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 ZrO_2-TiO_2 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 $g \cdot L^{-1}$, a mixture of BSA (10 $g \cdot L^{-1}$) and $CaCl_2$ (1.65 $g \cdot L^{-1}$) and whey protein concentrate (WPC) (total protein content 45 %) solutions at different concentrations (22.2, 33.3 and 150.0 $g \cdot L^{-1}$). 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.

Keywords: Ultrafiltration; membrane cleaning; electric fields; whey model solutions.

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, among others [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 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 $g \cdot L^{-1}$), BSA (10 $g \cdot L^{-1}$) with $CaCl_2$ (1.65 $g \cdot L^{-1}$) and WPC (22.2, 33.3 and 150.0 $g \cdot L^{-1}$) 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 ± 1.42 mV and 4.6 ± 0.47 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 ZrO_2-TiO_2 INSIDE CéRAM^{TM} 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 tank, a variable speed volumetric pump that allowed the crossflow velocity to be maintained constant, two manometers placed at the inlet and outlet streams of the membrane module to measure the transmembrane pressure, a temperature regulating system to control the temperature during the fouling and cleaning stages and a scale (±0.001 g accuracy) to gravimetrically determine the permeate flux.

The abovementioned membranes were placed in a Plexiglas GS® tubular membrane module (Metaval Abella S.L., Spain) and rolled on their external surface by a copper wire to ensure a constant potential distribution on this membrane side. Then, the external membrane surface was connected to the cathode. The second electrode (anode) consisted of a titanium electrode with an iridium coating (MAGNETO Special Anodes B.V., The Netherlands). The anode was placed inside the membrane, crossing it along the tubular channel. Both cathode and anode were connected to a direct current supplier (Konstanter SSP, Gossen, Germany). It is important to highlight that both electrodes were situated in

176 the position aforementioned in order to promote protein migration from the membrane
177 active layer to the bulk solution, due to the negative charge of most of the whey proteins in
178 the feed fouling solutions. Experiments with the electric fields were performed in
179 potentiostatic mode. The experimental set-up is shown in Fig. 2.
180
181 **2.4. Experimental procedure**
182
183 Firstly, membranes were fouled with the different feed solutions at a transmembrane
184 pressure of 2 bar, a crossflow velocity of 2 $m \cdot s^{-1}$ and a temperature of 25 ºC, according to
185 previous studies about protein UF [17, 23].
186
187 After the fouling step, membranes were rinsed with deionized water during 30 min at a
188 transmembrane pressure of 1 bar and 4.2 $m \cdot s^{-1}$. Several studies reported that low
189 transmembrane pressures and high crossflow velocities favour the removal of proteins
190 deposited on membrane surfaces [8, 24]. Then, a cleaning procedure was carried out at the
191 same transmembrane pressure and crossflow velocities as the rinsing step and varying the
192 applied potential (0, 15 and 30 V), the NaCl concentration (0 and 5 mM) and the
193 temperature of the cleaning solution (25-50 ºC) at a pH of 7. In order to avoid the use of
194 conventional cleaning agents during the cleaning step as much as possible, pH of the
195 cleaning solutions used (deionized water and NaCl solutions) was not adjusted nor varied.
196 These conditions were selected according to other works about membrane cleaning by
197 means of electric fields and saline solutions [12, 25]. Finally, membranes were rinsed again
198 with deionized water to remove the loose protein deposits from the membrane surface as
199 well as the cleaning agents. During all these steps, both the permeate flux and the hydraulic
200 resistance were determined.

An additional conventional cleaning step was performed when needed if the initial membrane hydraulic resistance was not completely recovered after the cleaning procedure. This step was performed with NaOH at a temperature of 50 ºC and pH values about 8.5-9. These conditions were selected to avoid damage of the electrodes or the membrane module.

2.5. Evaluation of the cleaning efficiency

Due to the destructive nature of the chemical methods to evaluate the efficiency of the cleaning procedure, which consist of the determination of chemical species on the membrane structure by spectroscopic techniques, a hydraulic method was used to calculate the efficiency of the cleaning protocol (HCE). Several authors reported different equations to determine the HCE from the resistance of the membrane after the rinsing and cleaning steps and to the original membrane resistance [9, 23, 26]. The values of the membrane resistances after the abovementioned steps were calculated by the Darcy's law (Eq. 1).

$$J = \frac{\Delta P}{\mu \cdot R_m} \qquad \text{Eq. 1}$$

The efficiency after the end of the cleaning protocol was estimated using Eq. 2.

$$\text{HCE}\,(\%) = \left(\frac{R_f - R_{r2}}{R_f - R_m} \right) \cdot 100 \qquad \text{Eq. 2}$$

Where HCE is the hydraulic cleaning efficiency, R_f is the resistance at the end of the fouling step, R_{r2} is the resistance at the end of the second rinsing and R_m is the resistance of the original membrane.

3. Results and discussion

3.1. Cleaning of membranes fouled with BSA solutions

3.1.1. Results of the 15 kDa membrane

Fig. 3 shows the values of HCE obtained for the 15 kDa membrane using two different cleaning solutions: deionized water at different temperatures (25 and 50 ºC) and NaCl solutions at a concentration of 5 mM and three different temperatures (25, 37.5 and 50 ºC). NaCl concentration was selected according to a previous work by the authors dealing with membrane cleaning by means of saline solutions. These experiments were performed at three different electric field potentials (0, 15 and 30 V) in order to check the influence of both temperature and applied potential on the HCE.

As it can be observed from Fig. 3, an increase in temperature and applied potential during the cleaning step resulted in an increase in the values of HCE achieved. This pattern was previously confirmed by other authors [12, 15, 25, 27, 28].

Tarazaga *et al.* [12] demonstrated that an increase in the electric field potential caused an increase in the permeate flux obtained during the membrane fouling with bovine plasma solutions. Chen *et al.* [27] reported that electric field strengths greater than 15 V resulted in

249 a dramatic decrease of the hydraulic resistance when membranes were fouled with sewage
250 water. This should be due to the greater amount of charged particles migrating from one
251 electrode to another when high electric field potential was applied. This effect was also
252 demonstrated by Shi *et al.* [15] and Huotari *et al.* [28] using oily wastewaters during the
253 membrane fouling. According to these authors, the electrophoretic forces increase as the
254 electric field potential increases. These forces are ascribed to the movement of charged
255 particles towards the electrode of opposite sign. In their works, this electrode is placed on
256 the bulk solution channel. For all these reasons, HCE increased as the electric potential
257 increased.
258
259 On the other hand, Corbatón-Báguena *et al.* [25] tested different temperatures during the
260 cleaning of several UF membranes with deionzed water as well as NaCl solutions. In all
261 cases, an increase in temperature resulted in an increase in the values of HCE achieved.
262 This trend was corroborated by Lee and Elimelech [10], who demonstrated that the mass
263 transfer process as well as the chemical reactions velocity increased when temperature
264 increased, favouring the weakness of the fouling layer on the membrane surface and easing
265 its removal. In addition, temperature has a great effect on solution viscosity [29, 30]. For
266 instance, Jawor and Hoek [30] demonstrated that an increase in temperature of the feed
267 solution from 25 to 35 ºC decreased solution viscosity while increased the diffusion rate of
268 salt ions through the membrane and back to the bulk solutions. This increase in
269 temperature also loosened membrane structure and thus, it became more permeable and
270 solute diffusion was enhanced. Therefore, concentration polarization effect was minimized
271 when increasing temperature and thus, membrane cleaning was favoured at high
272 temperatures. Regarding the effect of the saline solutions, results shown in Fig. 3 indicated
273 that higher HCE values were obtained when cleaning was carried out in presence of NaCl

at low concentrations (5 mM). When an electric field is applied on NaCl aqueous solutions, different cathodic and anodic reactions occurred on the electrode surfaces [31]:

Cathode: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ Eq. 3

Anode: $2Cl^- \rightarrow Cl_2 + 2e^-$ Eq. 4

$4OH^- \rightarrow O_2 + 2H_2O + 4e^-$

Regarding the electrochemical reactions of the electrolysis of water, the cathodic reaction of hydrogen formation (Eq. 3) is the only one that takes place in the system previously described. This means that the anodic reaction that preferentially occurs is the chlorine formation (Eq. 4). When chlorine molecules are in contact with water molecules, the formation of hypochlorite occurs, as in the following reaction [31]:

$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-$ Eq. 5

Hypochlorite formed in this last reaction oxidizes the organic pollutant species (proteins in this work), breaking their bonds to partially decompose them and favouring their removal from the system [32]. This technique is known as indirect electrochemical oxidation and has been successfully implemented in the treatment of different organic effluents [33, 34]. Therefore, the electrochemical effect due to the presence of NaCl 5 mM in the cleaning solution that enables the formation of chlorine in the anodic reaction (Eq. 4) and thus, the formation of hypochlorite (Eq. 5), is added to the electrophoretic effect due to the application of electric fields. As a consequence of the combination of both electrochemical and electrophoretic effects, the HCE values obtained using NaCl 5 mM as electrolyte were greater than those obtained with deionized water.

299

300 Therefore, the best operating conditions to clean the 15 kDa membrane fouled with BSA
301 solutions were a temperature of 37.5 ºC, an electric field potential of 30 V and a NaCl
302 concentration of 5 mM, taking into account the boundary experimental conditions tested.
303 When cleaning with NaCl solutions was performed without applying electric fields, the
304 salting-in behaviour of NaCl was the only effect that promotes the protein removal from
305 membrane surface [25]. For this reason, harder operating cleaning conditions in terms of
306 temperature (50 ºC) were required to achieve the same HCE than that obtained when
307 electric fields and salt addition were combined. This fact confirms that the combination of
308 electric fields and NaCl addition is a more efficient membrane cleaning procedure because
309 it has two main advantages: firstly, the application of an electric field promotes the
310 movement of charged proteins from the membrane surface to the bulk cleaning solution,
311 favouring their removal; and secondly, the in-situ formation of hypochlorite in the anode
312 (Eq. 5) due to the presence of NaCl in the cleaning solution results in the oxidation of such
313 proteins.

314

315 3.1.2. Results of the 50 kDa membrane

316

317 Fig. 4 shows the effect of temperature of the cleaning solution on the HCE values obtained
318 when the 50 kDa membrane was fouled with BSA solutions and cleaned with different
319 cleaning agents and different electric potentials: NaCl at a concentration of 7.5 mM at 0
320 and 30 V and NaOH at a concentration of 5 $g \cdot L^{-1}$. These cleaning agent concentrations
321 were selected according to the range of pH recommended by the manufacturer to clean this
322 membrane and previous works about salt cleaning of UF membranes by the authors [25].

323

Figs. 3b and 4 show the influence of temperature and electric field potential on the HCE for the 15 and 50 kDa membranes, respectively. Thus, the effect of the same parameters on the cleaning efficiency of both membranes can be compared. As it is shown in Fig. 3b, greater HCE was obtained for the 15 kDa membrane when the cleaning procedure was performed at 30 V and 37.5 ºC in presence of NaCl (HCE around 100 %) than that obtained for the 50 kDa membrane at the same electric field voltage and temperature, achieving HCE values of 85 % in this case (see Fig. 4). Therefore, harder experimental cleaning conditions in terms of electric field potential and temperature are necessary to clean the 50 kDa membrane. It is worthy to note here that, greater NaCl concentration was considered in the case of the 50 kDa membrane. As the authors reported in a previous work about membrane cleaning with salt solutions [25], the optimal NaCl concentration to clean the 15 kDa membrane fouled with 10 $g \cdot L^{-1}$ BSA solutions was between 2.5 and 5 mM. However, in the case of the 50 kDa membrane fouled with the same solutions, a NaCl concentration of 7.5 mM was required to clean the membrane. This difference in NaCl concentration is due to the more severe fouling that proteins cause on the 50 kDa membrane. The reason for that is the similar size between membrane pores (50 kDa) and BSA molecules (67 kDa), which favours that these molecules completely block the membrane pores and/or penetrate inside its porous structure, as it was reported by other authors [35]. However, permeate flux decline was much lower for the 15 kDa membrane [25]. Therefore, fouling was less severe for this membrane and easier to remove. For all these reasons, greater values of HCE were obtained for the 15 kDa membrane at the same electric field potential and temperature and at lower NaCl concentrations than in the case of the 50 kDa membrane.

As it can be observed, the highest cleaning efficiency was achieved when the conventional cleaning with NaOH solutions was performed. Regarding the cleaning with NaCl solutions, negligible differences in HCE were observed between the cleaning protocol carried out at 37.5 ºC and 30 V and that performed at 80 ºC and 0 V. This fact demonstrated the greater efficiency reached when electric fields were applied, as it was reported for the 15 kDa membrane (Fig. 3). Higher temperatures are required if no electric fields are used to reach the same HCE. Despite the good results obtained when the electrochemical oxidation took place, this technique was not as efficient as the conventional cleaning protocol, even when higher temperatures were used to facilitate protein removal. This may be due to the fact that the amount of hypochlorite formed when an electric field was applied was too low to completely clean this membrane. In a previous work by the authors where this membrane was used to ultrafilter BSA solutions, it was observed that, this membrane shows a very sharp permeate flux decrease at the beginning of the UF process, which indicates severe membrane fouling [25]. As it was above reported, fouling was less severe for the 15 kDa membrane and easier to remove in comparison with the 50 kDa membrane.

3.2. Cleaning of the 15 kDa membrane fouled with whey model solutions

As electric fields were not able to completely restore the initial membrane permeselective properties in the case of the 50 kDa membrane, only the 15 kDa membrane was used to test the effectiveness of the electrochemical process when whey model solutions (BSA with $CaCl_2$ and WPC solutions at different concentrations) were employed as feed during the fouling step. It is expected that, according to the pattern observed for HCE when the 15 kDa membrane was fouled with BSA + $CaCl_2$ and WPC solutions (see Fig. 5), even lower

373 values of HCE were going to be achieved if these feed solutions were used to foul the 50
374 kDa membrane. When these feed solutions (BSA + $CaCl_2$ and WPC solutions) are
375 ultrafiltered using the 50 kDa membrane, a more severe membrane fouling is expected to
376 occur due to the presence of inorganic salts and two major whey proteins (α-lactalbumin
377 and β-lactoglobulin). These proteins have molecular sizes of 14 and 18 kDa, respectively,
378 and tend to form dimers at the neutral pH of the WPC solutions tested [36, 37]. This fact
379 causes their molecular sizes to increase and thus, their size is more similar to that of the
380 membrane pores (50 kDa) and may seal the pore entrance or block the pores internally.
381 Therefore, the study of the influence of the feed fouling solution on HCE was not
382 continued with the 50 kDa membrane.
383
384 The same cleaning operating conditions that resulted in the best HCE values when the
385 membrane was fouled by BSA (1 bar, 4.2 $m \cdot s^{-1}$, 37.5 ºC, 30 V and a NaCl concentration of
386 5 mM) were tested with the 15 kDa membrane once it was fouled with BSA and $CaCl_2$ and
387 WPC (22.2 $g \cdot L^{-1}$) solutions. The results shown in Fig. 5 demonstrated that the maximum
388 HCE achieved was about 90 % in both cases at the experimental conditions tested. This is
389 due to the more severe membrane fouling caused when salts are introduced in the protein
390 solution. These salts can act as bridging agents between proteins, aggregating them and
391 favouring its deposition on the membrane surface. This behaviour was previously reported
392 by other authors [5, 38].
393
394 In order to improve the efficiency of the cleaning process, the 15 kDa membrane was
395 cleaned at three different temperatures within the range 37.5-50 ºC. Fig. 6 shows the
396 evolution of HCE with temperature for the BSA with $CaCl_2$ and WPC (22.2, 33.3 and
397 150.0 $g \cdot L^{-1}$) solutions.

As it can be observed, HCE values about 100 % were obtained at temperatures of 43.8 and 50 ºC. Therefore, an increase in the cleaning solution temperature caused an increase in the HCE as it had been previously observed when this membrane was fouled with BSA solutions (Fig. 3). Therefore, the best operating conditions to carry out the cleaning protocol of the 15 kDa membrane fouled with whey model solutions were 1 bar, 4.2 $m \cdot s^{-1}$, 30 V, 43.8 ºC and a NaCl concentration of 5 mM. As it was expected, the higher the protein concentration in the feed solution was, the more severe the membrane fouling was due to the greater protein aggregation and accumulation on the membrane surface. This fact was previously reported by the authors in works about membrane fouling characterization and modelling in the case of UF membranes fouled with whey model solutions [17]. Therefore, the more severe fouling caused by an increase in protein concentration decreased the HCE achieved with the cleaning procedure, requiring harsher operating conditions (higher temperature) to achieve similar HCE values [3].

3.3. Analysis of cost

In order to ensure that the proposed physical cleaning procedure by means of electric fields was competitive compared to a conventional chemical cleaning, an analysis of costs was performed for both cleaning methods. For the conventional cleaning, as it can be observed in Fig. 4, the best operating conditions (HCE values of around 100 %) were a transmembrane pressure of 1 bar, a crossflow velocity of 4.2 $m \cdot s^{-1}$, a NaOH concentration of 5 $g \cdot L^{-1}$ and a temperature of the NaOH solution of 50 ºC during an hour. These last conditions (NaOH concentration and temperature of the cleaning solution) were recommended by the membrane manufacturer to completely clean both ceramic

membranes (15 and 50 kDa). On the other hand, the optimal operating conditions to perform the physical cleaning by electric fields were an electric field potential of 30 V, a NaCl concentration of 5 mM and a temperature of the NaCl solution of 43.8 ºC as maximum. The rest of operating conditions remained the same (transmembrane pressure, crossflow velocity and duration of the cleaning step). Therefore, in order to compare both cleaning procedures, only the influence of cleaning agents, heating and electric fields generation costs were considered. It is worthy to note that the heating cost was calculated from the energy consumed by the electrical resistance to heat the feed solution from room temperature to the cleaning solution one (50 and 43.8 ºC for the conventional cleaning and physical cleaning, respectively). In the same way, the electric fields generation costs were determined considering the energy consumed to apply the selected electric field potential (30 V) and the maximum current intensity achieved at those conditions (2 A). The results of this comparison are summarized in Table 2. As it can be observed, the chemicals cost was the key item. As it was above mentioned, the NaOH concentration required to perform the conventional cleaning (5 $g \cdot L^{-1}$) was much higher than that of the NaCl needed to carry out the physical cleaning (5 mM, i.e. 0.24 $g \cdot L^{-1}$). In addition, the cost of NaOH is greater than that of NaCl, according to the provider of both chemicals. Therefore, performing a physical membrane cleaning by means of electric fields and using NaCl as electrolyte is a cost-effective procedure in comparison with the conventional NaOH membrane cleaning, at the experimental conditions considered for each type of cleaning.

4. Conclusions

- Cleaning by means of the application of electric fields combined with the addition of NaCl solutions was effective to completely restore the 15 kDa membrane initial

permeation properties when it was used to treat different whey model solutions. However, the 50 kDa membrane could not be completely cleaned by this cleaning procedure, probably due to the more severe fouling that proteins caused in this membrane.

- Results demonstrated that the higher the temperature of the cleaning solution as well as the electric potential were, the higher HCE values were achieved.
- The presence of NaCl at low concentrations (5 mM) favoured membrane cleaning, obtaining HCE values about 100 % at mild temperatures (37.5-50 ºC) for the 15 kDa membrane. This fact is due to the electrochemical oxidation process that occurs when NaCl is used as electrolyte and transformed to hypochlorite by the application of electric fields.
- The best operating conditions to clean the 15 kDa membrane fouled by whey model solutions were a NaCl concentration of 5 mM, a transmembrane pressure of 1 bar, a crossflow velocity of 4.2 $m \cdot s^{-1}$, a electric field potential of 30 V and a temperature around 43.8 ºC.

Acknowledgements

The authors of this work wish to gratefully acknowledge the financial support from the Spanish Ministry of Science and Innovation through the project CTM2010-20186 and the company MAGNETO Special Anodes B.V. for supplying the Ti-Ir electrode.

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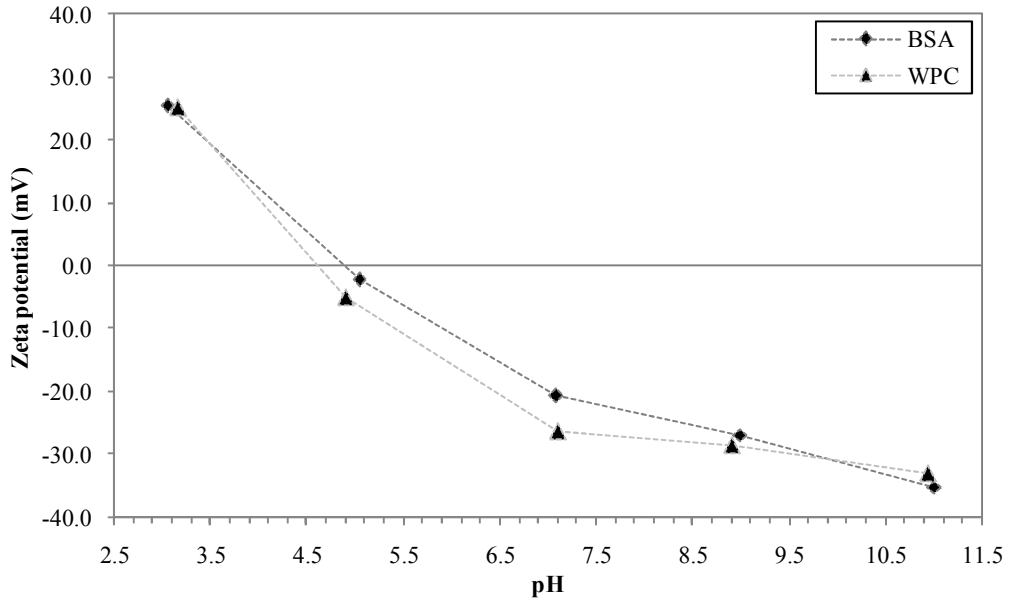


Fig. 1. Evolution of zeta potential with pH for BSA and WPC solutions.

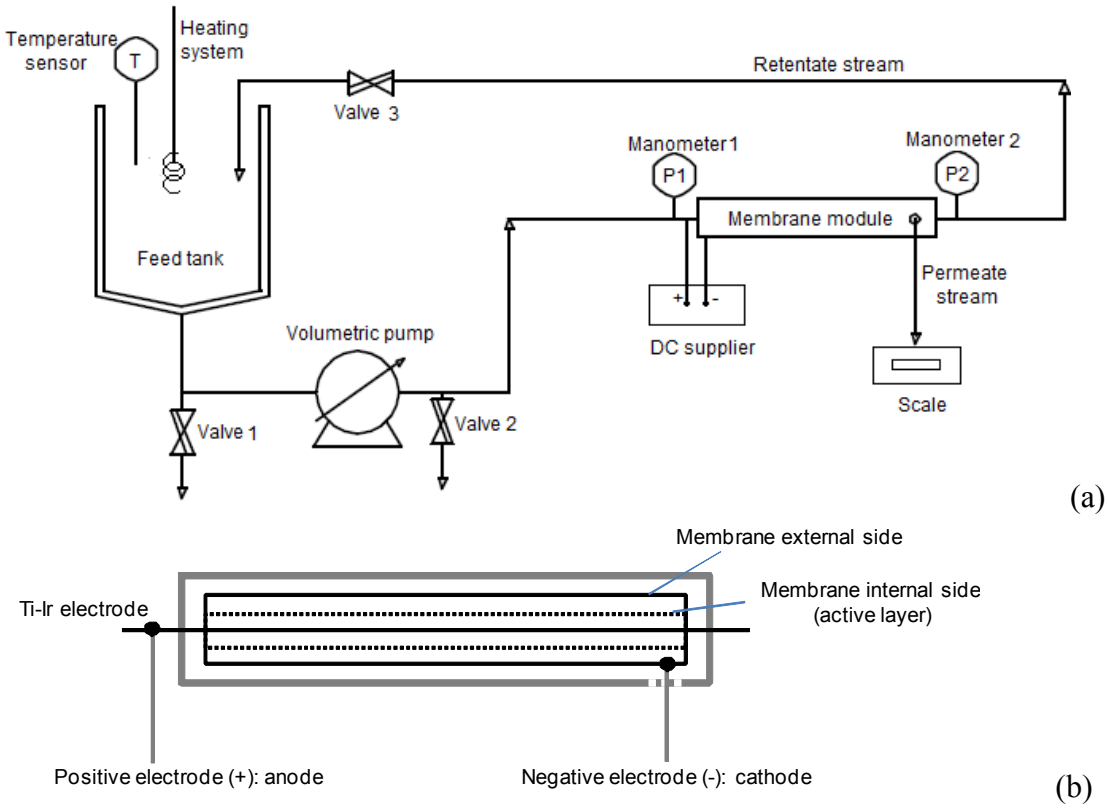
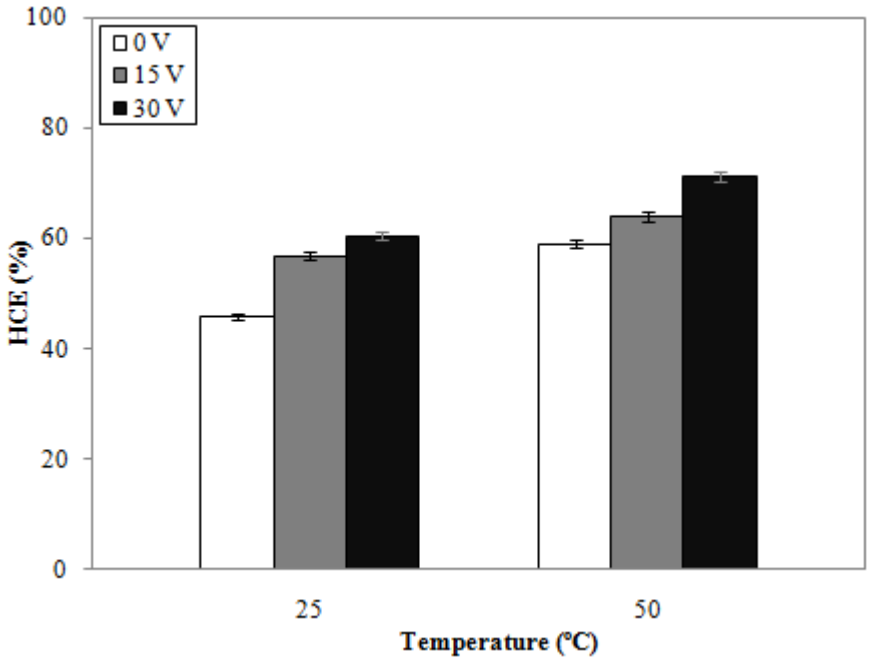
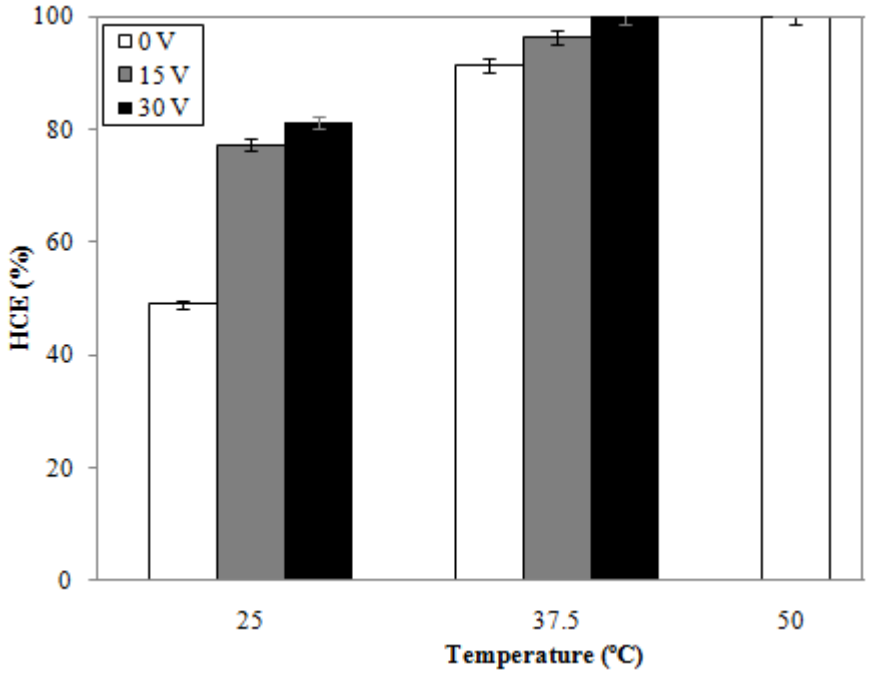


Fig. 2. Schematic representation of the VF-S11 UF plant connected to a direct current (DC) supplier (a) and electrodes connection in the membrane module (b).



(a)



(b)

Fig.3. Influence of temperature of the cleaning solution and electric field potential on HCE for the 15 kDa membrane using (a) deionized water and (b) NaCl at a concentration of 5 mM as cleaning solution (fouling solutions: BSA; operating conditions during cleaning: 1 bar and 4.2 $m \cdot s^{-1}$).

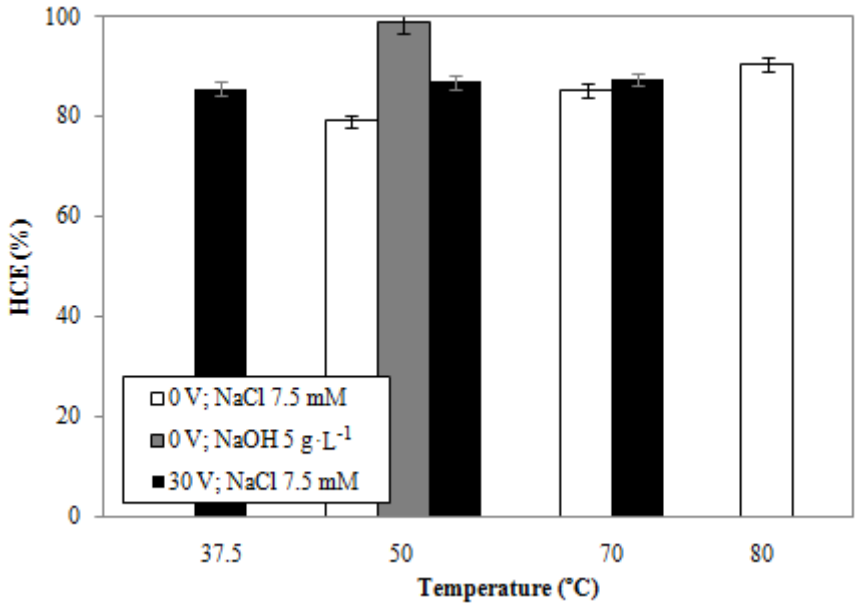


Fig. 4. Influence of temperature of the cleaning solution and electric field potential on HCE for the 50 kDa membrane using different cleaning agents (fouling solution: BSA; operating conditions during cleaning: 1 bar and 4.2 $m \cdot s^{-1}$).

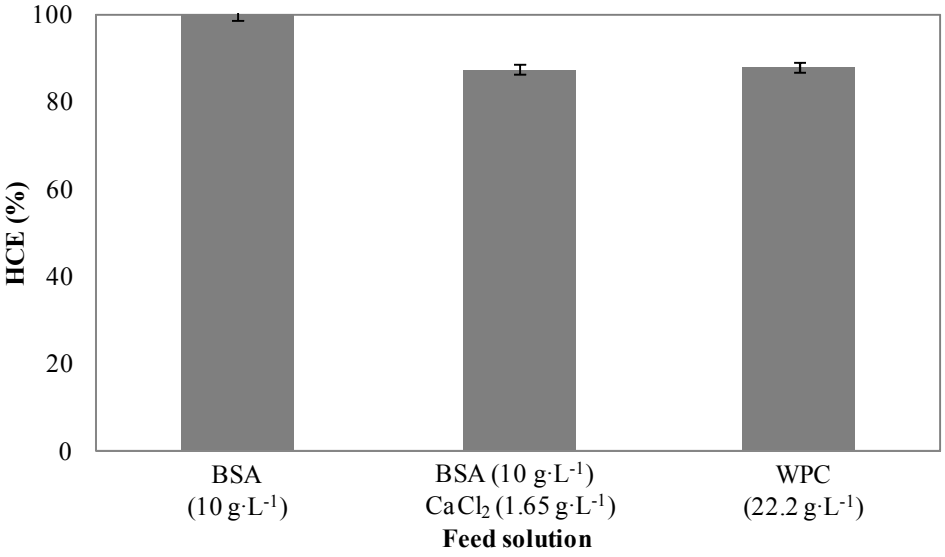


Fig. 5. Influence of feed solution composition during the fouling step on HCE for the 15 kDa membrane (operating conditions during cleaning: 1 bar, 4.2 $m \cdot s^{-1}$, 37.5 ºC, 30 V and 5 mM NaCl).

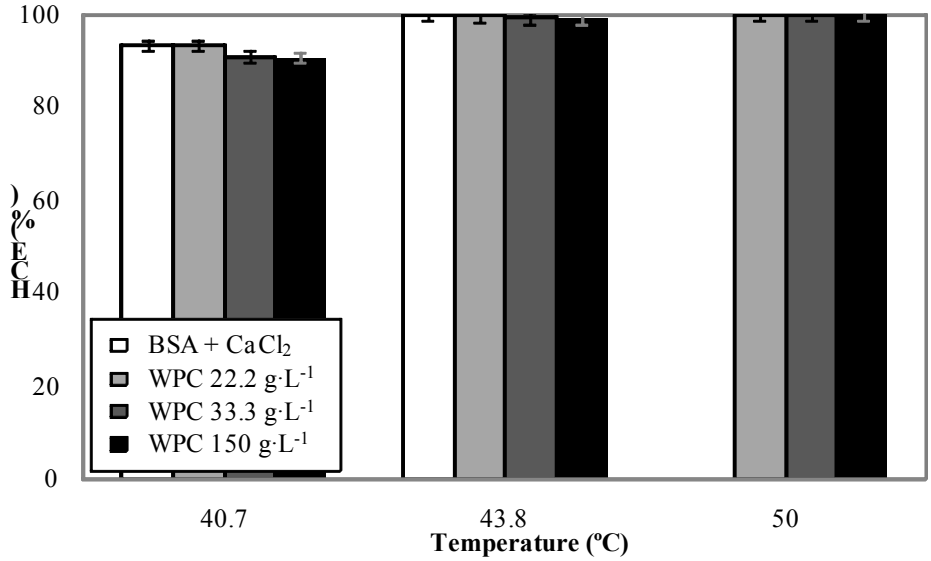


Fig. 6. Influence of temperature of the cleaning solution on HCE for the 15 kDa membrane (operating conditions during cleaning: 1 bar, 4.2 $m \cdot s^{-1}$, 30 V and 5 mM NaCl).

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
PO_4-*P*	0.37 ± 0.03

Table 2. Cost comparison between conventional chemical cleaning and physical cleaning

Item	Cost (€ per cleaning experiment)	
	Conventional chemical cleaning (NaOH 5 g/L at 50 ºC)	Physical cleaning by electric fields (NaCl 5 mM at 43.8 ºC and 30 V)
Chemicals	0.30	0.02
Heating	0.02	0.02
Electric field application	—	0.01
TOTAL	**0.32**	**0.05**