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UHT Condensate Recovery by Reverse Osmosis: A Pilot-Plant Study

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ABSTRACT: Real ultrahigh-temperature (UHT) flash cooler (FC) condensates obtained in the dairy industry were processed in situ using a reverse-osmosis (RO) membrane (Duratherm HWS 4040 HR; 8.2 m^2 filtration area). Semicontinuous trials were performed to estimate the maximum volume concentration rate (VCR) that fulfils the permeate specifications for reuse as boiler water. Then, long-term continuous experiments (100 h in duration) were carried out at a constant permeate flow rate (40 L h^{-1} m^{-2}), and the conductivity, chemical oxygen demand (COD), pH, and Ca^{2+} concentration were monitored. An activated carbon column (ACC) was used to reduce the organic charge of the permeate.

1. INTRODUCTION

The food industry is one of the most extensive water-using industries,[1] and the dairy industry can be considered one of the most polluting because of its high water consumption (up to 10 L of water per liter of processed milk) and its status as the largest producer of food processing wastewater.[2,3] These dairy wastewaters are characterized by wide fluctuations in pollutant charge and flow rate.[4,5] In most cases, wastewaters generated in different individual operations are mixed and neutralized prior to discharge into the municipal sewage system; however, it seems that segregating streams would enable optimal treatment for each type of wastewater, as well as energy and cost savings.[6,7]

Because of this extensive water consumption; stricter pollution control legislation regarding waste disposal; growing concerns for sustainable development; and the need for a safe, reliable, and affordable supply of quality water, interest in developing a zero-discharge process has gained relevance, and the practice of reconditioning, recycling, and reusing water is becoming an objective of study and research.[8−12] In this scenario, membrane processes are useful technologies in wastewater reclamation and reuse and are currently well integrated in the dairy industry, as the decreasing costs of installation and operation in continuous processing favor their use.[13−16] More specifically, reverse osmosis (RO) has been found to be a promising separation technique for the treatment of dairy wastewaters.[2,11,17−24]

Ultrahigh-temperature (UHT) treatment of milk and milk-based products involves a heating process that ensures a sterile product with a shelf life at room temperature of at least 6 months, while minimizing cooked flavors.[25,26] A typical UHT treatment consists of treatment at 140 °C for 4 s,[26] which could be achieved by direct or indirect heating. The benefit of the direct process lies in higher rates of heat transfer and lower changes in taste compared to the indirect process.[27] In direct steam injection (DSI) contact, the product is preheated and pressurized to approximately 4 bar, in order to prevent boiling of the liquid. After passing through the holding tube, the product is flash-cooled, homogenized, cooled again, and aseptically packaged.[28] The flash cooling takes place in an expansion chamber, known as the flash cooler (FC), which operates under a partial vacuum, obtained using a vacuum pump, where the pressure is controlled to restore the product's original dry matter.[29] Flash cooling also permits the removal of some volatile compounds.[30] As the FC operates at the liquid boiling point, when the pressure drops, some milk droplets could be entrained in the vapor phase, contributing to an increase in the pollution load of the condensates.

In the dairy company considered in this work as a case study, the vapor from the FCs is used to preheat the dairy product to be processed and subsequently condensed and collected after the vacuum pump. This rotary volumetric pump uses tap water as an auxiliary liquid that is mixed with the condensates to generate the vacuum. As a consequence, the feed wastewater used in the present work is a mixture of condensates from the FCs and tap water used in the vacuum pump. This effluent can be considered to have a low pollutant load.[21,31]

The amount of tap water used in the vacuum pump determines, to some extent, the characteristics of the stream that feeds the RO process. In general, a higher degree of vacuum requires greater consumption of tap water, which causes two effects: an increase in the pollution load (at a higher degree of vacuum, more milk drops pass into the vapor) and a dilution effect.

Among all possibilities for water reuse, including drinking-water standards, the most restrictive specifications are given by boiler-water usage [pH 7−10, conductivity at 25 °C < 40.0 μS/cm, chemical oxygen demand (COD) < 10 mg of O_2/L, total organic carbon (TOC) < 4 mg of O_2/L, Ca^{2+} concentration < 0.40 mg/L].[32,33] These parameters must be guaranteed to improve the efficiency and service lifetime of the boiler; control the blow-down frequency; and avoid potential scaling, corrosion, and formation of foams.

The aim of this study was to evaluate the effectiveness of an RO continuous pilot process at a moderate temperature to obtain high-quality water for boiler reuse from real dairy condensates of a UHT vacuum system. Preliminary tests were performed in semicontinuous mode to determine the optimum volume concentration rate (VCR) for long-term processing. An

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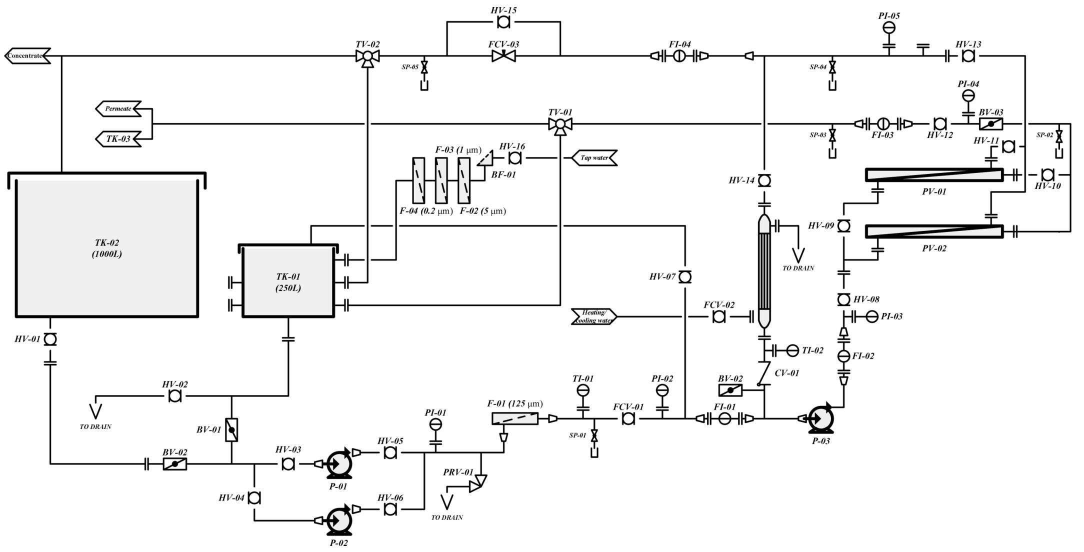

Figure 1. Scheme of the RO equipment.

activated carbon column (ACC) was coupled after the RO step to correct the limit on organic matter in the final permeate.

2. MATERIALS AND METHODS

2.1. Flash Cooler Performance. In the Spanish factory where the trials were carried out, four FCs operate simultaneously but at different stages of the production cycle. When working at the same time, they generate ~20 m^3 of condensates per hour with different pollution loads.

The UHT system works in a cyclic manner, although some steps can vary depending on production needs or breakdowns. Aseptic intermediate cleanings (AICs), consisting of the use of an alkaline-based detergent with two intermediate rinsing steps, are performed routinely. In the same way, depending on the variability of the production schedule, a final cleaning (alkaline and acid) is carried out. On the other hand, when the aseptic bottle-filling step cannot handle the production rate, the product is sent to an aseptic tank; when this tank becomes full, the system starts to work with sterile water until the aseptic packaging system can again allow product to enter.

Condensates are continuously produced during the cycle (even when the equipment is loaded with sterile water), except when final cleaning is performed. As a result, their characteristics are highly dependent on the instant in the cycle; for example, condensates collected during the intermediate cleaning show high pH, whereas those accumulated during the start of or after the AICs are more concentrated and have a higher organic load, and those occurring when sterile water is circulating have low pollution levels. Finally, the type of product sterilized (milk, shakes, etc.) and the sterilization parameters also have some influence on the condensate characteristics.

2.2. Analytical Methods. In this work, pH (accuracy ±0.05), electrical conductivity (accuracy ±0.5%), and COD (accuracy ±3 mg of O_2/L) were measured continuously, whereas residual hardness, expressed as Ca^{2+} concentration (accuracy ±0.10 mg/L), was analyzed occasionally. pH and electrical conductivity were measured by means of an HQ40d multimeter (Hach Lange, Mechelen, Belgium). Spectrophotometric measurements of COD (in which samples were oxidized with a hot sulfuric solution of potassium dichromate using silver sulfate as the catalyst and Cr^{3+} ions were then determined photometrically at 605 nm) and residual hardness (in which calcium and magnesium ions were reacted with phthalein purple in alkaline solution to form a violet colorant that was determined photometrically at 565 nm) were carried out by means of Spectroquant cell test kits (Merck, Darmstadt, Germany) using a NOVA 60 photometer (Merck, Darmstadt, Germany).

2.3. Reverse-Osmosis Pilot Plant. A scheme of the membrane pilot plant is shown in Figure 1. Although the plant had the capacity for two in-parallel 4040 spiral-wound membranes, only one was used, and the other pressure vessel was kept closed. Tank TK-01 was used exclusively for cleaning purposes, and an additional external 1000-L capacity tank (TK-02) was available to collect the condensates from the FC vacuum system. The RO rig was fed using two consecutive pumps [P-02 and P-03, 7.5 and 2.2 kW, respectively (Grundfos, Bjerringbro, Denmark)]. A 125-μm prefilter (F-01) was installed to remove coarse particles that could damage these pumps and the membrane. The pressure and feed flow rate were fixed by regulating a needle valve (FCV-03) placed on the retentate side. Transmembrane pressure (TMP) was controlled by means of manometers PI-03 and PI-05, whereas the feed, permeate, and concentrate fluxes were measured by flow meters FI-01, FI-03, and FI-04, respectively. Finally, to maintain low water silt density index (SDI) values (<3)[34] for the rinsing and cleaning steps, a prefiltration system consisting of a bag filter and 5-, 1-, and 0.2-μm cartridge prefilters was used.

2.4. Membrane. A spiral-wound thin-film (proprietary composite) RO Duratherm HWS 4040 HR membrane (GE Water & Process Technologies, Trevose, PA) was selected on the basis of the recommendation of the membrane manufacturer to treat the type of effluent studied. The specific membrane properties are listed in Table 1. The maximum TMP and permeate flow rate (J) were set to 30 bar and 40 L h^{-1} m^{-2}, respectively, according to the manufacturer.

The cleaning protocol followed was suggested by a detergent manufacturer (Diversey, Utrecht, The Netherlands) according to the characteristics of the wastewater processed. Standard membrane cleanings consisted of acid (Divos 2, pH 1.9−2.1, 20 min, 45 °C) and alkaline (Divos 123, pH 11.0−11.5, 45 min, 45 °C) steps with intermediate water rinsings. However, only

Table 1. Main Membrane Characteristics

RO Duratherm HWS 4040 HR	
average salt rejection[a] (%)	99.5
active area (m^2)	8.2
spacer thickness (mil)	31
maximum operating pressure (bar)	
5−50 °C	41.4
51−70 °C	20.7
maximum operating temperature (°C)	70
maximum cleaning temperature (°C)	50
pH operating range	4.0−11.0
pH cleaning range	2.0−11.5

[a]Testing conditions: 2000 mg/L NaCl solution at 15.5 bar operating pressure, 25 °C, and pH 7.5.

alkaline cleaning was carried out when intermediate cleanings were required. To avoid microbiological growth, the membrane was stored between runs in acid solution (Divos 2 at pH 3.0−4.0). The flux recovery was evaluated to determine the cleaning efficiency by measuring J at an inlet pressure of 10 bar, a feed flow rate of 1 m^3/h, and a temperature of approximately 15 °C with prefiltered tap water (SDI < 3).

2.5. Operating Procedure. Trials were carried out in two different modes of operation. In the first mode, after the 1000-L feed tank had been filled, semicontinuous operation of the plant was started, with the permeate being drained and the concentrate returned to the feed tank until the desired VCR was reached. Once this degree of concentration was achieved, the permeate stream was sent back to the feed tank, and the system began to run in a discontinuous closed loop for several hours. The VCR was estimated as

$$\mathrm{VCR} = \frac{V_f}{V_r} = \frac{V_f}{V_f - V_p} \tag{1}$$

where V_f, V_r, and V_p are the feed, retentate, and permeate volumes, respectively. The concentration tests in semicontinuous mode were performed at VCRs of 2, 6, and 10.

Regarding the VCR, the recovery rate (RR), defined as

$$\mathrm{RR}\ (\%) = \frac{\mathrm{VCR} - 1}{\mathrm{VCR}} \times 100 \tag{2}$$

is an essential parameter for determining the size and economy of a large-scale membrane installation.

In the second part of the work, the rig was operated in continuous mode. The degree of concentration in the feed tank was maintained by regulating the entrance of fresh feed and controlling the amount of concentrate returned to the rig. The surplus concentrate stream was drained out to the factory's sewage system. On the other hand, the permeate stream was continuously drained out. To verify the desired VCR, the conductivity inside the feed tank was continuously measured.

When working in continuous mode, a model FCA-355 activated carbon column (ACC; Sefiltra S.A., Madrid, Spain) with 110-L capacity was coupled to the permeate outlet to reduce the organic matter content (RO−ACC). This ACC, made of polyester reinforced with fiberglass and having a capacity of 1800 L/h, was filled with 50 kg of GAC MG 1050 activated carbon (density ≈ 0.45 g/cm^3) (Sefiltra S.A., Madrid, Spain).

In both semicontinuous and continuous trials, a cooler was used to regulate the inlet temperature, which was fixed at approximately 45 °C in all tests. The feed flow rate was maintained at 1000 L/h [with the cross-flow velocity (CFV) estimated to be 0.35 m/s], the permeate flow rate was fixed at 40 L h^{-1} m^{-2}, and the system was run at increasing TMP to compensate for fouling. Feed and permeate characteristics such as temperature, TMP, feed flow rate, and permeate flow rate (J) were continuously recorded, as was the membrane rejection (or reduction) (R) in terms of conductivity and COD, which can be calculated by means of the equation

$$R\ (\%) = \left(1 - \frac{C_p}{C_f}\right) \times 100 \tag{3}$$

where C_p and C_f are the permeate and feed (concentrated) values, respectively, of both parameters considered.

3. RESULTS AND DISCUSSION

3.1. Flash Cooler Performance and Condensate Characterization. A complete characterization of the condensates used in this work was previously published by Riera et al.[4] The values correspond to samples taken during several complete operating cycles of the FCs (eight cycles per FC), and their variability reflects the different steps explained before. The compositions of fresh condensates (and condensates at a VCR of 6) are presented in Table 2. As can be

Table 2. Compositions of Initial and Concentrated Feed Condensates for RO Processing

parameter	initial value range	average	concentration value range[a]	average
T (°C)	40.5−59.9	50.2	−	−
pH	5.31−6.49	5.83	6.34−8.24	7.14
conductivity (μS/cm)	76.1−245.0	184.1	447.0−1017.0	728.2
COD (mg of O_2/L)	4−240	70	30−360	153
TOC (mg of O_2/L)	−	−	32.1−85.4	56.8
Ca^{2+} concentration (mg/L)	−	−	1.22−2.38	1.67

[a]At VCR 6 for continuous processing.

seen, the condensates, before and after any previous concentration, did not fulfill the requirements for boiler water (pH 7−10, conductivity at 25 °C < 40.0 μS/cm, COD < 10 mg of O_2/L, TOC < 4 mg of O_2/L, Ca^{2+} concentration < 0.40 mg/L),[32,33] so its treatment by RO was justified.

The characteristics of the fresh condensates are in agreement with those reported by Chmiel et al.,[35] slightly higher than those published by Mavrov and Bélières,[21] and well above those described by Verheyen et al.[36,37] for steam condensates in the dairy industry. Detailed analyses of condensates from UHT FCs are scarce, although odors due to thermal treatment and produced by the presence of volatile compounds from milk fat and some protein degradation have previously been reported. However, in-depth analyses of these compounds are available. Some major organic compounds found in previous research suggested that acetone, ethanol, acetoine, dimethylsulfide, diacetyl, and so on are mainly responsible for organic matter and odors.[23,38−40]

3.2. Reverse-Osmosis Treatment. *3.2.1. Semicontinuous Trials.* The RO membrane was characterized using prefiltered tap water with an SDI of <3.[34] The membrane permeability at a feed flow rate of 1000 L/h, temperature of 40 °C, and TMP of

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4−19 bar was calculated to be 5.7 L h^{-1} m^{-2} bar^{-1}. After the temperature correction, the calculated value, 0.143 L h^{-1} m^{-2} bar^{-1} $°C^{-1}$, was comparable to those published by other authors who used RO for dairy wastewater processing.[2,17,19,20]

The water recovery capacity, that is, the amount of permeate recovered as a percentage of the volume of condensates fed, determines the size of a large-scale membrane installation and is dependent on the VCR. RO plants are designed in stages, and membranes face the highest concentration in the final stages. As the feed concentration increases, the permeate flow rate is reduced, the quality of the permeate is poorer, and the membrane cleaning frequency increases.[41] For this reason, the maximum VCR (or RR) at which the permeate satisfies the requirements must be estimated.

Semicontinuous trials were performed to determine the behavior of the RO membrane at different VCRs, for experiments that were 12 h in duration. The trials were carried out at a constant feed flow rate of 1000 L/h and a fixed permeate flow rate of 40 L h^{-1} m^{-2}, which was the value recommended by the membrane manufacturer. The TMP was increased during the experiments to compensate for the permeate flux decay due to polarization concentration and fouling. The temperature was maintained at an average of 45.2 ± 1.7 °C by using a cooler, as the condensates come from FCs at temperatures between 47.8 to 55.1 °C.

The permeate and feed conductivities and their R values are plotted for VCRs of 2, 6, and 10 in Figure 2 (the conductivity

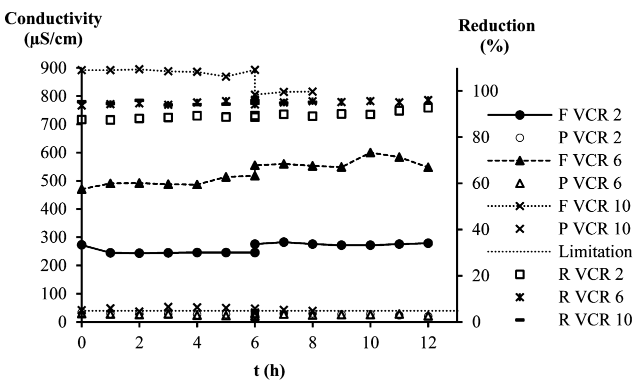

Figure 2. Semicontinuous RO. Feed (F) and permeate (P) conductivities (μS/cm) and conductivity reduction (R, %) at different operating VCRs. Boiler-water limit = 40.0 μS/cm.

requirement for boiler water is represented by the horizontal dotted line at 40 μS/cm). The experiments were conducted on different days, with different condensate characteristics, which explains the discontinuity in conductivity observed at 6 h. The R value for conductivity remained almost constant after 12 working hours, and the average values were estimated to be 89.5%, 95.0%, and 95.0% for VCRs of 2, 6, and 10, respectively; this means that the conductivity R value increased slightly with increasing VCR. At higher VCR, the difference in solute concentration across the two sides of the membrane increased, and as a result, the reduction increased as well (see eq 3).[42] The permeate conductivity varied from 20.3 to 53.5 μS/cm.

The conductivity requirements were satisfied (permeate values were lower than 30 μS/cm in most cases) at VCRs of 2 and 6. However, the limit was exceeded at a VCR of 10, reaching permeate values up to 53.5 μS/cm; for this reason, the trial was stopped after 8 h.

The evolutions of the TMP and COD were also studied as functions of time at the three VCRs tested (2, 6 and 10) (data not shown). The greater the VCR, the higher the pressure required to reach the same permeate flow rate, as the increase in the concentration of solutes had a negative impact on the performance of the process;[41] however, the difference was not significant (TMP increased from 8.1 to 10.1 bar as VCR was increased from 2 and 10). During these experiments, the TMP remained constant, and fouling effects were not observed.

Irrespective of the VCR, the permeate exhibited COD values between 0 and 14 mg of O_2/L, which exceeded the limit for boiler-water reuse (10 mg of O_2/L) in some cases. These results suggest the need to install an ACC to treat the RO permeate.

Considering that the permeate conductivities at a VCR of 10 exceeded the requirements for boiler water, continuous trials were designed at a VCR of 6, which means an RR value of 83.3% of the water fed.

3.2.2. Continuous Trials. Continuous 100-h long-term trials were performed at a constant VCR of 6 to simulate the last stages of an industrial RO installation. Such tests provide information about the membrane behavior in terms of rejection and fouling frequency. When the industrial UHT sterilization system was undergoing cleaning and no other FC was producing condensates, the RO pilot plant worked with condensates that had been previously accumulated in the 1000-L feed tank, which remained at a VCR of 6; during this period, both the permeate and the concentrate were sent back to the tank. The ACC coupled to the permeate line was not used when the permeate and concentrate were recirculated. The experiment was performed at a feed flow rate of 1000 L/h, a constant permeate flow rate of 40 L h^{-1} m^{-2}, and an increasing pressure to compensate for the flux decay. The feed temperature was maintained at an average of 44.9 ± 3.3 °C.

Figure 3 shows some of the results of the long-term continuous experiment, where the temperature, TMP, conductivity, and COD of the condensates fed are plotted as functions of time. The temperature variability during the first part of the experiment (first 10 h) was due to the mode of operation. During the periods in which the system was working in recirculation mode (see 6−10 h, 23 h, 49−51 h, etc.), the temperature control was difficult, and a drop in temperature was recorded. During these periods, the membrane perme-

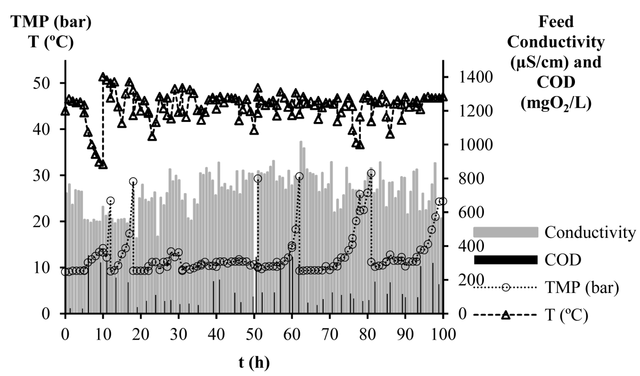

Figure 3. Long-term continuous RO−ACC test. Conditions: Constant permeate flow rate (J = 40 L h^{-1} m^{-2}), feed flow rate = 1000 L/h (CFV = 0.35 m/s). TMP (bar) and temperature (T, °C) vs time at a VCR of 6 and as functions of feed conductivity (μS/cm) and COD (mg of O_2/L).

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ability was lower because of an increase in fluid viscosity and a decrease of diffusivity; then, an increase in TMP was necessary to maintain a permeate flow rate of 40 L h^{-1} m^{-2}. Except in the situations described above, the average temperature was 45 °C, with the loss of temperature inside the equipment estimated to be 2.5 °C by comparing the inlet and RO−ACC outlet temperatures.

The evolution of TMP in Figure 3 provides information about membrane fouling. Each time the rig reached the maximum TMP (30 bar), an alkaline cleaning step was performed. Further, complete (alkaline- and acid-based) cleaning was carried out each weekend to avoid microbiological growth during the weekend shutdowns (the UHT system was stopped on weekends as well). During the entire continuous test, seven complete cleanings and three alkaline cleanings were performed. The original membrane permeate flow rate was recovered after both cleaning protocols.

The COD values in Figure 3 (black columns) demonstrate the heterogeneity of the inlet condensates, with values from 30 to 360 mg of O_2/L, corresponding to the original values (at a VCR of 1) of 4 and 117 mg of O_2/L, respectively. After 12, 18, 51, 62, and 81 h of operation, the maximum TMP was reached. However, the pressure peaks at 12, 18, 51, and 62 h were due to a momentary increase in concentrated feed COD (black bars). COD values higher than approximately 300 mg of O_2/L caused a very rapid increase in TMP. On the contrary, when COD remained low (as in the period from 67 to 81 h), the increase in TMP was slower, and the membrane worked for 14 h without any cleaning or rinsing step.

Values of concentrated feed conductivity during the test were approximately 728 ± 113 μS/cm. Conductivity did not show any influence on fouling as, in general, there was neither significant heterogeneity nor momentary peaks of high conductivity.

In Figure 4, the conductivity and COD values of the permeate before and after ACC treatment, as well as the reductions of these two parameters, are plotted as functions of time. As expected, the differences in conductivity before and after ACC were negligible (Figure 4a), and only some permeate samples showed values higher than the limit. Conductivity RO−ACC reductions were between 90.1% and 98.3% (average = 95.4%) and remained almost constant throughout the tests.

It has been well demonstrated that fouling affects membrane selectivity and that a rise in pressure both increases the hydraulic resistance of the membrane and decreases the membrane selectivity.[43] This behavior was observed from 76 to 81 h, when the permeate quality in terms of conductivity worsened as the TMP increased. As a result, the RO−ACC outlet conductivity changed from 18.6 to 47.1 μS/cm, and the reduction decreased from 97.5% to 94.4%. This behavior was also observed in the last part of the tests, from 90 h onward, although it is difficult to determine whether this stepwise increase was related to the previous statement or was influenced by the observed rise in feed conductivity, given that, although the permeate conductivity increased from 22.8 to 39.2 μS/cm, the decrease in reduction was negligible (0.5%).

With respect to the COD (Figure 4b), permeates showed significant differences with time due to the variability of the feed characteristics. (In Figure 4, if a permeate or an ACC vertical bar does not appear for a given feed, this means that the value reached was 0 mg of O_2/L.) In most cases, high COD values corresponded to momentary inlets of highly polluted organic matter, where the concentrated feed reached values

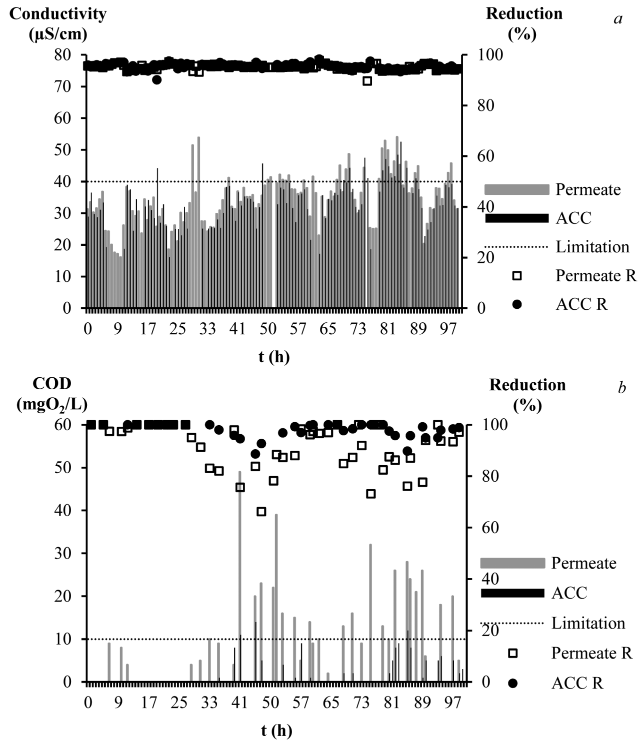

Figure 4. Long-term continuous RO−ACC processing. Conditions: Constant permeate flow rate (J = 40 L h^{-1} m^{-2}), feed flow rate = 1000 L/h (CFV = 0.35 m/s), temperature = 44.9 ± 3.3 °C. (a) Conductivity (μS/cm) and reduction (R, %) and (b) COD (mg of O_2/L) and reduction (R, %) at a VCR of 6 of permeate and after activated carbon (ACC) treatment.

between 185 and 360 mg of O_2/L, which are far from the average COD for concentrated feed, estimated to be 85 mg of O_2/L. As a result, these fleeting high feed CODs or the combination of various consecutive high values were strongly associated with the fouling observed and, thus, with the rapid increase in TMP. Regarding the nature of the fouling observed, it has been demonstrated that inorganic fouling is negligible at working pH values (neutral to alkaline), as soluble calcium, which appears to be the main promoter of inorganic fouling, is complexed with phosphate and citrate at these pH values.[44] On the other hand, organic fouling, mainly caused by proteins, is the major factor responsible for membrane fouling in dairy applications.[3,22,45] However, the TMP peak at 81 h was not due to an increase in the organic load of the feed. In that case, the membrane was run for 14 h without cleaning.

There were several significant drops in the COD reduction of the RO permeate that matched the fouling periods corresponding to peaks at 51 and 81 h. As reported by Brans et al.,[46] the fouling and increase in associated pressure reduced the selectivity of the membrane, leading to a lower permeate quality in terms of organic matter. Furthermore, in some cases, the decreases in COD reduction corresponded to peaks in feed COD, such as that at 41 h, where the R value was reduced by up to 75.6%, which matched with a momentary COD inlet of 201 mg of O_2/L. Figure 4b demonstrates the inability of RO membranes to completely remove the organic matter, as stated by Bódalo-Santoyo et al.,[47] who showed the limited ability of RO to reject low-molecular-weight organic compounds. On the other hand, the effectiveness of the ACC in reducing the organic charge of permeates is well demonstrated in the figure.

The maximum COD reduction achieved after the RO−ACC process was 100.0%, although the average was established as 97.9%.

During the continuous trial, 25.9 m^3 of permeate was fed into the ACC. No information was found with respect to the maximum organic load for the commercial column used, as it is highly dependent on the contaminant type, the characteristics of the effluent, the feed concentration, and the type of activated carbon.[48] The average COD removal of the ACC was calculated to be 76.8% by comparing the concentrations of the inlet (RO permeate) and outlet streams. The total organic load supported was 0.0057 kg of COD per kilogram of activated carbon. At this rate, the breakthrough for this particular case, defined as the point at which the COD in the RO−ACC outlet stream continuously exceeded the limit required and, therefore, the ACC exhaustion point, was not reached. The ACC required some maintenance during the trials: retro-washing with tap water at high flow rate was performed weekly. In the case of reaching irreversible saturation, the activated carbon should be regenerated in an oven or completely removed and replaced.

The average conductivity and COD of the RO−ACC outlet stream were 33.3 μS/cm and 3 mg of O_2/L, respectively. Some samples reached maximum values of 52.6 μS/cm and 14 mg of O_2/L, respectively. Although these values were above the restriction, the permeate obtained in an industrial plant will be diluted before entering the boiler. Permeates obtained in RO steps with high VCRs and those from low VCRs (with better permeate quality) will be mixed in the final current[49,50] and the RO−ACC outlet stream will be mixed with fresh demineralised water to satisfy the current necessity of the factory. This behavior could also be applied to Ca^{2+}, which showed values of up to 0.67 mg/L, even though the average, 0.36 mg/L, was located below the limit (0.40 mg/L).

The heterogeneity in organic matter found when processing wastewaters from the vacuum system of the UHT sterilization process by RO advised that possible momentary high-organic-load streams should be controlled and segregated to reduce the cleaning frequency and avoid unexpected rapid periods of fouling. If this segregation is not taken into account, the membrane behavior is not easily predictable and intermediate cleanings would take place when necessary, reducing the economic interest of the application.

Finally, the pH performance during the continuous test as a function of time for the concentrated feed, the permeate, and the ACC output stream are shown in Figure 5. The feed pH values (at a VCR of 1) varied between 5.3 and 6.9, and those for the concentrated feed (at a VCR of 6) showed values between 6.3 and 8.2. In the figure, some pH drops (clearly observed after 18, 40, 51, 78, and 93 h) can be noted due to the variability in the inlet waters (as a function of the degree of vacuum, the timing in the FC cycle, the product treated, and the sterilization parameters of the UHT system) for different working days. Permeate acidification with respect to the feed was previously observed by Suárez and Riera,[42] and it was attributed to the membrane and feed characteristics. The permeate was acidified, generally always to the same extent (it was reduced in all cases by 1.9 ± 0.2 pH units), as a result of electrostatic repulsion/attraction effects between the solutes and the membrane surface, which is negatively charged at working pH values, as are most RO membranes,[51] promoting the favorable passage of H^+ ions.[52] The acidification observed is common in RO wastewater processing.[53−55] Organic or inorganic compounds adsorbed on the membrane surface can influence its charge.[56] In these experiments, there was no evidence of this effect, as the pH of the permeate remained practically constant (5.3 ± 0.4). No significant differences were found between pH samples before and after the ACC.

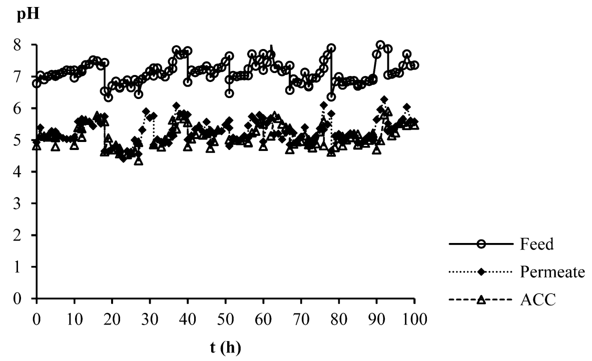

Figure 5. Long-term continuous RO−ACC processing. pH evolution with time of feed and permeate and after activated carbon (ACC) treatment. Conditions: Constant permeate flow rate (J = 40 L h^{-1} m^{-2}), feed flow rate = 1000 L/h (CFV = 0.35 m/s).

When permeate pH values were outside the range required for boiler-water reuse (7−10), then some treatments, such as degassing steps to reduce the content of CO_2 or the addition of alkalizing or antiscalant/antifouling chemicals, should be taken into consideration as a final step before the reuse of permeates as boiler waters.

4. CONCLUSIONS

The main difficulties when treating real UHT condensates in the dairy industry are related to the heterogeneity of the feed in terms of conductivity and, especially, COD. The conductivities of concentrated condensates treated when working in continuous mode (at a VCR of 6) varied between 447 and 1017 μS/cm (average = 728 μS/cm), and the reduction obtained was between 90.1% and 98.3% (average = 95.4%). In general, the permeate conductivities were lower than the fixed limit (40 μS/cm). Momentary values higher than the boiler-water requirement were obtained when the membrane was fouled (maximum TMP) as a result of an increase of organic matter in the feed. COD was considered to be the most important parameter that affected the permeate quality because, when the organic load was significant (about 300 mg of O_2/L in this work), rapid membrane fouling took place, and an additional cleaning had to be done. The feed COD at a VCR of 6 varied between 30 and 360 mg of O_2/L, and the RO membrane was not able to reduce the permeate content to satisfy the limit established when the feed presented high COD values. The ACC was demonstrated to be effective in reducing the organic load, and the RO−ACC outlet stream fulfilled the boiler-water specifications (10 mg of O_2/L) in most of the samples analyzed. When permeate pH was acidified (outside the limit), then a post-treatment was advisible to adjust it.

A general recommendation for treating these types of wastewaters with the objective of producing boiler water is to segregate condensates when COD reached values higher than 300 mg of O_2/L. In the rest of the cases, RO followed by ACC was shown to be effective.

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Notes

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NOMENCLATURE

Abbreviations

ACC = activated carbon column
AIC = aseptic intermediate cleaning
CFV = cross-flow velocity
COD = chemical oxygen demand
DSI = direct steam injection
F = filter
FC = flash cooler
FCV = flow control valve
FI = flow indicator
P = pump
PI = pressure indicator
RO = reverse osmosis
RR = recovery rate
SDI = silt density index
TK = tank
TMP = transmembrane pressure (bar)
TOC = total organic carbon
UHT = ultrahigh temperature
VCR = volume concentration rate

Symbols

C_f = feed concentration (μS/cm or mg of O_2/L)
C_p = permeate concentration (μS/cm or mg of O_2/L)
J = permeate flow rate (L h^{-1} m^{-2})
R = reduction (%)
V_f = feed volume (m^3 or m^3/h)
V_p = permeate volume (m^3 or m^3/h)
V_r = retentate volume (m^3 or m^3/h)

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