layers with a thickness depending on the process temperature; lower temperature resulted in thinner fouling layer. Most plausible reason is the lower driving force and transmembrane flux. These lab scale experiments showed promising fluxes, which still can be improved, that are a good starting point for the development of large scale production units.

Keywords: Membrane distillation; Milk; CLSM; Membrane fouling; Energy efficiency

INTRODUCTION

With the rapidly growing world population to feed, 8 billion in 2025 (United Nations, 2012), the need for efficient use of raw materials and energy sources is increasing. Sustainability has therefore become a major issue in the food industry, and the pressure of the society for sustainable food production has become larger. The EU works on the realization of their Europe 2020 strategy to achieve sustainable growth and discusses already the targets for 2050. Efficient use of water and energy have a key role in these strategies.

One of the food industries in which large quantities of water and energy are involved, is the dairy industry. Product concentration and drying are large energy consuming processes and release considerable amounts of water. Although new technologies have been introduced, the production process for milk powder has not changed radically over the last half a century. Nowadays concentration is mostly done by falling film evaporators, which concentrate the milk to a total solid content of approximately 50% (Walstra et al., 1999). By making use of a multiple stage evaporator and thermal or mechanical vapour recompression, the energy use can already be reduced to around 300 kJ/kg water removed (Ramírez et al., 2006). Together with the drying step, however, concentrating is still highly energy consuming.

Concentrating milk with reverse osmosis uses considerably less energy compared to concentration by evaporation (reverse osmosis: 14 – 36 kJ/kg water removed (Ramírez et al., 2006)). Due to increasing osmotic pressure and concentration polarisation with the degree of concentration, the maximum concentration by reverse osmosis is around 20% total solids, which is far from the 50% total solids required for spray drying. Nowadays reverse osmosis is, therefore, used as a pre-concentration step followed by evaporation which is used to reach the final solids concentration. An alternative membrane separation method is membrane distillation, which allows concentration to higher solid contents. This method is, other than reverse osmosis, not limited by the osmotic pressure and concentration polarization has a less significant role (Tijing et al., 2014). For example Laganà et al. (2000) managed to concentrate apple juice to a concentration of 64°Brix. Figure 1 gives a schematic overview of the membrane distillation process. Water vapour passes the membrane due the vapour pressure difference between the hot and the cold stream.

Advantages of this process, besides higher final solid concentrations, are the use of low quality energy (waste heat), and the water produced (permeate) has a high purity (Hausmann et al., 2014).

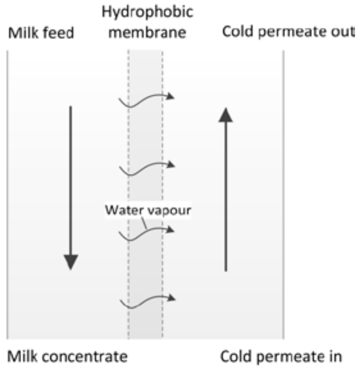

Figure 1. Schematic representation of the membrane distillation concept. The warm milk feed is separated by a hydrophobic membrane from the cold permeate stream. Due to the temperature difference, and thus the vapour pressure difference, water vapour moves from the hot to the cold side of the membrane.

The current knowledge of membrane distillation in relation to dairy processing is limited. Hausmann et al. (2011, 2013, and 2014) studied membrane distillation to concentrate skimmed milk, whole milk, whey and lactose solutions. For skimmed milk they achieved a total solids content of 43.5%. Optimization of the process conditions could increase the total solids content even further. A combination of reverse osmosis and membrane distillation seems a promising replacement for the conventional evaporation step for milk concentration. Further investigation is necessary to explore the potential of membrane distillation.

Fouling has a large influence on the flux decline in membrane processes (Gryta, 2008). It is, therefore, important to understand how operating conditions influence fouling formation. Confocal laser scanning microscopy (CLSM) is a technique to construct 3D images of labelled samples, allowing formation of cross sectional images. CLSM could be used in membrane distillation process to analyse fouling formation in membrane samples (Ferrando et al., 2005).

Our objective is to investigate the potential of membrane distillation for skimmed milk concentration. This work shows first lab scale results of skimmed milk concentration using membrane distillation, and the influence of operating conditions on fouling formation.

MATERIALS & METHODS

Membrane distillation

The membrane distillation system was set up by TNO, Zeist, based on the Memstill technology (Hanemaaijer et al. 2006). The system contains a hydrophobic flat-sheet membrane with similar properties to expanded PTFE membranes (but showing less compaction), a thickness of about 120 µm, nominal pore size of 0.2 µm, porosity of 80%, and an effective area of 0.05 m^2 (flow channel length 500 mm, width 100 mm, and height 2 mm). Temperature and conductivity were measured at every in- and outlet flow of the membrane module. Transmembrane flux was continuously measured by recording the weight gain of the permeate, which is re-circulated in a closed system.

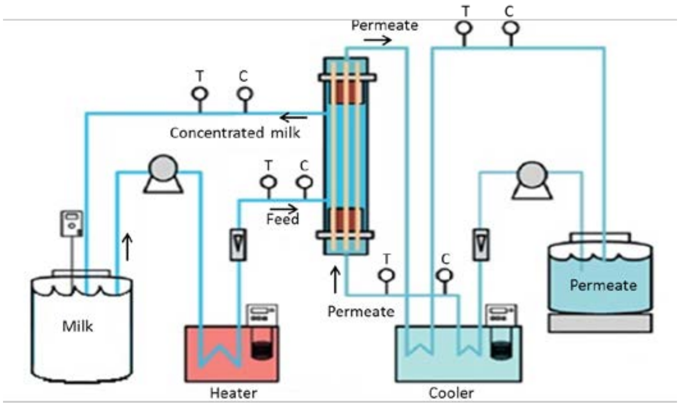

Figure 2. Experimental setup of the membrane distillation system. The milk feed and permeate are either heated or cooled before recycling. Temperature (T) and conductivity (C) are measured for at every in- and outlet of the membrane.

Experiments were either performed in continuous or in batch mode. In continuous mode the concentration of the milk feed was kept constant by re-adding permeate to the milk reservoir. In continuous mode three different feed temperatures were tested: 40°C, 50°C and 60°C. In the batch mode the milk was recirculated and concentrated from 12% through 20% total solids and from 25% trough 50%. Experimental settings during the experiments are in Table 1. Deionized water was used for the permeate flow.

Table 1. Operational settings for batch experiment with low and high final concentration, and continuous experiments with different feed temperatures.

Process parameters	Batch High	Batch Low	Continuous
Temperature feed	55°C	50°C	40°C, 50°C or 60 °C
Temperature permeate	13°C	15°C	13°C
Feed flow rate	3.5 L/min	3 L/min	3.5 L/min
Permeate flow rate	3 L/min	3 L/min	3 L/min
Initial concentration	25%	12%	25%
Final concentration	50%	20%	25%

Skimmed milk solutions of 12% and 25% total solids were prepared by rehydrating skimmed milk powder (ELK, Campina, the Netherlands).

Confocal laser scanning microscopy

A Zeiss LSM510 CLSM was used to create cross sectional images of membrane samples. The membrane samples were analyzed in fluorescents mode (with Nile red as fluorescents marker). All samples were analyzed with a 40x objective, and zoom magnification of 3. Samples were taken from the center of the membrane sheet.

RESULTS & DISCUSSION

Figure 3a shows the flux development during concentration of skimmed milk with the membrane distillation system. A concentration of 20% total solids (from 12% total solids) was achieved after 4 hours, with an initial flux of 25 kg/m^2 per hour, and final flux of around 5 kg/m^2 per hour. The flux reduction is fastest at the initial phase of the experiment. Figure 3b shows the results of the flux development where milk is concentrated from 25% through 50% total solids. Compared to Figure 3a the initial flux was much lower (around 11 kg/m^2 per hour) and the flux reduces faster in the initial period of concentration. The final flux stabilizes around 3 kg/m^2 per hour. After 19 hours a total solid content of 50% was achieved. In the continuous mode, operating at a constant feed concentration of 25% total solids, the flux declined in the initial period. After the initial period, however, the flux is almost constant around 8 kg/m^2 per hour (50°C in Figure 4).

Results of these experiments illustrate the influence of product concentration on the transmembrane flux. Higher solids concentration result in a reduced flux, because of higher amounts of solids present. Higher amounts of solids results in the formation of a thicker fouling layer. For a constant concentration the flux stabilizes i.e. most of the fouling is build-up in the initial phase. These results are in line with the work of Hausmann et al. (2014) although in the current work higher fluxes were achieved. These higher fluxes can possibly be attributed to the usage of different membrane and feed channel spacer.

Operational conditions for the experiment presented in Figure 3a were slightly different from the experiment presented in Figure 3b (see Table 1); slightly higher permeate and lower feed temperature, and lower feed flow rate have an effect on the flux. A larger temperature difference over the membrane has a positive effect on the flux. A lower flow rate, however, can influence the flux negatively, as the fouling layer is less disturbed by the feed flow.

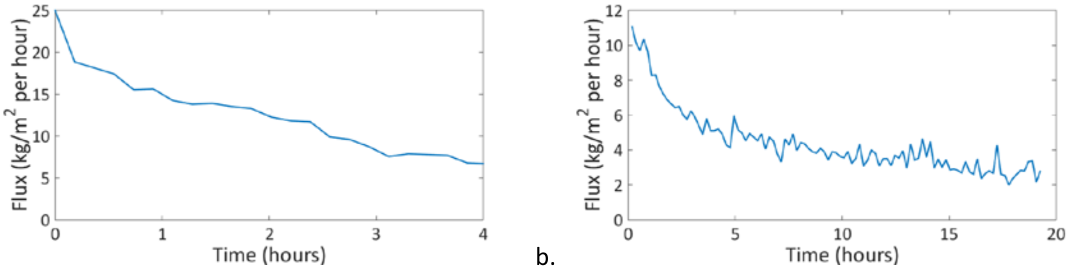

Figure 3. Flux during skimmed milk concentration by membrane distillation from 12% through 20% total solids in 4 hours (a), and from 25% trough 50% total solids in 19 hours (b).

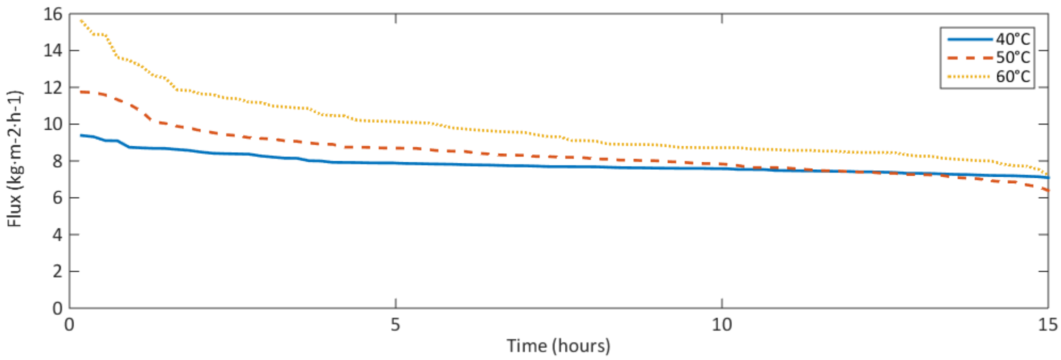

Figure 4. Flux during skimmed milk concentration by membrane distillation at different feed temperatures (40°C, 50°C and 60°C), while keeping the inlet milk concentration at 25% total solids.

Membrane distillation is driven by difference in vapour pressure. Feed temperature, therefore, has an influence on the performance. Figure 4 shows the influence of feed temperature on membrane performance. The temperature influence is the greatest in the initial phase. For a feed temperature of 60°C the initial flux is around 16 kg/m^2 per hour, while for a feed temperature of 40°C the initial flux is around 9 kg/m^2 per hour. The difference in initial flux is caused by the difference in temperature, and thus in driving force.

During the experiments the flux stabilizes between 7 kg/m^2 per hour and 9 kg/m^2 per hour for all different feed temperatures. This could be explained by a higher resistance over the membrane caused by a thicker fouling layer. Higher temperatures lead to higher fluxes, meaning more particles move towards the membrane. More particles moving towards the membrane might results in a thicker fouling layer that reduces the flux. In order to maintain higher fluxes, introducing a cleaning cycle might be a solution. Cleaning after a few hours will restore the flux to the initial flux, and improve overall performance.

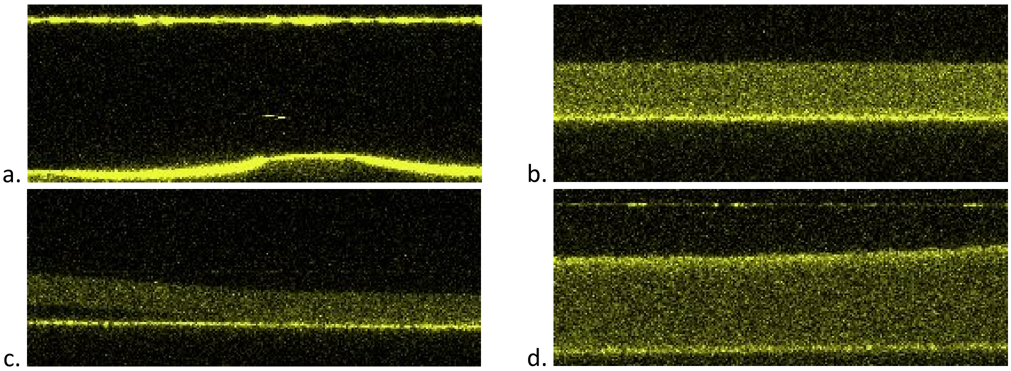

Figure 5. CLSM images of the XZ direction (75.8x28.0 µm) of a clean membrane sample(a), membrane sample after concentrating with feed temperature of 50°C (b), and of 40°C (c) both with constant inlet concentration of 25% solids. And a membrane sample after concentrating to high solids content (from 25% through 50%) with a feed temperature of 55°C (d).

In order to investigate the influence of the operational settings on fouling formation, CLSM was used to create cross sectional images of several membrane samples. Figure 5 shows the cross section results of the membrane samples. Figure 5a shows a clean membrane, in which the top fluorescence layer corresponds to the glass layer that covered the sample, and the lower layer is most likely the top layer of the membrane. Between these two layers the fouling layer can be seen in Figure 5b, c and d. In these figures a difference in thickness of this layer can be seen. This difference is likely caused by the different operational conditions. Increased temperature results in more fouling; i.e. the fouling layer is thicker at 50°C compared to 40°C (Figure 5b and c). This corresponds with the results in Figure 4. Concentrating milk to a solid content of 50% results in a fouling layer that is thicker (Figure 5d) compared to concentrating at lower solid content of 25% (Figure 5b and c).

CONCLUSION

This work shows the potential of using membrane distillation to concentrate skimmed milk. At a milk concentration of 25% total solids the highest fluxes (around 16 kg/m^2 per hour) are achieved with a feed temperature of 60°C. At the beginning of the experiment the flux drops exponentially. After this initial drop the flux reduces more linearly to around 8 kg/m^2 per hour after 15 hours. This behaviour is observed in all experiments. Higher feed temperatures result in higher initial fluxes. Higher fluxes at the same time lead to more fouling formation, resulting in a comparable final flux. Introducing a cleaning cycle after this phase is advisable to regain higher fluxes. CLSM images of the membrane confirm the negative influence of higher temperature and higher concentrations of solid content on fouling formation. Smaller fouling layer is observed at a feed temperature of 40°C. A 50% total solids concentration is achievable with membrane distillation. Higher concentrations, however, will lead to greater fouling formation.
These lab scale experiments show promising results. Transmembrane fluxes, however, can be further improved by optimization of operational conditions. Membrane distillation has the potential to drastically reduce the energy consumption in milk concentration, because no external high quality energy for phase change is required, and high concentrations can be achieved.

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