



Biofouling on RO-membranes used for water recovery in the dairy industry

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ABSTRACT

Recovery followed by re-use of process-water obtained from dairy effluents by means of reverse osmosis technology is one route that can provide the dairy industry with the possibility to reach sustainable water regimes. However, membrane fouling is a phenomenon that limits both the efficiency and increases the running costs of such reverse osmosis units and can potentially alter the quality characteristics of permeate water. In this paper, several industrial-scale RO membranes used for recovery of process-water from whey UF permeate have been examined for their fouling tendency. At the end of a complete clean-in-place (CIP) protocol based on alkaline-acid formulations, biofouling appears to be the main issue in the investigated RO-elements. Between 4.19 and 5.69 log₁₀ (CFU cm⁻²) of viable microorganisms still remained on the membrane retentate surface and, more surprisingly, evidence of significant contamination was found on permeate side of these particular membranes. Microbiological analysis indicate that minor loads of microorganisms do pass into the permeate streams but final UV treatments ensured final process-water with non-detectable levels. There is a need for optimization of cleaning procedures and finding the best compromise for achieving surface disinfection while still preserving membrane integrity and not compromising the water quality.

1. Introduction

Climate change responsibilities, fresh water shortage, strict discharge regulations and financial gains are all factors that drive industries to seek out more sustainable practices and possibilities of water recovery in order to lower their effluents and water intake. Over the years, reverse osmosis (RO) membrane technology has demonstrated to be an excellent platform for recovery of water with high quality characteristics. Acting as a barrier to nearly all pollutants, the RO membranes produce water which can fulfill the strict quality regulations for public health and environment protection [1].

The food industries depend upon clean fresh water for a wide range of processes and cleaning operations and the implementation of safe and efficient water recovery strategies has become a major priority for many stakeholders [2]. Several examples of water recycling or reuse within the food sector have been reviewed by Vourch et al. [3] and Casani et al. [4]. Emerging originally as the biggest consumer and wastewater producer within the food area, the dairy industry is becoming one of the leaders in water reuse practices [5]. This is enabled primarily by their vast experience in applying membrane technology for concentrating or fractionating different liquid streams, but also due

to the great potential of recovering water from different process streams such as milk or cheese whey [6,7]. This trend is also stimulated by the large volumes of water required for cleaning [8]. Raw whey was traditionally considered a big biological pollutant. However, using various membrane technologies in series, such as ultrafiltration (UF) followed by RO and possibly a RO-polisher, it can now be valorized by harvesting high nutritional products – whey and lactose powder – and ultimately the liquid can be recovered as water which can qualify for reuse in production steps such as heat exchangers and clean-in-place (CIP) operations [9].

RO units primarily use reverse osmosis membrane elements with a spiral wound configuration. The main reason for this is their compact format, advantageous price, high membrane surface area in relation to their volume, and fitness for multiple applications [10]. Unfortunately, when processing a product with increased organic loads, such as permeates from whey separation, organic fouling and more notable biofouling are major risks that need to be minimized. Next to the potential safety risks, the biofilm formation can have a serious negative impact on the performance of the filtration system by lowering the trans-membrane flux [11] and cause decreased salt rejection [12]. Most published works conclude that biofouling appears to be an inevitable

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phenomenon, which dictates the need to develop strategies to keep it under control. These may include development of novel, more easily cleanable membrane configurations and materials or control of fouling of existing technologies by optimizing the cleaning formulation and operational conditions in relation to the characteristics of the feed product. However, the operation and maintenance costs combined with the risk of destroying the membranes structural integrity will increase if too frequent or too *harsh* cleaning is applied. An additional sanitation-in-place (SIP) step, right after CIP using e.g. peroxides or chlorine containing formulations, could shorten the lifetime of membrane elements considerably and is thus limited to special occasions. Like any technological solution in the food industry, a positive cost-benefit assessment will make the difference, and running costs of the equipment is an important aspect in this.

In spite of cleaning and technological improvements of membrane properties aimed to mitigate the issue, biofouling is still reported on whey concentration RO membranes by e.g. Hassan et al. (2009) [13], Adnan et al. [14] and Tang et al. [15]. Their findings showed that CIP was not entirely efficient in removing the resident microflora and could in the worst case result in cross-contaminating of the concentrated product. Similarly, when a new water recovery mode is added to the functionality of existing RO systems processing whey UF permeate, it becomes important to understand the biofouling trends, and even more, the impact on the RO permeates quality. The published work of Meneses and Flores [16] supports the feasibility and safety aspects of using the recovered process-water for cleaning purposes. Depending on the specific industry and application, the process water treatment operations may have multiple RO units in series and this is generally followed by UV treatment to further safeguard the microbiological quality.

Here we focus on the extent of biofouling on industrial-scale RO membrane elements used in recovering process-water from whey UF permeate. Operational conditions in industrial set-ups are in general complex due to factors which may be hard to be controlled, such as variable quality of whey UF permeate (batch-to-batch and dairy-to-dairy variations), unforeseen break-downs, leakages, or simply due to parameters that are occasionally tuned based on production demands. And even on a short term (i.e. weeks) the semi-continuous production chain in RO (-production-CIP-production-CIP-) has many unpredictable dynamic patterns where the previous run(s) can leave an imprint on the elements, influencing the present performance [17]. All these render any industrial RO-membrane to be exposed to a large set of varying conditions during their life-time, which are impossible to simulate realistically in laboratory studies. It is therefore important to derive operational practices based on observations attained from industrial-scale RO-membranes and evaluate the efficiency of industrial cleaning in removing any type of fouling. To do so, membrane autopsy is the best tool to gain accurate insights concerning fouling composition, patterns and their residing communities. In this study the condition of used industrial scale reverse osmosis membranes on both retentate and permeate surfaces will be characterized from a biofouling perspective in order to provide new knowledge which may help to improve practices for reusing process-water.

2. Methods and materials

2.1. Collection of RO membrane

A total of six RO-elements have been subjected to membrane autopsy and examined for their fouling tendencies. Elements are denoted alphabetically in Table 1 where their membrane specific characteristics can be found as well. With one exception, namely RO-E, all elements originate from the same processing plant, which uses cheese whey UF permeate trucked in from different dairies as a feed product for a two stage reverse osmosis system (RO plus RO-polisher) operated in a recovery mode for process-water, see Fig. 1a. The elements investigated were collected in particular from the first stage – RO – of the reverse

osmosis process. The collection has been conducted in two steps, namely Phase I (RO-A and RO-B) and six months later Phase II (RO-BB, RO-C, RO-D and RO-E). Phase I was initiated as an exploratory stage, meant to assess the extent of fouling at this particular RO unit. In this phase the first interest was to determine the level of actual surface fouling at the end of a production cycle that a CIP protocol would have to remove or reduce. For that purpose, element RO-A was collected prior to cleaning, after flushing with water. Also in Phase I, element RO-B was collected (at a different location/loop), after subjecting the entire unit to the current CIP protocol, which at that time used an alkaline-acid cleaning formulation. This formulation had been in place for more than one year. In Phase II all elements have been collected after having completed their CIP cycle. It should be noted that due to a management decision, shortly after Phase I the routine CIP protocol for the RO units has been changed to a new alkaline-acid cleaning formulation. In Phase II it was also possible to inspect the surface of element RO-E from a different production plant. It was in particular interesting to examine RO-E due to the heat sanitation step included in the routine CIP-SIP regime. Such a SIP step, in particular via hydrogen peroxide or by short heat treatments is a known efficient way to counteract biofouling trends. To further explore this option, element RO-C was taken from production and moved to a pilot unit to undergo a heat sanitation step. Given their industrial history of use, distinct locations within the unit and the before-mentioned complex process dynamics of membrane operations, each inspected RO-element should be considered as a snapshot of its independent scenario. Direct, quantitative comparisons can thus not be made; this is only possible to a minor extent for RO-B, RO-BB, RO-C and RO-D due to their origin being the same pressure vessel. All RO-elements were transported in polyethylene bags from the production site to the university on the day of their removal, and stored at 5 °C for up to 5 days until autopsy and analysis.

2.2. Membrane configuration and autopsy procedure

The RO membrane itself is structured in a three layer configuration, namely a thin polyamide layer (< 200 nm), deposited on top of a polyethersulfone porous layer (about 50 µm), placed on top of a non-woven fabric support sheet, see Fig. 2b. The polyamide top layer is responsible for the permeability of water and rejection of dissolved impurities, whereas the other layers give the membrane mechanical strength. Therefore, the concentration of the feed product takes place at the side of the polyamide layer which will be further named as retentate surface side, while the bottom support layer represents the permeate surface side.

When constructing them in the spiral wound elements (Fig. 2a), several membrane sheets are laid out alternating their display, namely one facing inward while the next membrane faces outward. Following this arrangement in the whole stack, the retentate sides of consecutive membranes end up facing each other, and similar sorting happens for the permeate sides. The edges of consecutive membranes are glued by sealing their permeate sides together, with a fine permeate spacer in between, leaving only one side to serve as an exit for the permeate stream. The outer sides of the glued membrane envelopes (the retentate sides) are also separated, this time by coarser feed spacers in order to ensure flow of the feed product. Finally a central tube is attached to the exit side of each of the resulting envelopes and rolled around the tube creating the spiral shape, see Fig. 2a, which is further enclosed in a polypropylene hard outer shell for the elements investigated.

The autopsy was initiated by cutting the polypropylene hard outer shell and exposing the membranes by unrolling all leaves from their connection to the central permeate tube. For each RO-element, two non-consecutive envelopes with sufficient distance between each other were investigated. Next, a series of large coupons of 10 × 10 cm size were cut aseptically along each leaf surface, as illustrated in Fig. 1b, and inspected for biofilm presence by means of confocal laser scanning microscopy (CLSM). All coupons were stored at 5 °C until the following

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