Redesign of the milk powder production chain:
assessment of innovative technologies

Sanne N. Moejes

Propositions

1. Process design and optimisation at an early stage are essential for the viability of a processing chain.
   (this thesis)
2. In order to cut down on fossil fuels, investment in innovative technologies is a necessity.
   (this thesis)
3. Scientific advancement does not guarantee an increase in quality of life.
4. The continuous growth of computer power will make model-based solutions to scientific problems increasingly favourable over experimental approaches.
5. Daydreaming is both your best friend and worst enemy as a scientist.
6. To enhance healthy food choices later in life, it is beneficial to teach children about food and agricultural production at primary schools.
7. An organic lifestyle is a western privilege.

Proposition belonging to the thesis, entitled:

Redesign of the milk powder production chain: assessment of innovative technologies

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Thesis

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Table of contents

Chapter 1

Introduction

1 Global challenges

The world population has increased at a fast rate, and is predicted to continue; by 2050 the planet will be shared by 9.8 billion inhabitants (UN, 2017). This is twice the population from 1980. The growing number of people to feed, leads to an inevitable need for us to increase our food production in order to ensure food security. The production of most food commodities has already doubled since 1980 (see Figure 1). The increase in food production is not only needed to feed more people but also to cope with the increased consumption per person. The average caloric intake per person was in 2012 19% higher compared to 1980, which is an consequential result of the increase of overall consumption and shift towards western diets, and is only expected to increase further (FAO 2012). This all results in a demand for a more efficient use of available (food) resources if the impact on the environment is to be minimised. With this in mind, and to guarantee food security the EU formulated its Energy Road Map 2050 (EC, 2012).

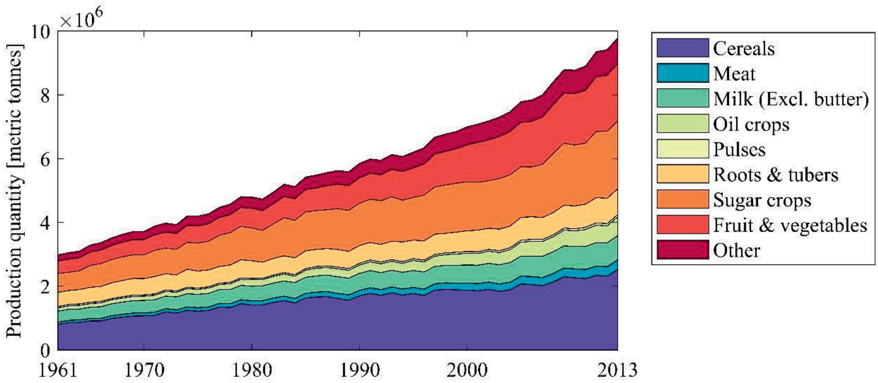

Figure 1. Increase in world production of primary food commodities from 1961 to 2013 (data source: FAO (2016)).

Within the energy road map, the EU set the goal to cut greenhouse gas emissions by 80 – 95% by 2050. In order to meet this goal, our current energy consumption has to be reduced. In 2013 the food industry was responsible for 26% of the total energy usage in the EU, 28% of which is allocated to food processing (Monforti-Ferrario et al., 2015). To achieve the goals on reducing energy usage and cutting down on greenhouse gas emissions, innovation in industrial processing is needed. Energy efficient processing systems with low environmental impact are essential to achieve these targets. For this reason, it is important to look for innovative alternative technologies. The work in this thesis focusses, for this reason, on the redesign of a food production process, and the impact and opportunities of different innovative technologies.

2 Energy consumption for dried food products

In food processing thermal processes, like drying, concentrating, and pasteurizing, are responsible for the main part of the energy consumption in food processing (Klemeš, Smith, & Kim, 2008; Ladha-Sabur, Bakalis, Fryer, & Lopez-Quiroga, 2019). Drying is the thermal removal of moisture or water from a solid, and is an important method to preserve food for a long time. Besides prolonging the shelf life of a food product, storage and transportation of dried products is more efficient due to the reduced volume and weight. Drying is an energy intensive process, and responsible for 10 – 25% of the total national energy consumption of western countries (Mujumdar, 2007).

Prior to drying liquid foods are normally concentrated. Due to the efficiency of evaporation with respect to drying, it is more effective to concentrate liquid foods first. Evaporation is used for the concentration of liquid products, like milk, from a dry matter concentration of around 5 – 10% to a final dry matter concentration of around 50 – 70%. Fast evaporation takes place at the boiling point of the liquid, and is controlled by heat transfer. In a single-stage evaporator approximately 2.3 MJ (latent heat of evaporation) are needed to evaporate 1 kilogram of water. By using multiple stages, the energy consumption is lowered to 2.3 MJ divided by the number of stages. The energy consumption of a multi-stage evaporator ranges between 0.3 – 1.2 MJ per kilogram water removal, depending on the number of stages (Ramirez, Patel, & Blok, 2006; Westergaard, 2004).

Drying, on the other hand, is controlled by mass transfer, and most dryers require more energy than just the latent heat of evaporation. The majority of industrial dryers are convective dryers with hot air as drying medium. Examples are: spray dryers, flash dryers, fluidized bed dryers, and conveyor dryers. The overall thermal efficiency of convective dryers is often less than 60%, resulting in an energy usage of 3.5 – 4.5 MJ for the removal of 1 kilogram of water (Kemp, 2005; Kudra, 2012).

2.1 Dairy industry

The dairy industry makes extensive use of evaporation and drying processes. Improving the energy efficiency of these process is of increasing interest at the moment, because of the ending of the EU milk quota in 2015. The EU milk quota, was established in 1984, and over the past decades the milk production was almost constant in Europe. Due to the restricted production high value specialties, like cheese, were favoured over commodity products (Kelly, 2006). Termination of the quota resulted in an expected increase of milk production, followed by an increase in commodity products like whole or skimmed milk powder (EC, 2018; Jansik, Irz, & Kuosmanen, 2014). At the same time the demand for milk powder is rising due its long shelf life and nutritious value. Milk powder is not only used as a substitute for fresh milk, but also applied in many types of infant formulas, desserts, bakery products, chocolates, soups, and other dairy products (Augustin & Margetts, 2003). Due to the shift in life style in especially Asian countries, the demand for infant formulas and health products containing milk powder is increasing. By 2030 the skimmed milk powder production from the EU is expected to rise by 15% compared to 2018. (EC, 2018)

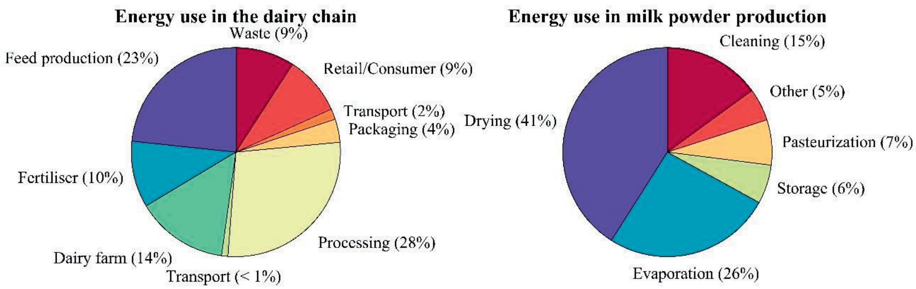

Figure 2. Distribution of energy usage along the dairy chain in the Netherlands (left), and an average milk powder processing plant (right) (Krebbekx, Lambregts, Wolf, & Seventer, 2011; Ramirez et al., 2006).

In the dairy chain, from feed production to consumer, most energy is used for feed production and dairy processing (Figure 2). The major part of raw milk is processed as fresh fluid milk (58 – 67%), the rest is further processed into a wide range of products like butter, condensed milk, and a large variety of cheeses and dried milk products (Flapper, 2009; Xu & Flapper, 2009). Due to the further processing, the energy consumption in the dairy industry is high, especially for dried products where large amounts of water have to be removed. The production process of milk powder requires 11.1 – 26.2 MJ per kilogram of powder, compared to fluid milk which requires 1.0 – 1.5 MJ per kilogram of product (Ramirez et al., 2006; Xu & Flapper, 2009; Yan & Holden, 2018).

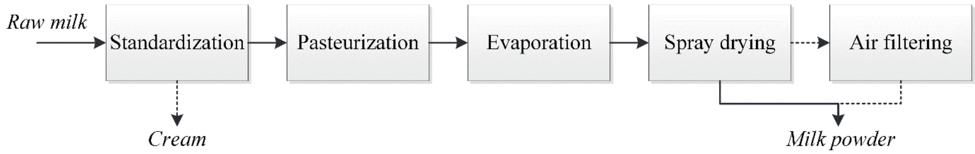

Figure 3. Schematic overview of the main processes in milk powder production, from raw milk to powder.

Milk powder production starts with the receiving of raw milk, which is subsequently standardized, pasteurized, concentrated and dried. The process steps involved are depicted in Figure 3. As discussed above, and shown in Figure 2, the concentration (evaporation) and drying are the most energy consuming processing steps. Over the past decades large efforts have already been made to increase energy efficiency of the traditionally used multi-stage evaporators and spray dryers. Introduction of thermal vapour recompression (TVR) and mechanical vapour recompression (MVR) decreased the energy use of evaporators significantly. When applying TVR on a five-stage evaporator the energy consumption is lowered from around 0.45 MJ to 0.3 MJ per kilogram water removed. Combining an evaporator with TVR, with an additional MVR unit lowers the energy consumption even further to around 0.05 – 0.1 MJ per kilogram of water removed. (Ramirez et al., 2006; Westergaard, 2004)

Despite the large potential, high investments costs are often a reason why the newest technologies are not yet implemented.

Following the concentration step, milk is dried. Spray drying is most applied in milk powder production, due to the relatively low product temperature in the dryer. Milk is a heat sensitive product, therefore, lower product temperatures are preferred. Unlike the large improvements in energy consumption for the evaporation process, achievements in spray drying have been less extensive. A two-stage drying process, where spray drying is combined with a fluidized bed dryer, increases the energy efficiency of the drying process by 13% (Ramirez et al., 2006; Walstra, Geurts, Noomen, Jellema, & Boekel, 1999; Westergaard, 2004). Furthermore, optimisation of the drying chamber and process settings is essential in process controllability and energy reduction (Baker & McKenzie, 2005; Kudra, 2012; Verdurmen et al., 2002). A major opportunity to increase the energy efficiency of dryers is the energy recovery of the latent and sensible heat of the exhaust air of the dryer. Several studies have already assessed the potential of heat recovery and integration of the dryer with other processes (Atkins, Walmsley, & Neale, 2011; Krokida & Bisharat, 2004; Walmsley, Walmsley, Atkins, Neale, & Tarighaleslami, 2015). The expected savings reported are in the range of 14 – 25% when using a heat exchanger.

In view of the global challenges we are facing, the EU has decided that large reductions in energy usage are required. To achieve these, breakthrough solutions like the ones mentioned above are necessary, rather than small incremental decreases. The potential of innovative technologies requires further investigation for milk powder production.

3 Research challenges

The introduction of new technologies and also modification of existing technologies in the milk powder production chain has an impact on up and downstream processes, the product properties, production costs, and environmental impact. For this reason, it is of importance in the development of new technologies, to assess its effect on the entire production chain by using a systematic modelling approach. This is the objective of the work presented in this thesis, which was used as contribution to the EU project ENTHALPY. Within the ENTHALPY project different innovative technologies were developed, with the goal to reduce the energy consumption of milk powder production by 63%.

This led to the first question to be answered; what are alternative technologies for milk powder production to reduce energy consumption? Thereafter, the question on how these technologies will be integrated with one another, and what would be the best configuration in terms of costs and environmental impact will be addressed. Additionally, the optimisation of the operational conditions is of major importance to reach the maximum impact.

3.1 Innovative technologies

Several studies have already pinpointed the large energy consumers within the milk powder chain: concentrating and drying (Ramirez et al., 2006; Xu & Flapper, 2009; Yildirim & Genc, 2017). The key bottleneck identified is the energy efficient removal of water.

Membrane processes have been successfully developed and used as a pre-concentration step in dairy processes. The application of reverse osmosis (RO) to concentrate milk to 15 – 20% solids before evaporation already lowers the energy consumption to 14 – 36 kJ per kilogram of water removed (Ramirez et al., 2006). However, RO as a pressure driven membrane process is limited by concentration polarisation. Hence, the maximum solids concentration lays around 20% (Walstra et al., 1999). An alternative membrane technology is membrane distillation (MD), which is a thermal driven technology and thus potentially less limited at high solids concentrations. Over the past decades, MD has been proven as a desalination technology, and is currently in the commercialisation phase as such (Lawson & Lloyd, 1997; Wang & Chung, 2015). For the concentration of food products like milk, some studies have been done, but applications are still limited (Hausmann et al., 2011; Nene, Kaur, Sumod, Joshi, & Raghavarao, 2002). An advantage of MD are the low processing temperatures (up to 60 – 70 °C) which provides more opportunities for the use of processing waste heat from other processes, compared to when using evaporators. The challenge of using MD for the concentration of milk, however, will be to limit the effect of fouling deposition of milk components on the membranes, as well as the development of suitable membranes.

In a conventional milk powder plant, most energy is lost in the form of heat in the exhaust air of the spray dryer (Atkins et al., 2011; Kudra, 2012). The exhaust air contains, in addition to water vapour, small powder particles (fines), which are mostly removed by cyclones and bag filters. Besides causing product losses, these fines cause fouling in heat recovery equipment, which makes heat recovery of the exhaust air challenging. Monodisperse droplet drying is one of the investigated technologies developed within the ENTHALPY project (the research described in this thesis is part of that project), which will eliminate the fines from the exhaust air (Deventer, Houben, & Koldeweij, 2013; Wu, Patel, Rogers, & Chen, 2007). By making use of nozzles based on inkjet technology, streams of spherical droplets are produced which have a very narrow size distribution. The mono-dispersity of the milk droplets allow for better process controllability, and avoidance of the production of fines.

Integrating the monodisperse droplet dryer with an adsorption system to recover the heat from the exhaust air, has the potential to increase the energy efficiency of the drying section to a large extent. Zeolite or silica adsorption systems are known to have a positive effect on the dryer efficiency (Boxtel, Boon, Deventer, & Bussmann, 2012). An alternative, not yet proven, air dehumidification technology is a membrane contactor system with a liquid desiccant. Membrane contactors are already used at lower temperatures mainly for air conditioning purposes or gas absorption and stripping (Klaassen, Feron, & Jansen, 2005; Woods, 2014;

Yang, Yuan, Gao, & Guo, 2013). At elevated air temperature the application of a gas-liquid membrane contactor is not developed yet. For this reason, the question which type of adsorption system has the most potential as a dehumidification technology will be investigated in this thesis.

3.2 Impact evaluation

In this thesis energy efficiency and environmental impact are key characteristics of a technology, but besides this, a technology should also be economically viable for industry to adopt it. The economic and environmental impacts of the proposed innovative technologies will be assessed and compared in order to select the optimal combination for a milk powder process with the least impact.

Life cycle assessment (LCA) is a widely used tool to evaluate the environmental impact of an existing production chain. Traditionally, in process design, operational conditions and equipment dimensions are optimized to maximize economic performance while meeting operational constraints to ensure product quality. The environmental impact is often assessed afterwards by LCA. Pieragostini et al. (2012) reviewed process optimisation combined with LCA methodology, and concluded that LCA in process design is increasing. Although some literature is available, the use of LCA in processes design is still limited (Azapagic & Clift, 1999a, 1999b; Gerber, Gassner, & Maréchal, 2011; Guillén-Gosálbez, Caballero, & Jiménez, 2008; Pieragostini et al., 2012; Zhou, Yin, & Hu, 2009). Instead of applying the LCA when the process design is completed and all production data is available, LCA in early stage process design can be powerful in the design of new production chains. The approach helps to identify bottlenecks in the chain which could be circumvented in an early stage. For this reason, in this thesis we aim for the integration of LCA in the process design and optimisation of the milk powder production chain.

4 Process design and simulation

The implementation of new technologies in production chains, will have impact on the overall process performance. Both up and downstream processes are affected by the replacement, change, or addition of another unit operation. Modelling and simulation of the entire production process can be used as a powerful tool to provide insights in potential environmental and economic savings.

The use of systematic computer aided methodologies to optimize the design, operation, or control of a process is often referred to as Process System Engineering (PSE). PSE is widely applied in the chemical, petrochemical, pharmaceutical, and also more and more in the food industry (Cameron et al., 2019). Tools used in PSE and applied in this work are the use of mixed-integer nonlinear programming for the optimisation and synthesis of process flows sheets or superstructures, pinch analysis for optimal heat integration, process scheduling, multi-objective optimisation, and sensitivity analysis. Multi-objective optimisation will be of interest to evaluate both the economic and environmental impact of alternative processing chains.

Process modelling can be done at many levels, from a black box model of an entire production facility to a detailed model of interactions at molecular level. In this work we will look at process modelling at different levels in order to gain a better understanding of the different processes, their optimal conditions, and optimal systems integration. Especially for innovative technologies, with a limited amount of available process data, simulations will help to assess the impact and identify what crucial parameters are.

5 Thesis aim and outline

Innovative technologies are a necessity to realize breakthrough steps in environmental impact reduction. The aim of this thesis is to use different modelling and optimisation tools to assess the potential of innovative technologies on lowering the energy usage and environmental impact in the milk powder production chain. This will lead to a redesign of the current milk powder production chain.

In Figure 4 the schematic overview of the thesis outline is given and highlights the focus areas of the different chapters. The work starts with the general evaluation of the milk powder production chain in **Chapter 2**. In this chapter both the state-of-the-art and available innovative technologies and their potential energy savings are described and discussed.

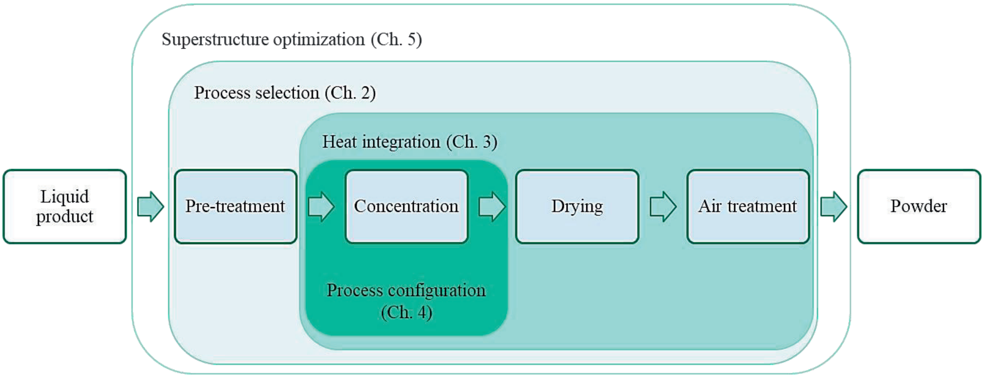

Figure 4. Schematic thesis outline.

Chapter 3 focuses on the energy reduction of the drying section by optimizing different configurations of a monodisperse closed-loop dryer. The aim is to recover the latent and sensible heat from the exhaust air by a dehumidification system. A closed-loop drying system has a surplus of heat which cannot be fully utilized in the dryer section; hence the concentration step is included here as a heat sink. The operational conditions and the heat integration network are simultaneously optimized in order to minimize the energy usage. **Chapter 4** discusses alternatives for the concentration step. Membrane distillation is proposed as an alternative for the multi-stage evaporation process. A combined membrane network of reverse osmosis and membrane distillation is simulated and optimized in order to minimize the economic and energy objectives.

Chapter 5 considers the whole processing chain from milk to powder. By creating a superstructure consisting of all potential processing units and all possible processing pathways, multi-objective optimisation is applied to minimize both environmental and economic impact. LCA is applied to cover the environmental impact at this stage of early process design, and circumvents the iterative loop after the design phase which is applied in the traditional process design approach. Chapter 5 results in the optimal processing chain for milk powder production.

Chapter 6 finalizes the thesis with the main findings of this work and an elaboration on future work.

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Chapter 2

Energy saving potential of emerging technologies in milk powder production

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Abstract

The food industry has a large potential for energy reduction which, with an eye on the future, has to be exploited. Milk powder production consists of many thermal processes and is responsible for 15% of the total energy use in the dairy industry. A reduction in energy consumption can be realized by using innovative technologies instead of realizing incremental process modifications. In this work first the current practice in milk powder production is described and analysed with respect to energy consumption. Then the potential of emerging technologies for milk processing like membrane distillation, monodisperse-droplet drying, air dehumidification, radio frequency heating, combined with renewable energy sources as solar thermal systems, are investigated. The combination of emerging technologies is able to reduce the operational energy consumption for milk powder production up to 60%, i.e. from 10 MJ/kg powder in current production to 4 – 5 MJ/kg powder. The implementation of these technologies and development of new production chains is essential to meet the future demand on energy efficient processing.

Keywords: Milk powder, energy consumption, membrane distillation, monodisperse-droplet drying, air dehumidification, radio frequency heating

1 Introduction

Society has an increased awareness on the limits in current energy usage and the consequence of emissions demands for sustainable production processes. This is reflected by the European Commission targets for 2050, i.e. cutting over 80% of the greenhouse gas emissions and moving to a carbon free energy system (European Commission, 2016). These targets can only be achieved by a significant improvement in energy efficiency of production processes and by moving to renewable energy sources. The costs aspects of energy have always been the important motive for energy reduction in industrial processing, but in most processes only incremental improvements have been realized.

A significant amount of the total energy consumption in the food industry is related to thermal processing. Okos et al. (1998) conclude that the contribution of thermal processing is around 29%. Wang (2008) estimates the contribution even at 59%. A sector with many heating and cooling processes is the dairy industry, which is in the Netherlands responsible for 15% of the total energy demand in the food sector (Ramirez, Patel, & Blok, 2006). Especially dehydration processes for the production of milk powder (generally divided in 35% whole and 65% skimmed milk powder (Eurostat, 2015)) require significant amounts of energy. According to Ramírez et al. (2006) the production of milk powder requires 11 MJ/kg powder, while the production of cheese requires 4 MJ/kg cheese. Evaporation and spray drying are, with 96% of the total energy used for milk powder production, responsible for this high energy consumption, consequently large potential for energy savings lies in these processes (Ramirez et al., 2006; L. Wang, 2008; Westergaard, 2004).

Although much attention has been given to product quality and process improvement, current milk powder production uses the same operational type of units as 50 years ago. Only a few breakthrough changes with respect to energy efficiency have been made: the implementation of reverse osmosis, and vapor recompression for example. Innovation in the dairy industry was mainly product driven instead of energy driven (Xu & Flapper, 2011). We, therefore, believe with an eye on the 2050 goals instead of incremental, large step improvements (aiming for at least a twofold energy reduction) are required.

Furthermore, world's milk production is still growing. The established quota system for the European dairy sector resulted in a nearly constant milk production in Europe over the past decades. As a result the production of commodity products like milk powder declined, while the amounts of high value specialties like cheese increased (Kelly, 2006). Countries which were not restricted by the milk quota, amongst which non-traditional milk producing countries like Brazil, Argentina and India, increased the capacity and production of commodity products (FAO, n.d.; Xu & Flapper, 2011). The end of the milk quota in the EU in 2015 will lead to an increase of milk production, followed by an increase in commodity products like milk powder (Jansik, Irz, & Kuosmanen, 2014).

In order to reduce the energy consumption of the dairy industry, breakthrough solutions are necessary; emerging processing technologies and alternative energy supply systems need to be investigated for milk powder production. This review discusses the state of the art in milk powder production and the opportunities of emerging technologies that have the potential to reduce the energy consumption up to 60% in milk powder production.

2 Current technologies

Figure 1 gives an overview of the main steps for milk powder production, which are categorised as pre-treatment (standardisation, homogenisation, and pasteurisation), concentration, drying including air filtering, and energy supply.

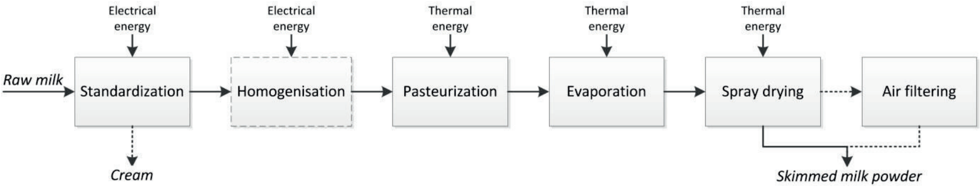

Figure 1. Overview of major unit operations in a skimmed milk powder production chain, from raw milk to skimmed milk powder.

2.1 Pre-treatment

2.1.1 Standardisation/ separation

Milk powder is produced with a specified ratio of fat to non-fat milk solids. This ratio is realised in during standardisation. Disc-stack bowl separators with high centrifugal forces are used to separate milk into cream and skimmed milk. After separation the skimmed milk and cream are mixed to the desired fat content. Both hot separation (50 – 52°C) and cold separation (4 – 20°C) are applied. With hot separation a higher separation efficiency is achieved, due to lower cream viscosity. The fat content of skimmed milk after hot separation is between 0.04% – 0.05%, while cold separation results in a fat content of 0.07% – 0.1%. Cold separation requires slightly more energy for separation due to the higher viscosity of milk at lower temperatures, but has the advantage of the absence of protein denaturation, saves energy as no heating is required, and microbial growth is inhibited, which improves the quality of the end product (GEA Westfalia, n.d.).

For hot separation a preheating step is required. In order to be energy efficient, hot separation can be integrated in the pasteurisation step. This integrated separation does not require preheating, and energy will be saved compared to a non-integrated hot separation system. The mechanical energy for the separation of milk varies from 0.04 to 0.11 MJ/kg milk powder (GEA Westfalia, n.d.).

2.1.2 Pasteurisation, sterilisation

Heat treatments are applied to reduce the number of bacteria in milk and dairy products. Pasteurisation, at 72°C for 15 – 30 s (Walstra, Geurts, Noomen, Jellema, & Boekel, 1999), is a continuous and widely used process. Pasteurisation equipment consists of three sections of plate heat exchangers: heating, regeneration, and cooling section, and a holding section where no heat is exchanged. Due to heat regeneration, 90% to 95% of the energy can be recovered, which makes pasteurisation a very energy efficient process. This results in an energy consumption of around 0.3 MJ/kg milk powder (Jong, 2013; Kessler, 1981; Ramirez et al., 2006).

Besides pasteurisation, sterilisation (10 – 45 min at 110 – 125°C) and UHT (0.4 – 4 s at 135°C or more) are also well-established heating treatments in the dairy industry (GEA, n.d.). These processes assure a longer shelf life, but for milk processed into powder, pasteurisation is sufficient.

2.1.3 Homogenisation

Homogenisation is used to reduce the size of the fat globules in milk. It results in an even distribution of fat in the milk, and thus also in whole milk powders. The optimal temperature for homogenisation is 60 – 70 °C. As this temperature is reached during pasteurisation and evaporation, homogenisation is usually integrated in one of these processes, consequently no extra heating is required. The applied pressure ranges between 150 and 1000 bar. The mechanical energy needed is 0.2 – 0.3 MJ/kg milk powder (Walstra et al., 1999). Skimmed milk contains only 0.1% fat, and homogenizing is not necessary for this product (U.S. Dairy Export Council, n.d.).

2.2 Concentration

2.2.1 Evaporation

Multiple-stage falling film evaporation is used to concentrate milk to a solid content of about 50%. Together with the solid content the viscosity of the product increases. The current atomiser equipment for spray drying is not able to process viscosities that belong to more concentrated solutions (Deventer, Houben, & Koldeweij, 2013). In the different evaporator stages steam surrounds a bundle of tubes with a flowing film of milk along the inner walls. Heat is transferred from the steam at one side of the tubes to the flowing milk film at the other side, and evaporates water from the milk. The vapor is separated from the concentrated milk and is used for evaporation at a lower pressure in the next stage. Using multiple-stage evaporators allows multiple reuse of vapor and saves large quantities of energy (Kessler, 1981; Ramirez et al., 2006). The more stages (up to 7 – 9) the larger the energy gain, but also the heat exchanging surface increases. Energy consumption of a 7 stage falling film evaporator is around 300 kJ/kg water removed (Pepper & Orchard, 1982; Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004). Milk is a heat sensitive product, therefore, denaturation of protein has to be minimised, which is possible with product temperatures in the first stage around 70°C and in the last stage between 40 to 50°C.

2.2.2 Vapor recompression

In addition to multiple-stage evaporation there are two options to increase the energy efficiency of the evaporator: 1) thermal vapor recompression and 2) mechanical vapor recompression. Thermal vapor recompression uses a steam jet booster to recompress vapor exiting one of the stages and results in increased temperature and pressure. In mechanical vapor recompression the vapor is compressed by a compressor driven by an electrical motor, gas engine or steam turbine. Electrical energy is the main energy input in mechanical vapor compression and is supported by a small amount of fresh steam to compensate for losses (GEA Wiegand Gmbh, n.d.). Vapor compression lowers the energy consumption of evaporation to 220 kJ/kg water removed for thermal vapor recompression (7-stage), and to 55kJ/kg water removed if mechanical vapor recompression is applied (European Commission, 2006). The choice for a compressing system depends on the energy price. Nowadays, the steam compressor is most applied, but the use of mechanical recompression is growing (Singh & Heldman, 2014).

2.2.3 Reverse osmosis

Osmosis is the diffusion of water from a low concentrated solution through a semipermeable membrane to a high concentrated solution. The flow of water depends on the osmotic pressure between the solutions. Reverse osmosis works in the opposite way. By applying a pressure above the osmotic pressure of the solution, water is forced from a high to a low concentrated solution. Protein molecules are fully rejected by the reverse osmosis membranes, lactose and salts have a high retention (Kessler, 1981). To avoid microbial growth the normal working temperature for milk concentration is 10°C, or in the range of 50 – 55°C which result in higher permeate fluxes (50 to 100% more compared to operating at 10°C). Disadvantage of operating at higher temperatures is the lower salt retention and required additional heating (Fellows, 2009; Membrane System Specialists Inc., n.d.).

Concentrating by reverse osmosis is interesting because of the low energy consumption compared to evaporation. Reverse osmosis is a pressure driven process instead of thermal to evaporate water. Evaporation requires 300 kJ/kg water removed, where the energy consumption for reverse osmosis is only 14 – 36 kJ/kg water removed (Pepper & Orchard, 1982; Ramirez et al., 2006). Concentration polarisation and the osmotic pressure of milk concentrate, however, limit the maximal total solid concentration for milk to 18 – 24%. Concentration of milk to 18% dry matter is commonly used as economical fluxes are guaranteed. For milk powder production, therefore, it is not sufficient to use only reverse osmosis for milk concentration, but it is used as a pre-concentration step before evaporation. Although pre-concentration of milk with reverse osmosis seems promising, it is not widely implemented for the concentration of milk (Poelarends, Slaghuis, & de Koning, 2009; Ramirez et al., 2006).

2.3 Drying

Spray drying is common practice in milk powder production. For example, in 2000 99.5% of all skimmed milk powder in Germany was produced via spray drying (Ramirez et al., 2006). Alternatives are fluidised bed drying, drum drying, vacuum and freeze drying. Although freeze drying result in high quality products, it is not suitable for bulk products. The long drying times, low processing capacities and high energy consumption, result in an expensive process (Barbosa-Cánovas, Ortega-Rivas, Juliano, & Yan, 2005; Evans, 2008). Drum drying has a better energy efficiency, making use of conductive heating, while spray drying uses convective heating. Drum drying, however, gives the powder a cooked caramel like flavour and some browning as a result of the Maillard reactions which is generally avoided in milk powder production (Bhandari, Bansal, Zhang, & Schuck, 2013).

2.3.1 Spray drying and fluidised bed drying

To minimise thermal damage of the product, spray drying is used because of the low product temperatures and short residence times compared to conductive heating processes. The process consists of three stages, product atomisation, moisture evaporation and separation of particles from the exhaust air.

Pre-concentrated milk is atomised at the top of the drying tower. Pressure nozzles and rotary wheel atomisers are most used due to their relative simplicity (Mujumdar, 2007). The wheel atomiser spins milk droplets away and creates a fine mist of droplets. Due to the tangential direction of the spray, wheel atomisation requires drying towers with a large diameter in order to prevent contact of the droplets with the drying tower walls. Pressure nozzles create droplets by forcing the milk through a small orifice. These nozzles have a relative small capacity and therefore multiple nozzles are used in large spray dryers (Walstra et al., 1999). The droplet direction is downward and compared to wheel atomisation, pressure nozzle dryers can have a smaller diameter but need a taller body.

Hot air (around 180 – 230°C) enters the drying tower in co–, counter–, or mixed current mode. The atomised droplets contact the hot air while exchanging heat and water. Dried powder leaves the tower at the bottom and depending on the design; the cooled and humidified air leaves the tower in the cone or at the top. The water content of the powder that leaves the drying tower is in the range 3 – 7% (Fellows, 2009; Kessler, 1981). Spray drying requires around 4.5 MJ/kg milk powder (Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004).

Spray drying is often combined with a second or even third drying stage to make the system more energy efficient, to give extra properties to the product, or for product cooling. Additional drying options are 1) an external fluidised bed, 2) an internal fluidised bed at the bottom of the drying tower, or 3) a filtermat dryer (Brennan, 1997; Schuck, 2002). In a multi-stage drying system the powder leaving the first drying stage has a water content in the range of 5 – 9%, and is dried in the second stage to the final water content. The air used in the second stage has a lower temperature, and to remove the last amounts of water a low air flow can be applied.

Energy requirements for a multi stage drying system is lowered to around 3.9 MJ/kg milk powder (Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004).

The size of powder particles depends on type of atomisation and drying conditions. The particle size range for milk powder obtained with rotating wheels is 1 – 600 μm, where pressure nozzles have of particle size distribution between 10 and 800 μm (Filková, Huang, & Mujumdar, 2006). Disadvantage of spray drying is the loss of latent heat in the exhaust air. Heat recovery is problematic due to fine powder particles (fines) entrained in the exhaust air flow, which will deposit on the heat exchanger surface (Atkins, Walmsley, & Neale, 2011).

2.3.2 Air filtering

For economical, safety, and emission reasons fines have to be removed from the exhaust air. As 10 – 20% of the powder leaves the tower as fines via the exhaust air, a significant amount of product and value loss is avoided by powder recovery (Gabites, Abrahamson, & Winchester, 2007; Westergaard, 2004).

The dairy industry uses two systems for fines recovery, i.e. cyclones and filter bags. Centripetal accelerations created in cyclones separate the fines from the air. Every type of cyclone has a specific range for the particles to be recovered and therefore an second cyclone or a filter bag is used after the first cyclone to remove the smallest particles (Kessler, 1981). Filter bags have a higher efficiency compared to a cyclone, but the drawback is the risk for microbial contamination due the long residence time in the filters. With the improvement of cleaning properties, however, filter bags are more often used (Gabites, Abrahamson, & Winchester, 2008).

The fines recovered by the cyclones can directly be added to the powder from the dryer, or returned to the spray dryer depending on the product quality requirements. Returned fines contact the droplets in the top of the tower and form agglomerates, which improve the solubility characteristics and flowability of the final powder (GEA Niro, n.d.-a; Kessler, 1981).

2.4 Energy system

The systems for steam generation, air heating and water supply are important units in a milk powder production plant. Moreover, any production facility needs electricity for overhead purposes as lighting, cooling/refrigerating, air conditioning and process equipment like pumps, fans and compressors etc. Electrical energy is the most versatile and flexible energy source. Large plants have electrical power generators which may be used as back-up when the normal supply from the grid is interrupted.

2.4.1 Boiler house

Evaporation and pasteurisation require steam for heating, furthermore steam is used to generate hot water for cleaning. The steam for the different unit operations is generated in a boiler house located at the production site. Two types of boiler houses are mainly applied: 1) fire-tube, and 2) water-tube boilers (Singh & Heldman, 2014). In a fire-tube boiler water is heated in a vessel by hot gasses which are circulated in tubes. Water-tube boilers on the other hand circulate water in tubes through the furnace, where hot gasses surround these tubes. These have larger capacities, and can be easily adjusted to the steam demand, therefore, more used in modern facilities. The phase change takes place within the tube, and is therefore considered safer compared to fire-tube boilers where this happens in a vessel. Fire-tube boilers have the advantage of easier accessible fires sides, resulting in lower operating costs. Efficiencies vary between 74 and 84% (Baker, 2013). In milk powder production only indirect heating is applied (except for cleaning); that means the condensate can be brought back to the boiler, and be reused in a closed-loop system, resulting in very low water loss (Walker, Lv, & Masanet, 2013).

2.4.2 Air heating

Hot air for spray drying is obtained by indirect heating from steam or fuel combustion. Steam heated air heaters have an efficiency of 98 – 99%, but should be corrected with the, above mentioned, efficiency of steam generation in the boiler house (GEA Niro, n.d.-b; Mujumdar, 2007). Maximum air temperature depends on the temperature of the steam (150 – 250°C). Fuel heated air heaters have an efficiency around 80%, similar to fuel heated boilers, and will reach air temperatures up to 400°C (GEA Niro, n.d.-b; Mujumdar, 2007). Natural gas, coal and petroleum products are the standard energy sources used for air heating. Natural gas has a relative clean combustion and large availability in Europe.

2.5 Current limitations and opportunities

In order to save energy in milk powder consumption we see three opportunities: 1) replacement of the traditional evaporator for concentration by membrane processes, 2) heat recovery from the dryer by closed loop drying, and 3) the introduction of alternative heating systems.

Membrane processes reduce the energy consumption for the concentration of liquids compared to conventional concentration by evaporators. The product concentration that can be achieved by pressure driven membrane process is limited by fouling and concentration polarisation (Koltuniewicz & Noworyta, 1994; Pepper & Orchard, 1982), therefore thermal driven membrane processes which are able to concentrate to higher solid contents, and exploit waste heat from other process units, may provide a solution.

The exhaust air from the spray dryer has a temperature in the range 60 – 95°C, and is saturated with water vapor (Walstra et al., 1999). Although several authors have proposed energy recovery from the exhaust air by heat integration and recirculation (Atkins et al., 2011; Golman & Julklang, 2014; Walmsley, Walmsley, Atkins, & Neale, 2013), heat recovery from the exhaust air still containing fines is not yet effective due to the energy losses in the filtering systems and fouling in the heat exchangers. Besides, fines deposition on the heat exchanger surface possibly generates caramel like off-flavours to the powder. Atomisation with no, or a minimum amount of, fines offers new possibilities for heat recovery and has the potential to make spray drying energy efficient.

Besides the reduction of energy consumption in milk powder production, renewable energy has potential to improve the energy consumption and related CO_2-exhaust. Steam and hot water supply could, for example, be generated with solar heat (Schweiger et al., 2000). At the moment electrical driven heating using electricity is not favourable, because of the low efficiency of electricity generation compared to boilers running on gas or oil. The use of photovoltaic cells, however, could cause a shift.

3 Emerging technologies

3.1 Pre-treatment

3.1.1 Radio frequency heating

Radio frequency heating is a direct heating system, generating heat within the product by using electromagnetic waves. Radio frequency uses frequencies in the range 1-200 MHz, which is below the frequency range used in microwave heating (from 300 MHz to 300 GHz). Radio frequency heating has therefore a deeper penetration in products compared to microwave heating (Awuah, Ramaswamy, Economides, & Mallikarjunan, 2005). The main potential of radio frequency heating in the food industry is to improve the quality of the product and to avoid fouling at heat transferring surfaces. It has currently industrial implementations for baking, blanching, drying, thawing and meat processing. (Fellows, 2009; Piyasena, Dussault, Koutchma, Ramaswamy, & Awuah, 2003)

The driving force in radio frequency heating is the transmission of electromagnetic waves, and not a temperature gradient as in thermal heat transfer. The heating is instantaneous; reported values are around 2°C/s (Awuah et al., 2005), but in industrial installations faster heating can be realised. Heating is uniform through the entire product mass and no gradients have been observed in heating of laminar flows. The absence of temperature gradients allows sterilisation at temperatures 15 – 20°C below the commercial standard sterilisation process. This can be extremely beneficial for the sensorial properties of liquids like milk, and to reduce fouling that occurs due to overheating of the product that contacts heat exchanging surfaces (Kudra, Voort, Raghavan, & Ramaswamy, 1991). As radio frequency heating in industrial installations is potentially 20 – 30 times faster compared to conventional heat exchangers, pasteurisation becomes more flexible and can be performed in equipment with smaller dimensions.

Contactless heating by radio frequency has considerably less fouling (residual deposits) compared to heating with heat exchangers. As a result, a lower number of cleaning cycles is required, and the capacity of the installation can be increased. Although radio frequency heaters can be integrated in existing plants the uptake by industry has not been large yet. Marra et al. explained the lack of uptake by industry on the higher need of understanding on the effectiveness of microorganism inactivation, as well as the needs in designing and up-scaling for industrial application (Marra, Zhang, & Lyng, 2009).

Radio frequency heating uses electricity as energy source, and eliminates steam consumption. This can, however, be considered as a disadvantage. The efficiency ratio of electricity and heat generation to primary energy use is 2.4:1 (Grubler et al., 2012). The energy needed for heating with radio frequency to a given temperature is comparable to that of heat exchangers. To heat the same amount of milk to the same process temperature with radio frequency heating requests therefore 2.4 times more primary energy compared to steam or hot water heated systems. Manufacturers of radio frequency heating systems suggest that with their systems lower operational temperatures can be applied and that the system has lower heat loss due to the fast heating. As a consequence, the amount of primary energy can be corrected downward. These benefits are, however, not quantified in literature. Local situations (the availability of hydropower, or electricity from solar cells) can give preference to radio frequency heating. In addition, because of the lower degree of fouling, energy can potentially be saved by a lower number and shorter cleaning cycles.

3.2 Concentration

3.2.1 Membrane distillation

Membrane distillation is an emerging technology to concentrate solutions. First reports date back to the 60's for desalination purposes, but it lasted till the late 80's before interest was picked up again due to the availability of membranes which increased fluxes (El-Bourawi, Ding, Ma, & Khayet, 2006). Although industrial applications are still limited, R&D interest is growing as well as the number of publications (El-Bourawi et al., 2006; Lawson & Lloyd, 1997). Interest for the use of membrane distillation in food application increased in the past few years. Results have been published for the concentration of fruit juices and dairy products (Alves & Coelhoso, 2006; Bagger-Jørgensen, Meyer, Varming, & Jonsson, 2004; Calabro, Jiao, & Drioli, 1994; Hausmann et al., 2011; Laganà, Barbieri, & Drioli, 2000; Nene, Kaur, Sumod, Joshi, & Raghavarao, 2002).

In contrast to membrane technologies like reverse osmosis, ultra- and micro filtration, membrane distillation is driven by a vapor pressure difference over a hydrophobic membrane. The different configurations are direct contact, air gap, sweep gas, and vacuum membrane distillation. Sweep gas and vacuum membrane distillation are often used for volatile organic compounds removal, where direct contact and air-gap membrane distillation are more common for water removal (P. Wang & Chung, 2015). In direct contact membrane distillation water vapor moves through the membrane due the vapor pressure difference between the hot feed and the cold permeate side (stripping water), which are separated by a hydrophobic membrane (see Figure 2a). Different from the direct contact system, in the air-gap membrane distillation a small air gap separates the membrane from the cold stream; the water vapor condenses on the cold side (see Figure 2b). Advantage of the air-gap membrane distillation is the possibility for internal heat integration, making this configuration more energy efficient compared to direct contact membrane distillation (Hausmann, Sanciolo, Vasiljevic, Weeks, & Duke, 2012; P. Wang & Chung, 2015). Downside is the lower fluxes due to increased vapor transport resistance. Fluxes up to 75 L/m2.h for direct contact membrane distillation (Schofield, Fane, Fell, & Macoun, 1990), and ranging from 1 to 4.5 L/m2.h for air-gap membrane distillation are reported (Duong, Duke, Gray, Cooper, & Nghiem, 2016; Kuipers et al., 2014).

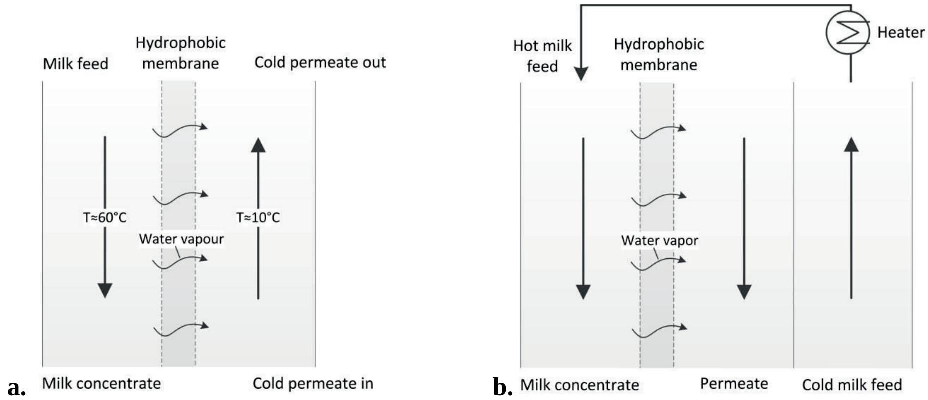

Figure 2. Schematic representation of two membrane distillation configurations. Direct contact membrane distillation (a), due to vapor pressure differences between hot and cold side, water evaporates from warm milk feed and passes a hydrophobic membrane to the colder permeate stream. In air gap membrane distillation (b), the water vapor condenses on the cold surface between the air gap and the cold milk feed. Due to the recycling of the milk feed, internal heat recovery is possible.

Concentration polarisation, which occurs in pressure driven membrane filtration, is not limiting and therefore high solid contents can be realised with membrane distillation (Tijing, Choi, Lee, Kim, & Shon, 2014). Publications on milk concentration by membrane distillation are limited. Hausmann et al. already showed the possibilities for the concentration of skimmed milk to a solid concentration of 40 – 43.5% (Hausmann, Sanciolo, Vasiljevic, Kulozik, & Duke, 2014; Hausmann et al., 2011) and recently it was found that concentration to 50% total solids is possible (Moejes et al., 2015). These solid contents allow the replacement of the evaporation process by membrane distillation. Like other membrane processes, membrane distillation is prone to fouling which result in a flux reduction over time, and at higher solid concentrations (Hausmann et al., 2013).

Main advantage of membrane distillation is the mild operational conditions. Product feed temperatures around 60°C are favourable to diminish protein denaturation and still give a good permeate flux. Membrane distillation is, therefore, able to use low grade heat obtained from waste heat of other processes or solar heat (Hanemaaijer et al., 2006). Dow et al. showed how integration of a direct contact membrane module in a power station for water production could run on the available waste heat (40°C), with an average energy consumption of 1500 kWh/m^3 (5.4 MJ/kg water removal) (Dow et al., 2016). Several pilot modules with internal heat recovery (air-gap membrane distillation) have been tested for desalination, resulting in energy consumptions ranging from 100 to 350 kWh/m^3 (0.4 – 1.3 MJ/kg water removal) (Duong et al., 2016; Guillén-Gosálbez, Caballero, & Jiménez, 2008; Koschikowski et al., 2009; Kuipers et al., 2014; Mar Camacho et al., 2013). Depending on the availability of waste heat and the desired fluxes either direct contact or air-gap membrane distillation could be favourable.

Although concentration of milk by membrane distillation is proven possible, further research is required, whereby the selection of membranes, which guarantee a high flux and minimal fouling formation, and optimisation of process configurations will be important for effective industrial performance.

3.3 Drying

3.3.1 Monodisperse drying

Patel et al. reviewed US patents to identify recent developments in spray drying of food, flavour and pharmaceutical applications (Patel, Patel, & Chakraborty, 2014). They showed that recent developments in these application fields have been focusing on microencapsulation and other product properties rather than energy saving purposes. An innovation with energy saving potential is the monodisperse-droplet atomiser, a concept which is already mentioned in the 90's for ink-jet printing (Brenn, Helpiö, & Durst, 1997). Compared to conventional systems, monodisperse-droplet atomisation yields droplets with identical shape and size (Deventer et al., 2013), resulting in powder particles with the same size, density, porosity, nutrient and moisture content. Identical droplets require the same drying time, and therefore no energy is lost due to overheating of smaller droplets. Enabling to control the right drying time for every droplet makes this technology interesting for thermosensitive products like milk. Uniform droplet drying can, furthermore, reduce the drying time with 50% under severely moist drying conditions, which results in either a smaller dryer and/or an equivalent increase in production capacity (Kosmodem'yanskii, Fokin, & Planovskii, 1968).

Monodisperse droplets are generated by gravitational and inertial forces based on the Rayleigh breakup of liquid streams. Additional electrical or mechanical forces in the nozzle speed-up the droplet breakup, and allows droplet size regulation. Most promising results are achieved by nozzles with piezo-electric materials transducers, where the vibrating force influences the droplet breakup (Wu, Patel, Rogers, & Chen, 2007). In recent years several results are published on monodisperse-droplet atomisers in spray drying systems (Deventer et al., 2013; Fu et al., 2011; Liu, Duo Wu, Selomulya, & Chen, 2012; Rogers, Fang, Qi Lin, Selomulya, & Dong Chen, 2012).

Figure 3 shows the difference in particle size rage between conventional and the monodisperse-droplet atomiser, before and after drying. Due to the absence of fines there is no powder in the exhaust air and the latent heat in the exhaust air can be recovered via air dehumidification. The dehumidified exhaust air can be recirculated, saving energy which is further discussed in section 4. The absence of fines also implies an increased product yield, as no powder is lost via the exhaust air through the filters.

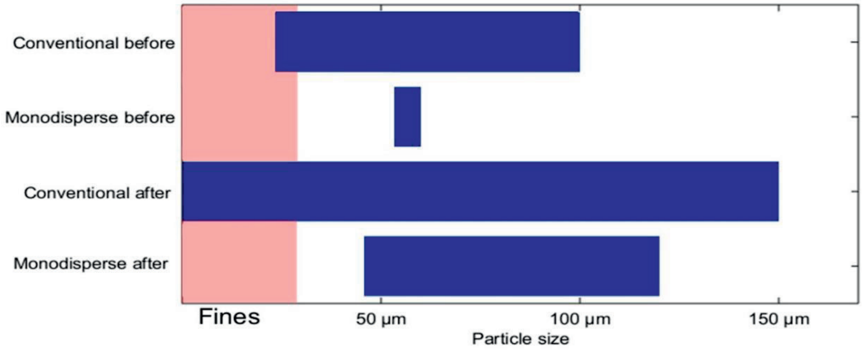

Figure 3. Comparison of particle size range between a conventional atomiser system and monodisperse-droplet atomiser. Both show particle size range direct after atomisation, and after drying. Drying enlarges the size distribution after monodisperse droplet generation, but shows to be out of the range of fines (highlighted area in the figure).

Monodisperse-droplet atomisers also have the potential to process fluids with high viscosities and enable to atomise milk concentrates with total solids contents in the range 50 – 60%. As concentration methods like evaporation and membrane distillation are more energy efficient compared to drying, a further reduction of energy consumption can be realised.

Feed flows between 100 and 250 L/h for monodisperse-droplet atomisers are mentioned in literature (Brenn et al., 1997; Deventer et al., 2013; FMP Technology Gmbh, 2011) which fit to the requirements for specialty products (for example infant formulas, flavourings, and microencapsulation's in the food and pharmaceutical industry). For capacities applied in the production of commodity products, like milk powder, multi-nozzle systems are required.

3.3.2 Air dehumidification

Applying a monodisperse-droplet atomiser results in a minimal number of fines, and fines are no longer a restriction in reusing the exhaust air. The exhaust air from the drying tower has a temperature between 60 – 95°C, but contains too much water vapor to be reused. Consequently, the air has to be dehumidified. For air dehumidification two systems are suitable: 1) a membrane contactor using liquid adsorbents (see next section), and 2) a contact sorption system with adsorbents like zeolite or silica gel. The adsorbents remove water from the dryer exhaust air while releasing heat of condensation/adsorption. The released heat benefits the operation of other units in the system. During dehumidification, the adsorbent is gradually saturated with water and needs to be regenerated. The energy for regeneration increases the energy consumption, nevertheless up to 50% energy recovery can be realised by exploiting surplus/waste heat from the regeneration system (Boxtel, Boon, Deventer, & Bussmann, 2012). The proposed closed-loop drying system is depicted in Figure 4.

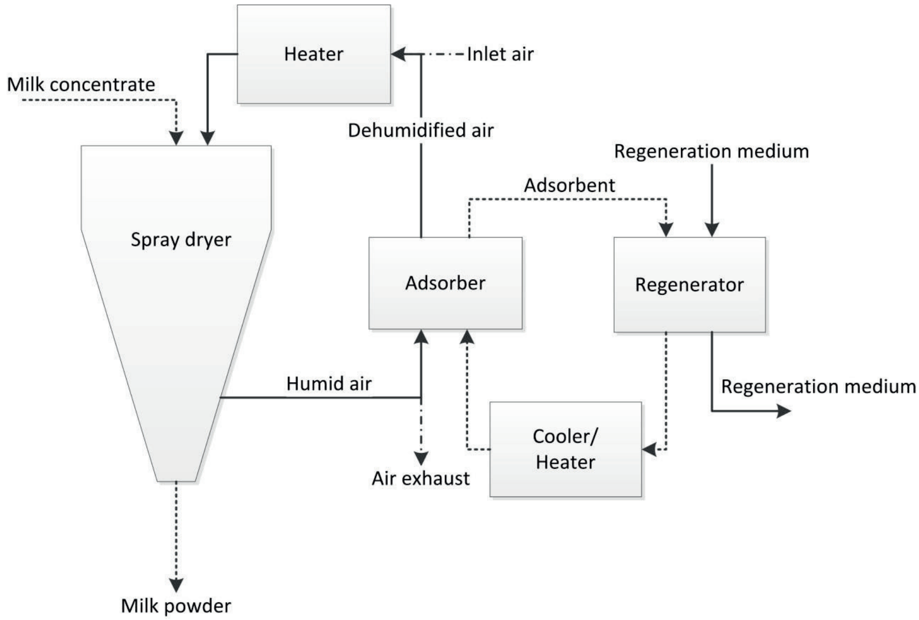

Figure 4. Proposed configuration of a closed-loop one-stage spray dryer with air dehumidification, including cooling or heating and regeneration of the adsorbent. The energy content of the exhausted regeneration medium can be used for heat integration with other processes like membrane distillation. In order to prevent accumulation of gasses in the system some air can be added to and exhausted from the air loop.

Membrane contactor

Membrane contactors are industrially used for degassing, for example the separation of CO_2 from gas streams (Li & Chen, 2005). More recently membrane contactors have become of interest for air dehumidification due to the energy efficiency, simple equipment without rotating parts, no direct contact between air and desiccant, and continuously operation mode (Yang, Yuan, Gao, & Guo, 2013). Most research is focused on dehumidification in air conditioning systems at ambient air temperatures, showing energy efficiency improvements up to 60% compared to conventional air conditioning systems (Bergero & Chiari, 2001; Isetti, Nannei, & Magrini, 1997; Jain, Tripathi, & Das, 2011; Kneifel et al., 2006). Despite published results as a proven technology for flue gas treatment (Li & Chen, 2005), air dehumidification with a membrane contactor at air temperatures in the range of 60 – 95°C is still limited.

The principle of the membrane contactor is the same as for membrane distillation; water vapor from the air passes a hydrophobic membrane and is adsorbed by a desiccant solution. The water vapor partial pressure difference between the moist air on one side of the membrane and desiccant solution on the other side is the driving force. Most used desiccants are lithium chloride (LiCl), lithium bromide (LiBr), magnesium chloride ($MgCl_2$), calcium chloride ($CaCl_2$) and triethylene glycol (TEG), or a combination of these (Abdel-Salam, Ge, & Simonson, 2013).

In the membrane contactor latent heat from the vapor in air is converted to sensible heat (raised temperature) in the desiccant solution, which can internally be recovered by a heat exchanger, generating hot water. Regeneration of the desiccant solution can be realised by using steam in an evaporator or hot air in an additional membrane contactor module. Energy recovery from the vapor of the regenerating evaporator is essential for the feasibility of the membrane contactor in the perspective of energy efficiency. Further research should focus on this as no literature is yet available.

Contact sorption system
Contact sorption systems make use of solid adsorbents with a high affinity for water, like zeolite or silica gel (Srivastava & Eames, 1998). The advantage of zeolite compared to silica is the ability for dehumidification and regeneration at elevated temperatures (Boxtel et al., 2012). Silica performs better at lower temperatures. The exhaust air of a spray dryer has temperatures in the range of 60 – 95°C, and therefore zeolite is preferred. Although additional energy is needed for regeneration, an energy reduction for regeneration with hot air of about 30 – 50 % is realised with zeolites (Boxtel et al., 2012; Djaeni, Bartels, Sanders, Straten, & Boxtel, 2007). The exhaust air from the regenerator has a temperature around 150°C, and allows heat application for other processes in the production chain. To increase the water loading capacity, the zeolite has to be cooled after regeneration. The energy obtained from cooling can be used to preheat ambient air that is used as regeneration medium.

Regeneration with superheated steam is an alternative for hot air regeneration. The advantage of this system is that the steam after regeneration has more options to be used at the production site. According to Bussmann et al. (Patent No. US 20060010713 A1, 2006) the energy costs for a dryer system with zeolite and a regeneration system with superheated steam, can be reduced up to 70% compared to a conventional dryer. Van Boxtel et al. (2012) showed that spray drying with air dehumidification by zeolite requires 96 kJ/kg air of which 46 kJ/kg air is recovered as steam, resulting in a 50% reduction compared to a conventional system requiring 92 kJ/kg air. Note that the humidity of the drying air will affect drying behaviour and consequently the product quality. Not all products will benefit from dry-air use in the spray dryer. For these products the dehumidified air can be mixed with a bypass of the non-dehumidified or ambient air.

3.4 Energy system / Renewable energy

3.4.1 Solar thermal system

Thermal energy takes the major part of the energy in milk powder production. Solar thermal systems collect solar radiation and transfer it into thermal energy. By making use of plates, mirrors or lenses solar energy is collected and stored as hot water or thermal oil with temperatures in the range 60 – 400°C (Kalogirou, 2004). These systems can be applied for domestic use, indoor heating and industrial thermal processes. Although there is a large potential for solar thermal systems, nowadays these systems are in an introduction phase and not yet used to a large extend. In 2010 the share of solar thermal systems for heat supply was in Germany only 0.4%, while there is a potential of 3.4% (Lauterbach, Schmitt, Jordan, & Vajen, 2012). A solar thermal system does not reduce the energy consumption in the production plant, but increases the contribution of renewable energy, and thus reduces the use of fossil energy carriers and the emission of greenhouse gasses.

Solar thermal collectors are classified in systems for direct heating of water and solar concentrators (H. Schweiger et al., 1999). Direct heating systems (flat plate and evacuated tube collectors) collect direct and diffuse/indirect solar radiation (Vannoni, Battisti, & Drigo, 2008). Solar concentration systems (Fresnel collectors, parabolic through collectors and mobile absorber collectors) on the other hand concentrate solar radiation and absorb only direct solar radiation (Vannoni et al., 2008).

Flat plate collectors are most used due to their efficiency and relatively simple construction. Main usage is for hot water generation and building heating, and up to a temperature of 100°C good efficiencies are reached (Kalogirou, 2003). The evacuated tube systems have a larger temperature range, and superheated water with temperatures up to 120ºC can be reached. The efficiencies are higher at low incidence angles, which give them an advantage over flat plate collectors especially in colder climates. Solar energy concentration systems always work with thermal fluids. The temperatures in the concentrator system can reach up to 400ºC. These systems, however, do not function in the absence of direct light (Kalogirou, 2004).

Radiation and user consumption profiles have a large influence on the effectiveness of solar systems. Daily variations in hot water uptake, periods of low processing capacities, or high consumption in periods with low radiation values will affect the design of the system. To manage variations in solar radiation and energy request, solar thermal systems need to be combined with auxiliary energy sources and an energy storage system (Tian & Zhao, 2013). There is potential for solar thermal systems, and the dairy industry is a good candidate due to the low to medium working temperatures (Lauterbach et al., 2012). The viability of solar heating systems depends largely on the local situation which is related to energy prices, weather conditions, consumption profiles, available space, and industrial acceptance. A guaranteed price of 0.05€/kWh for industrial use would make solar energy competitive to fossil energy in the current market. Pressure from policy makers to cut greenhouse gas emissions, and reduce the use fossil of energy sources is of huge importance as motivation for industrial uptake.

4 Potential for energy savings

In this review we presented emerging technologies to reduce the energy consumption of the production chain for heat sensitive dried products like milk powder. Energy savings can be realised with the implementation of the different proposed innovative technologies. Membrane distillation allows concentration at low temperatures and the usage of low-grade heat, resulting in a potential reduction of the energy consumption. Monodisperse drying in combination with air dehumidification and adsorbent regeneration enables the recovery of latent heat from the exhaust air, and closed-loop drying. Combination of the different emerging technologies results in the different possible production scenarios, given in Table 1.

Table 1. Overview of different combinations of process units in different scenarios for skimmed milk powder production.

Scenario	Process units involved
Benchmark	Standardisation – pasteurisation – preheating – evaporation – spray drying
Scenario 1	Standardisation – pasteurisation – reverse osmosis – preheating – evaporation – 2 stage spray drying
Scenario 2	Standardisation – pasteurisation – reverse osmosis – membrane distillation – 2 stage spray drying
Scenario 3	Standardisation – pasteurisation – reverse osmosis – membrane distillation (to 55% total solids) – 2 stage spray drying
Scenario 4	Standardisation – pasteurisation – reverse osmosis – membrane distillation – monodisperse drying with membrane contactor
Scenario 5	Standardisation – pasteurisation – reverse osmosis – membrane distillation – monodisperse drying with zeolite

Energy requirements for each process were estimated by using literature data and are summarised in Table 2. As stated before, membrane distillation with an air-gap configuration (with internal heat recovery) is more energy efficient compared to direct contact. For the concentration of milk, however, there is at this moment no data available of air-gap membrane distillation. The estimated energy consumption needs to be validated for milk, as the thermal heat transfer, and consequently the flux will be negatively influenced by the higher solid content of milk, as already shown for direct contact membrane distillation (Hausmann et al., 2014). The used energy requirements are based on air-gap membrane distillation for desalination, and are reported in a wide range; therefore, a low and a high value is included. The low value is based on an energy requirement of 0.4 MJ/kg water removed (Koschikowski et al., 2009). The high value is based on the 1.3 MJ/kg water removed as reported by Mar Camacho et al. (2013).

The energy requirements for monodisperse-droplet drying with air dehumidification by zeolite are based on a closed-loop spray drying system including regeneration by superheated steam. The adsorption-regeneration cycle with superheated steam results in excess steam of 150°C, which results in a 50% energy recovery (expressed as surplus) (van Boxtel et al. (2012)). For air dehumidification with a membrane contactor no data was available in terms of energy efficiency. An estimation was, therefore, based on mass and energy balances with the following settings: spray drying with air of 200°C, the membrane contactor works with a desiccant solution of 60% lithium bromide at 80°C with an internal heat exchanger in order to reuse the latent heat, and regeneration with a two-stage evaporator. This results in a hot water flow of 80°C containing the surplus heat. As the energy consumption is based on estimations there is uncertainty in these predictions, and further research is required to confirm these numbers. For drying systems with air dehumidification advantages of mono-dispersity in terms of drying rates were not taken into account.

Table 2. Average energy consumption of different processes in skimmed milk powder (SMP) production scenarios.

	Process	Energy consumption [MJ/kg SMP]	Process conditions	Sources
Pre-treatment	Pasteurisation	0.3	With 95% efficiency	(Jong, 2013; Kessler, 1981; Ramírez et al., 2006;)
	Standardisation	0.1		(GEA Westfalia, n.d.; Jong, 2013)
Concentration	Pre heating	2.7	9% total solids in skimmed milk, from 10 to 70°C, with c_p of 3.8 kJ/kg·°C	(Kessler, 1981; Pepper & Orchard, 1982; Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004)
		1.3	18% total solids after pre concentration, from 10 to 70°C	
	Evaporation	2.6	7 stage falling film, 9% to 50% total solids, 0.3 MJ/kg water removed	(Fellows, 2009; Kessler, 1981; Pepper & Orchard, 1982; Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004)
		1.0	7 stage falling film, 18% to 50% total solids, 0.3 MJ/kg water removed	
	RO	0.2	From 9% to 18% total solids. 36 kJ/kg water removed	(Fellows, 2009; Pepper & Orchard, 1982; SPX, 2012)
	Membrane distillation	3.2 – 11.1	From 9% to 50% total solids, air-gap configuration with energy range 0.4 – 1.3 MJ/kg water removed	(Guillén-Burrieza, Zaragoza, Miralles-Cuevas, & Blanco, 2012; Koschikowski et al., 2009; Mar Camacho et al., 2013)
		1.2 – 4.3	From 18% to 50% total solids	
		1.3 – 4.5	From 18% to 55% total solids	
Drying	Spray drying	4.5	Drying from 50 to 3.5% total solids	(Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004)
	2 stage spray drying	3.9	Drying from 50 to 3.5% total solids	(Ramirez et al., 2006; Walstra et al., 1999; Westergaard, 2004)
		3.6	Drying from 55 to 3.5% total solids	
	Monodisperse drying with membrane contactor	4.5 -1.6 (surplus)	Desiccant regeneration by 2 stage evaporation Hot water at 80°C	(Drioli, Criscuoli, & Curcio, 2006; Fellows, 2009; Gandhidasan, 2004; Patil, Tripathi, Pathak, & Katti, 1990)
	Monodisperse drying with zeolite	4.7 -2.4 (surplus)	Spray dryer with 50% recovery Superheated steam of 150°C	(Boxtel et al., 2012; Patent No. US 20060010713 A1, 2006)

The total energy consumption of the different scenarios, based on data from Table 2, is depicted in Figure 5. The benchmark, according to current practice, identifies concentration and drying as the energy hot spots in milk powder production. Pre-concentration with reverse osmosis results in a lower amount of product to be preheated and a lower energy use for evaporation (scenario 1). Compared to the benchmark the total energy consumption is reduced by about 30%. Reverse osmosis is, however, limited to a maximal concentration of around 18% total solids; therefore, membrane distillation could replace the additional evaporation step to concentrate to the final 50% total solids (scenario 2). Scenario 2 is given for the high and low level of energy consumption reported for membrane distillation (2H and 2L respectively). Replacement results in a total energy reduction of around 40% compared to the benchmark if the low-level energy consumption is realised. Scenario 2H, however, shows that using the high range value results in energy consumption 20% higher compared to scenario 1 (using reverse osmosis and evaporation). Advantage is still that waste heat could be used in the membrane distillation process which would reduce the external energy consumption. The concentration step is more energy efficient for water removal compared to the drying step. If spray drying of higher solid concentrations is possible, milk can be concentrated over 50%. Raising the milk concentration with 5% prior to drying will result in an energy reduction of 0.2 MJ/kg milk powder (scenario 3).

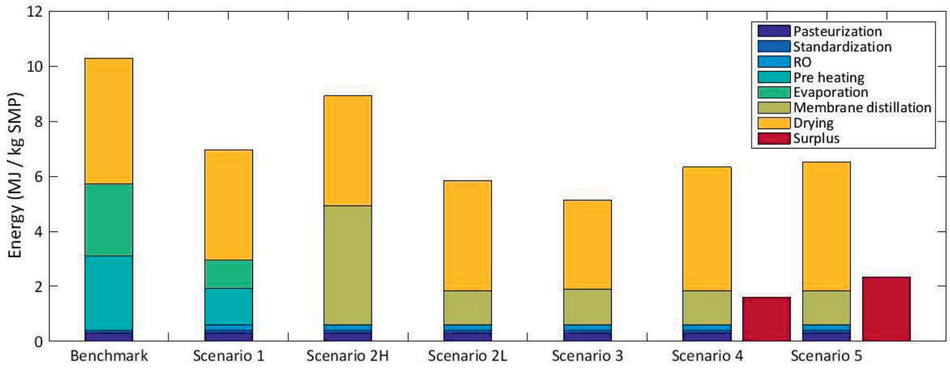

Figure 5. Energy use for the production of skimmed milk powder by different scenarios as listed in Table 1, single values are reported in Table 2. Scenario 2H includes to the high value for energy use of membrane distillation, where in scenario 2L the lower value was used. In scenario 3, 4, and 5 the low value was used. Regeneration of the dehumidification system in scenario 4 and 5 results in a surplus heat. The surplus heat can be used in other units in the milk powder production system, for example membrane distillation.

Monodisperse-droplet drying sustains the energy of the exhaust air after dehumidification in the drying system. However, the required energy for regeneration of the adsorbent on the dehumidification system cancels the benefit as is shown by scenario 4 and 5. The total energy consumption is higher than that of scenarios 2L and 3. In scenario 4 and 5, however, a surplus of useable energy is obtained from the regeneration systems. The energy gain in the closed-loop drying systems is therefore achieved by exploiting the surplus heat from the regeneration medium at other places in the production chain. This heat could, for example, be used in the membrane distillation unit. Usage of all surplus heat results in an overall energy reduction of almost 60%. Difference between the quality of the surplus heat is: scenario 4 generates hot water of 80°C, and scenario 5 generates steam of 150°C.

Presented numbers were based on literature data for the specific energy consumption, and mass and energy balances, of unit operations. The system is not yet optimised with respect to the operational conditions of the different units. It is expected that by optimizing the operational conditions an extra step forward in energy efficiency will be made. Additional benefit is the better controllability of drying uniform droplets from the monodisperse atomiser system. It is expected that this will lead to an additional reduction of the energy consumption required for spray drying. Furthermore, alternative energy and heating systems (e.g. solar thermal systems and radio frequency heating), and energy recovery systems between the different processing stages can further improve the energy efficiency of the production process.

4.1.1 Feasibility

The different technologies presented in this review are in different phases of maturity. Where radio frequency heating is already fully developed and implemented (Marra et al., 2009), its role in energy savings is mainly dependent on the local electrical energy situation. Renewable resources to produce electricity make this technology interesting. Current applications in food processing indicate that radio frequency heating can be constructed according safety and hygiene criteria.

Membrane distillation (only direct contact) and monodisperse-droplet drying are tested for milk processing on laboratory and small pilot plant scale. Larger scale testing, and in air-gap membrane distillation configuration, is necessary to prove their potentials. Gaining insight in the energy consumption of air-gap membrane distillation for milk is, as well as performance at high milk concentrations, of great importance for the viability of this process. Membrane units are relatively easy to scale up as more modules can be linked. Membrane distillation units are comparable to the current membrane systems (e.g. reverse osmosis and ultra-filtration). As these systems are accepted by the dairy industry, membrane distillation can certainly comply the safety and hygiene criteria.

The current monodisperse-droplet atomiser on the other hand faces more challenges. A multi-nozzle system has to be designed, and optimal positioning in the drying tower will be crucial for the drying behaviour of the uniform droplets. If droplets contact each other agglomerates will be formed, and mono-dispersity is lost. The effect of air flows on the droplet trajectories should be such that accumulation of powder at or around the nozzle is minimised to avoid risk of fire. Moreover, the droplets should not be sticky when reaching the dryer walls. These criteria may require adaptation of the drying chamber and air inlet system.

For air recirculation over the dryer, the air must be dehumidified. Zeolite adsorption wheels are already implemented in the drying industry for pre-dehumidifying ambient air (Boxtel et al., 2012). These systems proved in industrial tests to be reliable over longer periods. Regeneration conditions in the range of 250 – 300°C minimise microbial growth and the zeolite coatings on honeycomb carrier proved to be stable. Membrane contactors exist in climate control systems in buildings for example. Implementation of membrane contactors at elevated temperatures, as in spray drying operations, is new. Although the concept is proven in climate control, further investigation is necessary for implementation in the closed-loop milk powder drying, as well as a safety assessment for the use in a food grade plant. The main construction challenge for these systems is to guarantee that the desiccant solutions cannot have direct contact with the air recirculating over the dryer.

As Figure 5 already shows, benefits of the closed-loop drying depend on well-designed heat integration. Heat integration is thus essential for the implementation of the emerging technologies as next step in energy reduction.

4.1.2 Other applications

Although this review focusses on milk powder, the proposed technologies are not limited to this application. Most production chains of spray dried products are exceedingly similar. Except for other dairy products like whey and protein concentrates, these technologies could be implemented for solvent removal in chemical, biotechnology, agro and pharmaceutical industries.

5 Conclusion

The potential of the emerging technologies radio frequency heating, membrane distillation, monodisperse-droplet drying, and air dehumidification by zeolites, and membrane contactor, are discussed in this paper. Different production scenarios for skimmed milk powder are proposed, and the energy requirements are assessed.

The current energy consumption for skimmed milk powder production is around 10 MJ/kg powder. Combining reverse osmosis with membrane distillation, monodisperse-droplet drying, and zeolites have the prospective to reduce the energy consumption in milk powder production to 4 – 5 MJ/kg milk powder. Other combinations are also possible, but result in higher energy consumption in the range of 5 – 7 MJ/kg powder. Prerequisites for reducing the energy consumption are the elimination of fines by monodisperse-droplet drying, the exploitation of surplus heat generated in the air dehumidification step, and adequate energy efficiency and flux performance of membrane distillation at high milk concentrations.

To meet the future need for a significant reduction in industrial energy consumption, as formulated in the EU Energy Road Map 2050, incremental energy savings by modifications to current processes do not satisfy. New, innovative technologies are required. Although the proposed technologies are in different stages of development, it is essential to implement and further develop these technologies to make the next important step in energy reduction in milk powder production.

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Chapter 3

Closed-loop spray drying solutions for energy efficient powder production

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Abstract

Drying is an energy intensive operation in processing. To comply with the upcoming regulations that arise from the EU goals for sustainable development, the energy consumption of drying processes should be reduced drastically. Emerging technologies are the key for the next step in energy efficiency improvement. A closed-loop spray drying system for milk powder production is simulated and optimized in this work. The proposed technologies are: monodisperse droplet drying, membrane contactor and a zeolite wheel. By applying air dehumidification and heat integration the latent and sensible heat are recovered from the exhaust air. The heat integration solutions were obtained by simultaneous optimisation of the operational conditions and the heat exchanger network based on pinch analysis. The energy consumption for milk concentration and spray drying has the potential to be lowered from 8.4 to 4.9 MJ kg milk powder.

Keywords: Spray drying, milk powder, air dehumidification, zeolite, membrane contactor, pinch analysis.

1 Introduction

Thermal processes are responsible for 29% of the total energy consumption in the food industry (Okos, Rao, Drecher, Rode, & Kozak, 1998). Spray drying systems are the main energy consumer in powder production. The energy efficiency of spray drying has been improved over the last decades by the introduction of multi-stage drying with fluidized bed dryers, air pre-treatment, heat pumps, and the optimisation of the processes and operational conditions to a full extent (Ramirez, Patel, & Blok, 2006; Walstra, Geurts, Noomen, Jellema, & Boekel, 1999; Westergaard, 2004). Furthermore heat recovery and integration of current spray drying processes, like the production of milk powder, has been studied (Atkins, Walmsley, & Neale, 2012; Walmsley, Walmsley, Atkins, Neale, & Tarighaleslami, 2015). However, to reach the energy ambitions of the EU to reduce the energy consumption with 27% in 2030, and even more in the following decades (EC, 2012), incremental improvements in energy efficiency, by additional optimisation, do not satisfy this requirement. Large steps forwards, which can be achieved by introducing emerging technologies, are needed (Moejes & van Boxtel, 2017). An important opportunity for spray drying is to recover energy from the dryer exhaust air. The exhaust air has a temperature between 60 to 90°C and contains significant amounts of latent and sensible heat. Heat recovery from the exhaust air is, however, still a challenge due to the fine powder particles (fines) present in the exhaust air, which cause fouling in the heat exchangers used for heat recovery. Filter systems are needed but result in additional energy loss. Monodisperse droplet atomizers combined with proper airflow patterns and well-designed drying chambers, have the potential to operate without these fines. Both Deventer et al. (2013) and Rogers et al. (2012) showed that a spray drying system that uses monodisperse droplet atomizers based on inkjet technology results in a very narrow particle size distribution after drying. Monodisperse droplet drying is now applied at pilot plant scale (Debrauwer, 2016). With successful upscaling this technology offers the possibility for recirculation and recovery of heat from the dryer exhaust air in industrial installations.

Heat recovery from the exhaust has been proposed by Atkins et al. (2011) for spray drying; usage of heat exchangers and proper heat integration lead to a reduction of the hot utility up to 21%. Golman & Julklang (2014) investigated recirculation of the exhaust air over the dryer, and showed that partial recirculation of the exhaust air resulted in a reduced energy consumption for air heating, and a total energy reduction up to 20% was achieved. To improve the effectivity of air recirculation, the high moisture content of the spray dryer exhaust air has to be decreased by air dehumidification. Air dehumidification also enables the recovery of latent heat, and the dryer is operated at a constant (low) humidity of the inlet air. Dehumidification of the recycled air flow in closed-loop spray drying is an alternative which has not been discussed yet, and will be assessed in this study. Two technologies are proposed for air dehumidification in combination with closed-loop drying i.e., 1) contact-sorption system with a solid adsorbent (Atuonwu, van Straten, van Deventer, & van Boxtel, 2012), and 2) membrane contactor with a liquid desiccant (brine) (Isetti, Nannei, & Magrini, 1997). Both systems are already proven in other fields, and have potential to be implemented in closed-loop spray drying.

Contact-sorption systems use solid adsorbents with a high affinity for water. Zeolite and silica are the most used adsorbents for these systems. Since the spray dryer exhaust air has a temperature in the range of 60 to 90°C, zeolites are expected to be more effective compared to silica (Boxtel, Boon, Deventer, & Bussmann, 2012). Application of zeolites for air dehumidification in low-temperature dryers has been discussed before, and shows a significant potential for energy savings (Djaeni, van Straten, Bartels, Sanders, & van Boxtel, 2009; Goldsworthy, Alessandrini, & White, 2015). Likewise, zeolites are used for the pre-treatment (dehumidification) of ambient air prior to drying. Advantage of this pre-treatment is the increase in dryer capacity and improved controllability of the dryer conditions (Boxtel et al., 2012).

Membrane contactors are currently used for selective separation of gasses (Li & Chen, 2005) and in air conditioning systems (Bergero & Chiari, 2010; Kneifel et al., 2006). In air conditioning systems moist air is separated by a hydrophobic membrane from a saturated brine (i.e. lithium bromide, lithium chloride, magnesium chloride, calcium chloride, or a combination) (Abdel-Salam, Ge, & Simonson, 2013). The partial vapor pressure difference over the membrane is the driving force in these systems, and only water vapor passes through the membrane. Successful applications of membrane contactors for air dehumidification at ambient temperatures are already reported (Isetti et al., 1997; Jain, Tripathi, & Das, 2011; Kneifel et al., 2006), but the potential of these systems for air dehumidification at elevated temperatures has not previously been quantified.

The potential of zeolites and membrane contactors for the dehumidification of the recycled air in spray dryers is investigated in this work. Both dehumidification systems have in common that heat is released when water vapor is adsorbed, and external energy is required for the regeneration of the adsorbent. Air dehumidification is only effective when the heat released at adsorption and the remaining heat from the regeneration are used elsewhere in the system (Atuonwu et al., 2012; Djaeni, Bartels, Sanders, van Straten, & van Boxtel, 2007). This makes heat integration a prerequisite for these proposed configurations to be energy efficient.

Pinch analysis a well-established method for heat integration and the design of heat exchanger networks to minimise external utilities (Kemp, 2007). The pinch approach is a step-wise procedure in which operational conditions, like flows and temperatures, are optimized first. Subsequently, given those optimized conditions, a heat exchanger network is defined according to the pinch rules. The drawback of this approach is the optimized operational conditions are not necessarily the optimal conditions for the heat exchanger network with the minimal external energy requirements. Atuonwu et al. (2011) applied a simultaneous approach based on the work of Duran and Grossmann (1986), where pinch analysis and optimisation of operational conditions were combined in one step. By considering streams and temperatures as variables, the pinch point can be shifted resulting in an additional heat recovery. For a low-temperature drying system with zeolites the simultaneous optimisation resulted in a 13% improvement in energy consumption compared to the results obtained with a standard step-wise pinch analysis (Atuonwu et al., 2011). In line with this, Walmsley et al. (2013) found that applying variable temperatures in pinch analysis for spray drying systems specific heat recovery can be increased by 30%.

Next to the application of the existing methods for the reduction of energy, like multi-stage drying with fluidized bed, air pre-treatment, heat pumps, and heat exchange between inlet and exhaust air, emerging technologies are needed to further reduce the energy consumption in spray drying processes. In this work we discuss the potential for energy reduction by air dehumidification in closed-loop spray drying and compare the results with the common practice in milk powder production. By combining emerging technologies different new closed-loop spray drying configurations are proposed to increase energy efficiency. Simultaneous optimisation of the operational conditions and the heat exchanger network is applied to find an optimal process design.

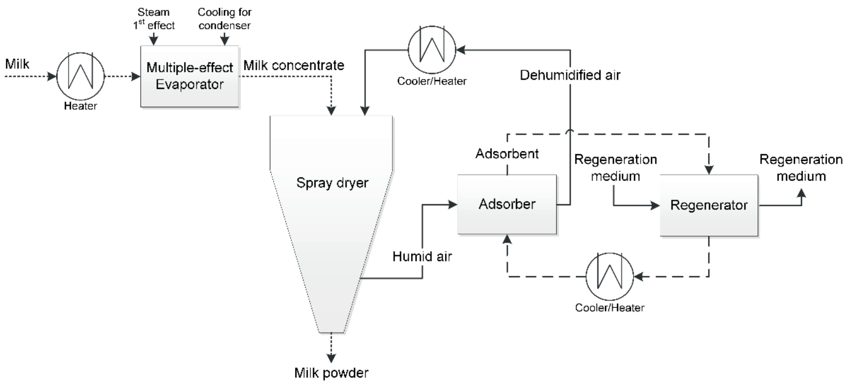

Figure 1. Schematic representation of a closed-loop dryer for milk powder production.

2 Process description

Spray drying systems are intensively used in the dairy industry. Milk powder is, therefore, used as model product. The processing steps for standardized milk powder production are heating, concentrating and drying. The focus in this study lays on the dryer section. In the closed-loop spray dryer system a surplus energy stream is created from the regeneration of the adsorbent. To be energy efficient the surplus energy has to be exploited elsewhere in the production process. Hence, a pre-heater and a multi-effect evaporator are included as a heat sink for the surplus energy of the drying process. In Figure 1. the dryer is given with the loops for air dehumidification, while the pre-heater and multi-effect evaporator are given as two-unit operations. The concentrated milk is atomized with a monodisperse nozzle and the spray dryer is operated in closed-loop with air dehumidification. The section for air dehumidification consists of an adsorber (either membrane contactor or zeolite wheel), regenerator, and a cooling/heating unit. The dehumidified air is heated/cooled to the drying temperature before re-entering the spray dryer.

The total system is split into subunits, as represented in Figure 1, and for each subunit overall steady-state mass and energy balances are used:

$$H_{l/s} = F_{l/s} \cdot (c_{p,l/s} \cdot x_{l/s} + c_{p,w} \cdot x_w)T_{l/s} \tag{1}$$

$$H_a = F_a\left(c_{p,a} + y_a \cdot c_{p,v}\right)T_a \tag{2}$$

where H is the enthalpy of the flows (kJ h^{-1}), F the mass flow of liquid (l), solids (s) and air (a) (kg h^{-1}), T the temperature of the flows (°C), x_w the water (kg kg^{-1}) and y_a the vapor content of the flows (kg kg dry air^{-1}), and c_p the heat capacities of water (w), vapor (v), liquid (l), solids (s), and air (a) (kJ kg^{-1} °C^{-1}).

2.1 Evaporator

Milk is first heated up in a pre-heater, and subsequently concentrated in a multi-effect evaporator. The energy requirements for heating follow from Eq. (1), and the size of the heat exchanger (A_{hex}, in m^2) is based on the following equation. In which Q_{hex} is the amount of energy exchanged, U is the heat transfer coefficient, and ΔT the temperature difference.

$$A_{hex} = \frac{Q_{hex}}{U\Delta T_{hex}} \tag{3}$$

The mass and component balances for the evaporator are:

$$F_m = F_{cm} + F_{v,1} + F_{v,2} + \ldots + F_{v,n} \tag{4}$$

$$F_m \cdot x_{m,in} = F_{cm} \cdot x_{m,out} \tag{5}$$

where F_m and F_{cm} are respectively the milk feed and concentrate flow (kg h^{-1}), x_{in} and x_{out} the solids concentration in the feed and concentrate (kg kg^{-1}), and $F_{v,i}$ the amount of evaporated water in each effect (kg h^{-1}). The number of effects (n) is set to 7, and milk is concentrated to 50% total solids.

The energy requirements for the evaporator result from the following equation (based on Eq. (1)):

$$H_{m,i-1} + H_{v,i-1} = H_{v,i} + H_{m,i} + H_{cw,i} \tag{6}$$

Where $H_{m,i-1}$ is the enthalpy flow of the milk feed to effect i, and $H_{m,i}$ the enthalpy flow of the concentrated milk flow leaving effect i, $H_{v,i}$ is the enthalpy flow of the vapor (or steam in case of the first effect), and $H_{cw,i}$ is the enthalpy flow of the condensate stream.

2.2 Monodisperse spray dryer

Water and energy are exchanged in the dryer due to the contact of hot air with atomized droplets. The mass balance for water is:

$$F_a \cdot y_{a,in} + F_{cm} \cdot x_{w,in} = F_a \cdot y_{a,out} + F_p \cdot x_{w,out} \tag{7}$$

where F_a and F_{cm} are the flows of dry air and concentrated milk (kg h^{-1}), y_a is the concentration of vapor in the air flow, and x_w is the water content of the milk flow (both in kg kg^{-1}). The moisture content of the inlet air ($y_{a,in}$) depends on the level of dehumidification achieved in the adsorber. The moisture content of the exhaust air ($y_{a,out}$) is related to the relative humidity of the exhaust air, which is set at 10%.

The overall energy balance for the inlet air ($H_{a,in}$) and milk concentrate (H_{cm}), and the exhausted air ($H_{a,out}$) and milk powder (H_p) is given by:

$$H_{a,in} + H_{cm} = H_{a,out} + H_p \tag{8}$$

3

The energy required for operating pumps, fans, nozzle, etc., and heat losses are not taken into account in this study.

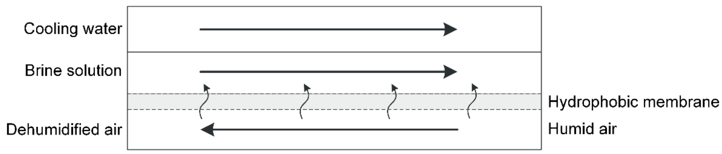

Figure 2. Schematic representation of a membrane contactor module with cooling water channel.

2.3 Membrane contactor

In the membrane contactor the air and brine streams are separated by a hydrophobic membrane, which is only permeable for water vapor. The difference in vapor pressure over the membrane is the driving force for vapor transport through the membrane. The brine heats up due to the released heat of condensation, and therefore a cooling system is integrated in the module as shown in Figure 2. The brine and cooling water flow are in co-current, while the brine and air are in counter current configuration. The applied mass and energy balances are:

$$F_a \cdot y_{a,in} + F_b \cdot x_{w,in} = F_a \cdot y_{a,out} + F_b \cdot x_{w,out} \tag{9}$$

$$H_{a,in} + H_{b,in} + H_{c,in} = H_{a,out} + H_{b,out} + H_{c,out} \tag{10}$$

$$P_{a,in} - P_{b,out} = P_{a,out} - P_{b,in} \tag{11}$$

where F_a and F_b are the flows of air and brine in the adsorber (kg h^{-1}), y_a and x_w the vapor and water concentrations (kg kg^{-1}), H_a, H_b, and H_c the enthalpy of air, brine and cooling water streams (kJ h^{-1}), P_a and P_b are the partial vapor pressures in the air and brine. The vapor pressure is based on the Antoine's equation (Patil, Tripathi, Pathak, & Katti, 1990).

The membrane contactor uses lithium bromide as brine solution, and the physical properties are taken from Florides et al. (2003), Iyoki et al. (1993), and Patil et al. (1990). Heat transfer between air, membrane, and brine, are based on the Maxwell-Stefan equation (Krishna & Wesselingh 1997), the membrane properties are taken from Zhang (2006). The membrane contactor is operated in a continuous mode with a moderate increment of the water concentration in the brine between in- and outlet of the module. Therefore, in the counter current operated membrane contactor the vapor pressure difference is assumed to be equal at every place. The brine temperature is related to the vapor pressure difference over the membrane (Eq.(11)).

After passage through the membrane contactor the brine is regenerated in a continuous operation. Two options for brine regeneration are considered: 1) water evaporation in a two-effect evaporator, or 2) by superheated steam. The energy needed for brine regeneration by the two-effect evaporator (H_{st}) is given by:

$$H_{st} = \frac{F_b \Delta x_{mc}}{1 + H_{v1}/H_{v2}} \cdot H_{v1}/F_{st} \tag{12}$$

where $H_{v,i}$ is the heat of evaporation (kJ h^{-1}) in effect 1 and 2, and depends on the temperatures in the effects. F_{st} is the amount of steam required for the first effect (kg h^{-1}), F_b the brine flow (kg h^{-1}), and Δx_{mc} is the concentration difference between the brine in and out. Boiling point elevation related to the lithium bromide concentration is taken into account.

For regeneration with superheated steam a second membrane contactor unit is needed, which operates in the same way as the membrane contactor for dehumidification, only without a cooling water section. Due to the low vapor pressure of the superheated steam, water vapor passes from the brine to the superheated steam. The energy balance for brine regeneration by superheated steam is:

$$H_{shs,in} + H_{b,in} = H_{shs,out} + H_{b,out} \tag{13}$$

where the enthalpy of the superheated steam (H_{rm}) depends on its temperature (T_{shs}) and pressure (Lachkov, Lysenkov, & Mamonov, 1999). After regeneration with superheated steam the temperature of the brine is adjusted to the optimal temperature for adsorption by either heating or cooling. The energy balance for the cooling/heating system is:

$$H_{b,in} + Q_{heat} = Q_{cool} + H_{b,out} \tag{14}$$

where Q_{heat} and Q_{cool} is the required energy for heating or cooling (kJ h^{-1}).

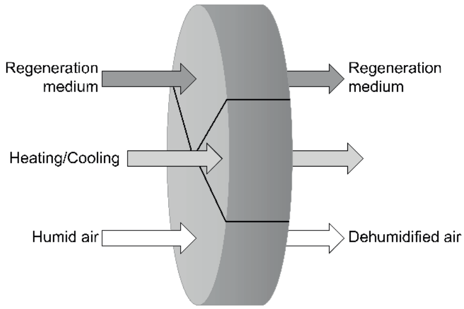

Figure 3. Schematic representation of a zeolite wheel with the adsorption, regeneration, and heating/cooling section.

2.4 Zeolite

The zeolite sorption system consists of a wheel system with three separated sections (see Figure 3): 1) adsorption, 2) regeneration, and 3) cooling or heating. Each section is modelled as an individual unit. For the adsorption section the following balances apply:

$$F_a \cdot y_{a,in} + F_z \cdot x_{w,in} = F_a \cdot y_{a,out} + F_z \cdot x_{w,out} \tag{15}$$

$$H_{a,in} + H_{z,in} - H_{des} = H_{a,out} + H_{z,out} \tag{16}$$

where F_a and F_z are the flows of air and zeolite (kg h^{-1}), y_a and x_w the vapor and water concentrations (kg kg^{-1}), H_a and H_z and the enthalpy of air and zeolite streams and H_{des} the desorption enthalpy (kJ h^{-1}). The sorption isotherm for a commercial zeolite (CeCA, 4A) is used (Boxtel et al., 2012).

Hot air or superheated steam can be applied for zeolite regeneration. The balances for regeneration of both regeneration systems are based on Eq. (16), and the balance for the cooling/heating is expressed in Eq. (14). The superheated steam flow is recycled after usage in the regenerator as shown in Figure 4. Part of this flow is upgraded and reused, the other part is a surplus flow and is exploited elsewhere in the system.

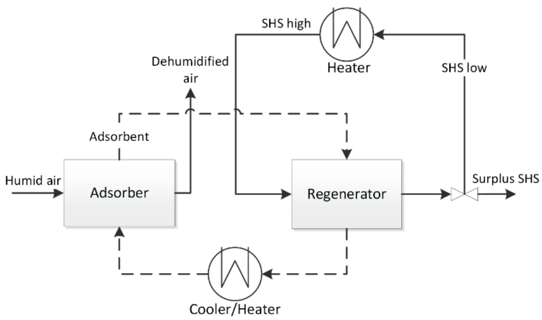

Figure 4. Superheated steam (SHS) cycle in adsorber/regenerator loop.

3 System optimisation

3.1 Process configurations

Four different configurations (see Table 1) are optimized, evaluated, and compared to a conventional spray dryer system without air dehumidification and recirculation. The energy consumption per kilogram produced milk powder for these configurations are compared to that of a conventional milk powder production system. In this conventional system a pre-heater, 7-stage evaporator and spray dryer are used and waste energy from the evaporator is recovered. The pre-heater and multi-stage evaporator are the same for all configurations. General data for the system is given in Table 2.

Table 1. Overview of the considered process configurations.

Configuration	Adsorber	Regeneration medium
Conventional	None	None
1	Membrane contactor	Evaporator
2	Membrane contactor	Superheated steam
3	Zeolite	Hot air
4	Zeolite	Superheated steam

Table 2. Fixed process variables.

Variable	Value
F_m (kg h^{-1})	100000
T_m (°C)	10
$x_{m,in}$ (kg kg^{-1})	0.09
$x_{m,out,evap}$ (kg kg^{-1})	0.5
$x_{w,out,dryer}$ (kg kg^{-1})	0.035

3.2 Operational conditions

Main objective for a closed-loop dryer is to minimise the external energy input. The decision variables that affect the energy input for all configurations are the operating temperatures of the evaporator ($T_{st,evap}$ and T_{cm}), the dryer inlet temperature ($T_{a,in}$), the moisture content of the air at the dryer inlet ($y_{a,in}$), and the temperatures of the regeneration medium entering and exiting the regenerator ($T_{rm,in}$, and $T_{rm,out}$). The concentration difference of the brine between the in- and outlet of the membrane contactor (Δx_b), and the temperature difference between the brine and the air side (ΔT_{mc}) are decision variables specific for the membrane contactor. The inlet temperature of the zeolite for the adsorption section ($T_{z,in}$) is of importance for the zeolite system. The upper and lower bounds for each decision variable are related to product and process constraints, and are listed in Table A.2.

3.3 Heat recovery design

In pinch analysis hot and cold composite curves are identified from the defined target temperatures and flows, and subsequently the heat recovery is estimated for a given temperature difference at the pinch point. In the applied procedure the target temperatures and flows are not fixed beforehand, but estimated in a simultaneous optimisation of the operational conditions and the heat exchanger network (Atuonwu et al., 2011). By altering the flows and temperatures in the system during the optimisation steps, the hot and cold composite curves are adjusted to minimise the energy consumption.

Table 3. Overview of all feasible hot and cold streams for the four configurations and the conventional configuration.

Hot streams		**1**	**2**	**3**	**4**	**Conventional**
H1	Vapor from the last effect of the milk evaporator	X	X	X	X	X
H2	Vapor from the last effect of evaporator for brine regeneration	X				
H3	Cooling water from the membrane contactor	X	X			
H4	Superheated steam surplus from regenerator		X		X	
H5	Hot air after regeneration			X		
Cold streams						
C1	Milk feed	X	X	X	X	X
C2	Steam to the 1st effect of the milk evaporator	X	X	X	X	X
C3	Steam to the 1st effect of the evaporator for brine regeneration	X				
C4	Superheated steam reheating after regenerator		X		X	
C5	Hot air for regenerator			X		
C6	Ambient air to the dryer					X
Hot/cold streams						
H6/C7	Air from adsorbent to the dryer	X	X	X	X	
H7/C8	Adsorbent from regenerator	X	X	X	X	

The hot and cold streams for all process configurations are given in Table 3. For each configuration a different number of hot and cold streams is available. It should be noted that, depending on the operational conditions in the system, some streams could then be either hot or cold streams. For example, depending on the operational conditions of the membrane contactor the brine needs cooling or heating after regeneration. The switches between heating and cooling are included in the procedure to optimise the operational conditions.

Figure 5 shows the generic heat exchanger network for the different configurations, including the switch streams. The number of hot and cold streams affects the size of the heat exchanger network. For the pinch point an individual minimum temperature difference was used, 10°C for gaseous streams and 5°C for liquid streams (Kemp, 2007).

The optimisation problem is defined as follows:

$$Q_{ex} = min\left(\sum_i Q_i^{HU}\right) \quad (17)$$

$$s.t.\, mass\ and\ energy\ balances\ (Eq. 1 - 15)$$

$$lower\ bound < decision\ variable < upper\ bound$$

where Q_{ex} is the total amount of required external heating (MJ kg milk powder^{-1}), which is based on the sum of hot utilities (Q_i^{HU}) of all the streams (i) and to total amount of product produced (F_p). Ambient air or ground water are sufficient for cooling, active chilling is not necessary, therefore cooling not included in the objective. Simulation and optimisation were performed in MATLAB R2014b using the genetic algorithm solver.

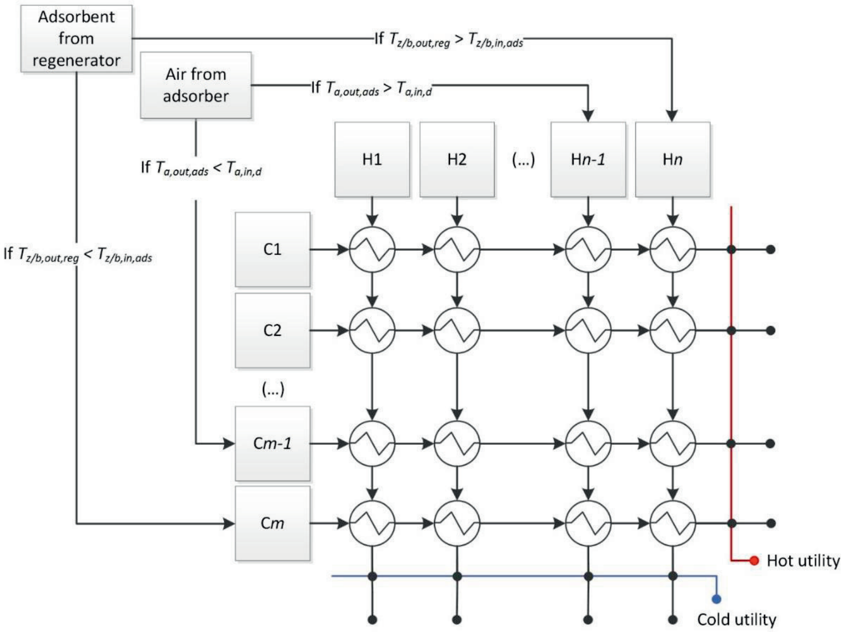

Figure 5. Generic heat exchanger network for the different configurations. The number of hot (n) and cold (m) streams depend on the configuration.

3.4 Economic evaluation

To address the industrial viability of the proposed technologies the utility and investment cost are estimated for the energy optimal scenarios. Each configuration is evaluated on the total annualized costs (TAC), consisting of the equipment costs (C_{equip}) and utility costs (C_{util}) for every process j and yearly operating time (t).

$$TAC = \sum_j C_{eq,j} + \sum_j C_{ut,j} t \tag{18}$$

$$C_{eq} = C_{inv} \cdot LF \cdot \frac{i(1+i)^{n_{life}}}{(1+i)^{n_{life}} - 1} \tag{19}$$

in which LF is the Lang factor to include the costs for building and installing the equipment, i is the interest rate, n_{life} is the lifetime, and C_{inv} is the equipment initial investment costs. The investment costs are derived from standard engineering data in Table 4. For up- and down scaling from the given equipment dimensions, the following equation is used:

$$C_{inv} = C_{ref} \left(\frac{A_{eq}}{A_{ref}}\right)^{n_{eq}} \tag{20}$$

with C_{ref} the costs for a reference installation with dimension A_{ref} as given in Table 4, A_{eq} the actual equipment dimension in required in the configurations, and n_{eq} the scaling factor. Membrane contactors operate on the same principle as membrane distillation units, therefore the membrane costs are based on a membrane distillation unit. Industrially used zeolite wheels have a diameter up to 4 meter, this means multiple wheels are placed if a larger area is required.

Table 4. Cost data for each operation.

Process	A_{ref}	C_{ref}	n_{eq}	**Life time**	**Reference**
Heat exchanger	80 m^2	€ 32800	0.68	20 years	(Smith, 2005)
Evaporator	7700 kg water h^{-1}	€ 830000	0.53	30 years	(Seider, Seader, Lewin, & Widagdo, 2010)
Spray dryer	400 kg water h^{-1}	€ 600000	0.29	30 years	(*APV Dryer Handbook*, 2000; Garrett, 1989)
MC (membrane)	1 m^2	€ 50	-	4	(Elsayed, Barrufet, & El-Halwagi, 2014)
MC (equipment)	1 m^2	€ 200	-	10	(Elsayed et al., 2014)
Zeolite wheel	1400 kg water h^{-1}	€250000	0.78	5	(Voogt, 2016)

The utility costs consist of the heating and cooling cost required for all processes:

$$C_{ut} = C_{heat} \cdot Q_{heat} + C_{cool} \cdot Q_{cool} \tag{21}$$

in which C_{heat} is the heating cost, and C_{cool} is the cost for cooling. Electrical usage of pumps, fans etc. is not taken into account. Table 5 lists the used economic data.

Table 5. Economic data.

Item	Value
Operational time (h year^{-1})	8000
Interest rate (%)	0.6
Lang Factor (-)	3.5
Cold utility cost (€ kJ^{-1})	0.1 x 10^{-5}
Hot utility cost (€ kJ^{-1})	1.3 x 10^{-5}

4 Results and discussion

4.1 Conventional configuration

The energy requirement for the optimized conventional configuration is 8.4 MJ per kg milk powder. For heat recovery vapor from the last evaporator effect is used to pre-heat the milk to the evaporator and to pre-heat the drying air. The energy consumption of the conventional configuration is in agreement with the values reported by Ramírez et al. (2006) for the same setup.

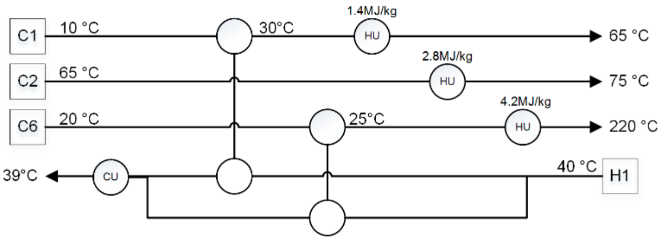

Figure 6. Heat exchanger network of the conventional configuration, including the hot and cold utilities (HU and CU respectively). The flow descriptions are given in Table A.3.

4.2 Configuration 1 – Membrane contactor with evaporator

In this optimized and heat integrated system a moderate energy increase in the air after dehumidification was achieved, and the dehumidified air needs further heating to the drying temperature. The other energy input in this system is required for the regeneration loop of the brine. The energy gain in this system is realized by recovery of the heat which is obtained at condensation of the water vapor in the brine in the membrane contactor. The temperature of the internal cooling water (H3), that is used to recover the heat, is raised to 92°C. This flow is used to pre-heat the milk feed before concentration (C1) and the brine after regeneration (C8). Figure 7 shows the optimal heat exchanger network for this configuration. The energy requirement for this optimized system is 7.3 MJ per kg of milk powder, which is a 14% improvement compared to the conventional configuration. The optimal values for the decision variables are listed in Table 6.

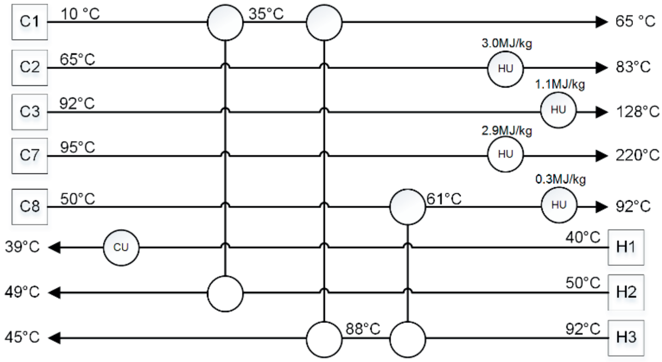

Figure 7. Heat exchanger network design for configuration 1. HU: hot utilities, CU: cold utilities, the flow descriptions can be found in Table A.4.

In this configuration the spray drying temperature ($T_{a,in}$) is the same as in the conventional configuration. The hot streams have a maximum temperature of 92°C, (see Figure 7) and cannot be used to heat the air after dehumidification, the spray dryer is most energy efficient at the highest temperature, i.e. 220°C. The other decision variables are all on, or close to, the boundaries for this system. To transfer most heat from the brine to the cooling water and air, the temperature of the evaporator ($T_{rm,in}$), and the temperature- and concentration difference (ΔT_{mc} and Δx_{mc}) over the membrane contactor are set to their maximum values.

3

Table 6. Optimal values for the decision variables for all configurations. T_{rm} is the regeneration medium temperature which can either be superheated steam, hot air or steam for the evaporator, depending on the configuration.

Decision variable	1	2	3	4	Conventional
$T_{a,in}$ [°C]	220	220	220	180	220
$y_{a,in}$ [kg kg dry air^{-1}]	0.010	0.012	0.002	0.002	-
Δx_{mc} [kg kg^{-1}]	0.099	0.014	-	-	-
ΔT_{mc} [°C]	1.2	1.0	-	-	-
$T_{z,in}$ [°C]	-	-	125	125	-
$T_{rm,in}$ [°C]	128	187	298	350	-
$T_{rm,out}$ [°C]	50	100	-	250	-
T_{steam} [°C]	83	88	87	91	75
T_{cond} [°C]	27	32	35	30	19

4.3 Configuration 2 – Membrane contactor with superheated steam

For this configuration the optimal conditions of the air loop over the dryer and the dehumidification unit are comparable to previous configuration. Most important difference is the high temperature of the superheated steam which changes the brine after regeneration into a hot stream (H7). The surplus superheated steam (H4) acts as a high-quality energy flow, and is used efficiently in heat integration. Related to this difference, the cooling water flow (H3) is of a lower quality, therefore, most of the latent heat released in the brine is transported to the regenerator via the brine flow, and a negligible amount to the cooling water system. This is achieved by a large brine flow and a very small cooling water flow, resulting from a small concentration difference of the brine between in- and outlet of the membrane contactor (Δx_{mc}). The heat exchanger network, as shown in Figure 8, does not include a heat exchanger connected to the cooling water stream (H3), as the energy content of this stream is so low it can be neglected (0.004 MJ per kg milk powder). The energy consumption for this optimized and heat integrated configuration is 4.9 MJ per kg milk powder, which is a large improvement compared to the conventional configuration (42%).

The relative high temperature of the superheated steam (over 187°C) is a possible drawback for this system. No literature is published on membrane contactors operating at these temperatures. Nevertheless, thermal stable membranes exist (Li & Chen, 2005), hence, this configuration could be feasible. As an alternative the temperature of the superheated steam could be reduced. Decreasing the temperature of the superheated steam from 187°C to 150°C results in an increase of energy consumption of less than 1%.

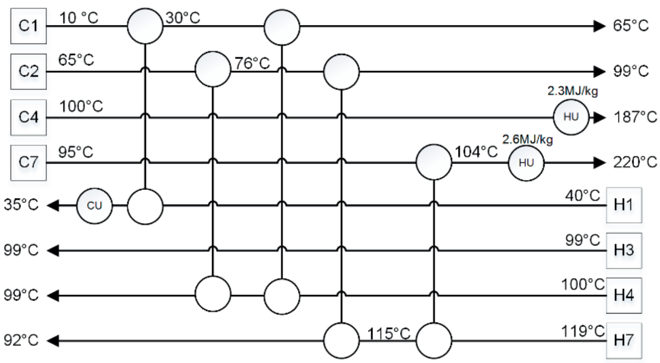

Figure 8. Heat exchanger network design for configuration 2. HU: hot utilities, CU: cold utilities, the flow descriptions can be found in Table A.5.

4.4 Configuration 3 – Zeolite with hot air

The energy consumption for this optimized and heat integrated configuration is 7.5 MJ per kg milk powder, which is 17% below the energy consumption in the optimized conventional system. The energy reduction is comparable to configuration 1. The optimal values for the decision variables are given in Table 6.

The dehumidified air (C6) exits the adsorber section of the zeolite with a temperature of 163°C, and to reach the drying temperature of 220°C a low amount of external energy is needed. This difference compared to the conventional configuration is the main contributor for the reduction of energy input. The configuration needs, however, a large amount of energy to heat the hot air for regeneration (from ambient temperature to 298°C), which can only partially be supplied from available hot streams. The regenerator exhaust air (H5) has a temperature of 125°C, and this stream can only be used to pre-heat the air for regeneration (C5), or to pre-heat the feed flow to the evaporator (C1). Figure 9 shows the optimal heat exchanger network for this configuration. A drawback of using hot air as regeneration medium is the lower quality compared to steam, and the need for air-air and air-liquid heat exchangers. This type of heat exchanger needs larger heat exchanging surfaces, and have therefore higher investment costs, than steam-heat exchangers.

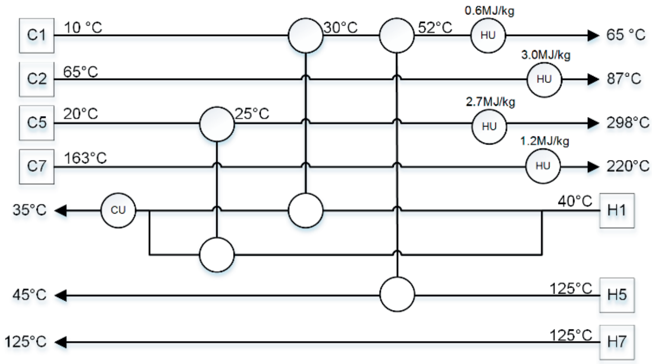

Figure 9. Heat exchanger network design for configuration 3. HU: hot utilities, CU: cold utilities, the flow descriptions can be found in Table A.6.

4.5 Configuration 4 – Zeolite with superheated steam

The energy consumption for this configuration is 5.2 MJ per kg milk powder. This is a large improvement in energy efficiency compared to configuration 3, and is similar to the energy consumption of configuration 2. Like in configuration 2, the energy reduction is possible due to the high quality of the surplus superheated steam after regeneration. The optimal temperature of the surplus superheated steam in this configuration has a temperature of 250°C, and can be applied for many purposes. In the optimized system the superheated steam (H4) is used to heat the dehumidified air (C7) to the drying temperature which is, in contrast to all other configurations, 180°C instead of 220°C. The zeolite requires cooling after regeneration (H7). The cold streams in the system, milk feed flow (C1) and evaporator steam flow (C2), are not suitable for direct cooling, hence the cooling energy from the zeolite cannot be recovered by heat integration. Cooling of the zeolites with ambient air is most advisable.

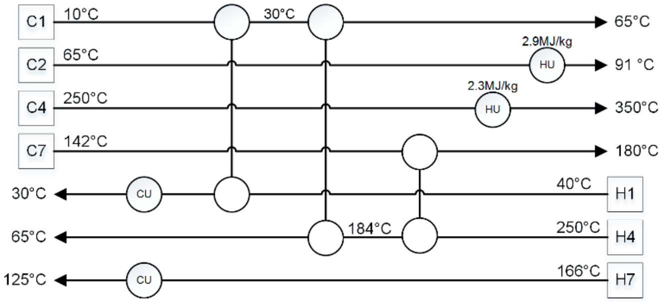

Figure 10. Heat exchanger network design for configuration 4. HU: hot utilities, CU: cold utilities.

4.6 Discussion of the different configurations

The energy requirements for the four configurations and the conventional configuration are summarized in Figure 11. The total energy savings range between 11 to 42% compared to the conventional configuration. Atkins et al. (2011) applied heat integration whereby the dryer exhaust air is used to pre-heat the inlet air and reached a reduction in energy consumption up to 20%. The configurations in this work with dehumidification realise higher energy savings.

The energy consumption of configurations 1 and 3 is higher than in configurations 2 and 4. The quality of the hot streams in configurations 1 and 3 is not sufficient to heat the dehumidified air to the optimal dryer temperature. The surplus of superheated steam in configuration 2 and 4 is a high-quality energy stream and is used efficiently; in configuration 4 even to heat the dehumidified air to the dryer temperature.

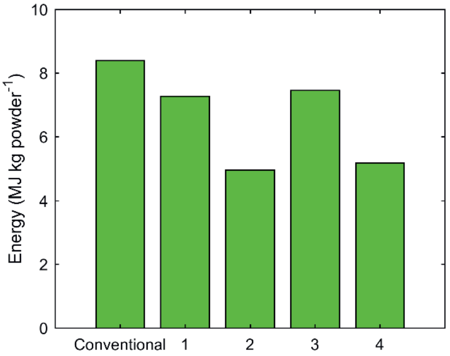

Figure 11. External energy requirements with and without heat integration for configurations 1 to 4 and the conventional configuration.

In configuration 2 more energy is used to operate the adsorber and regeneration section than in configuration 4, but in configuration 2 more energy is recovered by heat integration. There are two main causes for this effect. First, zeolite in configuration 4 has to be cooled after regeneration but the energy cannot be recovered, while the energy in the brine solution of configuration 2 is recovered by a cold stream. Secondly, the heat of adsorption in configuration

2 is released in the brine, whereas in configuration 4 the majority it is transferred directly to the dehumidified air. As a result, the dehumidified air (C7) in configuration 2 has a temperature of 95°C versus 142°C in configuration 4. Hence, the dehumidified air flow in configuration 2 needs more heating.

4.7 Sensitivity analysis

A sensitivity analysis on variations over the operational window, set by the bounds of the decision variables, showed for all systems that the energy consumption is almost constant. Examples of variations are given in Figure 12. Only at the edges of the operational window is the sensitivity meaningful. Variations in air temperature and humidity at the dryer inlet has a maximal effect of 4% on the total energy efficiency for all configurations (see Figure 12a and c for configuration 2 and 4). The dryer can, therefore, be operated at a broad range of operational conditions, without a large influence on the total energy efficiency. This low sensitivity allows tuning of the operational conditions to the product specifications rather than energy requirements. On the other hand Figure 12d illustrates that some combinations of superheated steam temperature for regeneration ($T_{rm,in}$) and zeolite temperature ($T_{z,in}$), in configuration 4 will increase the energy consumption significantly, however, there is still a large operational window where energy consumption is low. In contrast, for configuration 2 (Figure 12b), the energy consumption is hardly influenced a change in temperature of the superheated steam used for regeneration ($T_{rm,in}$).

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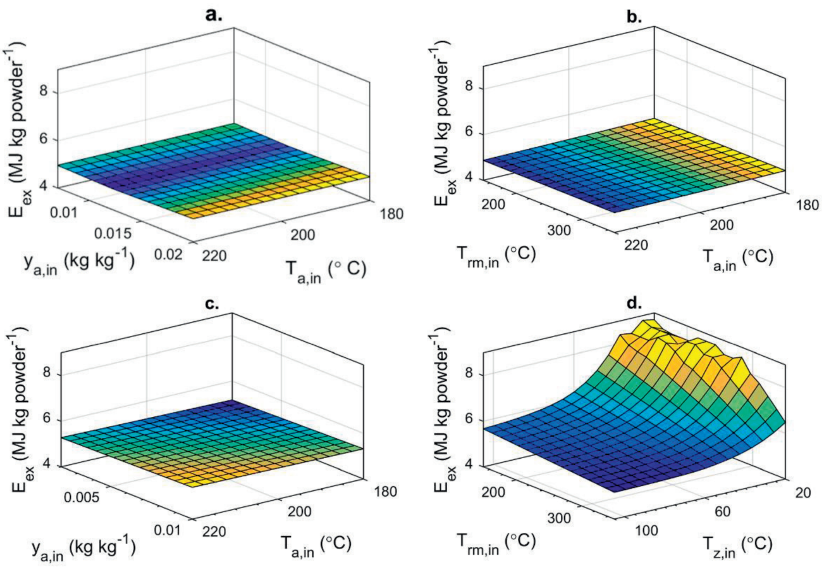

Figure 12. Energy consumption variation with different operational conditions for configuration 2 (a and b) and 4 (c and d).

In the presented results the inlet concentration of the milk for the dryer was set to 50% total solids, which is common in milk powder production. Increasing the inlet concentration reduces the total energy consumption (Walmsley, Atkins, Walmsley, Philipp, & Peesel, 2018). However, with increasing solids concentration the viscosity also increases, which may lead to clogging of the spray dryer nozzle. Advantage of the monodisperse atomisation nozzle is the better handling of high viscosity products (Deventer et al., 2013), which makes the increase of concentration possible. Figure 13 illustrates the effect of variations in dryer feed concentration on the energy consumption. All configurations show a decreasing energy consumption with increasing feed concentration. However, for configuration 2 the energy consumption is almost constant for feed concentrations above 45% total solids and for configuration 4 above 30% total solids. These configurations use superheated steam for regeneration and results in high quality heat, which is exploited effectively in the current system.

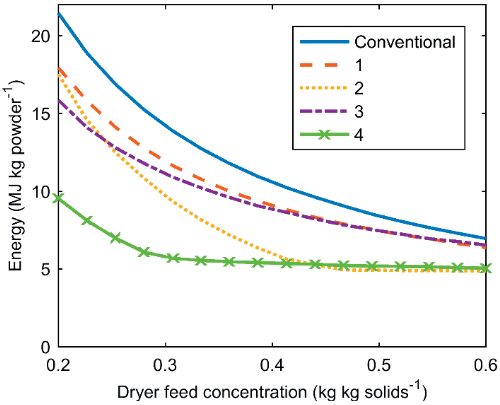

Figure 13. The effect of level of concentration before spray drying on the energy consumption is shown for the different configurations.

4.8 Used optimisation method

Simultaneous optimisation of operational conditions and heat integration resulted in an additional energy reduction varying between 1 to 11% for the different configurations. For configuration 1, 3, and 4 the additional energy reduction is minor (1 to 4%). These heat integrated systems already proved to be energy efficient and during optimisation the gain achieved for one stream is counteracted by extra energy needs for another. Configuration 2, however, has an additional energy reduction of 11%, and has the most advantage of the simultaneous approach. The gain for this configuration is the utilisation of the surplus superheated steam after regeneration, which has a high energy content to be reused for heating of other streams in the system. Atuonwu et al. (2011) achieved for low-temperature drying an improvement of 13% in energy reduction for the simultaneous optimisation, which is in line with the improvement found for configuration 2. The closed-loop spray dryer considered in this work operates at higher temperatures, which makes drying more energy efficient than low temperature drying (50°C). Although the simultaneous optimisation approach did not result in large energy reduction for all configurations, the approach allows an open search for solutions without an a priori choice of heat integration possibilities. In this work this approach proved to be powerful for systems with streams which, depending on the chosen operational conditions, can be used as either hot or cold streams.

4.9 Economic evaluation

The total annual costs in k€ year^{-1} (TAC) of the different configurations are summarized in Table 7. The costs of the pre-heater, evaporator, and spray dryer are the same for all configurations. The TAC for configurations 3 and 4, which use the zeolite systems, have comparable and significant lower costs than the conventional system. Configuration 1 and 2 need large membrane areas which result in high costs for the membrane contactor system. These high costs are result of the applied objective function that minimizes the energy consumption. Optimizing configuration 2 with respect to the TAC, instead of energy consumption, results in a 50% cost decrease, while the energy consumption is only increased by 4%. The resulting costs, 12.9 M€ year^{-1}, are still higher compared to the configurations with zeolites. However, with the development of new membranes the flux can be increased, which will decrease the costs drastically.

Table 7. Cost overview (k€ year^{-1}) for all different configurations.

	1	2	3	4	Conventional
Equipment costs	7573	18414	1636	1658	1135
Evaporator	739	739	739	739	739
Spray dryer	372	372	372	372	372
Zeolite			493	493	
MC	6014	4882			
MC regeneration	420	9174			
Heat exchangers	27	92	31	53	23
Utility costs	7287	4926	7397	5182	8286
TAC	14860	23340	9033	6841	9422

4.10 Discussion

Heat integration is essential for all configurations to realise the reduction in energy consumption. The realisation depends on the feasibility of the proposed heat exchanger network in industrial applications. Plant layout, possibilities for energy transport between units, and interference with other operations may limit the application of heat integration. The usage of heat pumps was not investigated in this study, but has the potential to further increase the recovery of the efficiency of heat recovery in spray drying systems (Jensen, Markussen, Reinholdt, & Elmegaard, 2015; Walmsley, Klemeš, Walmsley, Atkins, & Varbanov, 2017). In the current work the evaporator was used as a heat sink for the surplus heat. Heat integration with modern MVR installations is less effective and, in that case, the surplus heat has to be used for other processes/activities at the dairy plant.

The absence of fines in the dryer exhaust, realized by the monodisperse nozzle, is a main requirement for air dehumidification in the closed loop. The negative influence of fouling of the membranes or particle deposition in the zeolite, on the process performance should be avoided. Monodisperse droplet drying is proven at pilot scale (Debrauwer, 2016) and is under development for larger production capacities. The shown potential for energy reduction motivates further development of the monodisperse atomizers and the investigation of monodisperse powder properties. Monodisperse drying has also other energy efficiency advantages. When all particles are similar in size no overheating of smaller particles occurs, which leads to a better controllability of the dryer and a further reduction in energy consumption (Atuonwu & Stapley, 2017).

The application of membrane contactors is new in this sector, and dehumidification at elevated temperatures is still at technology development level, further research is required to prove the effectiveness and compatibility of this technology. Furthermore, although the brine is used in a closed loop and is isolated from the drying air by a membrane, the usage of a strong brine can be a hurdle for the acceptance of membrane contactors in the food industry. Zeolite dehumidification systems, on the other hand, are already tested in the food industry for the pre-treatment of the air prior to drying (Boxtel et al., 2012).

In 2015 the EU produced 2.9 million tons of skimmed and whole milk powder (Eurostat, 2016), which requires around 26 PJ for production. Implementation of the proposed closed-loop dryer system, with savings between 11 to 42% in energy consumption, results in an energy reduction of 4.3 to 11.6 PJ per year. The discussed emerging technologies are essential and the optimal systems surmount the 2030 EU goals on energy reduction. Although this study focusses on the production of milk powder, the proposed configurations are applicable to any spray drying process. Other industries like the pharmaceutical and chemical industry already use closed-loop spray drying in order to recover solvents and gasses (GEA Niro Pharmaceutical, n.d.). These industries, and also others, can now benefit from the discussed solutions to improve their energy efficiency.

5 Conclusion

Energy reduction by closed-loop drying is not possible in current spray drying systems. Energy for the dryer exhaust air cannot be recovered due to the fines present in the air. Application of monodisperse droplet atomizers result in exhaust air without fines, and hence closed-loop drying with energy recovery can be applied. Heat integration is then essential to make the system highly energy efficient. Four different configurations optimized in this work resulted in a potential energy savings of 11 to 42% energy compared to current practice of milk powder production. The configuration with the lowest external energy requirements, corresponding to a reduction of 42%, consists of the membrane contactor with superheated steam as regeneration medium (configuration 2). However, for air dehumidification at elevated temperatures zeolite adsorbent wheels are closer to commercial implementation compared to membrane contactors. A zeolite system with superheated steam as regeneration medium (configuration 4) results in an energy reduction of 39% compared to current practice. Furthermore, in this work simultaneous optimisation of the operational conditions and heat exchanger network proved to be an effective approach for additional energy reduction.

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Nomenclature

Variables

C	Costs (€)
c_p	Specific heat capacity (kJ kg^{-1}°C^{-1})
F	Flow (kg h^{-1})
H	Enthalpy flow (kJ h^{-1})
i	Interest rate (%)
LF	Lang factor (-)
P	Vapor pressure (Pa)
Q	Energy requirement (kJ kg milk powder^{-1})
Q_{hex}	Energy requirement (kJ
RH	Relative humidity (-)
T	Temperature (°C)
t	Operating time (h year^{-1})
TAC	Total annual costs (€ year^{-1})
U	Heat transfer coefficient (W m^{-2} °C^{-1})
x	Component content of stream (kg kg^{-1})
y	Moisture content of air stream (kg kg dry air^{-1})

Subscripts

a	Air
abs	Absolute
ads	Adsorber
b	Brine
c	Cooling water
cm	Milk concentrate
cw	Condensate
cool	Cooling requirement
d	Dryer
des	Desorption
eq	Equipment
evap	Evaporator
ex	External heating
heat	Heating requirement
hex	Heat exchanger
hu	Hot utility
in/out	Inlet or outlet stream
inv	Investment
l/s	Liquid or solid
life	Lifetime
m	Milk
p	Milk powder
reg	Regenerator
rm	Regeneration medium
st	Steam
shs	Superheated steam
ut	Utility
v	Vapor
w	Water
z	Zeolite

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3

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Appendix

Table A.1 Parameters used for optimization (Doran, 1995; Kreith, Manglik, & Bohn, 2012; Perry & Green, 1997)**.**

Parameter	**Value**
P_{abs} (Pa)	101325
$c_{p,a}$ (kJ kg^{-1} °C^{-1})	1
$c_{p,m}$ (kJ kg^{-1} °C^{-1})	1.54
$c_{p,z}$ (kJ kg^{-1} °C^{-1})	0.84
$c_{p,w}$ (kJ kg^{-1} °C^{-1})	4.18
$c_{p,v}$ (kJ kg^{-1} °C^{-1})	1.93
H_v (kJ kg^{-1})	2500
U_{a-l} (W m^{-2} °C^{-1})	170
U_{a-a} (W m^{-2} °C^{-1})	35
U_{a-st} (W m^{-2} °C^{-1})	200
U_{l-l} (W m^{-2} °C^{-1})	1200
U_{l-st} (W m^{-2} °C^{-1})	2200
U_{shs} (W m^{-2} °C^{-1})	100

Table A.2 Lower and upper boundary values for each decision variables. The boundaries for $T_{rm,in}$ depend on the used regeneration medium; [1] is for air and superheated steam, and [2] is for steam for the brine evaporator.

Variable	**Lower bound**	**Upper bound**
$T_{a,in}$ (°C)	180	220
$y_{a,in}$ (kg dry air kg^{-1})	0.002	0.1
$T_{st,evap}$ (°C)	75	100
T_{cw} (°C)	12	50
Δx_{mc} (kg kg^{-1})	0.01	0.1
ΔT_{mc} (°C)	1	20
$T_{z,in}$ (°C)	20	125
$T_{rm,in}$ (°C) [1]	200	350
$T_{rm,in}$ (°C) [2]	100	150
$T_{rm,out}$ (°C)	100	250

Table A.3 Stream data for the conventional configuration.

Stream name	ID	F (kg h^{-1})	T_s (°C)	T_t (°C)	Q (MJ h^{-1})
Milk feed	C1	100000	10	65	2.2×10^5
Steam to the 1st effect of the milk evaporator	C2	11968	65	75	2.8×10^5
Ambient air to the dryer	C6	211149	20	220	4.3×10^5
Vapor from the last effect of the milk evaporator	H1	11543	40	39	2.8×10^5

Table A.4 Stream data for configuration 1.

Stream name	ID	F (kg h^{-1})	T_s (°C)	T_t (°C)	Q (MJ h^{-1})
Milk feed	C1	100000	10	65	2.2×10^5
Steam to the 1st effect of the milk evaporator	C2	12069	65	83	2.8×10^5
Steam to the 1st effect of the evaporator for brine regeneration	C3	4692	92	128	1.1×10^5
Air from adsorbent to the dryer	C7	210205	95	220	2.7×10^5
Adsorbent from regenerator	C8	37987	50	92	4.4×10^5
Vapor from the last effect of the milk evaporator	H1	11543	40	39	2.8×10^5
Vapor from the last effect of evaporator for brine regeneration	H2	4223	50	49	1.0×10^5
Cooling water from the membrane contactor	H3	66113	92	40	1.4×10^5

Table A.5 Stream data for configuration 2.

Stream name	ID	F (kg h^{-1})	T_s (°C)	T_t (°C)	Q (MJ h^{-1})
Milk feed	C1	100000	10	65	2.2×10^5
Steam to the 1st effect of the milk evaporator	C2	12202	65	99	2.8×10^5
Superheated steam reheating after regenerator	C4	126131	100	187	2.2×10^5
Air from adsorbent to the dryer	C7	210219	95	220	2.7×10^5
Vapor from the last effect of the milk evaporator	H1	11543	40	35	2.8×10^5
Cooling water from the membrane contactor	H3	108	99	99	-
Superheated steam surplus from regenerator	H4	8674	100	99	2.3×10^5
Adsorbent from regenerator	H7	333252	119	92	2.2×10^5

Table A.6 Stream data for configuration 3.

Stream name	ID	F (kg h^{-1})	T_s (°C)	T_t (°C)	Q (MJ h^{-1})
Milk feed	C1	100000	10	65	2.2×10^5
Steam to the 1st effect of the milk evaporator	C2	12132	65	87	2.8×10^5
Hot air for regenerator	C5	90595	20	298	2.6×10^5
Air from adsorbent to the dryer	C7	202750	163	220	1.2×10^5
Vapor from the last effect of the milk evaporator	H1	11543	40	35	2.8×10^5
Hot air after regeneration	H5	90595	125	45	0.9×10^5
Adsorbent from regenerator	H7	57317	125	125	-

3

Table A.7 Stream data for configuration 4. The Zeolite stream after regeneration (H7) is cooled with ambient air, and is therefore not included as cold utility requirement.

Stream name	ID	F (kg h^{-1})	T_s (°C)	T_t (°C)	Q (MJ h^{-1})
Milk feed	C1	100000	10	65	2.2 x 10^5
Steam to the 1st effect of the milk evaporator	C2	12175	65	91	2.8 x 10^5
Superheated steam reheating after regenerator	C5	113000	250	350	2.2 x 10^5
Air from adsorbent to the dryer	C7	275000	142	180	1.1 x 10^5
Vapor from the last effect of the milk evaporator	H1	11500	40	30	2.8 x 10^5
Superheated steam surplus from regenerator	H4	8720	250	65	3.3 x 10^5
Adsorbent from regenerator	H7	64800	166	125	-

Chapter 4

Assessment of air gap membrane distillation networks for milk concentration

Abstract

Multi-effect evaporation is the state of the art for concentration of liquid food products to high solid content. Membrane technology with reverse-osmosis and membrane distillation offer an alternative. For the concentration of milk, a reverse osmosis and air-gap membrane distillation network was modelled and optimized. Fouling dynamics and scheduling are taken into account. Reverse osmosis is favourable until its maximum achievable concentration. Air gap membrane distillation is, despite the low operational temperatures, energy intensive for the concentration of milk. A large recirculation flow to keep sufficient cross flow has to be heated and cooled, and the costs for heating and cooling dominate the total costs for product concentration. Moreover, fouling increases the energy requirements. The optimal system for air gap membrane distillation has only one stage operating at a high concentration and relative low flux. Applying multiple stages reduces the investment costs due to smaller units, but the heating and cooling costs increase. Major opportunities to improve the performance of air gap membrane distillation for concentration of milk are: 1) increase the cold and hot side temperatures to their maximum acceptable values, 2) develop spacers that allow lower linear flow velocities in the system and thus lower recirculation rates, and 3) make use of available waste heat.

Keywords: Membrane distillation, milk, reverse osmosis, network optimisation, process design

1 Introduction

Increasing need to reduce energy consumption and to use sustainable energy resources result in a demand for alternative product processing methods in the food industry. Traditional multi-stage evaporators used to concentrate food products are energy intensive, and require around 300 kJ per kg water removed (Ramirez, Patel, & Blok, 2006). This energy efficiency has increased in last decades due to the introduction of thermal and mechanical vapor recompression. Concentration by pressure driven membrane filtration, however, only requires 14 – 36 kJ per kg water removed (Ramirez et al., 2006). The drawback of pressure driven membrane filtration is the achievable product concentration, which is limited due concentration polarization. For dairy products a maximum of 18% solids in the product stream is considered as economical feasible for reverse osmosis (RO) (Walstra, Geurts, Noomen, Jellema, & Boekel, 1999). Membrane distillation (MD) is an emerging technology with the potential to concentrate to high solid contents. MD was developed as a desalination process in the 60's, and with the further development of suitable membranes in the 80's, the interest in this technology increased (El-Bourawi, Ding, Ma, & Khayet, 2006). In more recent years MD gained attention for the concentration of food products, especially fruit juices and dairy products (Alves & Coelhoso, 2006; Hausmann et al., 2011; Nene, Kaur, Sumod, Joshi, & Raghavarao, 2002; Quist-Jensen et al., 2016).

In MD, a porous hydrophobic membrane separates the feed and permeate phases and allows only water vapour to diffuse through the membrane. The driving force for mass transport is the partial vapour pressure difference between feed and permeate, which is related to the temperature difference over the membrane. As a result, the retention rate is very high, and high-quality water is produced as permeate. These advantages are the reason for the interest of MD for desalination and waste water treatment (El-Bourawi et al., 2006). In contrast to other membrane processes, like reverse osmosis, ultrafiltration etc., MD is thermally driven instead of pressure driven. MD is therefore less affected by concentration polarisation (Tijing et al., 2014). For the concentration of milk a final solids concentration up to 45 – 50% is feasible by MD, which makes it a promising alternative for traditional evaporation (Hausmann, Sanciolo, Vasiljevic, Kulozik, & Duke, 2014; Hausmann et al., 2011; Moejes et al., 2015).

Direct contact membrane distillation (DCMD) and air gap membrane distillation (AGMD) are most used for desalination and food applications. In DCMD the hot feed is separated by a hydrophobic membrane from a cold permeate stream. Water evaporates at the feed-membrane interface, passes through the membrane, and condensates at the membrane-permeate interface. In an AGMD configuration, on the other hand, water vapour from the feed passes through the membrane into an air gap, which on the other side is separated by a plate from a coolant at which the vapour condensates.

Advantage of AGMD compared to DCMD is the possibility of internal heat recovery, which results in a higher energy efficiency (Summers, Arafat, & Lienhard V, 2012). Therefore, AGMD has potential to compete with multi-effect evaporation and is considered in this study. A drawback of AGMD, on the other hand, is the lower flux compared to DCMD due to the smaller vapour pressure gradient (El-Bourawi et al., 2006). Both systems operate at low temperatures, around 60°C, which makes MD processes interesting for heat sensitive products like fruit juices and dairy products. The thermal energy consumption is, however high compared to RO and modern multi-stage evaporators, with an energy consumption of 400 – 1300 kJ per kg water removed (Duong, Cooper, Nelemans, Cath, & Nghiem, 2016; González, Amigo, & Suárez, 2017; Koschikowski et al., 2009; Kuipers et al., 2014; Mar Camacho et al., 2013; Ramirez et al., 2006). The advantage of MD is in the low operating temperatures, which allow the usage of low-quality heat, for example waste heat of other processes. Several studies suggested and investigated the usage of waste heat for operating the MD process (Dow et al., 2016; Elsayed, Barrufet, & El-Halwagi, 2014; Hausmann, Sanciolo, Vasiljevic, Weeks, & Duke, 2012; Kuipers et al., 2014). In presence of abundant waste heat with temperatures of at least 40 – 70°C, MD might be an interesting alternative for traditional concentration methods like multi-stage evaporation.

For industrial applications MD will be applied in a network with RO network for pre-concentration. Both the RO and MD network consists of some concentration stages in series and in each stage a number of modules in parallel. Not only operational conditions, but also the configuration of a RO and MD network is crucial to guarantee a constant production, product quality, and minimum energy consumption. Several studies showed results on optimal membrane network designs, in which most focus on the synthesis of RO networks (Alnouri & Linke, 2012; El-Halwagi, 1992; Khor, Chachuat, & Shah, 2012; Srinivas & El-Halwagi, 1993; Zhu, 1997). González-Bravo et al. (2015) published the first results for the synthesis of a membrane distillation network for sea water desalination and dextrose syrup concentration. The main difference between desalination and concentration of food products, like milk, is that fouling plays a dominant role due a gradual decline of mass and heat transfer over time. In pressure driven membrane application the flux decline can be compensated by an increase in operational pressure, but this option is not available for MD. Several authors investigated the effect of fouling on the operation of a single MD unit (Hausmann et al., 2013a; Moejes et al., 2015; Ramezanianpour & Sivakumar, 2014; Tijing et al., 2014). However, the effect of fouling on the design of a MD network is yet to be investigated.

Network design implies decision making at two levels. First, the main task of the network is to reach the aimed concentration. The number of stages in series and the concentration applied in each stage are decision variables to reach this aim. Low concentrations in the stages imply a high flux and a lower fouling rate and thus increasing the number of stages will be beneficial. However, a too high number of stages results in a higher membrane surface (and thus investments) and higher energy costs for fluid recirculation over a stage, and therefore there is an optimum in the number of stages. Secondly, fouling in the MD unit results in a serious decline of the product flow, which is not accepted if the product is directly further processed in a dryer or other installation. To remove fouling and to guarantee microbial safety, the installation must be cleaned at regular time intervals during which the production is interrupted. To keep a constant product flow and to minimise the interruptions of the operation, the parallel modules in each stage are operated in an operation-cleaning schedule. I.e. most modules are active in the operation, while others are being cleaned. This approach needs extra membrane surface per stage which raises the costs of the operation (D'Souza & Mawson, 2005). Also, due to the simultaneous stop and start of fouled and cleaned modules the product flow and concentration vary. The challenge is to design a schedule that limits the variation in product flow and concentration at low operational costs. To solve such complex problem a numerical simulation model is used. The model is based on mass and energy balances, and the best available experimental results from literature. Assumptions in the model are evaluated by variations in the main parameters and the role of process variables is investigated by effect analysis. With the effect analysis the strengths, weaknesses a potential of the system are qualified. Zhu et al. (1997) approached this challenge for the design of a RO network and maintenance schedule for sea water desalination. The main differences with the current MD network design is that the RO flux was maintained constant over the operational period by increasing the pressure. Moreover, the operational window for cleaning was in the order of 50-100 days instead of 8-12 hours, which is needed for concentrating liquid food streams because of the stronger fouling rates and to prevent unacceptable growth of micro-organisms. This work presents a two-step approach whereby first the number of stages and membrane surface with the resulting concentrations in the succeeding RO and MD stages are obtained by mixed-integer non-linear optimisation (MINLP). Secondly the scheduling problem is solved, finding the optimal number of parallel modules in each stage, by minimizing the total annual costs in combination with constrained variation of the product concentration and flow rate.

2 Process models

2.1 Membrane distillation

Unlike the extensive literature available for RO process models, MD only recently gained more interest especially as desalination technique. Most models are based on DCMD, however, because of the internal heat recovery the AGMD system is investigated in this study. The overall schematic representation of the AGMD module is shown in Figure 1. The membrane unit itself consists of a hot feed channel (hot side), hydrophobic membrane (dashed line), air gap, condensation plate (solid line), and the cooling channel (cold side). Furthermore, the module consists of a mixer, splitter, two heat exchangers, and pumps.

Fresh product feed is mixed with the recirculation flow, and the mixture is cooled to a fixed temperature before entering the cold side, in order to realise sufficient driving force over the membrane. On the other side the product is heated to a set temperature. The product flow from the heater enters the feed channel (hot side) of the membrane unit and water evaporates through the membrane, as depicted in Figure 1. The water vapour condenses at the wall of the air gap (solid line) due to the lower temperature in the cooling channel. The released heat of condensation results in an increase in product temperature in the cold side. The concentrated product from the membrane unit is partly recirculated to obtain sufficient crossflow in the membrane unit, which enhances heat transfer and reduces fouling. The other part of the concentrate is fed to next stage.

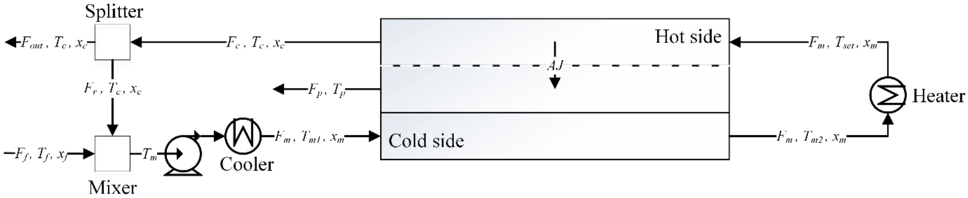

Figure 1. Schematic overview of a single AGMD module.

2.1.1 Mass transfer

In the mixer the feed is mixed with the recirculated concentrate. The general mass and energy balances over the MD module are:

$$F_m = F_f + F_r \tag{1}$$

$$F_m x_m = F_f x_f + F_r x_c \tag{2}$$

$$F_m c_{p,m} T_m = F_f c_{p,f} T_f + F_r c_{p,r} T_c \tag{3}$$

$$F_r = N_{MD} A_{channel} v_{lin} \rho \tag{4}$$

where F is the mass flowrate and x the concentration of solids, c_p the heat capacity of the flow, T the temperature of the flow, and subscripts m, f, r are denoting mix, feed, and recirculation loop respectively. The recirculation flow (F_r) is dependent on the linear flow velocity (v_{lin}), the number of parallel membrane units (N_{MD}), the cross-sectional area of the membrane channel ($A_{channel}$), and the density of the milk (ρ). The mass balance over the membrane unit itself is given by:

$$F_m x_m = F_c x_c \tag{5}$$

$$F_f = F_{out} + F_p = F_{out} + JA \tag{6}$$

where J is the water flux through the membrane and A the membrane surface area. According to Hausmann et al. (2013b) the retention rate of MD for dairy components ranges from 99 to 100%, therefore no component losses via the permeate are assumed in this work.

The water flux is based on the difference between the vapour pressures at the feed (P_f) and the condensing layer (P_{cl}), and the overall resistance. The vapour pressure is calculated based on the saturated vapour pressure, temperature and mole fraction of water.

$$J = \frac{P_f - P_{cl}}{R_{fl} + R_{mem} + R_{ag}} \tag{7}$$

The overall resistance consists of the resistance of the fouling layer (R_{fl}), the membrane (R_{mem}), and the air gap (R_{ag}). Figure 2 gives an overview of the different layers. The fouling resistance is discussed in section 2.1.3. Membrane resistance is described by the combined Knudsen and molecular diffusion model. The membrane and air gap resistance are based on the work of Drioli et al. (2015) and Hausmann (2014). The width of the air gap decreases with the increase of the condensing layer towards the outlet of the module, however, in this work the condensing layer is assumed to be equal over the whole length of the module.

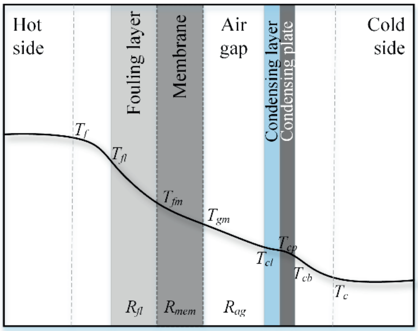

Figure 2. Schematic overview of the different resistance layers and temperature profile in the AGMD module.

General characteristics of milk like viscosity, density and specific heat capacity are influenced by the concentration and temperature. Viscosity estimations are based on Fernández-Martín (Fernández-Martín, 1972), equations for density and specific heat capacity are both taken from Choi et al. (Choi & Okos, 1986).

2.1.2 Heat transfer

The vapour pressure, and thus the flux, relies on the temperature (T_{fm}) at the membrane surface and condensing layer (T_{cl}) interface. The interfacial temperatures are calculated based on the overall heat transfer. Assuming the MD module operates at steady state without heat losses to the surroundings, the heat transfer per square meter of membrane equals:

$$\Delta Q = h_{bf}(T_f - T_{fl}) = \frac{\lambda_{fl}}{\delta_{fl}}(T_{fl} - T_{fm}) = J\Delta H_v + \frac{\lambda_{mem}}{\delta_{mem}}(T_{fm} - T_{gm}) = \frac{\lambda_{ag}}{\delta_{ag}}(T_{gm} - T_{cl}) + J\Delta H_v = \frac{\lambda_{cl}}{\delta_{cl}}(T_{cl} - T_{cp}) = \frac{\lambda_{cp}}{\delta_{cp}}(T_{cp} - T_{cb}) \tag{8}$$

in which ΔQ is the amount of transferred heat, T the temperatures at different locations, h is the heat transfer coefficient, δ the thickness, λ the conductivity of the specific layer, and ΔH_v the heat of evaporation. The different layers are visualised in Figure 2. Parameters and variables used are listed in Table A.1.

Since both the flux and the interfacial temperatures depend on each other, an iterative model is used to calculate the interfacial temperatures. The vapour flux is from the feed channel to the air gap. The energy required in the heater (Q_{heat}) and the energy transferred in the cooler (Q_{cool}) is given by the following energy balances.

$$Q_{heat} = F_m c_{p,m}(T_{set} - T_{m2}) \tag{9}$$

$$Q_{cool} = F_m c_{p,m}(T_{m1} - T_m) \tag{10}$$

in which T_{set} is the set operating temperature of the membrane module at the inlet of the hot side, T_m is the temperature after the mixer, and T_{m1} and T_{m2} are the temperature of the product flow at the in- and outlet of the cold side, respectively. T_{set} and T_{m1} are controlling parameters and fixed in the operational conditions.

Electrical energy, required for the pumps, is based on the size (F_m) and density (ρ_m) of the stream, the pressure drop over the system (ΔP_{drop}), and the energy efficiency of the pump (η_{pump}).

$$E_{pump} = \frac{F_m \Delta P_{drop}}{\eta_{pump}\rho_m} \tag{11}$$

2.1.3 Fouling model MD

Deposition of product components on the membrane over time results in a gradual increase of resistance for mass and energy transfer over the membrane. According to Hausmann et al. (2013a) the fouling mechanism of skim milk in membrane distillation relies on the interaction between milk proteins, caseins, and salts, which form a gel like layer. However, Tijing et al. (2014) pointed out, that the mechanism of fouling in membrane distillation is not yet extensively studied and as well understood as for pressure driven membrane processes. As fouling has a major impact on flux decline, and thus process performance, it is of importance to be included in process design and simulation.

A homogeneous fouling layer on top of the membrane is formed during the concentration of skim milk by MD, and to a lesser extent by adhesion inside the pores (Hausmann et al., 2013a). The formation of the fouling resistance can, therefore, be described by a cake filtration or gel layer model (Field, 2010). The linear relationship between the fouling resistance and the thickness of the fouling layer results in the following equation (van Boxtel, 1991), here expressed in mass flow, where the original was proposed in volumetric flow.

$$\frac{dR_{fl}}{dt} = \frac{\epsilon_{fl} c_b}{\rho} J - \epsilon_{fl} c_b k_f \ln\left(\frac{c_{fl}}{c_b}\right) \tag{12}$$

in which ϵ_{fl} is a constant for the resistance per unit of fouling layer thickness, c_b and c_{fl} are the concentration in the bulk and the fouling layer respectively, k_f is a mass transfer coefficient, and ρ the density. As we aim to study the effects of different levels of fouling on the organisation of the membrane system, and not to reveal the mechanism, the parameters in equation (12) are lumped, as suggested by van Boxtel et al. (1991). This results in the following semi-empirical equation:

$$\frac{dR_{fl}}{dt} = aJc_b - b \tag{13}$$

in which a and b are the lumped parameters and must be estimated from experimental data. Due to the lack of experimental data for the concentration of dairy or food products by AGMD, data for the concentration of skimmed milk by DCMD from Hausmann et all (2014) was used to estimate these constants. Data validation is listed in Appendix A.2. The thickness of the fouling layer (δ_{fl}) is estimated by a linear relationship to the fouling resistance (van Boxtel et al., 1991).

$$\delta_{fl} = \frac{R_{fl}(t)}{\epsilon_{fl}} \tag{14}$$

2.2 Reverse osmosis

The RO system (see Figure 3) consists of a high-pressure pump to pressurise the incoming feed to the desired operating pressure. Inside the apparatus the concentrate is to a large extend recirculated and mixed with the incoming feed to achieve high concentration factors and to have sufficient flow rate to prevent concentration polarisation. After mixing the feed and recirculation flow a booster pump will provide the extra pressure that was lost over the module and to ensure operating pressure is maintained. After passing the module the concentrate is split into a recycle flow and a concentrate flow which is fed to the next stage.

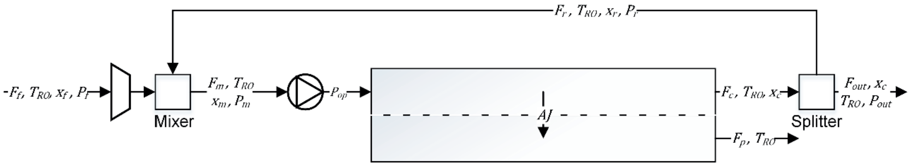

Figure 3. Schematic overview of the reverse osmosis modules.

2.2.1 Mass and heat transfer

The mathematical framework of the described system is based on the descriptions given in (Evangelista, 1985; Zhu, 1997). It is assumed that the feed flow (F_f) and the recirculation flow (F_r) are constant, and the recirculation flow is a fixed fraction (r_{RO}) of the feed flow.

$$F_m = F_f + F_r \tag{15}$$

$$F_m x_m = F_f x_f + F_r x_c \tag{16}$$

$$F_r = \frac{F_f}{1/r_{RO} - 1} \tag{17}$$

$$F_p = AJ = F_f - F_{out} \tag{18}$$

where A is the membrane area and J is the flux.

The flux in a RO unit is based on the pressure difference over the membrane and the overall resistance (R_{ov}). In this study it is assumed that there are no losses through the membrane.

$$J = \frac{P_{op} - P_p - \pi}{R_{ov}} \tag{19}$$

where P_{op} ,P_p and π are the feed pressure, the pressure at permeate side, and the osmotic pressure respectively. To guarantee a constant flux over time, the operating pressure is increased from 40 MPa to a maximum of 70 MPa to compensate for extra resistance due to fouling (Alnouri & Linke, 2012; Zhu, 1997). The osmotic pressure (π) is calculated as follows:

$$\pi = c_{mol,f} R_{gas} (T_{RO} + 273) \tag{20}$$

where $c_{mol,f}$ is the molar concentration of the feed, R_{gas} the universal gas constant, and T the temperature.

R_{ov} is the overall resistance consisting of the intrinsic membrane resistance (R_{mem}), the start-up resistance (R_p) and the fouling resistance (R_{fl}).

$$R_{ov} = R_{mem} + R_p + R_{fl} \tag{21}$$

$$R_{mem} = \frac{1}{D_w \gamma} \tag{22}$$

where D_w is the water permeability, and γ is a variable encompassing the membrane characteristics derived from Zhu (1997).

The energy requirements for a RO unit are based on the electrical energy used by the pumps. The energy usage of the high-pressure pump is:

$$E_{hp,pump} = \frac{F}{\eta_{hp} \rho} (P_{op} - P_{in}) \tag{23}$$

2.2.2 Fouling model RO

The used fouling model for RO is the same as for MD (Equation (12)). The vapour pressure difference used in the MD model as driving force is, however, replaced by the pressure difference over the membrane ($\Delta P = P_{op} - P_p - \pi$). The constants in the model are estimated by fitting the model to published data (Duclos-Orsello, Li, & Ho, 2006; van Boxtel et al., 1991). The parameters used for the RO model are given in Table A.1.

3 Approach and problem formulation

The combined model equations of the membrane system (Eq. 1-22) are non-linear, and the number of units are integer variables. Therefore, the optimisation of the network configuration of RO and MD is a mixed integer non-linear problem (MINLP). A downside of these problems is the complexity and the required computational time. The optimisation problem is, therefore, split into two parts: 1) the estimation of the optimal number of RO and MD modules in series (*N*) and their respective total membrane surfaces, and 2) the scheduling problem where the optimal number of parallel units (*M*) and scheduling strategy is derived. Figure 4 gives an example of the possible membrane network.

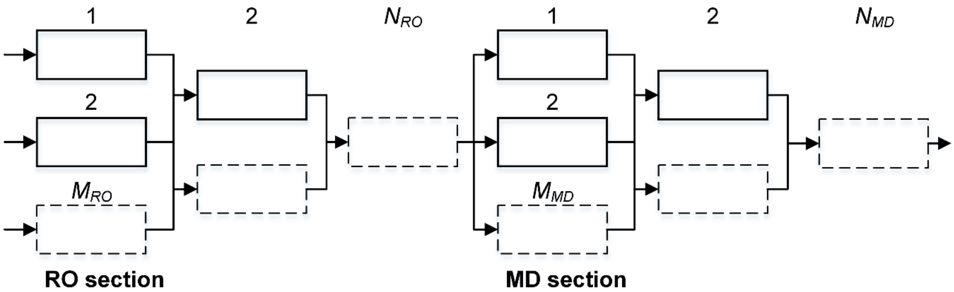

Figure 4. Schematic representation of a RO and MD network. N_{RO}, and N_{MD} the number of membrane stages in series for RO and MD, and M_{RO}, and M_{MD} the number of membrane unit in parallel for each RO and MD stage.

3.1 Stage optimisation

Each potential stage is considered as one large membrane module for which the total surface is estimated. The decision variable is the membrane surface in each stage, which results in a specific product concentration after each stage. The objective is to design a network with the lowest investment and operational costs that realizes a given final product concentration for a given feed rate. The objective function is formulated as:

$$min \left(\sum_{n=1}^{N_{RO}} C_{RO,inv}^{n} + \sum_{n=1}^{N_{RO}} C_{RO,op}^{n} + \sum_{n=1}^{N_{MD}} C_{MD,inv}^{n} + \sum_{n=1}^{N_{MD}} C_{MD,op}^{n} \right) \tag{24}$$

s.t. $c_{goal} \leq \bar{c}_{out,(n=N)}$

$Equations\ 1 - 23$

where C_{inv} are the investment costs and C_{op} are the operational costs for each stage n for the total number of stages N for both RO and MD. The operational conditions and process boundaries are listed in Table A.2.

The investment costs for RO consist of the equipment costs of the pumps, and the RO module which consists of the module costs ($C_{RO,mod}$) and the membrane costs ($C_{RO,mem}$), which both are linearly related to the surface membrane surface (A). The installation costs are covered by a Lang factor (L_f). The total costs are annualised by the life time of the equipment (LF) and the life time of the membranes LF_m. Subsequently all costs are expressed in euro per m^3 water removed.

$$C_{RO,inv} = \frac{\left(\frac{C_{pump,n}}{LF_{pump}} + \frac{A_n C_{RO,mod}}{LF_{mod}}\right) L_f + \frac{A_n C_{RO,mem}}{LF_{mem}}}{F_{p,a}} \tag{25}$$

The operational costs for RO contain the electrical cost for the pumps and the cleaning costs, both are annualized. Cleaning costs depend on the membrane surface (A_n) and the total cleaning time (t_{clean}).

$$C_{RO,op} = \frac{(E_{electric,n} C_e t_a + C_{clean} A_n)}{F_{p,a}} \tag{26}$$

Furthermore, the concentration of the last stage ($\bar{c}_{out}$) should not be lower than the set concentration (c_{goal}). The final concentration for RO is a decision variable but is limited to a final concentration of 18%. Output parameters (flow, concentration, and temperature) of the last stage of RO are the input parameters of the first stage of the MD section.

The MD investment costs and operational costs are formulated similar as for RO but contain additional components. In addition, the investment costs include the heat exchangers for heating and cooling. The membrane costs for MD are calculated in the same way as for RO. The operational costs also include the heating and cooling for every MD module, which results in the following equations.

$$C_{MD,inv} = \frac{\left(\frac{C_{pump,n} + C_{heater,n} + C_{cooler,n} + A_n C_{MD,mod}}{LF_{mod}}\right) L_f + \frac{A_n C_{MD,mem}}{LF_{mem}}}{F_{p,a}} \tag{27}$$

$$C_{MD,op} = \frac{\left(E_{electric,n} C_e + E_{heat,n} C_{heat} + E_{cool,n} C_{cool}\right) t_a + C_{clean} A_{tot} t_{clean}}{F_{p,a}} \tag{28}$$

MD is proposed as an alternative for multi-stage evaporation of milk, therefore, the final concentration of the last MD stage is fixed at 50% total solids. The resulting configuration is used as input for the scheduling optimisation. Figure 5 illustrates the total optimisation procedure, whereby Figure 5 part I represents the stage optimisation. To solve the series problem the fmincon function of MATLAB R2017b with the interior point method algorithm was used.

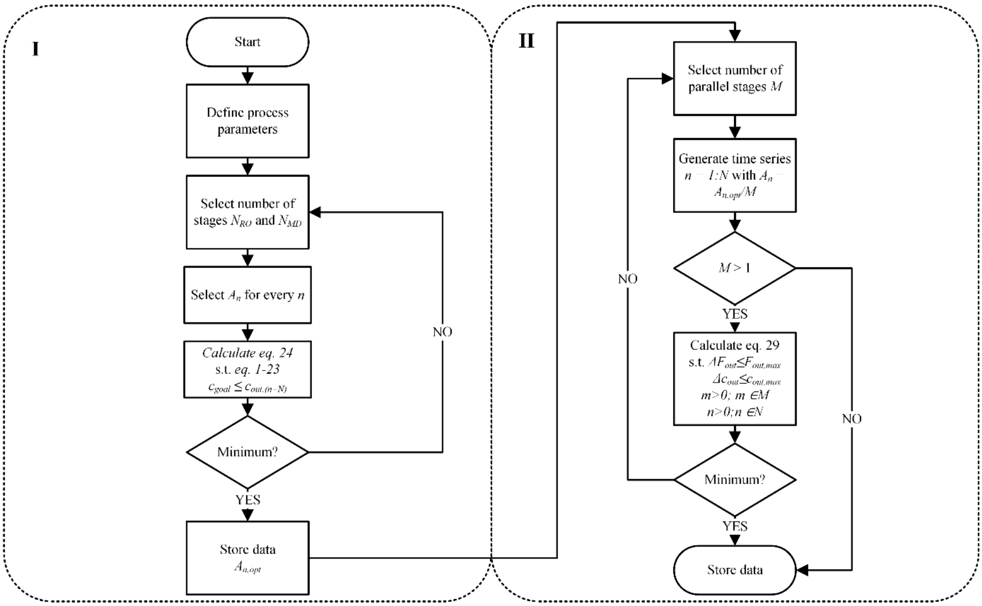

Figure 5. Solution strategy for an optimal membrane design. With I) the selection of the optimal number and area of RO and MD units in series, and II) the strategy for the scheduling problem and the selection of number of parallel units.

3.2 Scheduling strategy

The optimal scheduling strategy is based on the number of parallel units (M) and aims to guarantee a continuous production while minimizing the costs. For the economical evaluation of the different configurations the total costs are minimized. These consists of the annualized investment costs (C_{inv}) and the annual operational costs (C_{op}) of both the RO (C_{RO}) and the MD section (C_{md}). The operational conditions and process boundaries are listed in Table A.2.

$$min\left(\sum_{n=1}^{N_{RO}}\sum_{m=1}^{M_{RO}} C_{RO,inv}^{n,m} + \sum_{n=1}^{N_{RO}}\sum_{m=1}^{M_{RO}} C_{RO,op}^{n,m} + \sum_{n=1}^{N_{MD}}\sum_{m=1}^{M_{MD}} C_{md,inv}^{n,m} + \sum_{n=1}^{N_{MD}}\sum_{m=1}^{M_{MD}} C_{md,op}^{n,m}\right) \tag{29}$$

$$s.t.\ \Delta F_{out}^{n,m} \leq \Delta F_{out,max}$$

$$\Delta c_{out,nm}^{nm} \leq \Delta c_{out,max}$$

$$m > 0;\ m \in M$$

$$n > 0; n \in N$$

$$Equations\ 1-23$$

The next step in milk processing is spray drying, a unit operation that requires a constant feed rate and product concentration. Therefore, fluctuations in flow rate and concentrations have to be limited. For the RO section the deviation in outflow of every stage ($\Delta F_{out,max}$) may not be larger than 10%. In the MD section the final concentration will fluctuate due to the flux decline over time, therefore, the variation in final concentration ($\Delta c_{out,max}$) is restricted to 1.5% over the whole production period. These were set as constraints in the minimisation problem.

The investments costs consist of the cost of the membrane units, pumps and heat exchangers, which depends on the number of stages (N) and parallel units (M). The investment costs are annualized and corrected with a Lang factor (L_f) (Lu, Hu, Xu, & Wu, 2006).

$$C_{md,inv} = \frac{\frac{\left(C_{pump,mn} + C_{heater,mn} + C_{cooler,mn} + A_{mn}C_{md,mod}\right)}{LF_{mod}} L_f + \frac{A_n C_{md,mem}}{LF_{mem}}}{F_{p,a}} \tag{30}$$

$$C_{RO,inv} = \frac{\left(\frac{\left(C_{pump,mn} + C_{md,mn} + A_{mn}C_{RO,mod}\right)}{LF_{mod}} L_f + \frac{A_{mn}C_{RO,mem}}{LF_{mem}}\right)}{F_{p,a}} \tag{31}$$

The costs for the membrane distillation unit (C_{md}) consists of the membrane module (C_{mod}) and the membrane (C_{mem}) itself which all depend on the membrane area (A). The investment costs for the RO section are calculated in the same way, only without the heater.

The operational costs are based on the costs for electricity, heating, cooling, and cleaning. The cleaning consists of both thermal energy (E_{clean}) for cleaning and the material costs (C_{clean}), and depend on the number of operational hours (t_{op}).

$$C_{MD,op} = \frac{\left(E_{electric,mn}C_e + E_{clean,mn}C_{heat} + E_{heat,mn}C_{heat} + E_{cold,mn}C_{cold}\right)t_{op} + C_{clean}A_{tot}t_{clean}}{F_{p,a}} \tag{32}$$

$$C_{RO,op} = \frac{\left(E_{electric,mn}C_e + E_{clean,mn}C_{heat}\right)t_{op} + C_{clean}A_{tot}t_{clean}}{F_{p,a}} \tag{33}$$

Other auxiliary equipment, maintenance, and labour costs are not considered. To solve the scheduling problem the pattern search method of MATLAB R2017b was used.

For estimation of the number of parallel modules and the best scheduling strategy it was assumed that all modules have fixed production cycles of 7 hours followed by a 1-hour cleaning cycle, this to guarantee food safety. Furthermore, the membranes will operate at the same initial performance after every cleaning cycle. Additionally, it was assumed that the modules operate after cleaning immediately at steady-state, and the operating conditions of each parallel unit in the same stage are identical. Figure 5 part II shows the solution strategy. Data generated in the series configuration section is used as input for generating the time series which are the input for the scheduling problem. All cost parameters are listed in Table A.3. The effect of the usage of waste heat on the total costs are evaluated in additional optimisations, as well as the effect of the operational conditions on the total performance.

4 Results and discussion

4.1 Process design

The optimal process configuration to concentrate milk from 0.09 kg kg^{-1} to 0.5 kg kg^{-1} solids is by a two-stage RO section and a single-stage MD section. The optimal process configurations for the RO and MD section are shown in Figure 6, and details are displayed in Table 1. RO proved to be more cost efficient compared to MD. Milk is, therefore, concentrated by RO to the upper boundary of 0.18 kg kg^{-1} solids. A two-stage RO configuration is optimal for this case, which both consist of six parallel units. The energy consumption of the RO section resulted in 19 kJ per kg water removed, which is in line with reported values in literature (Ramirez et al., 2006).

Table 1. Results for the optimal total system with specifications of the configuration and performance of the RO and MD sections. RO concentrates milk from 9% to 18% dry matter and MD from 18 to 50% dry matter.

		Total system	**RO section**	**MD section**
Feed	tonne h^{-1}	25	25	12.5
Total membrane area	m^2	5300	1416	3884
Number of series	-	2 – 1	2	1
Number of parallel units in subsequent stages	-	6 – 6 – 4	6 - 6	4
Heating costs	€ m^{-3}	3.2	-	8.3
Cooling costs	€ m^{-3}	2.3	-	6.0
Electrical costs	€ m^{-3}	1.1	0.6	1.7
Equipment costs	€ m^{-3}	0.9	0.2	2.1
Cleaning costs	€ m^{-3}	0.5	0.2	1.0
Total costs	€ m^{-3}	8.1	1.0	19.1

Figure 6 shows the configuration for the RO and MD stages with operational conditions. In the figure the optimal MD configuration with one stage is given. Alternative, not optimal, MD configurations with operational conditions are given in appendix A.3.

The costs for the optimal RO and MD configurations and the combination of the two (total system) are listed in Table 1. To reach the end concentration of 0.5 kg kg^{-1} solids a single stage MD turned out to be best in terms of costs. This result was counter-intuitive as the single stage system operates at a high concentration with a low flux and stronger fouling compared to a multi-stage system. No advantage is taken from the higher fluxes and lower fouling rate in the first stages of a multi-stage system (see Figure 7 for a specification of the fluxes in subsequent stages). The required membrane area and thus investment costs of this single-stage system are therefore higher than that of a multi-stage MD system. Details of multi-stage MD systems are listed in Appendix A.3. The membrane area is, however, not the main cost driver, but the heating and cooling costs are (Table 1). Due to the required recirculation in each of the subsequent stages, to keep sufficient cross flow along the membrane surface, the increase of heating and cooling costs is larger than the reduction of the capital costs. Moreover, the flux decline over time plays an important role in the costs. Due to the flux decline the internal heat recovery decreases and as a consequence more heating and cooling is required in the recirculation loop during operation. Altogether, the costs of a two-stage system are 16% above those of a single-stage MD system.

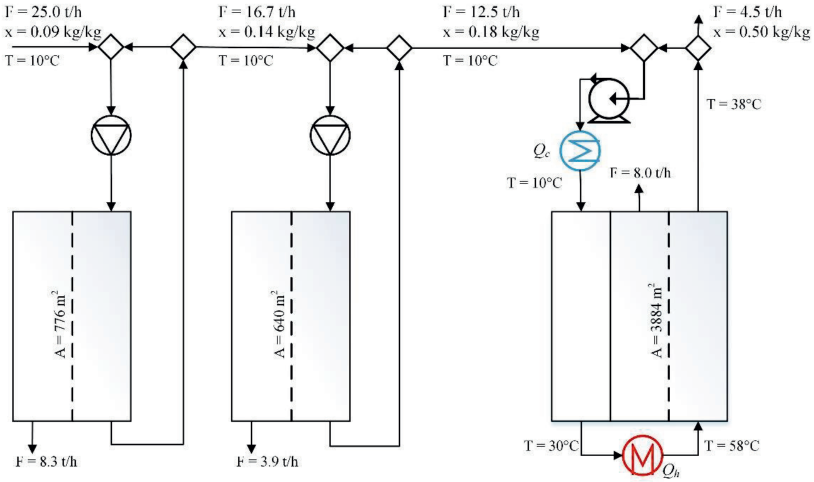

Figure 6. Optimal process configurations for the combined RO (first 2 stages) and MD (3rd stage) system, including average flows, concentrations, and temperatures.

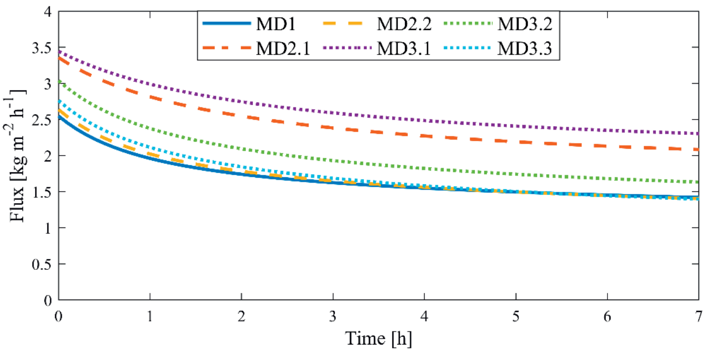

Figure 7. Flux profile over time in the different MD stages. Single stage: MD1, a two-stage: MD2 and a three-stage system: MD3.

Concentration of food products like milk by MD has the advantage of reaching a high final concentration by using a membrane system. However, the energy consumption is high compared to concentration by RO. For milk concentration, besides heating also cooling of the recirculation loop is necessary in order to maintain the driving force. The energy required for a single stage system is 2.6 MJ for heating and 2.5 MJ per kilogram water removed for cooling, which is much higher compared to previously reported values for MD on desalination (Duong et al., 2016; González et al., 2017; Koschikowski et al., 2009; Kuipers et al., 2014; Mar Camacho et al., 2013) and traditional multi-stage evaporator systems. Cooling is not required for desalination, as the permeate is the aimed product, and not the concentrate which is aimed for food products like milk. Reported costs values for desalination range between 0.3 and 5.1 euro per m^3 water removed (Elsayed et al., 2014; González-Bravo et al., 2015; Meindersma, Guijt, & de Haan, 2006). For milk the costs for MD are estimated at nearly 20 euro per m^3 water removed. However, when combining MD and RO the total costs are 8.1 euro per m^3 water removed. This is still higher when compared to reported desalination values but does include the effect of fouling and the additional energy costs caused by the high recirculation and for cooling.

Scheduling for the RO part is based on keeping the milk outflow within the fixed boundaries. This in order to minimise fluctuation in flow to the next unit, to guarantee continuous operation. The flux, and thus milk concentration, is kept constant by increasing the operating pressure over time. Due to the fixed 7 hours up and 1 hour down schedule a fluctuation is visible in the out flow, the more parallel units the smaller this difference will be. For RO in both stages six parallel modules resulted in the optimal solution. The cleaning period of all parallel modules is spaced out equally, which results in an outflow pattern as depicted in Figure 8.

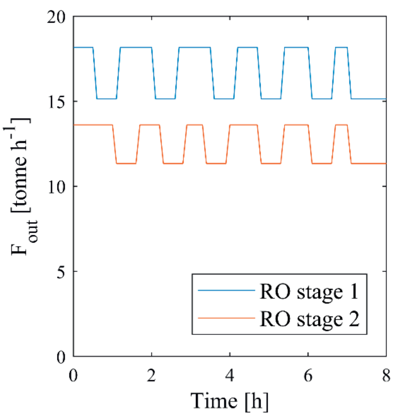

Figure 8. Effect of scheduling on the product out flow for a two stage RO system. At the high values all modules are in operation, at the low values one module is in the cleaning mode.

For the MD, four parallel modules for one stage system are enough to keep the final concentration and flow within the pre-defined operational boundaries (Figure 9). When comparing the profile of the MD to that of the RO, the effect of flux decline over time in MD is clearly visible (Figure 9b). The product outflow increases over time as the permeate flow decreases as a result of the flux decline (the recirculation and feed flow are kept constant) until the next cleaning cycle.

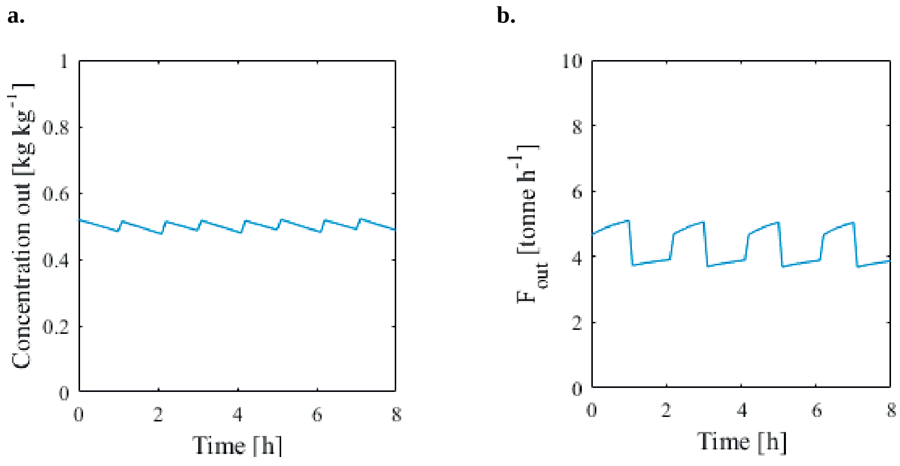

Figure 9. Effect of scheduling on MD for the product concentration (a) and product flow (b) of a one stage system. The gradual decline of concentration and increase of flow is result of the fouling.

4.2 Effect of fouling rate

Previous studies on MD featured a significantly lower fouling rate (Lu et al., 2006; See, Vassiliadis, & Wilson, 1999), or did not include the fouling dynamics at all (González-Bravo et al., 2015). Fouling, however, plays an important role in milk concentration by membrane processes. Although the fouling dynamics used in this study is based on assumptions, it gives an insight on the effect of fouling on the process configurations. To illustrate the effect of fouling, scenarios were simulated by varying the fouling rate (parameters a and b in Eq. (13)). The results for the one-stage MD system are given in Figure 10. All operating conditions were kept equal to previous simulations. At low fouling rates both the equipment costs and the utility costs decrease. There is no effect on the cleaning schedule.

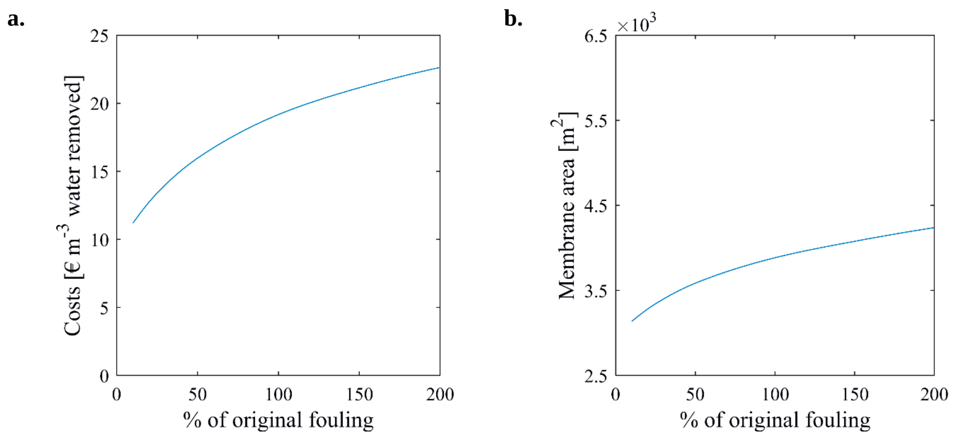

Figure 10. The effect of variation fouling for a one-stage MD system (100% standard fouling, 50% half of fouling rate, 200% doubled fouling rate) on the costs (a) and required membrane area (b).

With flux decline over time, the temperature change of the product in both the hot and the cold side decreases. As a result, the heating and cooling duties, which are the main cost drivers, increase. The heating and cooling duties overshadow the capital related costs. Reduction of the heating and cooling costs by a low fouling rate and keeping high fluxes over time is therefore crucial to make membrane distillation viable.

4.3 Influence of operational conditions on MD performance

Standard values for operational conditions were used for the discussed optimisation of the RO and MD network. These conditions, however, affect the outcomes. Variations of the key operational conditions and membrane properties, like temperature and recirculation settings, give information on the role of the operational conditions for further system improvement.

4.3.1 Effect of operating temperature

Heating and cooling demand are the main cost contributors in MD usage for the concentrating milk. Unlike MD for desalination, where the permeate is the main product (Meindersma et al., 2006), cooling is required in the product recirculation. In previous calculations, the temperature of the cold side was set at 10°C and the hot side temperature was set to 58°C. The effect of varying these temperatures on the costs and membrane surface is shown in Figure 11.

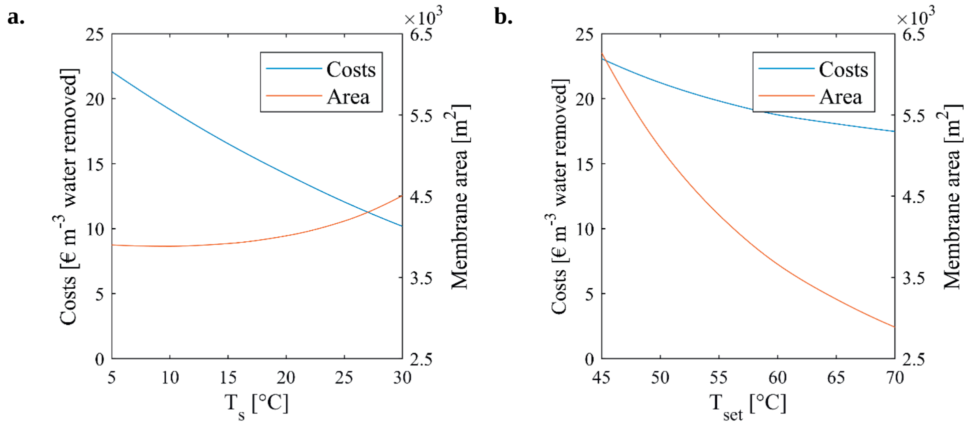

Figure 11. Variation analysis of temperature setpoints for a single-stage MD system. a) Effect of the cold side temperature, T_s, on the processing costs (left axis) and membrane surface (right axis). b) Effect of the hot side temperature, T_{set} , at the processing costs (left axis) and membrane surface (right axis).

Increasing the feed temperature (T_S) from 10 to 20°C reduces the cooling costs. The total membrane surface increases, due to the smaller temperature difference between the hot and cold side. Equipment costs have, compared to the heating and cooling, a small contribution to the costs. The costs decrease by 30% with a change of the feed temperature from 10 to 20°C on the cold side. It should be noted that raising the temperature may increase the risk of microbial contamination.

In previous calculations, the temperature of the hot side (T_{set}) was set to 58°C. Increasing the temperature results in a higher flux and reduces the required membrane area (Figure 11b). As a result, the costs drop due to the higher fluxes and increased heat transfer from the hot to the cold side. The hot side must be as warm as possible, the only limiting factor is the product quality. Temperatures over 60°C for a prolonged time are not desirable for milk due to protein denaturation (de Wit & Klarenbeek, 1984).

4.3.2 Effect of linear flow velocity

The product is recirculated over each module to ensure sufficient crossflow along the membrane. In the system optimisation the linear flow velocity was set to 0.05 ms^{-1}. To evaluate the effect on process performance the linear flow velocity was varied between 0.025 and 0.2 ms^{-1}. Increasing the velocity reduces the membrane surface due to higher fluxes (see Figure 12b). The same result was found by Hausmann et al. (2014), by showing that higher flow velocities have a positive effect on the flux which results in a smaller membrane surface. An extra advantage of high flow velocities is a lower fouling rate and less flux decline over time. This aspect is also a factor to reduce the required membrane surface. In contrast, Figure 12a gives the effect of varying the linear velocity on the costs, which decrease almost linear towards lower velocities. Lowering the flow velocity also reduces the recirculation rate and consequently the cooling and heating costs. Although also the flux reduces and thus heat transfer from the hot to cold side, the increase in recirculation rate causes a higher energy increase. These calculations do not fully cover the turbulence properties at low velocities. This is a strong assumption, but the results point to the importance of spacer optimisation to reduce the operational costs. Additionally, at higher solids concentrations higher cross flows might be desired, because of the increased viscosity and the shear thinning behaviour of milk (Morison, Phelan, & Bloore, 2013).

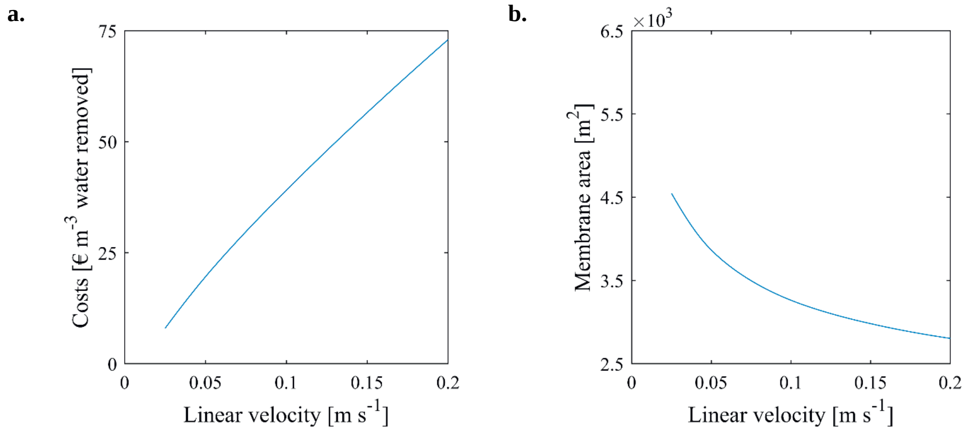

Figure 12. Effect of linear velocity on the costs (a) and required membrane area (b) for a single-stage MD system.

4.4 Use of waste heat

MD operates at a relative low operating temperature and, in contrast to multistage evaporators, MD can make use of low-quality energy streams. In food processing plants low-quality energy streams are often abundantly available (Papapetrou, Kosmadakis, Cipollina, La Commare, & Micale, 2018). The costs of the high heating demand for MD are reduced by using these low-quality energy streams. Several authors already exploited the potential of membrane distillation in combination with industrial waste heat (Elsayed et al., 2014; Hausmann et al., 2012; Meindersma et al., 2006). Dow et al. (2016) demonstrated the feasibility of operating a MD pilot plant by using waste heat from a gas fired power station. The temperature of the waste heat (less than 40°C was used) had a major influence on the flux of a direct contact membrane distillation unit. Also solar heat has potential as a heat source for membrane distillation (Chang, Wang, Chen, Li, & Chang, 2010). Higher waste heat temperatures result, as expected, in higher fluxes. Figure 11b illustrates the effect of varying set temperature on the required membrane surface and thus capital costs. More important is, however, the reduction of the costs for cooling and heating by using waste heat. Figure 13 gives the operational costs for water removal for different levels waste heat usage, ranging from zero to full replacement of the thermal energy demand by waste heat. Complete energy supply from waste heat, by heat integration with other processes, results in operational costs of 3.1 euro per m^3 water removed. This makes membrane distillation competitive with current concentration techniques. To reach these benefits, additional capital costs are required for waste heat integration. These costs are very case dependent, and will therefore have to be assessed case by case.

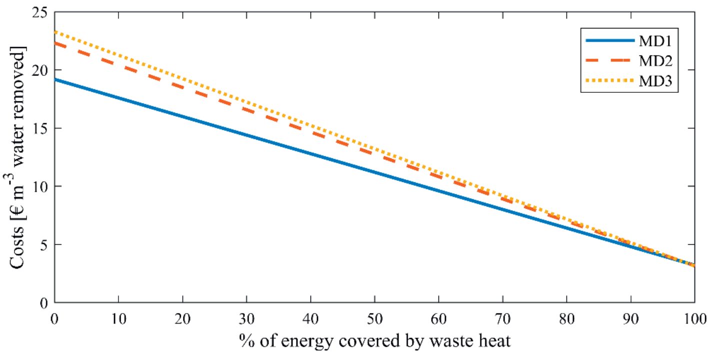

Figure 13. Operational costs as a function of % of required energy for heating and cooling covered by waste heat for MD installations with 1, 2 and 3 stages.

4.5 Membrane distillation system

Both the type of membrane distillation configuration, and the membrane specifications have an influence on its performance. Simulations were based on literature values for physical constants (heat and transfer coefficients and condensation layer thickness, see Table A1). Variations in the physical constants was not proven to effect on the optimisation results. In the previous sections the effect of variation of the most important other assumed variables (temperature, flow rate, fouling) was discussed. Moreover, the membrane properties used in this study are also based on reported literature. Experimental work is required to confirm these findings, and improve the used values for the membrane properties. Halving the membrane resistance (R_m) will, for example, increase the flux and will give a boost to the reduction of the heating and cooling duties. Both heating and cooling costs decrease with 18% for a one stage MD system, when the membrane resistance is halved. In order to achieve this, development and testing of new membranes is required.

The advantage of internal energy recovery in air gap membrane distillation (AGMD) was the reason to select this MD system for this work. Low fluxes and high recirculation rates proved to limit the internal heat recovery. In this light, direct contact membrane distillation (DCMD) could be a better option. In the AMGD the average temperature difference over the hot side is for the one-stage system 20°C (milk cools down from 58 to 38°C) while the temperature difference over the heater and cooler is 28°C (Figure 6). In this case it is more energy efficient to separate the heating and cooling circuited like in a DCMD. However, if the temperature difference of the hot side is larger compared to temperature difference over the heater, then the AGMD will be beneficial. These results point to the importance for higher fluxes to make the AGMD system viable for milk concentration.

Compared to permeate- or liquid gap membrane distillation (PGMD), the AMGD has a smaller temperature difference as driving force and thus lower flux. According to Swaminathan et al. (2016) the PGMD is 20% more energy efficient compared to AGMD. However, due to the liquid on the permeate side there is a higher chance of pore wetting (Drioli et al., 2015), which highly decreases the process performance and thus results in higher costs. Nonetheless, exploring PGMD as option for the concentration of food products is of interest.

5 Conclusion

Membrane distillation (MD) is an emerging technology for product concentration. In this work the potential of different process configurations for the concentration of milk by reverse osmosis (RO) and membrane distillation was assessed and investigated. Although milk was considered as feed, the findings of this work give also important information for application of MD for concentrating of other food products.

Due to the low costs, concentration of milk starts with RO to the maximal possible concentration of milk (18% solids). RO is followed by membrane distillation to concentrate milk to the final 50% solids. The used air gap membrane distillation (AGMD) has the advantage of internal heat recovery and is therefore often preferred over direct contact membrane distillation. Nevertheless, due to the high product recirculation to achieve sufficient cross flow along the membranes the energy costs of the AGMD unit are high. With the current available membranes and energy prices membrane distillation cannot compete with a multi-stage evaporator.

Gradual fouling during the operation has a large influence on process cost of MD, as fluxes decline so does heat transfer. Heating and cooling of product in each stage results in costs that overshadow the costs for membranes and equipment. The optimal configuration of the membrane distillation unit is therefore a single-stage unit that operates at high concentration and low flux. The effect analysis showed the following options for further improvement of the system:1) to increase the cold and hot side temperatures to their maximum acceptable values, 2) to develop spacers that allow lower cross flow velocities in the system and thus lower recirculation rates, and 3) make use of available waste heat.

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Nomenclature

A	Area (m^2)
a	Fouling rate parameter (Pa m^4 s kg^{-2})
b	Fouling rate parameter (Pa m^2 kg^{-1})
C	Costs (€ yr^{-1})
c	Concentration (kg kg^{-1})
c_{mol}	Molar concentration (mol m^{-3})
c_p	Specific heat capacity (J kg^{-1} K^{-1})
D_w	Membrane water permeability (kg s^{-1} N^{-1})
E	Energy requirement (J h^{-1})
F	Mass flow (kg s^{-1})
ΔH_v	Latent heat of evaporation (J kg^{-1})
h	Heat transfer coefficient (W m^{-2} K^{-1})
J	Water flux (kg m^{-2} s^{-1})
k_f	Mass transfer coefficient (m s^{-1})
L_f	Lang factor (-)
LF	Equipment life time (yr)
M	Number of modules in parallel (-)
N	Number of membrane stages (-)
P	Pressure (Pa)
Q	Heat flow (J s^{-1})
ΔQ_m	Heat transfer within the membrane (J m^{-2} s^{-1})
R	Mass transfer resistance (Pa s m^2 kg^{-1})
R_{gas}	Universal gas constant (J K^{-1} mol^{-1})
r_{RO}	Fixed recirculation fraction in RO module (-)
T	Temperature (°C)
t	Time (s)
v_{lin}	Linear velocity (m s^{-1})
x	Weight fraction (-)

Greek letters

η_{pump}	Energy efficiency of the pump (-)
γ	Variable encompassing the membrane characteristics (-)
δ	Thickness (m)
λ	Conductivity (W m^{-1} K^{-1})
π	Osmotic pressure (Pa)
ρ	Density (kg m^{-3})
ϵ	Constant for the resistance per unit of fouling layer thickness (Pa s m kg^{-1})
μ	Viscosity (Pa s^{-1})

Subscripts

a	Annual
ag	Air gap
b	Bulk
c	Concentrate
cb	Condensing plate – bulk

cl	Condensing layer
cp	Condensing plate
f	Feed
fl	Fouling layer
fm	Fouling – membrane
gm	Air gap – membrane
inv	Investment
m	Mix of feed and recirculated product
mem	Membrane
mod	Membrane module
op	Operational
ov	Overall
p	Permeate
r	Recirculation
s	Cold side variable
set	Hot side variable

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Appendix

A.1 Process data

Table A.1 Membrane specific process parameters.

Membrane specifications	**Value**
RO spiral wound	
Water permeability (kg s^{-1} N^{-1})	8×10^{-10}
Fixed recirculation fraction in RO module	0.95
Membrane resistance (Pa s m^{-1})	1.27×10^{12}
Start-up resistance (Pa s m^{-1})	2.13×10^{10}
MD flat sheets	
Air gap width (m)	1×10^{-3}
Condensation layer thickness (m)	1×10^{-4}
Membrane thickness (m)	6×10^{-5}
Condensation plate thickness (m)	6×10^{-5}
Global mass transfer coefficient (m)	5×10^{-7}
Cross sectional area channel (m2)	1.5×10^{-3}
Heat transfer coefficient membrane (J s^{-1} m^{-2} K^{-1})	385
Resistance per unit of fouling layer (Pa s m^{-1})	8×10^{-11}
Membrane resistance (Pa s m^{-1})	1.6×10^{5}
Thermal conductivity condensation layer (W m^{-1} $°C^{-1}$)	0.58
Thermal conductivity condensation plate (W m^{-1} $°C^{-1}$)	24
Thermal conductivity membrane (W m^{-1} $°C^{-1}$)	1.2
Thermal conductivity fouling layer (W m^{-1} $°C^{-1}$)	0.23
Thermal conductivity air (W m^{-1} $°C^{-1}$)	0.027
Latent heat of evaporation (J kg^{-1})	2257×10^{-3}
Gas constant (J K^{-1} mol^{-1})	8.314

Table A.2 Operating and optimisation conditions for both RO and MD process.

Parameter	**RO**	**MD**
Starting temperature feed (°C)	10	10
Temperature permeate (°C)	10	-
Pressure feed (Pa)	40×10^{5}	10^{5}
Pressure drop over the module (Pa)	0.22×10^{5}	0.2×10^{5}
Pressure permeate (Pa)	10^{5}	-
Starting pressure feed (Pa)	10^{5}	-
Feed flow (kg h^{-1})	25000	25000 – 12500
Starting concentration (w/w)	0.09	≤ 0.18
Final concentration (w/w)	≤ 0.18	0.50
Linear velocity (m s^{-1})	2	0.049
Annual operating time (h)	8000	8000
Cleaning cycle time (h)	1	1
Operating cycle time (h)	7	7
Hourly feed (tonne h^{-1})	25	12.5
Number of stages, N (-)	1 – 5	1 – 5
Equipment life time (year)	15	15
Membrane life time (year)	4	4

Table A.3 Economic data.

Parameter	Value
Investment cost pump (€)	$2590(P_{pump})^{0.79}$
Pump efficiency	0.85
Investment cost heater/cooler (€)	$1115F_{tot}$
Lang factor	1.4
Equipment lifetime (y)	15
MD module costs (€ m^{-2})	58.5
MD membrane costs (€ m^{-2})	100
RO module costs (€ m^{-2})	58.5
RO membrane costs (€ m^{-2})	17.75
Heating costs (€ GJ^{-1})	4.0
Cooling costs (€ GJ^{-1})	3.0
Electrical costs (€ kWh^{-1})	0.12
Cleaning cost (€m^{-2} $hour^{-1}$)	0.017

A.2 MD fouling model validation

In order to include fouling in the MD model, an estimate for the fouling resistance was made. Figure A.1 shows the fitting of the lumped parameters a and b for the estimation of the fouling resistance (R_f) as given in Eq. (13). Data from Hausmann et al (2014) was used the estimate the lumped parameters a and b. Initial fouling build up is well fitted as can be seen in the comparison between the simulation and the data in the figure below. When the fouling layer build up stabilises (roughly after 8 hours) the flux is underestimated for low concentrations (20%) and overestimated at high concentrations (40%).

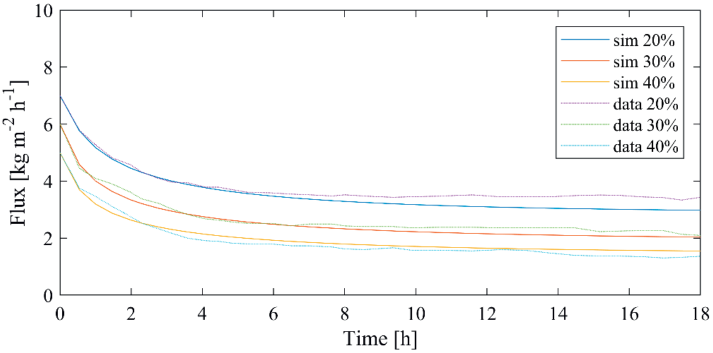

Figure A.1. Result of the fitting of the flux at different concentrations 20, 30 and 40% dry matter. The dotted lines are the actual data (data) (Hausmann et al., 2014) and the solid lines are the fitted simulations (sim).

The resistance over the air gap is based on the molecular diffusion model (Drioli et al., 2015):

$$R_{ag} = \left(\frac{\epsilon D P_t}{\delta_{ag} P_{ag,log}} \frac{M_v}{R(T_{ag,avg} + 273.15)}\right)^{-1} \tag{A.1}$$

where $T_{ag,avg}$ is the mean temperature in the air gap, $P_{ag,log}$ is the log mean pressure in the gap, P_t is the total pressure, ϵ is the membrane porosity, M_v the molar mass of water molecules, D is the water vapor diffusion coefficient through air.

A.3 Alternative MD configurations

In this section the results are presented for the other membrane distillation configurations.

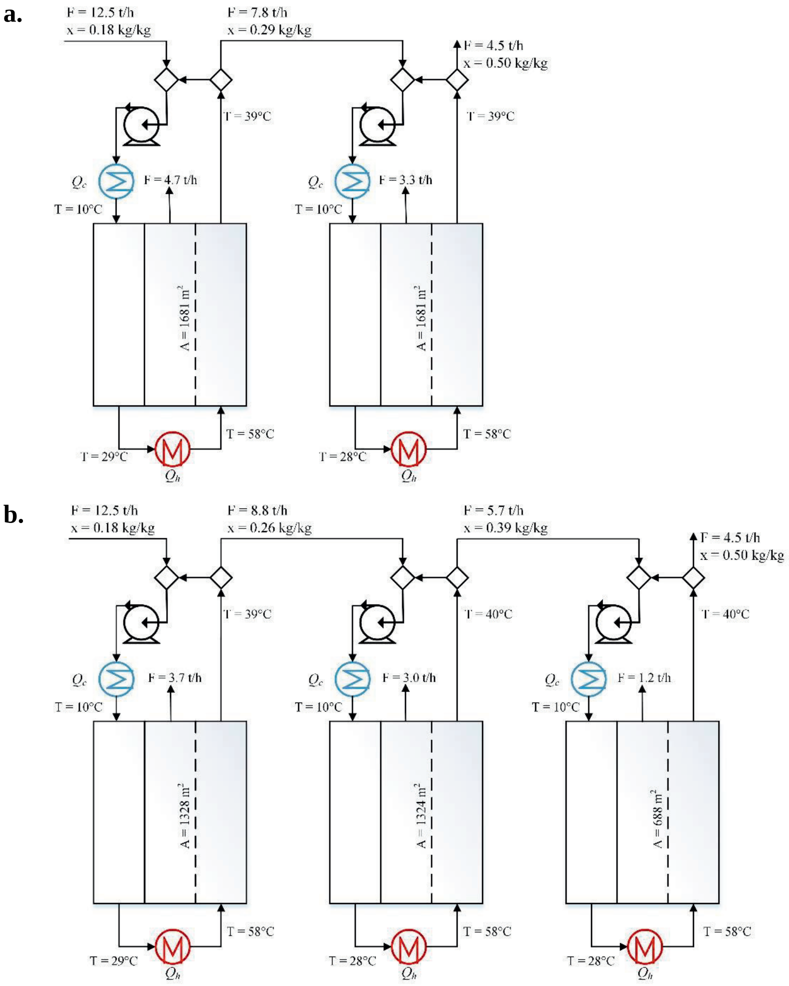

Figure A.2. Optimal process configurations for the MD configuration with 2 (a) and 3 (b) stages, including average flows, concentrations, and temperatures.

Table A. 4. Results for other MD configurations with 2, 3, 4, or 5 stages in series.

		MD 2	**MD 3**	**MD 4**	**MD 5**
Feed	tonne h^{-1}	12.5	12.5	12.5	12.5
Total membrane area	m^2	3448	3340	3329	3299
Number of stages	-	2	3	4	5
Number of parallel units in subsequent stages	-	3-3	3-3-3	3-3-3-3	3-3-3-3-3
Heating costs	€ m^{-3}	10.3	10.9	10.9	10.8
Cooling costs	€ m^{-3}	7.2	7.5	7.3	7.1
Electrical costs	€ m^{-3}	2.0	2.1	2.1	2.2
Equipment costs	€ m^{-3}	1.9	1.9	1.9	1.9
Cleaning costs	€ m^{-3}	0.9	0.9	0.9	0.9
Total costs	€ m^{-3}	22.3	23.2	23.1	22.9

Chapter 5

Towards optimal milk powder production by early stage life cycle assessment

Abstract

The environmental impact of industrial processes must be reduced in order to face the global climate challenge. Innovative technologies have a key role in this task. The environmental impact of a new, or existing process, is often assessed in an LCA study after completion of the design. In this work LCA was incorporated in the early stage of process design for a new milk powder production chain. By combining conventional and innovative technologies in a superstructure, all potential processing scenarios were optimised with respect to environmental and economic impact. Most promising process configurations consist of reverse osmosis combined with multi-stage evaporation and a closed-loop spray drying system with a zeolite sorption system. The surplus heat recovered in the drying section should be optimally utilised. Heat integration is, therefore, essential in the reduction of both environmental impact and costs. The energy consumption proved to be dominant in both the operational costs and environmental impact. Hence, process designs for milk powder production which are optimised with respect to energy consumption meet both minimise environmental and economic impact.

Keywords: LCA, milk powder, multi-objective optimisation, process design, superstructure.

1 Introduction

Milk powder production is energy intensive process (Ramirez, Patel, & Blok, 2006). In order to save energy and lower the environmental impact, alternative technologies need to be investigated. Promising innovative technologies are radio frequency heating as an alternative for pasteurisation with steam, and reverse osmosis or membrane distillation to concentrate milk instead of using evaporators. Furthermore, monodisperse droplet drying combined with heat recovery by an adsorbent wheel or membrane contactor is an alternative for the current spray drying system (Moejes & van Boxtel, 2017). Traditionally, process design is driven by product requirements and processing costs, but with the need for sustainable solutions, environmental impact is gaining equal importance. Redesigning of the milk powder production process with innovative technologies must therefore be based on both environmental and economic impacts while meeting product quality standards.

In current practice, first a process design resulting in a choice for a processing system consisting of a combination of unit operations and a set of specifications for the operational conditions is made. After that, a life cycle assessment (LCA) is performed of the selected processing system to evaluate the process' environmental impact. LCA identifies the hot spots in the production chain, and results in an identification of adjustments needed to improve the process. This iterative work flow is repeated until the decision maker is satisfied with the results. This approach may lead to a sub-optimal design in terms of LCA (Azapagic, Millington, & Collett, 2006). Potentially only marginal improvements will be achieved by adapting process conditions, instead of considering alternative processing pathways or using other alternative solutions.

Incorporating LCA aspects in the initial phase of process design can circumvent or automate the iterative loop. Such an approach requests for a different work flow and a multi-objective optimisation approach whereby both economic and the different LCA impacts are minimised, while satisfying the production and product constraints (Burgess & Brennan, 2001). Several authors have applied this approach for chemical processes (Azapagic & Clift, 1999a; Brunet, Reyes-Labarta, Guillén-Gosálbez, Jiménez, & Boer, 2012). In this paper we will use an integrated approach by combining environmental and economic impact while satisfying product requirement, for the selection of innovative technologies to redesign the milk powder production process.

2 Combined work flow for process design and LCA

For both process design and LCA workflows are well defined in a specified number of steps. The steps in early stage process design are: 1) definition of production goals, 2) definition of the required functions to achieve the production goals, 3) definition of all flows of materials, energy and kinetics, 4) selection of unit operations, 5) derive operational conditions by process simulation, 6) interpretation and evaluation of the design and improve, and 7) finalise the design by documentation (Chen & Shonnard, 2004). The resulting design from this phase is subsequently subject to detailed design, engineering and construction. This procedure for early stage process design is iterative over the steps 3 to 6, whereby the choices are reassessed and where alternative solutions and operational conditions are considered.

The LCA workflow consists of the following four steps: 1) goal and scope definition, 2) inventory analysis, 3) impact assessment, and 4) the interpretation. LCA is a well-established and internationally recognised way to assess the environmental impact of a product, process, or activity, stipulated in an ISO standard (Bauman & Tillman, 2012). A main weakness of the LCA methodology is that the required data is gathered after completion of the process design to compare the impact of different processing conditions (Azapagic et al., 2006). The procedure does not help to generate better processing pathways, for instance to make choices between emerging technologies. In order to find optimal process configurations in terms of environmental impact, LCA aspects should be included in the iterative loop of the early process design. Although a few studies proposed such an approach for chemical processes (Azapagic & Clift, 1999b; Brunet, Cortés, Guillén-Gosálbez, Jiménez, & Boer, 2012; Guillén-Gosálbez, Caballero, & Jiménez, 2008), LCA is not integrated in the design procedures for food processing systems; it is only used to assess available process designs.

The work flows for combined process design and LCA consist of the following steps:

1. Simultaneous definition of the system and formulation of the production objectives with respect to the product properties, LCA, and economics.
2. Definition of process functions and selection of all possible unit operations for the required functions. Development of a superstructure for selecting the unit operations. Set-up models for mass and energy balances of all unit operations together with collecting impact inventory data of input-output streams of each unit operation.
3. Apply simultaneous process simulation and impact assessment. Apply multi-objective optimisation for the selection of unit operations and operational conditions for all objectives.
4. Joint interpretation of process performance and LCA impact.

The combined activities 1 to 4 results in a forward work flow (Figure 1) where the interpretation and improvement of the process design and impact assessment is automated. In step 3, multi-objective optimisation (MOO) is applied to the multiple LCA criteria, the economic objectives and the product requirements.

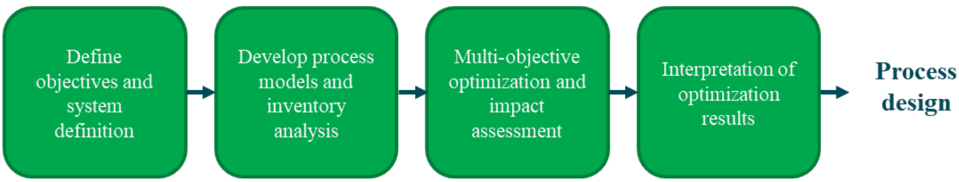

Figure 1. Work flow for integrated LCA and process design steps.

In the selection of emerging technologies, it is essential that process operations are not a priori rejected. Therefore, in step 2 of the work flow, all potential unit operations that can satisfy the process functions are combined in a superstructure. The path through the network and operational conditions to be applied in this superstructure are obtained by mixed integer non-linear programming (MINLP) multi-objective optimisation or by optimisation of each individual path in the superstructure (Kocis & Grossmann, 1989; Yeomans & Grossmann, 1999).

2.1 Objective and system definition

In this study we focus on the processing part of raw milk to powder, this is a gate-to-gate approach. The system starts with the transfer of raw milk to the first processing unit and ends with the delivery of dried product from the dryer. The processing steps consist of a pre-treatment, a concentration step, a drying step, and air treatment step. The inputs to the system are the milk and utilities used. Outputs are, besides the milk powder, the cream, emissions to the air, waste water and the other effluents. The system boundaries are illustrated in Figure 2.

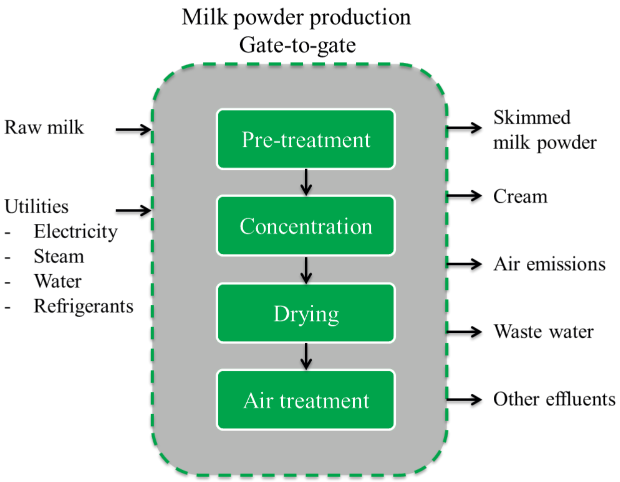

Figure 2. Overview of the system boundaries, inputs and outputs, and the main processes of a milk powder production plant.

Besides defining the system boundaries, it is essential to define a functional unit in order to express the objective function in quantitative terms (Bauman & Tillman, 2012). The functional unit used is one ton of skimmed milk powder with a moisture content of 3.5%. The starting material is raw milk, and for the production of skimmed milk powder cream is separated. This means that two products are produced, cream and skimmed milk powder. As cream is separated after the first operation, the main impact and costs concern the powder production. Therefore, it was decided to allocate the impacts fully to the skimmed milk powder. Moreover, costs which are similar for each system (labour, management and buildings) are excluded from the cost calculations.

The objective for the process design is the production of skimmed milk powder (from here on referred to as milk powder) at a capacity of 10 tonne of raw milk per hour during 8000 hours a year. The production should be efficient, both in terms of production costs that arise from investments and utilities, as well as the environmental impact of the production process.

2.2 Process models and inventory analysis

The following step in the combined LCA and process design approach consisted of three activities: 1) superstructure construction, 2) modelling of all processes, and 3) data collection.

2.2.1 Superstructure construction

Within the given boundaries a superstructure of the system was constructed. A superstructure is a network representation of all possible production routes (Kocis & Grossmann, 1989; Yeomans & Grossmann, 1999). It is essential to identify unit operations, here all emerging technologies, for each process function and to define the flows between the different unit operations. During superstructure optimisation, the potential costs and LCA impacts of all possible production routes are evaluated. The superstructure for milk powder production was based on the review of conventional and emerging technologies given by (Moejes & van Boxtel, 2017), and is shown in Figure 3.

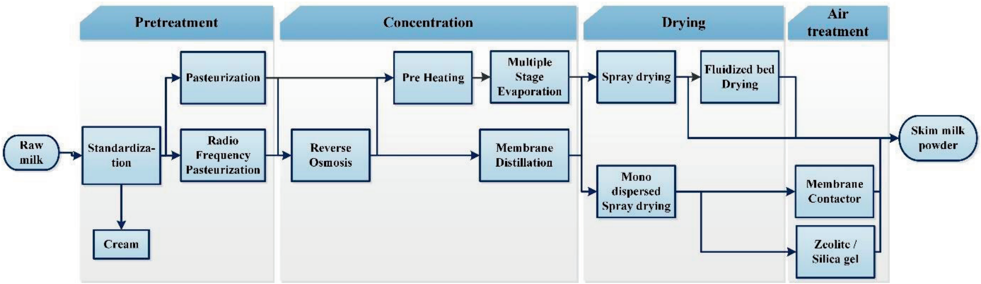

Figure 3. Superstructure representation of the milk powder process, with all possible production routes using conventional and emerging technologies.

2.2.2 Process models and decision variables

For each unit operation mass and energy balance models were used (see Appendix 1). All these models have the same in- and outputs in order to link the unit operations in the superstructure and to automate route searching. The models, which are linked to the operational conditions, are modelled in MATLAB 2017b. The choice for the operational conditions affects the performance of a production chain, the production costs, the energy consumption and environmental impact. Therefore, operational conditions were also used as decision variables (listed in Appendix 2). Moreover, upper and lower boundary constraints were set on the decision variables, and on conditions to which the product can be exposed to guaranty product quality. For example, the inlet air temperature for the spray dryer was constrained at 180 and 220°C.

2.2.3 Data collection

Process simulation results in the definition of all internal process flows, and in- and output of materials and energy. These inputs and outputs were linked to the LCA data for the inputs and outputs. Moreover, data on the performance and costs of the unit operations was collected from literature and engineering databases (listed in Appendix 1 and 4).

The individual environmental impacts for the different material and energy inputs and outputs were based on the food database of GaBi 6 and are listed in Appendix 3. To assess the economic impact, reference costs for both utilities and equipment were defined, see Appendix 4. Data collection is essential for the usability of the results, and is case depended.

2.3 Multi-objective optimisation and impact assessment

2.3.1 Multi-objective optimisation

For sustainable processes, a low environmental impact and a high economic performance has to be achieved. These objectives are combined in a multi-objective optimisation (MOO) problem. Two most used MOO methods are the weighted-sum method, and the ϵ-constraint method. The weighted-sum method combines multiple objectives into one objective function, by assigning weights to each objective (Marler & Arora, 2010). The ϵ-constraint method minimises one of the objective functions, while the other objective function(s) are incorporated in the constraint section of the model and limited to a maximum value (ϵ_j).

The strong influence of the assigned weights on the obtained solutions is the main disadvantage of the weighted-sum method. This drawback does not exists for the ϵ-constraint method as a set of solutions are generated (Mavrotas, 2009). The optimal solutions that represent the trade-off between objectives are visualised in Pareto plots. The optimisation problem is formulated as:

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$$
\begin{aligned}
& min\, f_j(x) && (1)\\
s.t.\quad & f_i(x) \leq \epsilon_j \\
& c(x) \leq 0 \\
& c_{eq}(x) = 0 \\
& Ax \leq b \\
& A_{eq}x = b_{eq} \\
& lower\ bound\ \leq x \leq upper\ bound \\
& x \in \mathbb{R}
\end{aligned}
$$

where $f_j(x)$ and $f_i(x)$ are the objective functions. In this study the objective functions are: 1) the environmental impact and 2) the economic impact. The objective functions are affected by the decision variables x (i.e. the operational variables). The operational variables are process or product related, and an example in this case is to what concentration the milk is concentrated by reverse osmosis prior to evaporation. Another example is the temperature at which the dryer operates. To minimise the objective value(s), a set of constraints should be satisfied. c represents the non-linear inequality and equality constraints on the decision variables x. A and b concern the linear equality and inequality constraints. Examples of constraints are the production target, and process limitations. The decision variables x are restricted by its lower and upper bounds. These are mainly set to ensure the product properties and quality, for example to make sure that the product is not exposed too high temperatures. A list of all decision variables and constraints is given in Appendix 2. Besides optimisation of the operational conditions, all possible routes from the superstructure were evaluated. The models were developed in MATLAB 2017b.

2.3.2 LCA impact assessment

After defining the goal and scope the inventory analysis of the system will result in a large amount of data on amounts of resources used and pollutant emissions related to the functional unit (e.g. 1 tonne of milk powder). The inventory results are characterised by sub groups, so called impact categories. LCA has multiple impact categories and using them all together with the cost objective and product constraints, would lead to a complex problem. For this reason, the different impact categories are combined into a single impact. Combing them into a single impact is achieved by normalizing and assigning weights to the results. To go from the inventory results into a single impact can be summarised into the following steps: classification, characterisation, normalisation, and weighting (Bauman & Tillman, 2012). These steps are summarised in Figure 4, and discussed in further detail in the next sections.

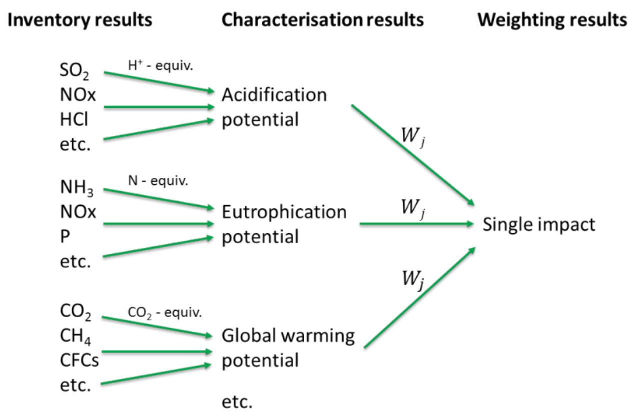

Figure 4. Step wise aggregation of information in LCA. From inventory results to a single impact by weighting the characterisation results with factor W_j per impact category j (adapted from Bauman and Tillman (2012)).

Classification and characterisation

The LCA impact categories relevant for the system were selected (characterised) (see Table 1), and values were assigned to each energy and material in- and outputs defined in the superstructure from the GaBi database. The impact categories were based on the inventory analysis given by Taxiarchou et al. (2015). The impact for all categories for each production route was derived from the size of in- and output streams and their contributions to the impact categories:

$$I_{i,j} = \sum_{x} R_{i,x,j} \cdot I_{x,j} \tag{2}$$

in which I is the impact of category j for processing route i in the superstructure, and x is the resources type. R is the amount of resources used during production. For each route the total environmental impact of each impact category was estimated.

Table 1. Selected impact categories with their units and the applied European normalisation factors (N_j) from Sleeswijk et al. (2008), * marked values are taken from (Benini et al., 2014)**. Weighting factors** (W_j) **are based on Huppes et al. (2012).**

Impact category	Unit	Normalisation factor	Weighting factor
Global Warming Potential (GWP)	kg CO_2-eq.	4.49E+12	28
Ozone Layer Depletion Potential (ODP)	kg R11-eq.	6.79E+06	5
Particulate Matter Formation (PMF)	kg PM10-eq.	8.12E+09	8
Acidification Potential (AP)	kg SO_2-eq.	2.84E+10	5
Resource depletion, mineral and fossil (ARD)	kg Sb eq.	7.23E+11	8
Ecotoxicity	CTUe	6.80E+12 *	13
Human Toxicity (carcinogenic)	CTUh	2.63E+05 *	7
Human Toxicity (non-carcinogenic)	CTUh	4.42E+05 *	5
Photochemical Oxidant Formation (POF)	kg NMOVC eq.	2.80E+10	6
Eutrophication Potential (freshwater)	kg P eq.	3.47E+08	3
Eutrophication Potential (marine)	kg N eq.	5.89E+09	3
Terrestrial Eutrophication	mole N eq.	9.04E+10 *	3
Water depletion (WD)	m^3 eq.	4.06E+10 *	6

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Normalisation

The magnitude and units of the different impact categories differ. Moreover, the importance of the impact categories varies. To compare impact categories of alternative production systems a normalisation step was applied on the impact categories. By normalising the data, the impact of each impact category is translated into a relative impact on national, regional or even global level (Bauman & Tillman, 2012). Sleeswijk et al. (2008) derived normalisation factors for an European and a global system (Sleeswijk et al., 2008). The normalisation factors refer to the reference situation of the extractions and emissions in the year 2000. Not all factors were covered by Sleeswijk et al., the missing factors were taken from Benini et al. (2014) (see Table 1).

Weighting

To compare the LCA results to the production costs of the process, the results for the impact categories were combined into a single score as expressed in Equation (3). Hereby, weighting factors were assigned to each individual impact category. The combined LCA score is defined as:

$$I_{single,i} = \sum_{j} \left(\frac{I_{i,j}}{N_j} \cdot W_j \right) \tag{3}$$

in which I_{single} is the combined result of the environmental impact for each process route i in the superstructure, and W_j is the weighting factor of impact of category j.

The weighting factor expresses the importance of an impact category relative to the others. The choice of weighting and normalisation factors has a large influence on the final score (Shen, Worrell, & Patel, 2010)). Several approaches have been developed in the past years to assign objective weighting factors, examples are the Ecoindicator99, Nogepa, BEES and EPA (Finnveden, Eldh, & Johansson, 2006; Goedkoop & Spriensma, 2001; G. Huppes et al., 2007; Lippiatt, 2007). Huppes et al. (2012) did a large analytic survey on weighting environmental impacts, combining the different available approaches. The resulting weighting factors are listed in Table 1. In contrast to the work of Huppes et al. land use and ionising radiation were not considered as relevant impact factors in our study. Whereas for eutrophication three categories were used in this study; freshwater, marine, and terrestrial eutrophication. Therefore, the weighting factor Huppes et al. assigned to eutrophication are divided equally over the three eutrophication categories used.

2.3.3 Economic impact assessment

The economic indicator used in our study is the Total Annual Costs (TAC). The TAC consists of both investment for equipment and utility costs (energy, water, etc.). For the ranking of production scenarios, the costs for labour, cleaning, laboratory and overhead are assumed to be similar for the scenarios and therefore not included. Detailed description of the economic indicator is listed in Appendix 4.

3 Results and discussion

3.1 Optimal process configurations

The superstructure resulted in 32 different process routes and a list of these scenarios is provided in Appendix Table A. 1. This number of process scenarios is still relatively low and therefore all scenarios are discussed in this section. All 32 scenarios were optimized for both the costs (TAC) and combined LCA score. Figure 5 (top) gives the optimisation results of all scenarios for both the combined LCA score and the TAC. Figure 5 (bottom) is an enlarged figure of the results for cluster I.

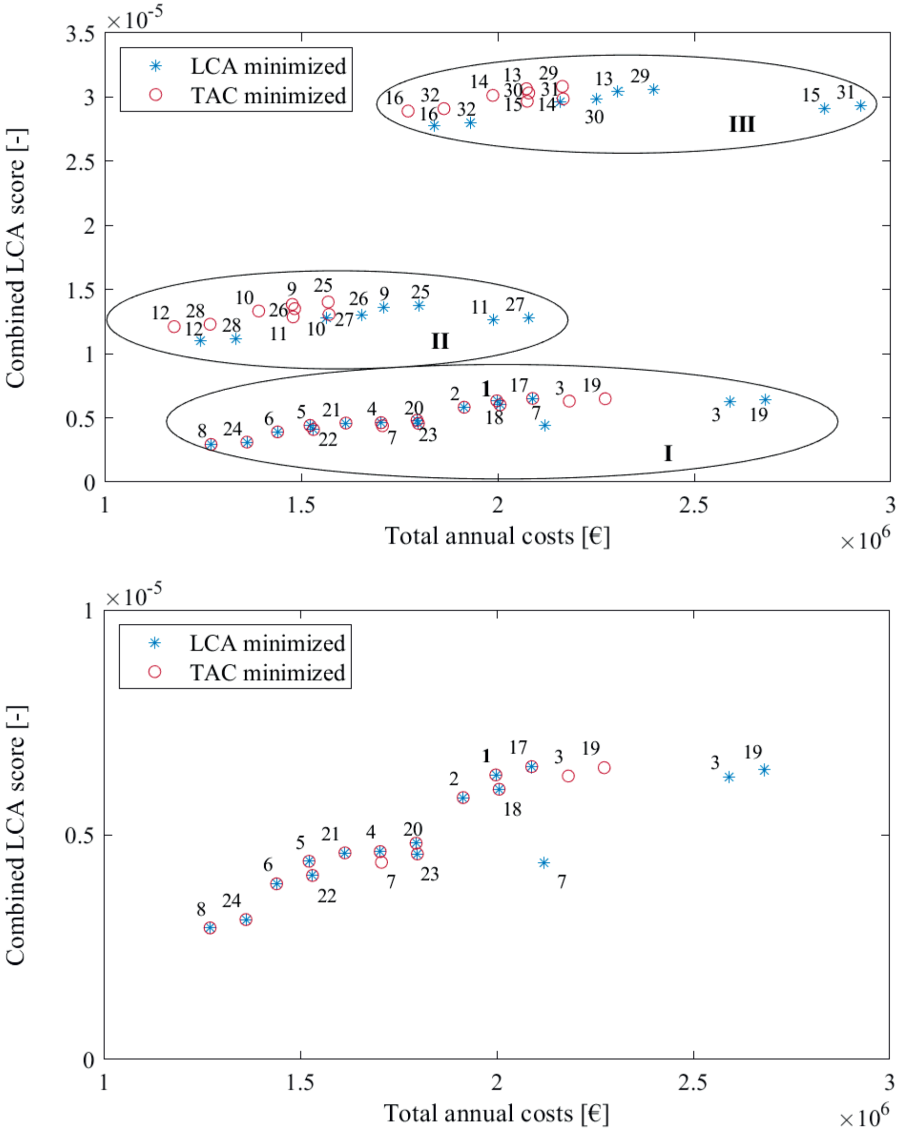

Figure 5. Combined results on minimised LCA score and TAC for all scenarios (top). 3 clusters (I, II and III) are circled. The difference between the clusters is caused by the usage of membrane distillation as concentration technology (see text below). The bottom graph is zoomed in on the results of cluster I.

Scenario 1 represents the state-of-the-art milk powder production system, and is regarded as the reference production system. It consists of centrifugation, standardisation, pasteurisation, a 7-stage evaporator, and a spray dryer. In terms of the combined LCA score, scenario 8 gives the best results. Scenario 8 differs from scenario 1 by the application of pre-concentration by reverse osmosis (RO) prior to the evaporator, and the traditional spray dryer is replaced with a monodisperse droplet dryer from which the energy is recovered with a zeolite adsorption system. The main advantage of this system is that recovered heat is applied for the steam production required for the multi-stage evaporation system. This leads to a lower total energy consumption and consequently to a low LCA score. Scenario 8 scores second best with respect to the total annual costs (TAC).

The best scenario in terms of costs is scenario 12. This system is similar to scenario 8, but the multi-stage evaporator is replaced by membrane distillation (MD). MD is a thermal driven membrane process, and has the potential to concentrate milk to higher concentrations than pressure driven membranes (Hausmann, Sanciolo, Vasiljevic, Kulozik, & Duke, 2014). The advantage of MD, compared to an evaporator, is the lower processing temperatures (around 60°C) enabling to reuse more of the surplus heat of the drying section. A drawback of MD is the required active cooling (Moejes, Wonderen, Bitter, & van Boxtel, 2019) which results in the large increase of the combined LCA score when comparing scenario 12 to 8 or the reference scenario 1.

Three clusters are marked in Figure 5 (top). The differences between these clusters are a result of the usage of MD. The process configurations in cluster I are without MD. In cluster II, MD is combined with pre-concentration by RO. This application doubles the LCA score compared to the results in cluster I. The scenarios in cluster III have LCA scores which are four times higher compared to cluster I, due to complete concentrating milk by MD. Despite the fact the heating demand for the MD unit is fully covered by waste heat from the spray dryer. The active cooling, of milk to 10°C, is responsible for the increased combined LCA score. The environmental impact of active cooling is high compared to steam usage or cooling with a ground water source (see Appendix Table A. 4).

Minimizing the combined LCA score and TAC is a multi-objective problem, which by using different weights to both objectives would result in a Pareto front. The bottom graph of Figure 5 focusses on cluster I, and shows, however, that with exception of three scenarios (7, 3, and 19), the minima for TAC and the combined LCA score (i.e. the extremes of the Pareto front) are the same. For these scenarios both objectives are dominated by the energy requirements, which is the main contributor in both costs and environmental impact. The scenarios for which the results are not the same for both objectives (scenarios 7 and 23, and 3 and 19) are processes where spray drying is combined with air dehumidification via a membrane contactor (MC). Increasing the membrane area of the membrane contactor, results in a small reduction in energy usage. For example, in scenario 19, an energy gain of 1.7% is achieved by decreasing the spray dryer temperature to 180°C, which results in an increase of the membrane surface by a tenfold. This leads to an increase of the investment costs of nearly 17.9% against a decrease of 0.7% of the LCA score. Minimizing the costs results in an inlet air temperature of 220°C, which reduces the membrane surface compared to the LCA score minimisation

Table 2 Comparison of the different scenarios to reference scenario 1. = means ± 1%, ↓ means a 1 – 50% lowering, ↓↓ means a lowering of >50%, ↑ means a 1 – 50% increase, and ↑↑ means an increase of >50% compared to reference (Ref.). The centrifugation and standardisation step are the same for all scenarios and therefore not specifically mentioned in the list.

Scenario	Process units	LCA score	TAC
1	Pasteurisation – Pre heater – Evaporator – Spray dryer	Ref.	Ref.
2	Pasteurisation – Pre heater – Evaporator – Spray dryer – FB dryer	↓	↓
3	Pasteurisation – Pre heater – Evaporator – Monodisperse dryer with MC	=	↑
4	Pasteurisation – Pre heater – Evaporator – Monodisperse dryer with zeolite	↓	↓
5	Pasteurisation – RO – Pre heater – Evaporator – Spray dryer	↓	↓
6	Pasteurisation – RO – Pre heater – Evaporator – Spray dryer – FB dryer	↓	↓
7	Pasteurisation – RO – Pre heater – Evaporator – Monodisperse dryer with MC	↓	↓
8	Pasteurisation – RO – Pre heater – Evaporator – Monodisperse dryer with zeolite	↓↓	↓
9	Pasteurisation – RO – MD – Spray dryer	↑↑	↓
10	Pasteurisation – RO – MD – Spray dryer – FB dryer	↑↑	↓
11	Pasteurisation – RO – MD – Monodisperse dryer with MC	↑↑	↓
12	Pasteurisation – RO – MD – Monodisperse dryer with zeolite	↑↑	↓
13	Pasteurisation – MD – Spray dryer	↑↑	↑
14	Pasteurisation – MD – Spray dryer – FB dryer	↑↑	=
15	Pasteurisation – MD – Monodisperse dryer with MC	↑↑	↑
16	Pasteurisation – MD – Monodisperse dryer with zeolite	↑↑	↓
17	RF pasteurisation – Pre heater – Evaporator – Spray dryer	↑	↑
18	RF Pasteurisation – Pre heater – Evaporator – Spray dryer – FB dryer	=	=
19	RF Pasteurisation – Pre heater – Evaporator – Monodisperse dryer with MC	↑	↑
20	RF Pasteurisation – Pre heater – Evaporator – Monodisperse dryer with zeolite	↓	↓
21	RF Pasteurisation – RO – Pre heater – Evaporator – Spray dryer	↓	↓
22	RF Pasteurisation – RO – Pre heater – Evaporator – Spray dryer – FB dryer	↓	↓
23	RF Pasteurisation – RO – Pre heater – Evaporator – Monodisperse dryer with MC	↓	↓
24	RF Pasteurisation – RO – Pre heater – Evaporator – Monodisp. dryer with zeolite	↓↓	↓
25	RF Pasteurisation – RO – MD – Spray dryer	↑↑	↓
26	RF Pasteurisation – RO – MD – Spray dryer – FB dryer	↑↑	↓
27	RF Pasteurisation – RO – MD – Monodisperse dryer with MC	↑↑	↓
28	RF Pasteurisation – RO – MD – Monodisperse dryer with zeolite	↑↑	↓
29	RF Pasteurisation – MD – Spray dryer	↑↑	↑
30	RF Pasteurisation – MD – Spray dryer – FB dryer	↑↑	↑
31	RF Pasteurisation – MD – Monodisperse dryer with MC	↑↑	↑
32	RF Pasteurisation – MD – Monodisperse dryer with zeolite	↑↑	↓

3.2 Most promising technologies and opportunities

A comparison of all scenarios to the reference scenario 1 is listed in Table 2. The results as depicted in Figure 5 are translated to improvements or deteriorations from scenario 1 for both the TAC and the LCA results. The largest savings, both environmentally and economic, are in the monodisperse droplet drying process with air dehumidification and heat recovery by a zeolite adsorption wheel (scenario 8 and 24). The combination with monodisperse droplet drying, to limit the number of fines in the exhaust air is essential for the heat recovery with the adsorption wheel. The zeolite wheel is currently preferred over the application of a membrane contactor (MC) for air dehumidification, because zeolite adsorption wheels perform better in terms of both costs and environmental impact. Furthermore, zeolite adsorption wheels are a proven technology and already industrially implemented (Boxtel, Boon, Deventer, & Bussmann, 2012). The MC works with a brine solution to generate a vapour pressure difference over the membrane, which facilitates mass transport of water vapour from the humid air on one side to the brine solution on the other side of the membrane. The brine solution becomes saturated and the performance of the MC will drop. Regeneration of the brine solution is therefore needed. In the current configuration the brine solution in the MC is regenerated in a two-stage evaporator. From an energy point of view, regeneration by superheated steam in a second membrane contactor has the potential to further improve the energy efficiency (Moejes, Visser, Bitter, & van Boxtel, 2018). In order to achieve this, further development of the membranes is required, and therefore for current implementation is not yet preferred.

The implementation of RO in the dairy industry is not new, and already known for a long time, however it still offers large opportunities for factories that did not yet implement RO (Poelarends, Slaghuis, & de Koning, 2009; Ramirez et al., 2006). RO is more energy efficient compared to traditional evaporators (scenario 5 versus 1), and more energy efficient compared to MD (scenario 5 versus 13). Especially with respect to the environmental impact, implementation of RO will become more important. Besides RO, the additional fluidized bed after the spray dryer is an already conventional process which improves both costs and environmental impact (scenario 2 versus 1).

The difference between scenarios 1 – 16 and 17 – 32 is the usage of normal pasteurisation (1 – 16) and pasteurisation using radio frequency (RF) heating (17 – 32). The effect of RF heating on the costs and LCA score is shown in Figure 5 and Table 2. For example, comparison of scenario 8 and 24, where the use of RF is the difference. The TAC and combined LCA score for Scenario 24 (with RF) are lower compared to scenario 8 (without RF). Therefore, the usage of RF heating for pasteurisation has a negative effect on both the combined LCA score and the costs. RF has, however, the advantage of fast and homogeneous heating, which may improve product quality and reduce fouling (Awuah, Ramaswamy, & Tang, 2014; Kudra, Voort, Raghavan, & Ramaswamy, 1991). Industrial implementation is, however, still low (Marra, Zhang, & Lyng, 2009). The main opportunity for RF is on the other hand, the current shift to electrification of industrial processing. Where electrical energy generated by fossil fuels has a high environmental impact due to the low efficiency ratio (Grubler et al., 2012), the environmental impact of electricity based on solar energy will reduce this impact (see Table A. 3).

Membrane distillation (MD) proved to be cost efficient in combination with heat recovery from the drying process and pre-concentration by RO (scenario 12). MD equipment is relatively cost efficient as no high pressure is required. The opportunities for MD increase when the fluxes improve due to better membranes (Moejes et al., 2019). Furthermore, the combined LCA score is highly dominated by the usage of active cooling. By increasing the cold side temperature cooling with ground water becomes feasible and the environmental impact will decrease.

3.3 Evaluation of the environmental impact

The individual environmental impact categories of the scenarios of cluster 1 are plotted in Figure 6. In some this radar plot some distinction can be observed between the different scenarios for the different impact categories. Scenario 8 (bottom left in Figure 5) scores well in all impact categories, where scenario 19 performs the worst, which could also be concluded from Figure 5 (bottom). For most impact categories the difference between the scenarios is relatively equally distributed, except for the categories water depletion (WD) and eutrophication (freshwater). For these two categories the scenarios are grouped into two groups. Difference between these two groups is the use of RO as pre-concentration step, and is a direct result of the energy reduction.

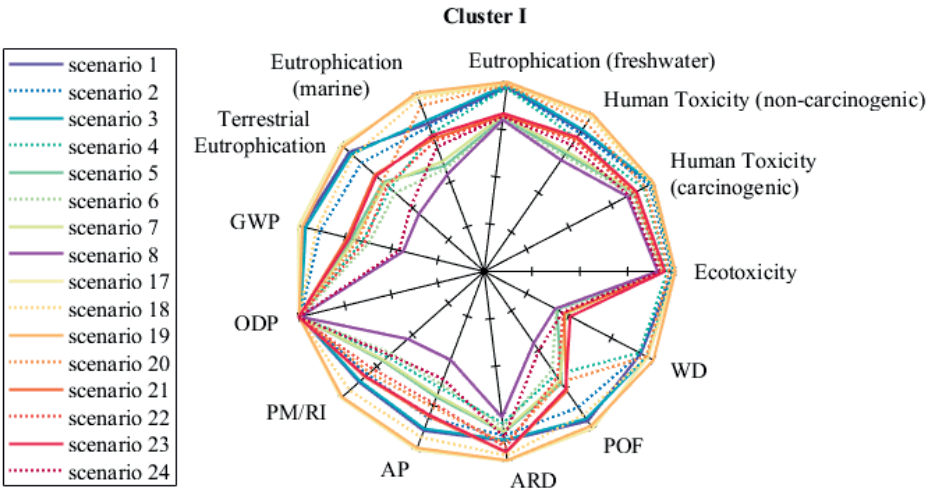

Figure 6. Radar plot of the environmental impact (scaled to the maximal value) per category for each scenario of cluster I, with minimizing the combined LCA score as objective. Global Warming Potential (GWP), Ozone Layer Depletion Potential (ODP), Particulate Matter Formation (PM/RI), Acidification potential (AP), Resource depletion, mineral and fossil (ARD), Photochemical Oxidant Formation (POF), and Water depletion (WD). The scenarios are listed in Table 2.

In Figure 7 the results of the individual impact categories for the scenarios with the lowest combined LCA score of all three clusters (scenario 8, 12 and 16) and the reference scenario 1. When comparing the different scenarios, the different impact categories are ranked in the same order. Scenario 8 scores better in all impact categories, and scenario 16 the worst of these four. The most contributing impact categories for all scenarios are global warming potential (GWP)

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and water depletion, followed by eutrophication terrestrial, acidification potential and photochemical oxidant formation.

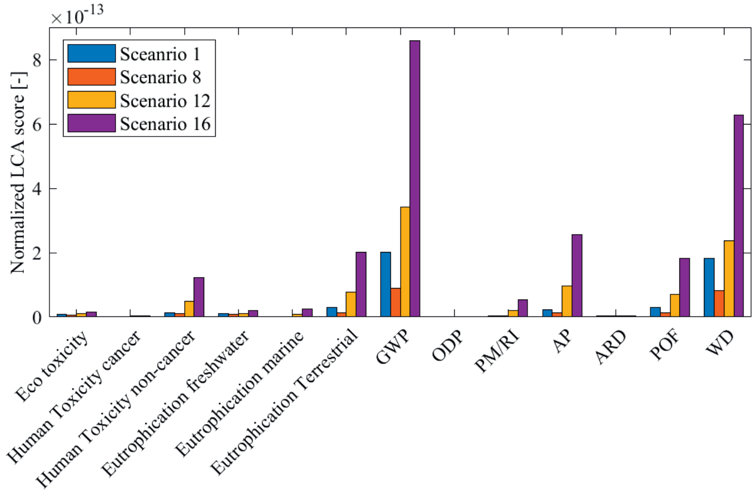

Figure 7. Overview of all individual impact categories for scenario 1, 8, 12, and 16. The impact categories are normalised, but no weights are applied.

The high impact of GWP is a direct link to fossil-based energy consumption, which is the main consumable in milk powder production (Taxiarchou et al., 2015). To reduce the GWP the energy consumption must be reduced. Furthermore, the remaining energy consumption could be replaced by energy from a renewable energy source. In Appendix Table A. 3 the environmental impact of electricity from photovoltaic and heat from solar collectors is listed, both show over a factor 10 in reduction of GWP for the same amount of energy usage.

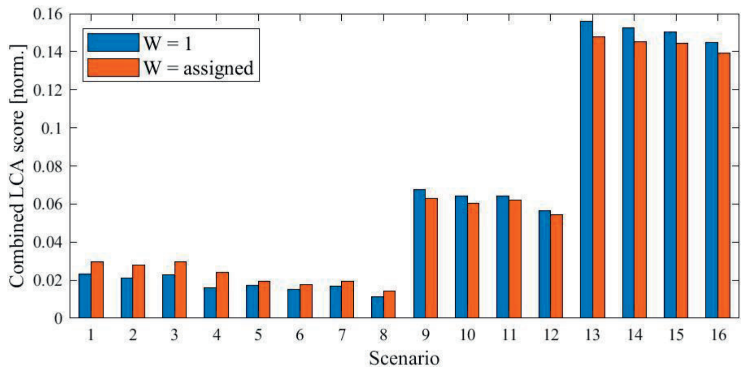

Figure 8. Normalized result of the effect of assigning different weights (W = assigned, see Table 1) or no weights (W =1) on the combined LCA score for the first 16 scenarios. Scenario 17 – 32 use RF pasteurisation instead of normal pasteurisation, and results follow the same trend, hence not shown here.

The impact of water depletion is paradoxical since water is a side product from milk powder production. After the waste water treatment of the condensation water, the water can be returned to the water cycle. Water removed by membrane separation systems (RO and MD) has a high quality (Karakulski, Gryta, & Morawski, 2002), and can actually directly be returned to the water cycle to diminish water depletion.

To combine the values of the different LCA categories, weighting factors (Table 1) were used in equation 3. The effect of the assigned different weights was examined by comparing the optimisation results obtained with the assigned weights to the results with no weights (W=1). Figure 8 shows that the applied weight had a minor effect on the combined LCA score, and did not affect the ranking of the different scenarios. This outcome is not unexpected, as Figure 7 shows that all individual impact categories have a similar distribution and same ranking between the different scenarios.

3.4 Applied objective functions

The combined LCA scores for three different objective functions are given in Figure 9. In this figure the earlier discussed objectives TAC and the combined LCA score are compared to a third objective: the primary energy usage. Figure 9 shows that the three optimized scores for each scenario are very close. The relative standard deviation is 0.4%. Minimizing the total energy consumption or TAC results thus also in the best LCA scores and environmental impact. This outcome is consequence of energy consumption being the main consumable in milk powder production. Hence, for milk powder production and other energy intensive operations in the food industry the reduction of environmental impact and costs is strongly related to energy reduction. Djekic et al. (2014) performed an LCA study for several dairy products (excluding milk powder), and also concluded that the main environmental impact of dairy processing plants is caused by their energy consumption. Larger differences between the objectives will arise when the consumption of chemicals in the production system increase. Chemicals have a major up and downstream environmental impact, and recycle loops can be expensive and increase the operational costs.

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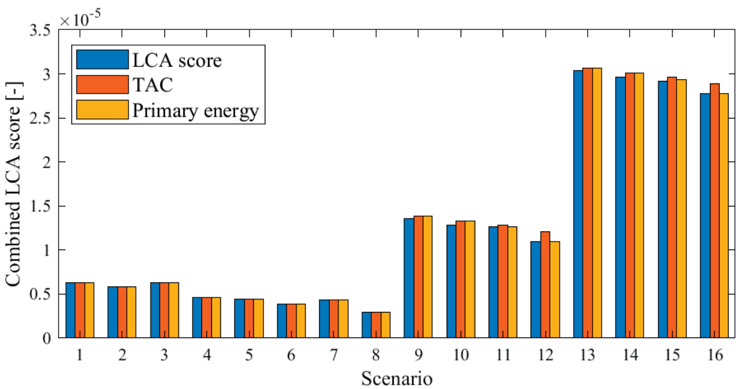

Figure 9. Combined LCA score results for 3 different objectives: combined LCA score, TAC, and primary energy, TAC, for the first 16 scenarios.

3.5 LCA in early stage process design

In this work LCA is applied in early stage of process design to select innovative technologies for future milk powder production. The effect of alternative unit operations and their operational conditions on the environmental impacts was obtained by evaluation of each potential chain individually. The common iterative two-step approach in process design is circumvented by a simultaneous evaluation and optimisation of process performance and environmental impacts. The strengths and weaknesses of the integrated method are summarized in Table 3.

Table 3. Overview of strengths and weaknesses of LCA driven process design

Strength	Weakness
- Process design with minimal environmental impact - Alternative production routes are generated and considered in decision making - Quick evaluation of alternatives - Visualises trade-off between LCA and cost - Hotspots in the production process are highlighted	- A single score method has to be applied - Low level of process detail, as not all inventory and data are known in the early design phase, so assumptions have to be made - Recycling of water is not covered

LCA concerns multiple environmental impact categories, but for evaluation and optimisation the impact categories have to be combined into a single score. This step implies the assignment of weights to the different impact categories, which affects the objectiveness of the evaluation. On the other hand, it also allows the decision maker to distinguish which impact categories are of more importance. Furthermore, in early stages of process design, especially with emerging technologies, process performances and specifications are not always available, and assumptions must be made. As a result, the LCA and process design calculations are approximative. By using justified assumptions and performing a sensitivity analysis, however, good estimates for ranking process options can be obtained.

4 Conclusion

With the necessity to reduce global warming, it is essential to introduce innovative technologies in industrial food processing. For the production of milk powder, the environmental and economic impact of new production systems were optimized and analysed in an early stage of process design. Combining conventional processes with innovative technologies in a superstructure resulted in 32 different processing scenarios. Most promising technologies are a combination of monodisperse droplet spray drying and a zeolite adsorption wheel for energy recovery from the exhaust air. Furthermore, reverse osmosis as a pre-concentration step lowers both the environmental and economic impact. The high cooling demand in membrane distillation has a negative effect on the environmental impact, and requires further development in order to be competitive with the currently used evaporation process. The best processing scenario consists of pasteurisation, RO, multi-stage evaporation, monodisperse droplet spray drying combined with a zeolite adsorption wheel for energy recovery.

The application of LCA as a single score objective as studied in this work, resulted for most scenarios to the same results as minimizing the economic impact (TAC). It was found that scenarios with the lowest energy consumption were the same as the process scenarios with the lowest costs and the lowest combined environmental impact. The main reason for this outcome is that energy consumption has a dominant role in both TAC and LCA. It is expected that for production processes with consumption of chemicals and expensive chemical recycle loops, there costs and environmental impact need to be balanced and the applied method will provide more insight in potential process designs.

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Nomenclature

I	Environmental impact (see Table 1)
R	Amount of resources used (see Table A.3)
N	Normalisation factor (see Table 1)
W	Weighting factor (-)
T	Temperature (°C)
ρ	Density (kg m^{-3})
c_p	Specific heat (J kg^{-1} $°C^{-1}$)
P	Pressure (Pa)
F	Mass flow (kg s^{-1})
Q	Energy (W)
A	Surface area (m^2)
η	Efficiency factor (-)
V	Volume flow (m^3 s^{-1})
t	Time (s)
Subscripts	
i	Processing route
j	Environmental impact category
x	Resource type
$single$	Combined LCA score
m	Milk
sep	Separation
reg	Regeneration
pas	Pasteurisation

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5

Appendix 1 Process models

The processes are modelled by mass and energy balances, and shortly described in this section. The combination the processes resulting in the 32 different scenarios are listed in Table A. 1. Cleaning and waste water treatment were not considered in this study.

A.1.1 Standardisation

Raw milk (4% fat content) is separated into cream and skimmed milk with a disk centrifuge. The fat content of the separated milk depends on the temperature of separation. Hot separation (50°C) leads to a lower fat content of the separated milk than in cold separation (4 – 10°C), but requires pre-heating. The fat content of the cream is 40%. The efficiency of separation depends on the fat content of the separated milk. After separation the milk has to be adjusted to a specific fat content (0.15%) in the skimmed milk by adding a low flow cream; i.e. standardisation. The remaining amount of cream is further processed for other applications not further considered in this study. The centrifuge requires electricity, cooling, and processing water. The electric demand depends on the separation pressure (E_{sep}=1467 W/m^3 (GEA Westfalia, n.d.)) and the milk volume.

$$Q_{p,sep} = E_{sep} \cdot \frac{F_m}{\rho} \tag{A.1}$$

A.1.2 Pasteurisation

Pasteurisation units consist of a heating, regeneration, cooling and a holding section. The heated milk is held for 15 seconds at 72°C (holder) and is used to heat the cold stream in regeneration section. Due to the temperature difference between hot and cold stream in the regeneration section, additional heating and cooling is required. Heat regeneration makes pasteurisation efficient, efficiencies (η_{reg}) up to 96% are common for pasteurisers. The temperature of the milk streams over the regeneration part are calculated by:

$$\Delta T = \eta_{reg} \cdot (T_{pas} - T_{in}) \tag{A.2}$$

$$T_{reg1} = T_{in} + \Delta T \tag{A.3}$$

$$T_{reg2} = T_{pas} - \Delta T \tag{A.4}$$

After the heater additional heating (steam) is required to reach the pasteurisation temperature. Heat loss in the holder is minimal and set to zero. The heat requirement of the heater is:

$$Q_{heat,pas} = F_{m,in} \cdot c_{p,m} \cdot (T_{pas} - T_{reg1}) \tag{A.5}$$

$$F_{steam} = \frac{Q_{heat,pas,}}{\Delta H_{steam}} \tag{A.6}$$

For temporarily storage the milk is cooled in the final part of the pasteuriser with a brine solution. The cooling capacity is:

$$Q_{cool,pas} = F_{sm} \cdot c_{p,sm} \cdot (T_{reg2} - T_{out}) \tag{A.7}$$

$$F_{brine} = \frac{Q_{cool,pas}}{c_{p,brine} \cdot (T_{in} - T_{out})} \tag{A.8}$$

The heat exchanger surface depends on the amount of heat exchanged and the section of the pasteurisation unit. Each section has a different heat transfer coefficient (Kessler, 1981); the heat exchanging surface of each section follows from:

$$Q_{pas,section} = k_{section} \cdot A_{pas,section} \cdot \Delta T_{section} \tag{A.9}$$

The holding section consists of holding tubes which area follows from (Bylund, 2003):

$$V_{pas} = \frac{\frac{F_m}{\rho_m} \cdot t_{pas}}{3600 \cdot \eta_{hold}} \tag{A.10}$$

$$A_{pas,hold} = \frac{V_{pas} \cdot 4}{d_{hold}} \tag{A.11}$$

A.1.3 Radio frequency pasteurisation

Radio frequency (RF) heating is an instantaneous and uniform heating method, resulting in less fouling in heating equipment and less heat damage to the product. The energy required to increase the temperature of the product for a specified temperature difference is given by (Fellows, 2009; Kudra et al., 1991):

$$Q_{RF} = \frac{F_m \cdot \Delta T_{RF} \cdot c_{p,m}}{863 \cdot \eta_{RF}} \tag{A.12}$$

A.1.4 Reverse osmosis

Reverse osmosis (RO) is able to remove water from the milk feed at operational pressures which are above the osmotic pressure of the feed. In chapter 4 the RO model is described in more detail.

A.1.5 Pre-heating and Evaporation

Heating of the milk before concentrated in a multi-effect evaporator. In Moejes et al. (2018) the mass and energy balances of the heating and evaporation process are described. The number of evaporative stages is a decision variable. The pre-heating is a plate heat exchanger.

A.1.6 Membrane distillation

Membrane distillation (MD) is a mass transfer process driven by the difference in vapour pressure between the hot (feed) and cold side (permeate or distillate) of the membrane. The vapour pressure difference is caused by the difference in temperature at both sides. Advantage of the system is that the performance is less limited by feed concentration, and it is possible to concentrate milk to 50% total solids. The process and model are described in detail in Chapter 4. In this study we made use of steady state models, and did not take scheduling into account. Therefore, a constant average fouling is used which results in a flux that does not fluctuate over time.

A.1.7 Spray drying

Milk is atomised into fine droplets at the top of the spray dryer, after which it gets into contact with the hot air. The hot air transfers heat to the milk droplets, water evaporates from the milk droplets and water vapour leaves the dryer with the outgoing air. Depending on the dryer design, a small part of the milk powder particles leaves the dryer with the air flow, and must be recovered by a cyclone or bag filter. Mass and energy balances are equal to those of the monodisperse droplet dryer and described in Moejes et al. (2018).

A.1.8 Fluidised bed drying

The fluidised bed dryer can be placed after the spray dryer to improve energy efficiency and for final drying and product cooling. The operational temperatures are in the range 60 – 100 °C for drying and ambient temperature for cooling. The use of the additional fluidised bed dryer increases also the capacity of the total system. Milk powder leaves the spray dryer with a moisture content of 6 – 9%, and is further dried in the fluidised bed to the final moisture content.

The mass and energy balances for the drying part of the fluidised bed are equal to the ones of the spray dryer. The fluidised bed consists of a heating and cooling section with equal dimensions. The energy required for the fans is given by (Kessler, 1981; van't Land, 2012; Westergaard, 2004):

$$P_{loss,bed} = \rho_{smp} \cdot h_{bed} \cdot g \tag{A.13}$$

$$P_{loss,dis} = \frac{1}{2} \cdot P_{loss,bed} \tag{A.14}$$

$$Q_{p,fan} = V_{air} \cdot \frac{P_{loss,bed} + P_{loss,dis}}{\eta_{fan}} \tag{A.15}$$

A.1.9 Monodisperse droplet spray drying and air dehumidification

Due to the use of monodisperse droplet drying it is possible to recover the latent and sensible heat from the exhaust air. Two options are used: a membrane contactor (MC) making use of a liquid desiccant (brine), and the other option is a zeolite sorption system. These processes are described in detail in Moejes et al. (2018). Regeneration of the brine solution is done with a two-stage evaporator. For the zeolite regeneration, superheated steam is used as this is already proven technology and both economical and energy efficient.

Table A. 1. Overview of all possible processing scenarios from the superstructure.

Scenario	Process units							
1	Centrifugation	Standardisation	Pasteurisation	Pre heater	Evaporator	Spray dryer		
2	Centrifugation	Standardisation	Pasteurisation	Pre heater	Evaporator	Spray dryer	Fluidised bed dryer	
3	Centrifugation	Standardisation	Pasteurisation	Pre heater	Evaporator	Monodisperse dryer with membrane contactor		
4	Centrifugation	Standardisation	Pasteurisation	Pre heater	Evaporator	Monodisperse dryer with zeolite		
5	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Pre heater	Evaporator	Spray dryer	
6	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Pre heater	Evaporator	Spray dryer	Fluidised bed dryer
7	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Pre heater	Evaporator	Monodisperse dryer with membrane contactor	
8	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Pre heater	Evaporator	Monodisperse dryer with zeolite	
9	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Membrane distillation	Spray dryer		
10	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Membrane distillation	Spray dryer	Fluidised bed dryer	
11	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Membrane distillation	Monodisperse dryer with membrane contactor		
12	Centrifugation	Standardisation	Pasteurisation	Reverse osmosis	Membrane distillation	Monodisperse dryer with zeolite		
13	Centrifugation	Standardisation	Pasteurisation	Membrane distillation		Spray dryer		
14	Centrifugation	Standardisation	Pasteurisation	Membrane distillation		Spray dryer	Fluidised bed dryer	
15	Centrifugation	Standardisation	Pasteurisation	Membrane distillation	Monodisperse dryer with membrane contactor			
16	Centrifugation	Standardisation	Pasteurisation	Membrane distillation	Monodisperse dryer with zeolite			
17	Centrifugation	Standardisation	RF pasteurisation	Pre heater	Evaporator	Spray dryer		
18	Centrifugation	Standardisation	RF pasteurisation	Pre heater	Evaporator	Spray dryer	Fluidised bed dryer	
19	Centrifugation	Standardisation	RF pasteurisation	Pre heater	Evaporator	Monodisperse dryer with membrane contactor		
20	Centrifugation	Standardisation	RF pasteurisation	Pre heater	Evaporator	Monodisperse dryer with zeolite		
21	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Pre heater	Evaporator	Spray dryer	
22	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Pre heater	Evaporator	Spray dryer	Fluidised bed dryer
23	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Pre heater	Evaporator	Monodisperse dryer with membrane contactor	
24	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Pre heater	Evaporator	Monodisperse dryer with zeolite	
25	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Membrane distillation	Spray dryer		
26	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Membrane distillation	Spray dryer	Fluidised bed dryer	
27	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Membrane distillation	Monodisperse dryer with membrane contactor		
28	Centrifugation	Standardisation	RF pasteurisation	Reverse osmosis	Membrane distillation	Monodisperse dryer with zeolite		
29	Centrifugation	Standardisation	RF pasteurisation	Membrane distillation		Spray dryer		
30	Centrifugation	Standardisation	RF pasteurisation	Membrane distillation		Spray dryer	Fluidised bed dryer	
31	Centrifugation	Standardisation	RF pasteurisation	Membrane distillation	Monodisperse dryer with membrane contactor			
32	Centrifugation	Standardisation	RF pasteurisation	Membrane distillation	Monodisperse dryer with zeolite			

Appendix 2 Decision variables and constraints

The unit operations are optimised with respect to different decision variables. Table A.2 gives an overview of all operational/decision variables for the total system. Each operational/decision variable is constrained by upper and lower bounds which are based on best current practice with respect to equipment or product limitations. The upper and lower bounds are explained in the foot notes below the table.

Table A.2. List of all operational/decision variables, their lower and upper bound values, the initial value is used as starting point of the optimisation routine. Product and equipment limitations are discussed in the foot note of the table

Variable	Lower bound	Upper bound	Unit
Milk temperature in evaporator [1]	60	70	°C
Milk concentration after RO [2]	0.1	0.18	kg/kg
Milk concentration after evaporator [3]	0.4	0.5	kg/kg
Air temperature spray dryer [4]	180	210	°C
Water content powder after spray drying (in 2 stage dryer) [4]	0.035	0.09	kg/kg
Air temperature fluidised bed dryer [4]	80	100	°C
Milk concentration after membrane distillation [5]	0.4	0.5	kg/kg
Water content air dryer in [6]	0.001	0.02	kg/kg dry air
Concentration difference over membrane contactor [7]	0.01	0.1	kg/kg LiBr
Temperature difference over membrane contactor [7]	1	20	°C
Steam temperature regeneration evaporator [7]	100	150	°C
Vapour temperature out regeneration evaporator [7]	20	50	°C
Zeolite temperature adsorbent phase in [8]	20	100	°C
Superheated steam temperature regeneration in [8]	150	250	°C
Superheated steam temperature regeneration out [8]	150	200	°C

Footnotes for Table A.2:

1) *A 7-stage evaporator was used. To prevent protein denaturation the temperature is constrained to 70°C. The lower bound was set in order to have enough driving force between the stages.*
2) *The driving force for permeation reduces significantly for product concentrations above 18%, which is considered as the maximal feasible with reverse osmosis on commercial scale.*
3) *The viscosity of concentrated milk by evaporation increases exponentially around 0.5 kg/kg and in practice 0.5 kg/kg is considered as a feasible value for conventional atomisation systems in spray dryers.*
4) *Standard ranges for spray drying and fluidised bed drying.*
5) *Membrane distillation can achieve milk concentrations of 0.5 kg/kg.*
6) *Values for water vapour content in air after zeolite and membrane contactor dehumidification.*
7) *Operational range for membrane contactor and brine regenerator (evaporator), derived from model simulations.*
8) *Operational range for zeolite dehumidification, regenerator and superheated steam system, derived from model simulations.*

5

Appendix 3 LCA data

Table A. 3. List of individual environmental impact categories for each resource used in the milk powder production chain, from the Food database of GaBi 6 (Politis, 2016)**.**

	Eco toxicity (CTUe)	**Human Toxicity cancer (CTUh)**	**Human Toxicity non-cancer (CTUh)**	**Eutrophication freshwater (kg $P_{eq.}$)**	**Eutrophication marine (kg $N_{eq.}$)**	**Eutrophication Terrestrial (mole of $N_{eq.}$)**
1 MJ Electrical energy	0.00281	3.14e-11	2.82e-9	2.69e-7	8.14e-6	7.91e-4
1 MJ Thermal energy	0.000387	6.27e-12	8.08e-11	4.64e-9	1.73e-7	1.85e-4
1 MJ of cooling energy	0.3	2.9e-9	1.97e-8	1.54e-5	3.78e-6	2.95e-4
1 MJ of processing steam (85% eff)	0.000455	7.37e-12	9.42e-11	5.43e-9	2.03e-7	2.17e-4
1 kg deionised water	0.00051	1.31e-11	1.91e-10	1.79e-7	1.19e-6	3.92e-5
1 kg tap water	0.0000789	1.77e-12	3.86e-11	2.68e-8	1.33e-7	2.94e-6
1 kg sodium hydroxide	0.0148	2.63e-10	1.53e-8	3.26e-6	7.02e-5	5.25e-3
1 kg nitric acid	0.00613	7.88e-11	1.64e-9	1.01e-6	2.23e-4	3.71e-3
1 kg phosphoric acid	0.235	2.12e-9	1.48e-7	1.22e-5	1.03e-4	2.19e-2
1 kg hydrated lime	0.16	2.22e-9	2.12e-8	1.09e-5	1.14e-4	2.21e-3
1 kg of iron chloride 40%	10.6	8.77e-8	8.98e-7	6.74e-4	1.94e-4	8.23e-3
1 kg of treated waste water	0.84159	1.7802e-8	1.469e-7	2.44e-5	2.00e-4	3.07e-5
1 MJ electricity from photovoltaics	6.7328e-3	8.9895e-12	3.2328e-9	4.58e-8	2.33e-6	9.48e-5
1 MJ heat from hot water from solar collectors	0.18426	1.0336e-9	1.1663e-8	6.81e-6	1.77e-6	5.64e-5

Table A. 3. Continued.

	GWP (kg $CO_{2\text{-eq.}}$)	ODP (kg $R_{11\text{ eq.}}$)	PM/RI (kg $PM_{2.5\text{-eq.}}$)	AP (kg $SO_{2\text{-eq.}}$)	ARD (kg $Sb_{\text{eq.}}$)	POF (kg NMVOC)	Primary energy (MJ)	WD ($m^3_{\text{eq.}}$)
1 MJ Electrical energy	0.129	9.16e-11	2.17e-5	0.000359	4.55e-7	0.000211	3	1.4157e-3
1 MJ Thermal energy	0.0678	1.18e-12	1.81e-6	4.09e-5	2.18e-8	5.85e-5	1.23	1.9831e-5
1 MJ of cooling energy	0.121	1.66e-8	2.59e-5	0.000233	3.65e-6	0.00016	2.21	6.1023e-4
1 MJ of processing steam (85% eff)	0.0798	1.38e-12	2.13e-6	4.81e-5	2.56e-8	6.88e-5	1.45	2.3268e-5
1 kg deionised water	0.00478	1.03e-12	6.55e-7	1.01e-5	6.82e-9	9.38e-6	0.0905	2.1786e-4
1 kg tap water	0.000571	2.35e-14	7.1e-8	8.58e-7	3.02e-11	7.52e-7	0.00664	0.00016605
1 kg sodium hydroxide	0.627	3.06e-11	7.74e-5	0.00116	1.29e-6	0.00122	11.51	0.0052413
1 kg nitric acid	1.15	3.72e-12	1.37e-5	0.00063	2.75e-7	0.000943	9.32	0.0012878
1 kg phosphoric acid	2.03	8.06e-11	0.00141	0.0236	3.25e-5	0.00724	56.9	0.00721232
1 kg hydrated lime	0.754	4.69e-8	7.87e-5	0.000863	2.72e-6	0.000855	4.83	0.00084066
1 kg of iron chloride 40%	0.762	4.7e-7	5.7e-4	0.0049	1.62e-4	0.00237	14.8	0.017609
1 kg tap water	0.010227	6.0044e-10	5.5506e-7	8.7445e-6	2.6881e-8	9.3203e-6	0.17204	7.5671e-6
1 MJ electricity from photovoltaics	0.012636	3.1705e-12	1.1277e-5	4.4882e-5	4.4095e-6	3.4094e-5	7.7607	1.2562e-4
1 MJ heat from hot water from solar collectors	3.936e-3	3.1975e-10	6.094e-6	4.997e-5	8.5739e-7	1.7183e-5	1.189	3.7659e-5

5

Table A. 4. Reference of the environmental impacts from the GaBi 6 database presented in Table A. 3 (Politis, 2016).

Utility	Gabi process reference
1 MJ Electrical energy	EU-27: Electrical Grid mix
1 MJ Thermal energy	EU-27: Thermal energy from natural gas
1 MJ of cooling energy	CH: Cooling energy, from natural gas, at co-gen. unit with absorption chiller 100kW
1 MJ of processing steam (85% eff)	EU-27: Process steam from natural gas 85%
1 kg deionised water	EU-27: Water (deionised)
1 kg tap water	EU-27: Tap water
1 kg sodium hydroxide	DE: Sodium hydroxide mix (50%)
1 kg nitric acid	DE: Nitric acid (60%)
1 kg phosphoric acid	DE: Phosphoric acid (100%) (wet process)
1 kg hydrated lime	CH: lime production, hydrated, packed
1 kg of iron chloride 40%	CH: iron (III) chloride production, product in 40% solution state
1 kg of treated waste water	EU-27: Waste water treatment (adopted for dairy factory)
1 MJ electricity from photovoltaic	NL: Electricity from photovoltaic
1 MJ heat from hot water from solar collectors	RoW: operation, solar collector system, Cu flat plate collector, multiple dwelling, for hot water ecoinvent

Appendix 4 Cost calculation

The economic indicator used in this study is the Total Annual Costs (TAC). The TAC consists of both investment ($C_{equipment}$) and operational costs (energy, water, etc.) ($C_{operational}$). For the ranking of production scenarios, the costs for labour, cleaning, laboratory and overhead are assumed to be similar for the scenarios and therefore not included in this study.

$$TAC = \sum C_{equipment} + \sum C_{operational} \tag{A.16}$$

The equipment costs are a function of lifetime (n_{life}), Lang factor (LF), and interest rate i to take the time value of money into account ($i = 0.06$ which corresponds to 6%) (Seider, Seader, Lewin, & Widagdo, 2010).

$$C_{equipment} = C_{invest} \cdot LF \cdot \frac{i(1+i)^{n_{life}}}{(1+i)^{n_{life}} - 1} \tag{A.17}$$

where C_{invest} are the equipment purchasing costs. Equipment costs depend on the dimensions/capacity. The equipment investment costs for a specific capacity (A_{eq}) are estimated from standard equipment with given dimensions/capacity by using process sizing:

$$C_{invest} = C_{ref} \left(\frac{A_{eq}}{A_{ref}} \right)^{n_{eq}} \tag{A.18}$$

where C_{ref} are the costs for a reference installation with dimension/capacity A_{ref} and n_{eq} the scaling factor for the equipment. Reference costs, size and scaling factors are discussed in Appendix 4.

The operational cost is a combination of costs for heating, cooling and the use of other resources:

$$C_{operational} = C_{heating} \cdot Q_{heating} + C_{cooling} \cdot Q_{cooling} + \sum C_{resource} \quad \text{(A.19)}$$

in which $C_{resource}$ costs for processing water, chemicals, etc. The prices for the used utilities are summarised in Appendix Table A. 5.

Table A. 5. Overview of process and cost related variables (DACE, 2014; Eurostat, 2015; Hausmann et al., 2014; Houben, 2016; Kessler, 1981; Seider et al., 2010).

Variable	Value
Natural gas (Nm^3)	€ 0,046
Electricity (kWh)	€ 0,12
Tap water (m3)	€ 1,00
Cooling water (kWh)	€ 0,033
Boiler efficiency (-)	0.80
Air heater efficiency	0.78
RF efficiency	0.65
Lang factor (-)	3.5
Interest rate	0.1
Operations hours (h/year)	8000
Chiller efficiency (-)	0.3
Membrane flux RO (kg/m^3h)	12
Membrane flux MD (kg/m^3h)	4
Membrane flux MC (kg/m^3h)	0.5
Tap water temperature (°C)	12
Ambient air temperature (°C)	20
Vapour content ambient air (kg/kg dry air)	0.011
Moisture content raw milk (kg/kg)	0.09
Final moisture content milk powder (kg/kg)	0.035

Table A. 6. Investment cost estimation of different processing equipment.

Unit operation		Reference costs (C_{ref})	Reference size (A_{ref})		Scaling factor (n_{eq})	Life time (years)	Reference
Disk bowl centrifuge		€ 150,000	25	m^3	0.6	20	(Piek & Telgenkamp, 2015)
Heat exchanger		$€11{,}546 + €192.6 * A_{eq}$					(based on: DACE, 2014)
RF unit		€ 2,312	1	kW			(Koral, 2013)
RO	non-membrane eq.	€ 300	1	m^2		15	(Elsayed, Barrufet, & El-Halwagi, 2014; González-Bravo et al., 2015; Houben, 2016; Medevoort, 2016; Meindersma, Guijt, & de Haan, 2006; Petrides, 2013)
	membrane	€ 50	1	m^2		5	
MD	non-membrane eq.	€ 2.50	1	m^2		15	(Elsayed et al., 2014; González-Bravo et al., 2015; Houben, 2016; Medevoort, 2016; Meindersma et al., 2006; Petrides, 2013)
	membrane	€ 100	1	m^2		5	
Evaporator		€ 830,000	7700	kg water removed/h	0.53	30	(Garrett, 1989; Petrides, 2013)
Spray dryer		€ 600,000	400	kg water removed/h	0.29	30	(*APV Dryer Handbook*, 2000; Garrett, 1989)
MC	non-membrane eq.	€ 300	1	m^2		10	(Medevoort, 2016; Petrides, 2013)
	membrane	€ 100	1	m^2		50	
Zeolite wheel		€ 250,000	1400	kg water removed/h	0.78	10	(Voogt, 2016)

Chapter 6

General discussion

1 Introduction

The world population is expected to grow to 9.8 billion by 2050 (UN, 2017), and with this growth the consumption of food will also increase. As a result, the energy consumption in cultivation, transport, and processing of food products is expected to raise when no counter measures are taken. Currently, the food industry in Europe consumes 26% of the total energy usage in the EU, 28% of which is used for the industrial processing of food (Monforti-Ferrario et al., 2015). The majority of this energy originates from fossil fuels, resulting in a negative environmental impact and depletion of resources. Reducing the environmental impact requires a two-part solution; firstly, a reduction of energy consumption should be achieved, and secondly renewable energy sources should be used for the remaining energy requirements. For the first part of the solution i.e., to decrease the energy consumption of the food industry, innovative food processing technologies have to be implemented. The work described in this thesis focusses on the redesign of the milk powder production chain in order to decrease energy consumption, and thereby lower the environmental impact. To that end the selection, evaluation, and optimisation of innovative technologies have been described in the previous chapters of this thesis. This chapter is a reflection on the work presented in this thesis complemented with topics which are not yet (fully) addressed and is complemented with elaborations on a future outlook.

2 Conclusions and perspectives

2.1 Selection of innovative technologies

To assess the potential of innovative technologies for milk powder production and to redesign the current milk powder production chain a systematic modelling approach has been applied. The first step for each process innovation is a selection of process technologies and to quantify their impact in the chain. Chapter 2 gives an overview of the state-of-the-art and innovative technologies relevant for milk powder production. In that chapter a first assessment of the alternative technologies is given. The opportunities were identified as:

- Alternative heating technology for pasteurisation
- Heat recovery of the dryer exhaust air by air dehumidification
- Complete replacement of evaporation by membrane-based process

A promising alternative for steam heat exchangers used for pasteurisation, is radio frequency (RF) heating. Disadvantage of traditional pasteurisation by steam heating is the environmental impact caused by the use of fossil resources. Moreover, fouling of the heat exchanger surfaces caused by milk components that denature and deposit at the heat exchanging surfaces and, therefore, limit heat transfer. The main advantage of RF heating is the rapid and homogeneous heating due to the direct heating system. This results in less fouling compared to a steam heat exchanger. Due to the homogeneous heating and less fouling, lower processing temperatures are needed, which besides reducing energy use also improves product quality. The energy conversion from fossil fuel to electricity is still low and makes that RF has a higher environmental impact than steam-based pasteurisation. Nevertheless, with the increasing availability of renewable energy and the current industrial shift to electrification RF can become an interesting technology.

Following pasteurisation, milk is currently concentrated to around 50% dry matter by multi-stage evaporation. Alternatively, water can be removed by the use of membranes. Most membrane technologies are, however, not able to replace the evaporation process completely as they cannot reach the required final dry matter concentration of 50%. Membrane distillation, however, is an emerging membrane technology which is able to reach these high solids concentrations and which has the potential to replace the traditional evaporators. Advantage of membrane distillation are the low working temperatures (around 50 – 70°C), enabling the use of hot water as heating medium instead of steam. These low temperatures enable more opportunities for the reuse of waste heat, for example from the drying section. Based on a literature review an estimate was made that the energy consumption for membrane distillation is similar to a seven-stage evaporator, however, could be completely run on waste heat from the drying section.

After concentration, milk is dried by spray drying. This is where the prospect of heat recovery becomes interesting. Potential improvements in the spray dryer are achieved by a two-part solution: 1) use of a monodisperse droplet atomizer and 2) air dehumidification. The combination of monodisperse drying and air dehumidification technology results in a closed loop drying system, in which the exhaust air is recycled over the dryer. Monodisperse droplet drying eliminates the small particles from the exhaust air, which allows heat recovery from the exhaust air as no powder particles will cause fouling problems. The latent and sensible heat from the exhaust air is recovered either by a contact sorption system or a membrane contactor with a liquid desiccant. The heat recovered from the dryer exhaust is enough to provide sufficient heat for the membrane distillation process.

Based on numbers reported in literature, successful implementation of the proposed technologies could lead to a 60% energy saving for milk powder production. The energy consumption for the complete chain would be reduced from 10 MJ per kilogram powder to 4 – 5 MJ per kilogram powder.

2.2 Optimal heat recovery in closed-loop spray drying

Chapter 3 focusses on the heat recovery of the spray dryer exhaust air by air dehumidification. The proposed technologies are: monodisperse droplet drying combined with a contact sorption system (zeolite) or a membrane contactor (MC) with liquid desiccant (chapter 2). Four different closed-loop dryer configurations were simulated and their operational conditions were optimized to minimize costs and energy consumption. The configurations consist of the two different dehumidification technologies (the zeolite sorption system and MC), and for each dehumidification technology two regeneration methods were investigated namely hot air and superheated steam for the zeolite system, and evaporation and superheated steam for the MC system.

Not all heat recovered from the drying process can successfully be utilized in the drying process itself. For this reason, to make optimal use of the recovered heat, the concentration step (evaporator) was included as a heat sink. The optimisation of the operational conditions was combined with Pinch analysis with the aim to optimize the operational conditions at the same time as the heat integration network.

The closed-loop dryer configuration with MC and regeneration by superheated steam proved to have the lowest energy consumption. The energy consumption for concentration and drying was lowered from 8.9 MJ per kilogram milk powder to 4.9 MJ per kilogram milk powder for the concentrating and drying step. From this work is concluded that energy savings ranging from 11 to 42% compared to current milk powder production are feasible.

An MC system as dehumidification technology proved to have a larger energy savings potential compared to a zeolite sorption system (42 and 39% energy reduction resp.). Zeolite sorption wheels, however, are already commercially available, while MCs designed for elevated temperatures are still at an early stage of development. The highest energy savings are achieved with an MC in which the liquid desiccant was regenerated with superheated steam in a second MC. The challenge for MC regeneration system is the application of the high temperatures of superheated steam (over 180°C) to the membranes. Further development of MCs as a dehumidification technology at higher temperatures, should therefore focus on the use of membranes to regenerate the liquid desiccant at temperature of over 180°C. With the process performance as calculated in chapter 3, the MC is a viable technology for air dehumidification in a closed-loop spray drying system. To meet the predicted process performance (e.g. predicted fluxes, temperature resistance, etc.) further development and testing of an MC system is required.

2.3 Membrane distillation networks for milk concentration

The feasibility to replace the evaporation process by a membrane system was addressed in chapter 4. Two membrane processes, reverse osmosis and membrane distillation, were modelled including their fouling dynamics. Moreover, the network configuration of the different membrane modules for scheduling of operation and cleaning was optimised.

Reverse osmosis (RO) is a proven membrane technology in dairy processing. Limited by the osmotic pressure and fouling, the dry matter content that can be reached by RO is 18-24%. Therefore, RO needs to be combined with an additional concentration step to reach the desired 50% dry matter for the drying process. Membrane distillation (MD) can reach that high solid content and is therefore of interest to replace traditional evaporators. RO and MD are both membrane processes, with the difference that RO is pressure driven and MD is thermally driven. The advantage of pressure driven membranes is the possibility to increase the operational pressure to compensate for reducing membrane fluxes caused by gradual fouling. This is not possible for MD. The fouling was included in our work for both RO and MD, and is based on the work of Hausmann et al. (2014) and van Boxtel et al. (1991).

MD is available in different configurations. In chapter 4 an air gap membrane distillation (AGMD) module was modelled. AGMD differs from the standard direct contact membrane distillation module by an air gap between the hot and cold side in which the permeate is collected. AGMD has the advantage of internal heat recovery and was therefore assessed in this thesis.

The optimisation discussed in chapter 4 resulted in a network configuration of two consecutive stages of RO followed by one MD stage. RO proved more energy and cost efficient compared to MD, and milk is therefore pre-concentrated to the maximum concentration by RO (which was set to 18% dry matter). MD concentrates milk from 18 to 50% dry matter in one stage. The heating and cooling requirements for MD are high, due to the low membrane fluxes and large recirculation flow. Although AGMD has internal heat recovery, it was insufficient due to the low heat transfer as a result of low fluxes. In order to exploit the benefit of internal heat recovery in the AGMD configuration, the temperature difference over the hot side of the module (between feed inlet and outlet) has to be larger than the temperature difference over the heater and cooler (see chapter 4). These conditions could not be achieved with the fluxes obtained in this study. A direct contact membrane distillation will probably perform better under these conditions.

In chapter 4 was concluded that a combination of RO and MD is technical able to replace the traditional evaporation process. However, due to the high energy consumption the combined system is not yet a viable option. For MD the fluxes have to be increased and the recirculation flow has to be reduced.

2.4 Environmental and economic optimisation

The main objective of chapters 2, 3, and 4 was to minimize energy consumption and/or costs for the production of milk powder. To minimize the environmental impact, life cycle assessment (LCA) was integrated with the early steps in process design in chapter 5. Multi-objective optimisation was applied to a superstructure. This approach resulted in a selection of technologies and their operational conditions with the lowest costs and environmental impact. Environmental impact involves a series of impact categories, for example global warming potential, water depletion, eutrophication, and ecotoxicity. The result for each impact category was normalised and weighted, in order to make a combination into a single score (referred to as the combined LCA score) possible.

The selection and optimisation of technologies was approached by evaluating all possible processing scenarios with the pre-selected technologies. From all optimised processing scenarios, a combination of traditional pasteurisation, multi-stage evaporation, monodisperse droplet drying, and zeolite sorption wheel proved best in terms of the combined LCA score, and thus environmental impact. The largest energy savings were achieved with heat recovery from the dryer. The surplus heat from the regeneration of the zeolite sorption system with superheated steam is effectively reused in the concentration process (as predicted in chapter 3). This process configuration reduced the combined LCA score by 50%, and total costs by almost 40% compared to the state-of-art production system.

The results for minimisation of the combined LCA score were similar to the results obtained from minimisation of the total cost or the energy usage. Energy (electric and thermal) is the main consumable in milk powder production, and therefore this outcome was not surprising. Finnegan et al. (2017) also concluded that energy contributes on average for 89% of the total global warming potential, the other 11% were caused by milk transportation, packaging, and waste treatment. These processes were not included in our study, and therefore energy consumption is the sole contributor. The integration of LCA in early stage of process design will therefore be of more importance for processes with extensive use of chemical compounds.

In chapter 2, energy savings around 60% were predicted. These results were based on the best possible values reported in literature. In chapter 3 and 4 a further in-depth analysis of the options for energy recovery from the dryer and the concentration step with MD was made. The results from these chapters, together with the other proposed technologies (chapter 2), were combined in the final superstructure optimisation in chapter 5. At the end of this thesis it is concluded that with the proposed milk powder production chain an energy reduction can be realised of 50%. The discrepancy with the prediction in chapter 2, is caused by the lower performance of the MD compared to first expectations.

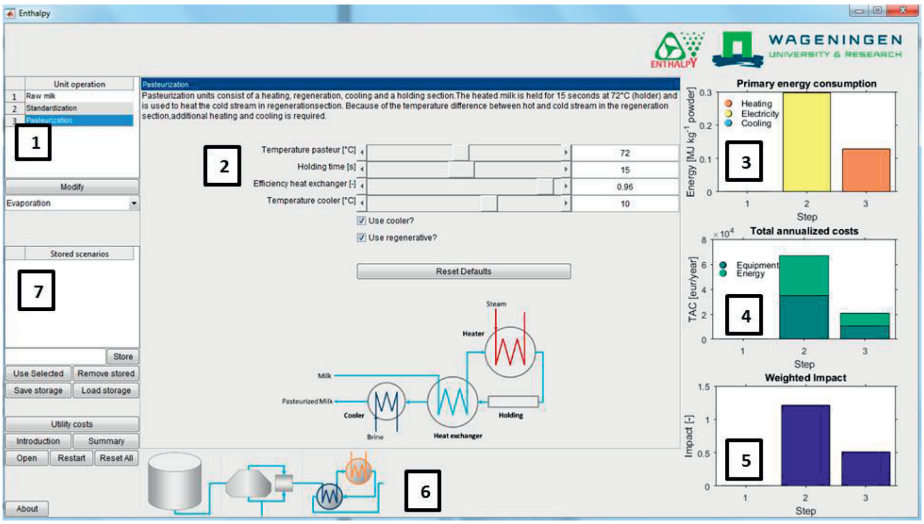

Figure 1. Screenshot of the GUI. 1) holds the field to select the unit operations in the process chain, in 2) the settings of the operational conditions can be adapted, 3) displays the graph with energy usage per processing step, 4) shows the costs per unit operation, 5) shows the combined score for environmental impact, 6) shows process configuration of the selected processes, and 7) contains options for saving and loading scenarios.

To make the developed process models, results, and gained insights easily available for the other project partners in the ENTHALPY project, a flexible simulation tool with graphical user interface (GUI) was developed. The goal was to allow the project partners to simulate different production processing chains by combining conventional and innovative technologies. Figure 1 shows a screenshot of the developed GUI. The simulated process scenario is evaluated on three objectives; energy consumption, total costs, and environmental impact. It allows the user to evaluate different scenarios and to compare the scenarios with respect to the three objectives. In this way it is easy to simulate and compare a variety of different production scenarios, and to evaluate the effect of process changes.

3 Additional opportunities and insights

In chapter 1, drying and evaporation were highlighted as the main energy consumers. Therefore, this work focussed on those two processes. Other opportunities for emerging technologies are discussed in this section, i.e. enzymatic cleaning of equipment, and the use of renewable energy sources. These aspects have not yet been extensively mentioned in this work so far, but have potential to reduce the energy consumption in industrial processing.

3.1 Enzymatic cleaning

The cleaning of process equipment is responsible for 10 – 26% of the overall energy consumption in milk production (Krebbekx, Lambregts, Wolf, & Seventer, 2011; Ramirez, Patel, & Blok, 2006), and for this reason alternative cleaning methods deserve attention. In dairy processing, cleaning is applied to guarantee microbial safety of the product and to maintain process performance. Also fouling on heat exchangers, pipes, and membranes by proteins, minerals, and fat can occur (Jeurnink & Brinkman, 1994). As a result of fouling an adherence surface for microorganisms in milk is formed which in turn results in the formation of unwanted biofilms. Furthermore, heat transfer is affected, and pasteurisation temperatures might not be reached, posing another potential safety problem (Flint, Bremer, & Brooks, 1997). For this reason, regular cleaning is of high importance in milk processing. In Chapter 4 the scheduling of cleaning, and effect on process performance was included. However, opportunities for energy reduction in cleaning are not yet addressed in this thesis.

The main sources for energy consumption in cleaning are: 1) heat loss from the storage tank for cleaning agents, 2) heat loss from the processing equipment, and piping towards/from the processing equipment, and 3) energy required for heating fresh cleaning solutions to the required temperature. Potential ways to reduce energy consumption in cleaning are 1) reducing temperature, 2) reducing the number of cleaning steps, 3) reduction of the length of the cleaning steps, and 4) using alternative cleaning agents. Traditionally cleaning consist of a combination of alkaline and acidic cleaning at temperatures ranging from 70 – 90°C (Timmerman, Mogensen, & Graßhoff, 2016). As part of the EU project ENTHALPY, a new enzymatic cleaning method was developed (Guerrero-Navarro et al., 2019). Advantage of using enzymes for cleaning are: the lower working temperatures (around 50°C), the reduced amounts of chemical waste, and a reduction in water consumption. These all contribute to a reduced environmental impact.

Model calculations showed that if the traditional two-step alkaline/acidic cleaning is replaced by a one-step enzymatic cleaning (while keeping the rinsing steps the same), the energy consumption of the cleaning step can be reduced by 70%, and water consumption can be reduced by 50% (assuming lowering process temperature from 70 to 50°C, and reducing the amount cleaning steps from 5 to 3). Although these are basic calculations, these numbers highlight the large potential of enzymatic cleaning.

An additional advantage for the reduction of environmental impact is the potential to reduce the water consumption for cleaning. The reuse of the permeate produced by for example membrane distillation (chapter 4). The retention of membranes used in MD is 99 – 100%, and the permeate is therefore directly suitable for cleaning processes (Hausmann et al., 2013). Further integration of the water cycles has not been investigated in this thesis, but provides additional opportunities for environmental impact reduction.

3.2 Renewable energy sources

Besides the reduction of energy consumption, renewable energy sources can provide part of the solution to reduce the environmental impact of a production chain by reducing the consumption of fossil fuels. In a continuous production processes, it is essential to have a stable and reliable network of renewable energy which does not affect the product or continuity of the processes.

3.2.1 Photovoltaic cells

In chapter 5 radio frequency heating (RF) was included as an innovative technology for milk pasteurisation. RF is electricity driven and if the electricity is generated by a non-renewable energy source, RF cannot compete with traditional pasteurisation based on steam heating. With the use of electricity from photovoltaic cells (PV's) RF becomes an attractive alternative. It has been observed that each time the total number of PVs manufactured doubles, the cost of PV cell drops 20% (Swanson, 2006). With the current focus on renewable energy sources it is expected that the shipping volume will continue to increase. This trend will lead to a reduction in PV installation cost, and a change in the balance with steam heating.

6

For a small model milk powder factory (10,000 kg raw milk/h) at least 1 ha of PVs is needed to fulfil the electrical demand on a bright day (assuming: maximum solar intensity of at least 600 W/m^2, a solar cell efficiency of 20%, and 10% of factory energy demand is electric). With the increasing number of domestically installed PVs, and their production being out of synchronisation with domestic demand, usage of their production peaks is of interest (Timilsina, Kurdgelashvili, & Narbel, 2011). Industries, like a milk powder plant, could utilize those peaks and adapt their consumption and production for optimal integration. This is an alternative for the grid, instead to invest in storage technologies for the excess energy produced.

3.2.2 Solar heating system

Milk powder production requires mainly thermal energy rather than electrical. In this sense, the use of solar heating is of more interest than PV's. The working temperatures in milk powder production make the use of solar thermal energy possible (Lauterbach, Schmitt, Jordan, & Vajen, 2012). Just like with PV's, is the integration with a day round production scheme a challenge, which can be met with hybrid boiler systems. For a large-scale factory (100,000 kg raw milk/h) the contribution of a solar heating field of 1 ha and bright radiation (maximum solar intensity of at least 600 W/m^2) would lead to a 10 – 20% steam reduction. For smaller factories this share increases. Depending on the location, and thus solar radiation, of the factory, the applicability of solar heating will be of interest as a hybrid boiler system. The energy demand used in this example is of a state-of-the-art milk powder plant without the previously discussed improvements. By applying the technologies discussed in this thesis, the contribution of solar powered energy will increase.

3.2.3 Opportunities for heat pumps

In food production most of the available waste heat has a relative low temperature (below 100°C) (Hammond & Norman, 2014). These low temperatures make heat recovery often difficult and inefficient. For this reason, heat pumps are of interest, by increasing the temperature of waste heat to a usable temperature level for other processes. Heat pumps can be used in addition to the dehumidification technologies as proposed in chapter 3. Krokida et al. (2004) already showed that the integration of a heat pump to the heat recovery from the dryer exhaust increased the total heat recovery by 15%. Walmsley et al. (2017) modelled a hybrid heat pump for a spray dryer system, which resulted in a total potential energy reduction of 47%.

The challenge for heat pump technology is the low efficiency at elevated temperatures. For spray drying, temperatures between 180 and 220°C are needed (chapter 3). This level cannot be reached by the currently available heat pumps with a maximum at 160°C, and additional heating is necessary (Arpagaus, Bless, Uhlmann, Schiffmann, & Bertsch, 2018). High temperature heat pumps are still in development and of growing importance with the increasing availability of renewable electricity.

By means of a next step in the development of an integrated process design, the superstructure optimisation as applied in chapter 5, should be extended by the integration of renewable energy sources, leading to a complete industrial site optimisation. Walmsley et al. (2018) proposed a total site heat integration approach, which should be extended with alternative energy sources and environmental impact evaluation.

4 Life cycle assessment in early stage of process design

Life cycle assessment is a widely used tool to assess the environmental impact of a process. In research projects funded with public money, LCA gains a prominent role and is often even a requirement. In the early phases of these projects the technical and product knowledge is under development and the data to be used for an LCA is not yet complete and uncertain. However, this is also the phase where many decisions are made which will influence the results of a final LCA. Bhander et al. (2003), and later Poudelet et al. (2012) referred to this problem as the environmentally-conscious or eco-design paradox. They encountered the difficulty of using LCA in product design. At early stages in product design aspects like composition and shape are unknown. The further the design get towards implementation, the more details are known, but less can be changed in order to improve the environmental impact of the product.

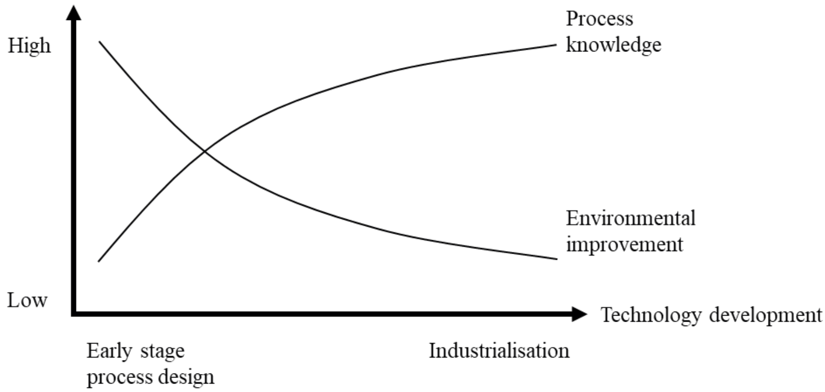

Figure 2. The eco-design paradox for process design (adapted from Poudelet et al. (2012) and Bhander et al. (2003)).

Process design meets the same paradox (Figure 2). Industrialised processes are completely developed and allow only small changes to improve environmental impact. In contrast, the early stages of a process design offer the best opportunities to make decisions for improvement on environmental impact. However, information in this stage is not yet fully accurate to perform an LCA. Thus, a major challenge of performing LCA at an early stage process design is how to deal with a limited amount of data available.

A way to deal with uncertainty of data is to use sensitivity analysis. Sensitivity analysis allows the assessment of varying operational conditions and process parameters and to identify which have a critical impact on the process performance. Although in the early stages of technology development literature data and assumptions have to be used, simulation of alternative processing scenarios provides insight which operational parameters are essential and indicate the operational window of the technology of interest. A common Dutch adage 'meten is weten' is literally translated as 'measuring is knowing', but an equivalent important statement is 'modelling is knowing'. Especially at the early stages of process design, the feasibility of innovative technologies can be quantified by simulations. By making use of justified assumptions and performing a sensitivity analysis, different process scenario can be ranked and compared to each other. This will allow a decision maker to choose which combination of technologies is of interest, and under which conditions.

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Summary

With the increasing world population and global warming challenges, it is of importance to reduce the energy consumption and environmental impact of food production. The dairy industry is an energy intensive industry and is expected to grow in the coming decades. Milk powder production has an especially high energy demand due to the concentrating and drying steps. Over the past decades the currently used processes have already been optimized to a large extent. For this reason, innovative technologies are needed to realize breakthrough solutions to reduce the environmental impact of food production. The aim of this thesis is to use different modelling and optimization tools to assess the potential of innovative technologies to lower the energy usage and reduce environmental impact in the milk powder production chain. The outcome of this thesis is a proposal to redesign of the current milk powder production process.

Chapter 2 provides a literature review of the state-of-the-art milk powder technologies, and alternative emerging technologies with the potential to replace current technologies. The main areas of interest are the replacement of the multi-stage evaporator by a membrane process, and to recover the latent and sensible heat from the spray dryer exhaust air. The emerging technologies discussed are: membrane distillation, monodisperse-droplet drying, air dehumidification and, radio frequency heating for milk processing. An assessment is made of the current energy usage for the production of skimmed milk powder, which is 10 MJ/kg powder with state-of-the-art technologies. The energy consumption of the innovative technologies is estimated based on literature review. In this chapter a 60% energy reduction is predicted, which lowers the energy consumption to 4 – 5 MJ/kg of milk powder depending on the process configuration.

In **chapter 3** the recovery of latent and sensible heat of the spray dryer exhaust air is further investigated. In this chapter a close-loop dryer system consisting of a monodisperse droplet atomizer and an adsorbent system is modelled and optimised. The monodisperse droplet atomizer limits the amount of fine powder particles in the exhaust air, reducing fouling problems on heat recovery equipment. Two adsorbent systems for air dehumidification are discussed; a membrane contactor with a liquid desiccant, and a zeolite sorption system. For regeneration of the zeolite both hot air and superheated steam are considered. For the regeneration of the liquid desiccant from the membrane contactor system, a two-stage evaporator is used and as alternative a second membrane contactor in which the liquid desiccant is regenerated with superheated steam is considered. The two adsorbent systems each with two regeneration methods lead to four different closed-loop spray drying configurations. The recovered heat from the drying section needs to be effectively utilised, for this reason the concentration step, i.e. seven-stage evaporator, is added as a heat sink. Each configuration is simulated and compared to a reference scenario consisting of a tradition spray dryer.

The operational conditions are optimised in one step together with the heat integration, using Pinch analysis. A configuration with a membrane contactor and regeneration with superheated steam proved to have the lowest energy consumption. The energy consumption of the milk concentrating and drying step was lowered from 8.4 to 4.9 MJ/kg milk powder, which is an improvement of 42% (only for concentrating and drying, further pre-treatment was excluded). The energy reduction for the other configurations ranged from 11 – 39% compared to the reference.

Prior to drying, milk is concentrated to a dry matter content of 50%. Currently milk is concentrated in multi-stage evaporators. In **chapter 4** the replacement of evaporation by membrane technology is further assessed. Membrane distillation is an emerging technology mainly used for desalination and waste water processes. The system is a thermally driven membrane technology, and can reach high solids concentrations. A reverse osmosis and air gap membrane distillation network is optimised in order to concentrate milk from 9 to 50% dry matter. For both reverse osmosis and air gap membrane distillation a process model is made based on mass and energy balances, and a membrane fouling model was included. Furthermore, scheduling of the cleaning cycles for the parallel membrane units was included to limit the effect of process fluctuations. Optimization of the number of stages and parallel membrane units resulted in a membrane network of 2 stages of reverse osmosis, followed by 1 stage of membrane distillation. Reverse osmosis is more energy and cost efficient compared to membrane distillation. Membrane distillation proved to be energy intensive, despite the low operating temperatures (58°C). The large recirculation flow, which is needed to keep sufficient cross flow, has to be heated and cooled. For this reason, the optimal system for membrane distillation has only one stage operating at a high concentration and relative low flux. Major opportunities to improve the performance of membrane distillation for the concentration of milk are: 1) increase the cold and hot side temperatures to their maximum acceptable values, 2) develop spacers that allow lower linear flow velocities in the system and thus lower recirculation rates, and 3) make use of available waste heat. Due to the low operating temperatures of membrane distillation, the usage of waste heat from other processes is its major opportunity to replace the traditional evaporators.

In **chapter 5** all technologies proposed in the previous chapters are combined with the state-of-the-art technologies in a superstructure. This allows the evaluation of all possible processing scenarios for milk powder production. In previous chapters the energy consumption and/or costs were the main objectives. In order to also assess the environmental impact of the different scenarios, life cycle assessment and cost optimization were combined as a multi-objective optimisation problem. To integrate life cycle assessment at this early stage of process design, a single score approach that combines all environmental impact categories was used. Most promising process scenario in terms of environmental impact consist of reverse osmosis combined with multi-stage evaporation and closed-loop spray drying. In the evaluation of all scenarios, the energy consumption proved to be dominant in both the operational costs and the environmental impact.

In **chapter 6** the main findings and achievements of this work are discussed together with the potential for further improvement. From this work is concluded that the heat recovery of the dryer exhaust air from a mono-disperse droplet dryer is the largest step in the reduction of energy consumption for milk powder production.

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About the author

Sanne Nadine Moejes was born on the 24th of September 1989 in Amsterdam, the Netherlands. She grew up in Amersfoort where she attended het Nieuwe Eemland College, and obtained her VWO diploma in 2007. Always fascinated by food she started a bachelor in Food Technology at Wageningen University, and graduated in 2010 with a minor in Operations Research and Logistics. The following year she used to travel, and work in a skiing area in Austria. In September 2011 she started her master in Food Technology with a specialisation in Sustainable Food Process Engineering. Sanne worked on a master thesis entitled: Sustainability evaluation of the bread supply chain in the Netherlands using MCDM methods and exergy analysis. This was a combined project between the Operation Research and Logistics group and the Laboratory of Food Process Engineering. She finalised her studies with an internship at DICTUC S.A., where she looked into the sweetening properties of stevia. The project was conducted within the Centro de Aromas y Sabores in Santiago, Chile. In 2013 she started her PhD project within the Biobased Chemistry and Technology group at Wageningen University. The research she started was part of the EU project ENTHALPY, and focused on the modelling and optimization of innovative technologies for the milk powder production chain. The result lead to this thesis. Sanne is currently working as a process engineer energy at Cosun R&D in Dinteloord.

List of publications

Zisopoulos, F. K., **Moejes, S. N.**, Rossier-Miranda, F. J., van der Goot, A. J., & Boom, R. M. (2015). Exergetic comparison of food waste valorization in industrial bread production. *Energy*, *82*, 640–649. https://doi.org/10.1016/j.energy.2015.01.073

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Overview of completed training activities

Discipline specific courses and activities

Numerical Methods for Chemical Engineers (OPST), Eindhoven, the Netherlands, 2014

Summer school HIntBioref (Erasmus), Lisbon, Portugal, 2014

Process Economics and cost Engineering (OSPT), Eindhoven, the Netherlands, 2015

NPS Symposium 2014 (poster presentation), Utrecht, the Netherlands, 2014

29th EFFoST conference (2 oral presentations), Athens, Greece, 2015

The 20th International Drying Symposium (oral presentation), Gifu, Japan, 2016

ENTHALPY Final Conference (oral presentation), Girona, Spain, 2016

NWGD Yearly Symposium (oral presentation), Wageningen, the Netherlands, 2016

General courses

VLAG PhD week, Baarlo, the Netherlands, 2014

Project and time management (WSG), Wageningen, the Netherlands, 2014

Competence Assessment (VLAG), Wageningen, the Netherlands, 2014

Voice Matters (VLAG), Wageningen, the Netherlands, 2015

Teaching and supervising thesis students (DOI), Wageningen, the Netherlands, 2015

Techniques for writing and presenting a scientific paper (WGS), Wageningen, the Netherlands, 2015

PhD Workshop Carousel (WGS), Wageningen, the Netherlands, 2015 and 2016

Career assessment (WGS), Wageningen, the Netherlands, 2017

Career orientation (WGS), Wageningen, the Netherlands, 2017

Additional activities

Writing and updating of research proposal, 2014

PhD study trip to Beijing and Shanghai, China, 2015

Weekly group meetings, 2013 – 2017

Project meetings (ENTHALPY), 2013 – 2016

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