



Application of electric fields to clean ultrafiltration membranes fouled with whey model solutions



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ABSTRACT

In this work, the effectiveness of electric fields to clean two ZrO₂-TiO₂ ultrafiltration (UF) membranes fouled with three types of whey model solutions was investigated. Membranes tested had different molecular weight cut-offs (MWCOs) (15 and 50 kDa). Whey model solutions consisted of aqueous solutions of bovine serum albumin (BSA) at 10 g L⁻¹, a mixture of BSA (10 g L⁻¹) and CaCl₂ (1.65 g L⁻¹) and whey protein concentrate (WPC) (total protein content 45%) solutions at different concentrations (22.2, 33.3 and 150.0 g L⁻¹). The hydraulic cleaning efficiency (HCE) achieved by means of the application of the electric fields was evaluated as a function of the membrane MWCO and the operating conditions of the cleaning technique (applied potential, temperature of the cleaning solution and concentration of NaCl). The results demonstrated that the presence of NaCl favoured the removal of protein deposits on the membrane layer. On the other hand, the higher the temperature of the cleaning solution and the applied potential were, the higher HCE was achieved. Regarding the membrane MWCO, the permselective properties of the 15 kDa membrane were completely recovered after the cleaning procedure by electric field for all the feed fouling solutions tested, whereas this technique could not completely remove the protein deposits on the 50 kDa membrane when BSA solutions were used as feed.

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1. Introduction

Ultrafiltration (UF) is one of the most widely used techniques in dairy industries to dehydrate milk, concentrate whey and fractionate and purify proteins [1,2]. However, the implementation of membrane separation processes at industrial scale has a major limitation: membrane fouling. This drawback is due to the combination of several phenomena, such as concentration polarization, pore blocking or cake formation [3].

In dairy industries, proteins are one of the compounds mainly responsible for membrane fouling, because they can deposit on membrane surface and also, be adsorbed inside the membrane porous structure [4]. In addition, when whey and WPC solutions are ultrafiltered, the salts present in these solutions (especially calcium salts) can act as binding agents between proteins, favouring their aggregation and accumulation onto the membrane surface [5]. In order to minimize membrane fouling, several researchers

have investigated the interaction among proteins, between proteins and membranes and also, protein–inorganic compounds interactions [4–6]. Other authors studied different pretreatments focused on increasing protein solubility and limiting salt–protein bridging during the UF process [7].

Since pretreating the feed solutions used during the UF may not be enough to completely avoid membrane fouling, membranes have to be cleaned to remove the foulant deposits and restore their initial permeation properties. The conventional cleaning protocol employed when treating dairy solutions includes an alkali cleaning step followed by an acid cleaning stage. If this cleaning procedure cannot completely remove the protein deposits, a subsequent cleaning step using sodium hypochlorite or sodium dodecyl sulphate can be carried out [1,2,4]. However, as these procedures may be performed even once per day in dairy industries [8], the abovementioned conventional cleaning agents may damage the membranes, reducing their lifetime and causing morphological modifications. In addition, the discharge of these chemicals as wastewaters results in a negative environmental impact. For all these reasons, during the

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last years several researchers have focused their studies on the development and implementation of non conventional cleaning techniques, for instance, ultrasounds [9], saline solutions [10,11] or electric fields.

This last technique, the application of electric fields, has been used by other authors to improve permeate flux during the UF of different feed solutions. They demonstrated that the total hydraulic resistance achieved at the end of this process is reduced and concentration polarization is minimized [3,12–14]. This technique is based on two electrokinetic phenomena: on one hand, the charged particles move towards the electrode with opposite sign when the electric field is applied (electrophoresis) and, on the other hand, a liquid (usually water, as most of the times aqueous solutions are ultrafiltered) is forced to move to a charged surface (for example, the membrane pores), which is known as electro-osmosis. Both effects, electrophoresis and electro-osmosis, are achieved by placing two electrodes at both sides of the membrane or using only one electrode, being the membrane the other one. This last case is very often used in the case of ceramic membranes, as they are made of electrically conductive materials [15].

Zumbusch et al. [3] investigated the utilization of alternating electrical fields to reduce membrane fouling during the UF of biological suspensions and studied the effect of several operating conditions (field strength, protein concentration and conductivity) on fouling decrease. Although both direct and alternating current can be used, the former is suitable only when the particles in the feed fouling solution have a uniform charge. They reported that high field strength and an increase in conductivity up to the limiting electrolytic current led to a more effective cleaning procedure. However, the increase in protein concentration reduced the effect of the electric field applied. Tarazaga et al. [12] used electric field pulses of 2–3 min to restore the initial membrane permeate flux during the filtration of bovine plasma at a concentration of 0.5% w/w at a pH of 7.8. They applied three different potentials (10, 15 and 30 V) and demonstrated that the higher the electric potential was, the greater the permeate flux was after the electric pulses. Holder et al. [14] investigated the effect of electric fields on the fractionation of bio-functional peptides from micellar casein hydrolysate. After the UF experiments, these authors reversed the polarity of the electrodes in order to study the effectiveness of electric fields to clean the membranes. They indicated that this technique was able to completely remove some peptides deposited on membrane surfaces because Van der Waals forces also influenced the fouling process.

Although there are several works available in the literature focused on the application of electric fields, they applied electric pulses during the feed solution filtration to recover the permeate flux once it decreased up to a certain value or to minimize the concentration polarization phenomenon. However, only a few papers deal with the application of this technique during the cleaning step, i.e. after the membrane was fouled by the feed solution treatment [14]. The main goal of this work is to evaluate the effectiveness of a physical cleaning procedure based on the application of electric fields to clean membranes previously fouled with whey model solutions. In addition, the effect of different cleaning operating conditions, such as applied potential, temperature of the cleaning solution and concentration of NaCl used as electrolyte, on the efficiency of the cleaning procedure was determined. The novelty of this work lies in the application of the electric fields during the cleaning step in order to remove the irreversible fouling caused on the membranes and not during the fouling stage as other authors reported to minimize fouling and the concentration polarization phenomena [12,16].

2. Materials and methods

2.1. Chemicals

Whey model solutions used during the fouling step consisted of BSA (10 g L^{-1}), BSA (10 g L^{-1}) with CaCl_2 (1.65 g L^{-1}) and WPC (22.2 , 33.3 and 150.0 g L^{-1}) aqueous solutions. As these products were supplied in powder form, a certain amount was weighted and dissolved in deionized water until the desired concentration was achieved. Renylat WPC solutions were supplied by Industrias Lácteas Asturianas S.A., Spain, BSA (lyophilized powder after heat shock fractionation, 98% purity, A3733) was provided by Sigma-Aldrich (Germany) and CaCl_2 (95% purity) was purchased from Panreac (Spain). The main components of the WPC used are shown in Table 1. The methods employed for determining the concentration of each component are described elsewhere [17]. The evolution of zeta potential with pH is depicted in Fig. 1 for both BSA and WPC solutions. As it can be inferred from this figure, the isoelectric points of BSA and WPC are, respectively, $4.9 \pm 1.42 \text{ mV}$ and $4.6 \pm 0.47 \text{ mV}$. These values are in a very good agreement with those reported by the BSA manufacturer and in the literature for both solutes [18–20]. As it can also be observed from Fig. 1, BSA and the main proteins in WPC were negatively charged at the pH values of the feed solutions used in the experiments (around 7).

Previous authors [21,22] reported the utilization of BSA and WPC solutions as whey model solutions for UF tests. In order to study the influence of salt presence on protein behaviour, CaCl_2 was one of the salts most often used as calcium ion favours protein–protein interactions and Cl^- is the main anion in whey and WPC [5,6,11].

Finally, NaCl (Panreac, Spain) aqueous solutions were used to clean the membranes in combination with the application of electric fields. In addition, NaOH (98% purity, Panreac, Spain) aqueous solutions were used to clean the UF membranes if the permselective properties of the original membranes were not recovered at the end of the cleaning protocol.

2.2. Membranes

Two monotubular $\text{ZrO}_2\text{-TiO}_2$ INSIDE CÉRAM™ membranes of 15 and 50 kDa (TAMI Industries, France) were used to perform the experiments. The dimensions of these membranes were a length of 20 cm, an internal diameter of 0.6 cm and an external diameter of 1 cm. Their effective area was 35.5 cm^2 . It is important to highlight that these membranes acted as a cathode during the cleaning step.

2.3. Experimental set-up

All fouling and cleaning tests were carried out in a VF-S11 UF plant (Orelis, France). This plant was equipped with a 10 L feed

Table 1
Main components of the Renylat WPC used as feed solution.

Component	Dry basis concentration (% w/w)
Dry matter	93.66 ± 0.95
Proteins	40.74 ± 0.79
Lactose	38.27 ± 0.49
Fat	8.14 ± 0.20
Ash	7.85 ± 0.07
Ca	0.79 ± 0.06
Na	1.21 ± 0.09
K	1.42 ± 0.02
Cl	4.07 ± 0.24
$\text{PO}_4\text{-P}$	0.37 ± 0.03

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