


# Rethinking Membrane Processes for Food: From Particle Behavior to Innovative Membrane Cascades

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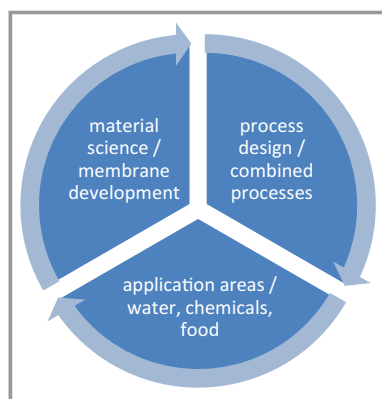
Within the food industry, membrane separation is commonplace due to its relatively low energy consumption. It allows fractionation of various feeds (e.g., milk) into starting materials for food design. We feel that considerable progress can still be made. For this, the specific properties of the components of interest would need to be taken into account, such as their mobility in flow, and their deformability in relation to the actual membrane structure. Furthermore, improvements are possible through cascaded use of membrane processes, and upgrading waste streams, which leads to new opportunities.

**Keywords:** Energy use, Membrane cascades, Membrane structure, Particle deformability, Particle migration

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## 1 Introduction


Unlike other separation processes, membrane separation, due to its flexible nature of operation, and modular design, is quite ultimately suited to fractionate the complex feed streams that may form the basis of a future bio-based circular economy [1]. Within the field of food, membrane fractionation is already commonplace [2–5], and the learnings taken from this field will be instrumental in gearing other fields in that direction [6–9]. Currently, membrane process design revolves around three aspects that are schematically shown in Fig. 1. In practice, material science/membrane development, (overall) process design, and the specific context in which the membranes are to be applied can be rather isolated worlds.



**Figure 1.** The inter-connectedness of the three fields that currently are distinguished, and together lead to successful membrane processes.

We feel that development of membrane processes does not necessarily revolve around an ideal membrane to do the job, since many good membranes are already available, and effects that occur due to process conditions used are much more influential on the success of operation. Furthermore, we note that within application fields, many accepted best practices exist but these may obscure other solutions that are available and have been presented decades ago. For example, the use of high turbulence to enhance mass transfer takes away attention from other solutions that lead to deposition free filtration, such as the critical flux concept [10], and does not take into account differentiation between concentration polarization and cake formation [11].

Membrane process design as currently carried out revolves around the evolution of flux as function of time, and the rejection/transmission of the components of interest. This leads to interpretation of effects that occur near the membrane. To substantiate that, some observation methods have been developed that allow analysis during filtration [12–14], and also considerable theoretical efforts have been done [15, 16]. Furthermore, miniature structures that either contain membranes, or emulate their behavior

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have been presented in literature, and reviewed [17, 18]. Interesting progress is in their use for solid [19, 20], as well as deformable particles [21–24], and 3D arrangement of membrane structures [25].

Within this paper, we will highlight aspects beyond the classical three from Fig. 1, and that in our view are currently underused in the field of food, more specifically:

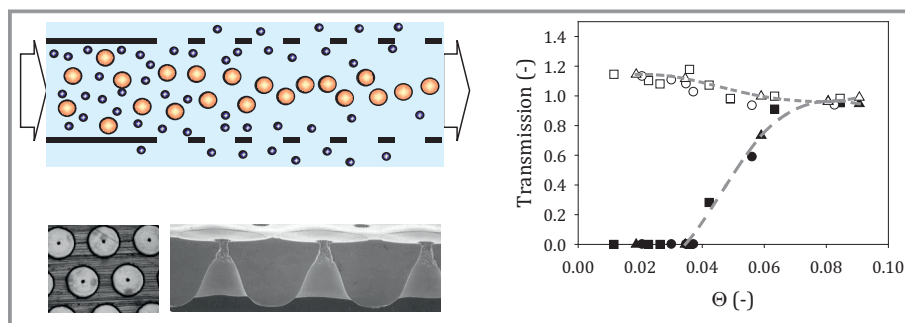
- particle behavior in flow can be used to improve filtration,
- deformability of components; not considered yet in membrane process design,
- cascaded membrane use; upgrading current processes in terms of energy,
- use of what are now considered waste streams.

These points will be addressed in the next sections, after which an outlook section follows with suggestions for improved membrane process design, both in terms of membrane, as well as process.

## 2 Particle Behavior in Flow

Earlier reviews summarized how particle migration may be used as a basis for innovative separation technologies [26–29]. For components relevant for food (typically micrometer-sized and smaller), three migration behaviors are relevant: molecular diffusion, shear induced diffusion, and inertial lift [30–38]. These effects have been reported long time ago, as is also evident from the years in which the works were written. Still, application of this knowledge is only very sparingly trickling down to the field of membrane separation. In contrast to that, particle migration is quite a vivid subject of investigation in the field of microfluidics, as summarized nicely in the reviews of [27, 39], and besides also used in other analytical techniques [40]. In the review of Kulrattanarak et al. [39], a comparison between microfluidic techniques and microfiltration is given in terms of volumetric productivity.

In our labs at University of Twente and Wageningen University, we investigated if effects related to particle migration, most notably shear induced diffusion, could be used to enhance filtration processes. At first, we used closed channels to investigate how migration took place in a mono-disperse dispersions [41], and later on also in bi-disperse systems [42]. We found that as theory predicts, the larger particles that we considered (size range 0.5–10 mm) migrate faster to the center of a channel compared to their smaller counterparts, as is schematically depicted in Fig. 2 (top left). This also implies that if this is the case, a ‘membrane’ with



**Figure 2.** Top left: schematic representation of shear induced diffusion, with larger particle moving faster to the center of a membrane channel. Bottom left: top view, and cross section of metal sieves used for filtration of emulsions that have 20 μm pores (top). Right: the filtration result for droplets of 2.7 μm (open symbols), and 5.3 μm (filled symbols) as function of the ratio cross flow versus transmembrane velocity ( $Q$ ), illustrating that droplets can be effectively separated based on size using their migration behavior [43, 44].

relatively large pores can be used to separate smaller particles from bigger ones, as long as the migration effect exceeds that of permeation.

We tested this further, and as model membranes we used metal sieve (from Stork VECO) with uniform pore size of 20 μm (see Fig. 2, bottom left entries, for a top view of the circular pores, and a cross section of their tapered substructure). We used these sieves under laminar flow conditions to allow shear induced diffusion to take place, and as feed we used, amongst others, emulsions of different sizes. The results shown in Fig. 2 (right panel), relate to emulsions with average droplet sizes of 2.7 μm (open symbols), and 5.3 μm (filled symbols). When filtration is carried out at low transmembrane pressure compared to the cross-flow velocity (low  $Q$ ), the concentration of small droplets is even higher than their concentration in the feed (transmission  $\sim 1.2$ ), while large droplets do not end up in the permeate at all. Upon increasing the transmembrane pressure eventually all droplets move through the sieve at the concentration in which they are present in the feed (Transmission = 1). This nicely illustrates that migration behavior can be used in separation processes, albeit rather different process conditions would need to be considered compared to what is nowadays standardly used in order to get this to work.

## 3 Deformability of Components

The particles/components that are present in a food product (ingredient) are typically flexible in their shape and size. They can swell or deswell, thus change their size without changing their shape, and in some cases also change their shape while keeping their volume (oil droplets), or even deform and loose or increase their volume (gels). These aspects are generally not taken into account when choosing a membrane for a specific separation. Mostly, the average membrane pore size relative to the size of the component of interest while dissolved in water determines this choice. If more information were available, this would put membrane

selection, and related to that the design of a membrane structure, at a next level.

In order to understand dynamic particle behavior during filtration better, a number of microfluidic techniques have been suggested as summarized in [18], as well as other non-invasive analytical tools [12,45]. In this extended abstract we summarize the work that was done using microfluidic devices containing 20 parallel pores with the same average size of 20 mm, and different entrance angles relative to the direction of permeation [20], (see Fig. 3, left bottom panel for the most extreme angles tested). These devices were used to evaluate the filtration of highly deformable polyacrylamide (pAAm) microgel particles with sizes ranging from 24 to 50 mm.

For single particle permeation, the result shown in Fig. 3, right panel, was obtained. For relatively small particles that are between 24 and 26 mm, but already considerably bigger than the pore (20 mm), the sphericity ( $\psi$ ) during permeation is close to 1, which implies that the particles remain spherical, but lose volume by deswelling. This occurs irrespective of the pore entrance angle; for a schematic depiction of the shape of the particles during permeation please see the left part of Fig. 3. For particles that are  $> 30$  mm the sphericity factor  $\psi$  is close to 0.9, which implies that these particles assume a dumbbell shape during permeation. They do so by first deswelling, and next deforming, and this occurs irrespective of the entrance angle of the pore. For intermediately sized particles we observe an effect of the entrance angle, with low entrance angles leading to lower sphericity, and higher entrance angles to higher sphericity, and less deformation as was the case at lower entrance angles. This nicely illustrates that deformability can have an extensive effect on permeation of particles, and thus also on the effectivity of a pore size to keep particles from permeating during a filtration process. It is clear that a size that is

measured in bulk liquid is not that informative in regard to permeation behavior [21,23].

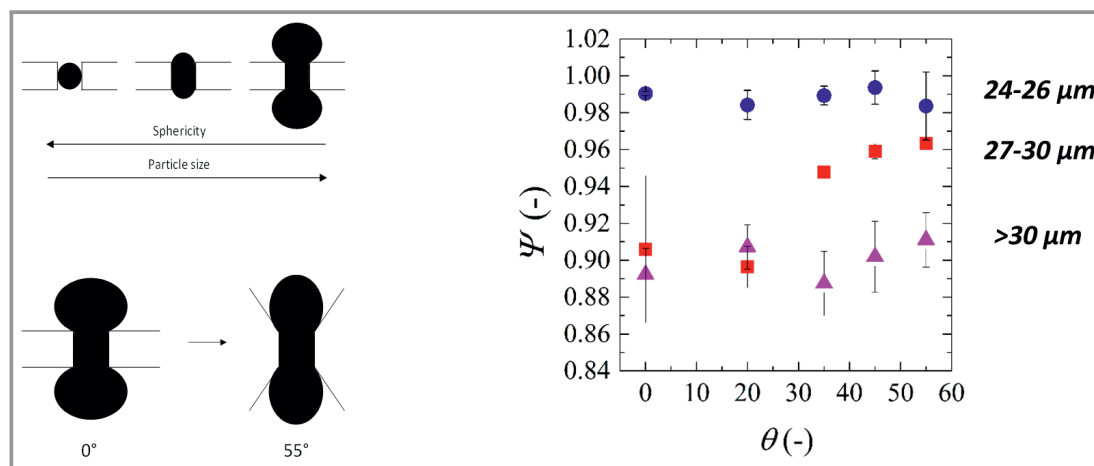
Furthermore, collective particle behavior has been investigated [20,22,24], also under cross-flow conditions, albeit not for soft particles [19], and it illustrates the flexibility of design of microfluidics to highlight various aspects related to filtration.

#### 4 Cascaded Membrane Use

We now move toward a different level at which membrane processes can be considered for fractionation of diverse feed flows. Depending on the composition of the feed, and clever arrangement of membrane separation systems, it is possible to obtain many different starting materials from different feeds such as milk amongst others for food production [5,46,47]. This actually holds much more generally for any bio-based feed stock, leading to many starting materials that fit into a circular economy approach, as well as isolation of valuable components, e.g., from a fermentation broth [3, 4, 6, 7, 9].

It is common practice to use membrane processes in close connection to each other to optimize productivity, reduce losses, and currently also more and more to reduce energy use. An interesting approach is the use of membranes in a cascaded setup in which the feeds and permeates are tuned in such a way that they optimally contribute to overall productivity. The concept of the ideal membrane cascade was proposed by Lightfoot [48] and applied in the food field amongst others for whey protein [49], and oligosaccharides [50].

Besides this, membranes can also be used to circumvent energy consuming heating steps, e.g., by removing bacteria from a product of interest. The bactocatch system that was



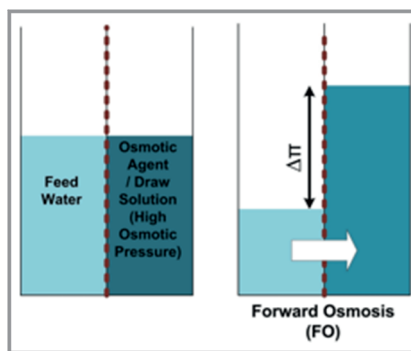
**Figure 3.** Left top: overview of observed behavior of deformable particles moving through pores perpendicular to their direction of movement, ranging from a sphericity factor of 1 (round) to 0.9 (extreme dumbbell shape). Left bottom: effect of entrance angle on deformation behavior. Right: overall sphericity behavior ( $\psi$ ) of deformable particles of various sizes moving through 20 mm pores with different entrance angles ( $\theta$ ) [21, 23].

already patented in the 1980ies is still going strong and has been shown to be an energetically (and exergetically) relevant method [51]. The results from an investigation from our own lab on the use of a combination of membranes to remove bacteria early on revolves around microfiltration and reverse osmosis [52]; results are summarized in Tab. 1 for a concentration factor of 2. We compared reverse osmosis that would remove bacteria completely, with a combination of microfiltration (higher flux and removal of bacteria) and reverse osmosis carrier out at two different temperatures. The addition of the microfiltration membrane leads to a small additional energy use when carried out at 15 °C but leads to energy reduction when used at 50 °C, mostly because of the redundancy of the heat exchanger. This leads to a lower energy requirement per liter permeate, simply because the process can be carried out faster. The resulting milk powder has similar quality as that obtained through regular processing, and was microbially safe, which nicely illustrates that smart combinations of membranes do pay off.

## 5 Use of Waste Streams

As a last innovative opportunity, we have considered the use of waste streams, in this case delactosed whey from a dairy plant [53], to concentrate skim milk through forward osmosis. For this relatively new concentration method a highly concentrated draw solution is used to remove water from a feed stream (Fig. 4, left panel). During this operation components from the draw solution are known to end up in the feed, and furthermore, the draw solution needs to be concentrated after use, which can be rather energy consuming.

In a study carried out at Teagasc (Ireland), two temperatures were tested, 10 and 30 °C, using equal amounts of feed and draw solution. As the concentration process proceeds, the driving force for water transfer reduces, and that is also seen in the increased energy consumption (pumps etc) at higher mass concentration factor (Fig. 4, right panel). The higher the mass concentration factor, the lower the remaining driving force, and the longer the process needs to be carried out to remove an additional liter of permeate. At lower temperature, the water flux is lower, and the process needs to be carried out for longer time, again leading to higher energy consumption per liter permeate. Further-



**Figure 4.** Left: schematic illustration of the forward osmosis principle. Right: result obtained with delactosed whey as draw solution expressed as energy consumption per kg water removed [53].

more, the energy consumption is lower for the higher temperature, and that is due to the much lower viscosity at this temperature. In this way, an interesting alternative for concentration was found that is low in energy usage compared to the results presented in the previous section [52], and also those of multi-stage evaporation. Because a waste stream is used, contamination of the feed is not an issue since all components are naturally occurring in milk. Besides, the diluted delactosed whey obtained after forward osmosis is at the right concentration to be used for ethanol fermentation. This shows that interesting options are available when looking at membrane processes from an overall perspective [53].

## 6 Conclusions and Outlook

There still seems a lot to be gained in the field of membrane separation. We have illustrated this by showing a number of examples that range from particle behavior in flow, particle behavior during filtration, smart combinations of membranes, and also making use of waste stream.

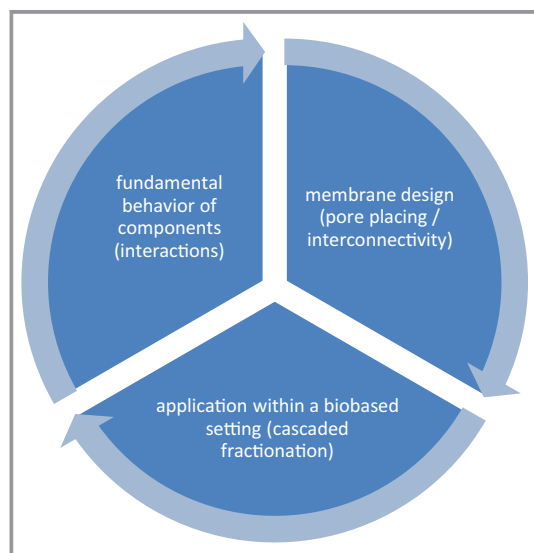
These are all examples that we have worked on over the years, and that may be a bit at the edge of current membrane research. Understanding particle behavior, for example, is expected to be very influential on how membrane processes need to be designed. Not only their behavior, but also their interactions, both between components and with the membrane. To get to these understandings at conditions

**Table 1.** Total power and energy consumption for reverse osmosis (RO), microfiltration (MF), and MF/RO combinations at 15 and 50 °C for a concentration factor of 2 for skim milk.

Membranes used	Temperature [°C]	Feed pump [kW]	Recirculation pump [kW]	Booster pump [kW]	Heat exchanger [kW]	Total power [kW]	Energy consumption [kJ L <sup>-1</sup> permeate]
RO	15	0.50	1.14	3.67	2.84	8.15	~400
MF/RO	15	0.52	1.23	3.87	2.94	8.56	~420
MF/RO hot	50	0.56	1.27	3.70	–	5.53	~178

relevant to the filtration system is essential, and for this microfluidic micromembranes could be of great help. This knowledge can subsequently be used to design membranes in terms of pore size, pore spacing, interconnectivity, etc., and thus used to tailor filtration processes to particle behavior.

In the end, taking into account not only the process in which membranes will be used but also the peculiarities and uniqueness of each component being separated can result in much more efficient processes, as illustrated in Fig. 5. We feel that adding these aspects to how we work as membrane technologists could lead to considerable gains, especially when viewed from within a circular bio-based economy setting.



**Figure 5.** The inter-connectedness of the three fields that are expected to lead to successful membrane processes bio-based circular processes.

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