

## Post-treatment of dairy wastewater by activated sludge-ultrafiltration for water reuse

Temesgen Sibhatu Habtu, Magdalena Zielińska\*

University of Warmia and Mazury in Olsztyn, Department of Environmental Biotechnology, 45G Słoneczna St, 10-709 Olsztyn, Poland, Tel. +48 89 523 41 85, email: temienebsi33@gmail.com (T.S. Habtu), magdalena.zielinska@uwm.edu.pl (M. Zielińska)

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### ABSTRACT

This study evaluated the efficiency of using aerobic treatment in a sequencing batch reactor (SBR) combined with ultrafiltration (UF) for post-treatment of anaerobically treated dairy wastewater to produce effluent for reuse. In the SBR, the removal efficiencies were as follows: 93.9% (chemical oxygen demand, COD), 78.8% (total nitrogen), and 88.0% (total phosphorus). Although the effluent met criteria for wastewater discharge into an aquatic environment, it did not meet criteria for water reuse. Therefore, further treatment using membranes was conducted. A UF system with a ceramic membrane was operated at trans-membrane pressures (TMPs) of 0.2, 0.3, 0.4 and 0.6 MPa. At all the TMPs, the permeates were completely free of suspended solids, and 88.9% of COD and 90.9% of biological oxygen demand (BOD) were removed. In the batch tests of UF, there was a linear relationship between permeate flux and pressure at 0.2–0.4 MPa. At a TMP of 0.6 MPa, the initial permeate flux was the highest, but permeate flux declined rapidly by about 43% in less than 1 h due to more serious membrane fouling. The final effluents from the SBR-UF system can be reused for irrigation, cooling and external cleaning, construction, and flushing.

*Keywords:* Dairy wastewater; UF; Ceramic membrane

### 1. Introduction

The dairy industry is considered the most polluting of the food industries because it consumes a large amount of water and generates large amounts of liquid waste that contains a high concentration of dissolved organic materials, inorganic solids, suspended oil/grease, nitrogen and phosphorus [1]. A number of researchers have reported that anaerobic treatment is appropriate for wastewater with a COD between 3000 and 40,000 mg/L [2], thus it is an advantageous method when treating dairy wastewater. Apart from its advantages of effective organics removal and low sludge production, anaerobic fermentation generates bio gas, which may be used for the production of heat and/or power.

However, disadvantages of the anaerobic process for dairy wastewater include high production of scum, poor

settle ability of the biomass, low resistance to organic shock loads, calcium precipitation and problems with the degradation of fats, oils and other specific types of pollutants [3–5]. In addition, anaerobic treatment results in a relatively low efficiency (up to 10%) of nutrient removal [6]. For these reasons, anaerobic treatment of dairy wastewater should be followed by aerobic treatment in intermittently aerated reactors consisting of alternate anoxic/anaerobic and aerobic phases [7]. To overcome the problems of the sensitivity of aerobic reactors to variations in COD and BOD loading and of the large area required for the installations, sequencing batch reactors (SBR) have been suggested as a highly flexible solution for the biological treatment of industrial waste waters [8]. Although SBRs can remove nitrogen with an efficiency above 75% [9], the activated sludge is prone to bulking due to the overgrowth of filamentous bacteria in SBRs treating dairy wastewater [10]. This overgrowth takes place because the

\*Corresponding author.

acidic pH of dairy wastewater favors the growth of filamentous bacteria [11], and this type of wastewater has a low buffering capacity [12].

Currently, there are a number of reasons for recycling or reusing industrial wastewater. These reasons include the need to lower the consumption of fresh water due to the reduced availability and quality of water resources, and increasingly restrictive requirements for wastewater discharge. Thus, the reuse of wastewater has become an environmentally and economically viable option for industry. This is particularly true in the dairy industry because dairy wastewater does not contain toxic chemicals [13]. This water could be reused for cooling or heating systems and for good manufacturing practices, such as washing floors and trucks and rinsing external areas [14]. However, biological treatment alone of dairy wastewater cannot produce effluent that satisfies the standards for industrial reuse, which means that further treatment is necessary and should be investigated. In this context, the food industry, including the dairy industry, is investigating membrane separation of wastewater, which may help to produce water of sufficient quality for recycle or reuse [15,16].

Of the various membrane filtration processes, nanofiltration (NF) and reverse osmosis (RO) produce permeate of a very high quality. Although the microfiltration (MF) and ultrafiltration (UF) membranes that are used to separate solids from a liquid do not reduce COD as well as NF and RO do, and do not concentrate small solutes like lactose [17,18], they yield a high flux of permeate at low TMP [19], which means that these techniques have lower energy costs than NF or RO. Because MF and UF permeates contain too much lactose [17], these techniques should not be used alone, but instead, they should follow biological treatment, for example by using membrane bioreactors (MBRs) that are compact and modular systems, produce little sludge and can completely remove suspended solids independently of the sediment ability of the biomass [20].

Non-potable water reuse, particularly for agricultural irrigation, is gaining popularity in many developing communities [21]. Sadr et al. [22] found that for non-potable water reuse in a water-stressed developed community, the most preferable option is primary treatment + MBR (aerobic + MF/UF) + disinfection. These technologies are effective for producing effluent that can be used for landscape and restricted agricultural irrigation, and for industrial reuse (e.g. cooling towers and washing). However, for non-potable water reuse in a developing, water-stressed community, the preferred option is primary treatment + conventional activated sludge process (anoxic + aerobic) + MF/UF + disinfection because this option is less costly than MBRs.

However, a drawback to the use of membrane processes for wastewater treatment is permeate flux decline due to concentration polarization and fouling [23]. The proteinaceous compounds of dairy wastewater foul existing membrane materials [24]. Ceramic membranes are less prone to fouling and can reach much higher flux than polymer membranes because of weaker bonding between foulants and the membranes [25]. Thus, in this study, ceramic membranes were used that have good thermal and chemical stability, and high resistance to corrosion, abrasion and fouling, all of which lead to high efficiency of backwashing and make ceramic membranes more durable [26].

The aim of this work was to study the applicability of a technological system consisting of an activated sludge process and UF for the post-treatment of anaerobically treated dairy wastewater to evaluate the possibility of reusing the final effluent for different purposes, such as irrigation, external cleaning and cooling water. Because the effect of operational conditions on the aerobic treatment of dairy wastewater has been established [e.g. 27], the aim of the activated sludge operation in this study was only to produce effluent that would serve as a feed solution for membrane filtration. While the literature on the use of membranes in dairy wastewater treatment is relatively abundant, a large proportion of those studies have employed polymer membranes, whereas a smaller number have used ceramic modules [28]. Moreover, the majority of studies on dairy wastewater treatment by membrane filtration used MBR technology, in which the membranes are immersed in an anaerobic or aerobic bioreactor. In this study, a tubular multichannel ceramic membrane was tested for filtration of the effluent from the aerobic bioreactor, which created totally different fouling conditions from those in MBRs. In addition to the efficiency of removal of organic compounds (COD), suspended solids, total nitrogen and total phosphorus, the effect of the TMP on the susceptibility of the membrane to fouling was investigated.

## 2. Materials and methods

### 2.1. Wastewater

In this experiment, the effluent from the anaerobic treatment of dairy wastewater collected in a pilot-scale plant was used. In this plant, dairy wastewater ( $30,000 \pm 760$  mg COD/L,  $20,560 \pm 420$  mg BOD/L,  $823 \pm 62$  mg N/L,  $278 \pm 44$  mg P/L, pH  $7.2 \pm 0.2$ ) was treated under mesophilic conditions in an anaerobic plug-flow reactor with a total volume of 100 L. This was a two-stage reactor in which the hydrolyzing and methanogenic phases were separated (Fig. 1A). The reactor had a circular cross-section and consisted of concentric chambers: an internal hydrolyzing chamber (20 L), into which wastewater was introduced from the bottom, and an external methanogenic chamber (80 L), divided equally into two concentric zones. The effluent was collected from a decanter placed in the upper part of the outer methanogenic zone. This construction enabled labyrinth flow of wastewater and good mixing conditions. The reactor was inoculated by granular sludge taken from a full-scale anaerobic dairy wastewater treatment facility. The reactor was operated without pH correction, at an organic loading rate (OLR) of 3 kg COD/(m<sup>3</sup>·d) and a hydraulic retention time (HRT) of 10 d.

### 2.2. Biological treatment

The two SBRs had working volumes of 6 L and were each equipped with an air diffuser and a mechanical stirrer. The SBRs were operated at 16.6–17.8°C and pH of 7.8–8.2. Because there are many studies on aerobic treatment of dairy wastewater, the aim of the SBR operation was only to produce effluent that would be post-treated by membrane filtration. Thus, in the SBRs, the experiment was performed

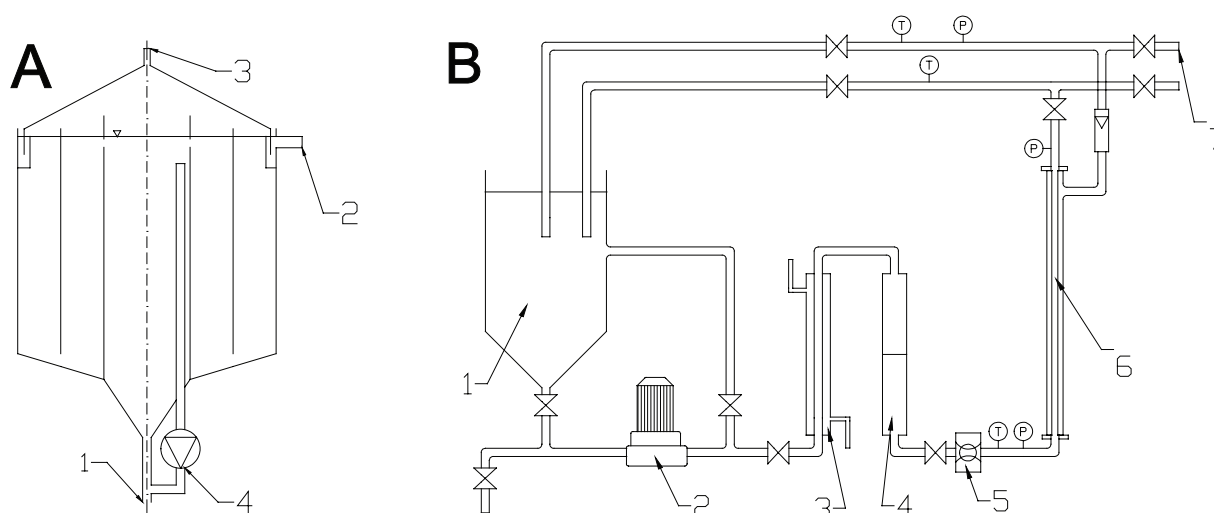


Fig. 1. A) Schematic diagram of the anaerobic plug-flow reactor: 1 – wastewater influent, 2 – wastewater effluent, 3 – biogas effluent, 4 – wastewater recirculation; B) Schematic diagram of the membrane installation: 1 – process tank, 2 – pump, 3 – heat exchanger, 4 – prefilter, 5 – flow control, 6 – membrane module, 7 – permeate sampling point, T – thermometer, P – manometer.

at an HRT of 4 d, similar to that given in the literature [27]. The other operational parameters are given in Table 1. The length of the cycle was 24 h, in which the length of each phase was as follows: 0.5 h of filling; 22 h of reaction, including 2 h of anoxic phase and 20 h of aeration phase; 1 h of sedimentation; and 0.5 h of decanting. In the anoxic phase, the dissolved oxygen (DO) concentration was below 0.2 mg/L; in the aeration phase, it was about 6 mg/L. The SBRs were inoculated with activated sludge from the aerobic chamber of a municipal wastewater treatment plant. The adaptation of biomass to the experimental conditions lasted about 20 days, at which point the concentrations of COD and  $\text{NH}_4\text{-N}$  in the effluent had not changed by more than 15% over the course of seven days. The two SBRs were operated in parallel; the results are presented as the arithmetic mean of data from both SBRs.

### 2.3. Membrane filtration

The effluent from the SBRs collected in the decanting phase was the feed solution for membrane filtration. The installation for membrane filtration is shown in Fig. 1B. The ceramic membrane that was used and the operational parameters of this installation were described in Zielińska and Galik [29]. In the installation, a high-pressure pump transported the feed solution throughout the system and ensured cross-flow of the permeate through the membrane. The membrane in the filtration unit had a filtration area (A) of 0.1 m<sup>2</sup> and a molecular weight *cut-off* of 150 kDa, which puts it in the range of UF. According to the producer, the pure water permeability of this membrane was 450 L/(m<sup>2</sup>·h·bar). The installation was equipped with two pressure gauges (at the feed solution and retentate streams) that supplied the data on operational pressures used to calculate the TMP. Four experimental series were conducted that differed in their TMP: 0.2, 0.3, 0.4 and 0.6 MPa. The filtrations were performed at 21 ± 2°C with an initial cross flow velocity of about 15 L/min. During the filtration cycle, the permeate

Table 1  
Operational conditions of the SBR and technological results

Parameter	Value
OLR, kg COD/(m <sup>3</sup> ·d)	0.61 ± 0.08
F/M, kg COD/(kg MLSS·d)	0.13 ± 0.03
MLSS, g/L	4.8 ± 0.4
MLVSS/MLSS, %	72 ± 2
SRT, d	42
Efficiency of COD removal, %	93.9
Efficiency of nitrification, %	80.4
Efficiency of denitrification, %	89.4
Efficiency of nitrogen removal, %	78.8
Efficiency of phosphorus removal, %	88.0
$k_{\text{COD}}$ , h <sup>-1</sup>	0.98
$r_{\text{COD}}$ , mg/(g MLVSS·h)	120.20
$k_{\text{NH}_4\text{-N}}$ , mg/(L·h)	4.34
$r_{\text{NH}_4\text{-N}}$ , mg/(g MLVSS·h)	1.26
OUR <sub>1</sub> , mg O <sub>2</sub> /(L·h)	25.74
OUR <sub>2</sub> , mg O <sub>2</sub> /(L·h)	2.70
OUR <sub>3</sub> , mg O <sub>2</sub> /(L·h)	15.66
SOUR <sub>1</sub> , mg O <sub>2</sub> /(g MLVSS·h)	7.45
SOUR <sub>2</sub> , mg O <sub>2</sub> /(g MLVSS·h)	0.78
SOUR <sub>3</sub> , mg O <sub>2</sub> /(g MLVSS·h)	4.53

F/M – food/microorganisms ratio, MLSS – mixed liquor suspended sludge, MLVSS – mixed liquor volatile suspended sludge, SRT – solids retention time,  $k_{\text{COD}}$  – rate constant for COD removal,  $r_{\text{COD}}$  – rate of COD removal,  $k_{\text{NH}_4\text{-N}}$  – rate constant for ammonia removal,  $r_{\text{NH}_4\text{-N}}$  – rate of ammonia removal, OUR<sub>1</sub> – oxygen uptake rate in exogenous respiration, OUR<sub>2</sub> – oxygen uptake rate in nitrification, OUR<sub>3</sub> – oxygen uptake rate in endogenous respiration, SOUR<sub>1</sub> – specific oxygen uptake rate in exogenous respiration, SOUR<sub>2</sub> – specific oxygen uptake rate in nitrification, SOUR<sub>3</sub> – specific oxygen uptake rate in endogenous respiration.

was received from the installation and the retentate was constantly circulated back to the feed tank. Thus, this was in fact the velocity of both the feed and the retentate that circulated in the loop. During the filtration cycle, permeation (batch) tests were conducted. These tests relied on the measurement of the time (t) necessary for collecting known volumes of permeate. Due to the fact that the membrane installation was not equipped with automatic backwashing, the permeation tests were carried out until the membrane was totally clogged and no permeate flow was obtained. Then, the membrane installation was washed according to the producer's instructions. This procedure included washing in the following sequence: with deionized water, then with 2% NaOH solution, next with deionized water to obtain neutral pH, next with 1% HNO<sub>3</sub> solution, and finally with deionized water to obtain neutral pH. After washing, the permeation flux of deionized water ( $J_w$ ) was measured. Each time, the recovery of the initial membrane flux was 95–97%. Directly before the next permeation test, the installation was operated to eliminate about 1 L of deionized water that was present in the system as dead volume.

The volumes of feed ( $V_f$ ), permeates ( $V_p$ ) and retentates ( $V_r$ ) obtained during the permeation tests were used to calculate the basic hydraulic parameters of the membrane, such as the permeate flux ( $J_v$ ), normalized flux ( $\alpha$ ), permeate recovery (Y), volumetric concentration factor (VCF), and the total membrane resistance ( $R_m$ ). These parameters were calculated with the following equations:

$$J_v = \frac{V_p}{t \cdot A} \quad (\text{L}/(\text{m}^2 \cdot \text{h}))$$

$$\alpha = \frac{J_v}{J_w} \quad (-)$$

$$Y = \frac{V_p}{V_f} \cdot 100 \quad (\%)$$

$$\text{VCF} = \frac{V_f}{V_r} \quad (-)$$

$$R_m = \frac{\text{TMP}}{J_v} \quad ((\text{MPa} \cdot \text{s})/\text{m})$$

#### 2.4. Analytical methods

The concentrations of COD, nitrogen and phosphorus in the influent and effluent of the SBR and in the permeates were measured spectro photometrically with cuvette tests (Hach Lange GmbH, Germany). BOD<sub>5</sub> was measured with a WTW Oxi Top BOD meter (Germany). The pH of wastewater was measured with an HI 2210 pH-meter (Hanna Instruments). Total solids (TS) and total suspended solids (TSS) were measured according to APHA [30]. Conductivity and total dissolved solids (TDS) were measured with a Hanna conductivity meter (HI 8733). DO concentration in the SBR was measured with a Pro ODO optical oxygen meter (YSI Environmental). In the SBR, the sludge volumetric index (SVI), MLSS and MLVSS were measured according to APHA [30]. At the end of the experiment in the SBRs, COD and ammonia concentrations were measured during

the cycle to investigate the changes in concentrations of these pollutants over time and to determine the kinetic parameters of removal, i.e. the rate constants and rates of COD and ammonia removal. The microbial activity of the activated sludge was measured using an Oxi Top control respirometric unit (OC 110, WTW), according to Zielińska et al. [31], to determine the specific oxygen uptake rates of exogenous respiration, nitrification and endogenous respiration. To determine the significance of differences between the series and for calculations of kinetic parameters of COD, ammonia removal and oxygen uptake, Statistica 13.1 was used.

### 3. Results and discussion

#### 3.1. Anaerobic treatment

In the anaerobic reactor treating raw dairy wastewater, 350 L of biogas was produced per 1 kg of COD removed. The biogas contained 55% methane. The main characteristics of the effluent from this reactor were as follows: 2150–2620 mg COD/L, 360–380 mg BOD/L, 100.0–100.8 mg total Kjeldahl nitrogen (TKN)/L, 60–70 mg NH<sub>4</sub>-N/L, 35.0–37.5 mg total phosphorus (TP)/L, 2350–2600 mg TS/L, 1710–1880 mg TDS/L, 380–580 mg TSS/L, and pH 6.5–6.7. This effluent served as the inflow into the SBR.

#### 3.2. Performance of the SBR

In the SBR effluent, the concentrations of pollutants were as follows: 132.0 ± 5.0 mg COD/L, 40.0 ± 2.0 mg BOD/L, 12.2 ± 2.0 mg TKN/L, 1.3 ± 0.2 mg NH<sub>4</sub>-N/L, 0.113 ± 0.003 mg NO<sub>2</sub>-N/L, 6.2 ± 0.8 mg NO<sub>3</sub>-N/L, 3.8 ± 0.3 mg P/L, 30.0 ± 3.0 mg TSS/L; the pH was 8.0 ± 0.3. In Table 1, the efficiencies of pollutant removal in the SBR are given. The nearly complete removal of ammonia in the SBR was due to the operational conditions, such as the long SRT and the long aeration period, which promoted the growth of nitrifying bacteria. The total efficiency of ammonia removal was 97%, due to nitrification and ammonia uptake for biomass synthesis. 80.4% of all ammonia removed was oxidized in nitrification. The introduction of an anoxic period resulted in high denitrification efficiency: 89.4% of all the oxidized nitrogen was reduced. Total nitrogen removal, including denitrification and biomass synthesis, was 78.8%. The unexpectedly high efficiency of phosphorus removal (88%) may have been due to the fact that a defined anaerobic zone is not necessarily required for the putative growth of phosphate accumulating microorganisms because phosphate storage may provide a selective advantage in fulfilling cell maintenance requirements in substrate-limited conditions (low F/M) [32]. These performance indicators are similar to those obtained in other experiments on dairy wastewater treatment. For example, Mohseni-Bandpi and Bazari [33] achieved a COD removal efficiency of above 90% at 2500 mg COD/L in the influent, using an SBR under DO concentrations of 3, 5, 6.5, and 7.5 mg/L. Lateef et al. [27] reported that 5 d was the most advantageous HRT for maximum efficiency of COD and BOD removal (96% at a DO concentration between 3 and 4.2 mg/L). Tawfik et al. [34] reported removal efficiencies of BOD and COD in an

up-flow anaerobic sludge blanket reactor followed by an aerobic activated sludge system of 98.8% and 97.4%, respectively, with an effluent quality (7.1 mg BOD/L and 35.0 mg COD/L) sufficient to discharge it into agricultural drains. The final quality of the effluent in the present study was not as good, probably because of the composition of the influent; in the studies by Tawfik et al. [34], mixed dairy and domestic wastewater was treated. In the present study, the effluent concentrations met the Polish standards for wastewater discharge into an aquatic environment for the smallest wastewater treatment plants (people equivalent < 2000).

The activated sludge was characterized by good settling properties as indicated by an average SVI ranging from 98 to 80 mL/g. The settling ability was the main indicator that was used to set the volumetric exchange ratio in the SBR. The initial experiments were conducted at ratios of 50% and 30%; however, this resulted in SVIs of 200 and 143 mL/g, respectively, washout of the biomass from the reactor and low quality of the effluent. For this reason, a volumetric exchange ratio of 25% was used because it was the highest tested ratio at which the quality of the SBR effluent was high enough to be used as a feed solution for the subsequent membrane filtration. Although the settling ability of activated sludge is not important for the overall performance of, for example, membrane bioreactors [20], a high concentration of suspended solids in the feed solution would negatively affect the hydraulic capacity of the membrane.

In the SBR cycle, COD concentrations decreased by almost 25% in the anoxic phase, and then under aerobic conditions, these changes in concentration were described by a first-order equation (Fig. 2). About half of the COD was consumed during the first hour of the aerobic phase. The rate of COD removal in this phase was 120.2 mg/(g MLVSS·h) (Table 1). During the anoxic phase, the concentration of ammonia was almost unchanging, whereas in the aerobic phase it was removed at a rate of 1.26 mg/(g MLVSS·h). Ammonia removal, up to the point that its final concentration in the effluent was reached, lasted about 6–7 h, which means that this reaction period permits safe operation of the SBR in case of a sudden increase in ammonia load in the influent. However, the short time that was necessary for ammonia oxidation indicates that it would be possible to shorten the reaction period by about 25% and still maintain efficient and fast ammonia oxidation. The reason for these high rates of removal could be the fact that activated sludge was characterized by high stability as indicated by the

MLVSS/MLSS ratio of 0.72. In addition, oxygen consumed by biomass for oxidation of ammonia and organic matter (exogenous respiration), and for endogenous respiration, indicated that the biomass was active. The oxygen uptake rates were described by first-order kinetics. The SOURs for exogenous respiration, nitrification and endogenous respiration were 7.45, 0.78 and 4.53 mg O<sub>2</sub>/(g MLVSS·h), respectively (Table 1). These values indicate stability and proper performance of activated sludge, in which the oxygen uptake for organics oxidation is higher than its uptake for nitrification because the growth rate of heterotrophic bacteria is higher than that of autotrophic bacteria.

Although the residual concentrations of pollutants met the criteria for wastewater discharge into an aquatic environment, the results of the present study indicate that the aerobic post-treatment of anaerobically treated dairy wastewater does not satisfy the limits established for water reuse. Therefore, further treatment using membranes is required, which was the major focus of this study.

### 3.3. Membrane filtration

Permeation tests showed the changes in volumetric permeate flux ( $J_v$ ) over time and were the bases for determination of the hydraulic capacity of the UF installation. In the study, the initial  $J_v$  was about 16 L/(m<sup>2</sup>·h) at TMPs of 0.2 and 0.3 MPa (Fig. 3). An increase in pressure is one method of improving flux, hence the TMP was increased to 0.4 and 0.6 MPa, which resulted in an increase in  $J_v$  to 23.0 and 28.5 L/(m<sup>2</sup>·h), respectively. Higher cross flow velocity could create turbulence in a membrane module and decrease the deposition of particles on the membrane surface. However, as can be seen from Fig. 3,  $J_v$  decreased with the time of filtration because of concentration polarization and fouling of the membrane with pollutants that were present in the feed solution, which are the main limitations of membrane processes. The main contributors to UF membrane fouling are the residual organic materials that are present in secondary effluents, like total suspended solids, organic colloids and exogenous polymers [35]. In addition, when treating dairy wastewater that contains calcium, carbonates and phosphates, membrane blocking by inorganic compounds should be considered. If these compounds are present as ions, they are unlikely to cause membrane fouling themselves, because they are smaller than the membrane pores. However, positively charged calcium ions cover negatively

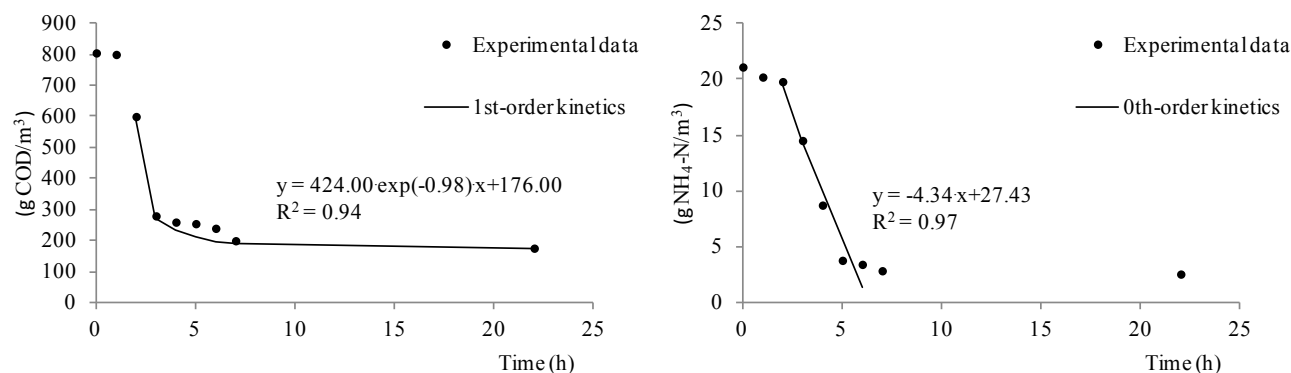


Fig. 2. Changes in COD and ammonia concentrations in the SBR cycle.

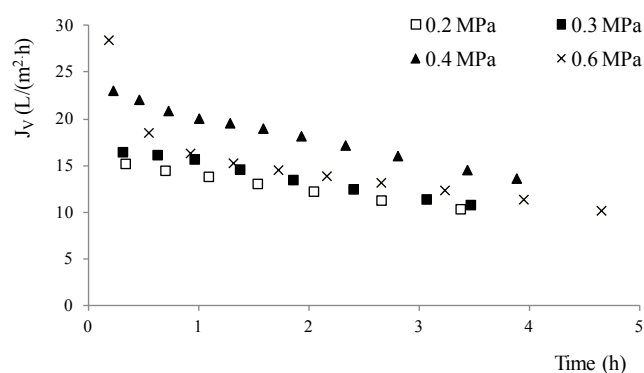


Fig. 3. Changes in  $J_v$  over time.

charged functional groups in extracellular polymers, which plays a major role in biofloculation and also increases the size of particles [36]. Next, during anaerobic treatment, calcium could interact with carbonates and phosphates and precipitate, which would increase the size of particles and lead to membrane scaling [37]; however, these inorganic precipitates form a layer on the membrane surface, which reduces the flux, instead of blocking the membrane pores.

In Fig. 3, the last point on the graphs indicates the moment at which the last portion of permeate was collected just before the permeate stopped flowing. This phenomenon was observed after about 3.5 h of filtration at TMPs of 0.2 and 0.3 MPa, and after over 4.5 h at 0.6 MPa. These working periods affect the frequency of membrane washing. At TMPs of 0.2, 0.3 and 0.4 MPa, the flux decline was almost the same and it decreased gradually with time. Although, at a TMP of 0.6 MPa, the initial  $J_v$  was the highest and the flow lasted for the longest time, the membrane showed a rapid initial decline in permeate flux of about 43% within less than 1 h. This rapid decline was due to more serious membrane fouling caused by the higher pressure. Such a decline is likely due to pore blocking, but a slight flux decline, as observed at lower TMPs, could be caused by both pore blocking and concentration polarization [38].

There was a linear relationship between average permeate flux and TMP in the range of 0.2–0.4 MPa. The average permeate flux was significantly higher at 0.4 MPa than at 0.2 MPa ( $p = 0.023$ ) (Table 2). A further increase to 0.6 MPa resulted in a decline in flux. The fraction of the feed flow which passes through the membrane ( $Y$ ) was also highest at a TMP of 0.4 MPa (Table 2). As opposed to 53% recovery at a TMP of 0.4 MPa, it took about 1 h longer to get 48% recovery at 0.6 MPa. This could be due to the accumulation of solutes near the membrane surface that could plug the pores [38]. Under these conditions, the percentage of adsorption on a membrane would be higher: pollutants would form a gel layer and would be highly compressed on the membrane because of the pressure [39]. As a consequence, the  $R_m$  increases with the TMP, causing the flux to decline due to pore blocking and saturation of the membrane with pollutants. An increase in pressure from 0.4 to 0.6 MPa nearly doubled the  $R_m$  (Table 2).

In this study, the constant outflow of permeate from the system during filtration and the constant circulation of the retentate to the feed tank caused the feed solution to become more concentrated. In Fig. 4 it can be seen that

Table 2

Mean hydraulic parameters of the membrane installation

Parameter	0.2 MPa	0.3 MPa	0.4 MPa	0.6 MPa
$J_v$ , L/(m <sup>2</sup> ·h)	12.96	13.92	18.60	14.55
$Y$ , %	35	38	53	48
VCF, –	1.54	1.60	2.13	1.90
$\alpha$ , –	0.86	0.62	0.62	0.65
$R_m$ , (MPa·s)/m	55,537	77,560	77,417	148,441

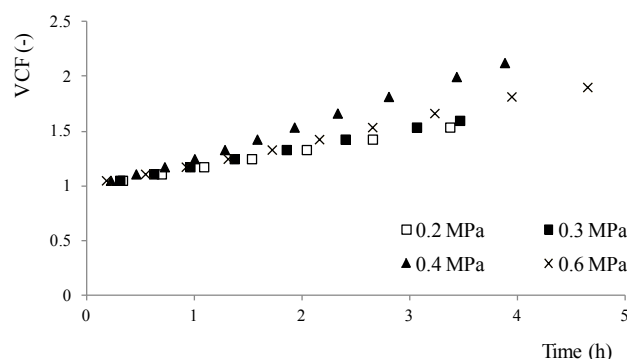


Fig. 4. VCF in relation to TMP.

at each TMP the volumetric concentration factor increased with time, up to about 2.0. The higher concentration of the solution in the feed tank increased membrane plugging and decreased the flux.

With the membrane, the average permeation flux of deionized water ( $J_w$ ) was 1350 L/(m<sup>2</sup>·h). The decrease in  $J_v$  was larger than that of  $J_w$  as can be seen from the values of normalized flux (Table 2). In all the experimental series,  $\alpha$  was below 1, which suggests membrane fouling. However, these values are above 0.5, suggesting that the fouling was not fast. According to some authors, it is not so much the size of the pollutant particles that is responsible for membrane plugging, but the ratio between the size of the particles and that of the pores [40]. Particles close to or smaller than the pore diameter clog the pores and membrane surface more quickly than larger particles.

In this study, the efficiency of membrane filtration was evaluated in terms of COD, BOD and TSS removal. The retention of turbidity was the highest of all retained pollutants, as the permeate was completely free of suspended solids. The retention of organic compounds was not sensitive to the pressure changes, despite the fact that an increase in the TMP leads to an increase in the permeate flux at lower solute concentrations, resulting in a more diluted permeate stream [38]. In the present study, 88.9% of COD and 90.9% of BOD were removed. These rejection coefficients contributed to the total efficiencies of the system with aerobic biological treatment and membrane filtration, which were 100% of TSS, 99% of COD and 99% of BOD.

Although the cut-off of the UF membrane was probably larger than the size of some dissolved organic compounds in wastewater, the concentrations of COD and BOD in the permeates were very low (Table 3). Independently of the TMP, COD and BOD concentrations in the permeates were about

Table 3  
Concentrations of pollutants in the permeates

Pollutant indicator	0.2 MPa	0.3 MPa	0.4 MPa	0.6 MPa
COD, mg/L	14.4 ± 2.1	14.6 ± 2.2	14.9 ± 2.2	14.5 ± 2.1
BOD, mg/L	3.6 ± 0.8	3.7 ± 1.2	3.7 ± 1.4	3.6 ± 1.4
Total hardness, mg CaCO <sub>3</sub> /L	275.0 ± 22.1	275.1 ± 13.5	275.2 ± 21.4	275.0 ± 18.5
Conductivity, µS/cm	310.0 ± 0.2	310.1 ± 0.4	310.2 ± 0.2	310.0 ± 0.3
TSS, mg/L	nd	nd	nd	nd
TDS, mg/L	198 ± 23	195 ± 15	203 ± 20	190 ± 35
NO <sub>3</sub> -N, mg/L	4.4 ± 0.5	4.2 ± 0.8	4.6 ± 0.5	4.1 ± 0.4
TP, mg/L	0.4 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.4 ± 0.1
pH	8.5 ± 0.1	8.5 ± 0.1	8.5 ± 0.1	8.5 ± 0.2

nd – not detected

14.5 mg/L and 3.6 mg/L, respectively. This indicates that retention depended not only on sieve retention but also on adsorption of pollutants on the membrane surface. This fouling reduces the nominal diameter of the membrane pores to the so-called “effective diameter”, making possible the rejection of particles smaller than the membrane cut-off [41].

The above results are very promising with regard to the possibility of reusing the treated water. For both environmental and economical considerations, industries have to find solutions to effectively treat effluents and reuse them at the head of production processes. It should be noted that different industries have their own criteria for reused water. In the permeates obtained in the present study, the conductivity, hardness, and TDS were about 310 µS/cm, 275 mg CaCO<sub>3</sub>/L and 196 mg/L, respectively (Table 3). Taking into account the following requirements for permeate use, it is possible to reuse the permeate obtained in the present study for irrigation, cooling and external cleaning (washing cars, floors and some external surfaces), construction, and flushing:

- i) requirements for irrigation: TSS < 10 mg/L, COD < 100 mg/L, conductivity 250–750 mg/L, TDS < 450 mg/L, NO<sub>3</sub>-N 5–30 mg/L, pH 6.5–8.3 [42];
- ii) requirements for cooling water: COD 75 mg/L, pH 6.9–9.0, TDS 500 mg/L, hardness 650 mg CaCO<sub>3</sub>/L [43];
- iii) external cleaning, construction, and flushing require lower quality water than the permeates obtained in the current study.

Effluent from sequencing biological treatment and MF or UF has been directly reused for irrigation [16] or for recreational purposes after removal of the residual color [44]. However, because very low concentrations of pathogens are required for agricultural use and the microbiological characteristics of the permeates in the present study were not determined, disinfection might be required before using these permeates in this manner. Although UF membranes are generally considered an absolute barrier to all pathogens, Falsanisi et al. [45] identified a small amount of coliform microbes in a study on the reuse in agriculture of municipal wastewater treated with UF. However, the requirements for different industrial uses depend on the

site-specific end use. The specific requirements given by the US Environmental Protection Agency (EPA 2012) concern municipal wastewater.

Final concentrations of nitrate and total phosphorus were 4.4 mg/L and 0.4 mg/L, respectively. This was not an expected result; however, according to a study by Kim et al. [46] on the pre-treatment of secondary effluent with a UF membrane, nitrate and phosphate were reduced by 20% and 26%, respectively. The reason for the unexpectedly low concentrations in the present study could be the nature of the wastewater and its composition. Dairy wastewater has a high content of proteins, fats, grease and lipids. Therefore, some residuals could be present even after biological treatment, for example, long chain fatty acids, as they are less biodegradable. Fatty acids are adsorbed on the membrane surface. Hence, they can restrict the passage of nitrogen and phosphorus through the membrane by blocking the membrane. Another possible reason is that phosphorus was biologically assimilated by phosphate accumulating microorganisms and therefore bound to suspended solids. Because UF membranes reject suspended solids, the phosphorus could also be rejected. In addition, as can be seen from the decrease in phosphorus content by about 87% during anaerobic treatment in the present study, phosphorus could have precipitated as CaPO<sub>4</sub> and been rejected by the membrane.

Membrane filtration is a mass transport process, so the concentration of dissolved organic matter, suspended solids, microorganisms and pathogens in the retentate depends on permeate recovery and rejection. Based on experimental data on pollutant loadings in the feed solution and permeates in the present study, the mass balance was calculated, indicating that the loadings of pollutants in the retentate were 0.3 g TSS, 1.3 g COD, and 0.4 g BOD per filtration cycle. Studies that investigate how to manage retentate are scarce. Due to its high concentration of organic matter, retentate can serve as an excellent source for biogas production. The residual organic matter in the effluent after biological treatment is less biodegradable, but during the UF process, the high pressure may disrupt the particles, increasing their biodegradability. Therefore, the retentate could be managed by recycling it into the anaerobic bioreactor, in which energy could be recovered from this concentrated stream.

#### 4. Conclusions

This study revealed that an activated sludge-UF system can effectively post-treat anaerobically treated dairy wastewater, achieving total removal efficiencies of 100% of TSS, and 99% of COD and BOD. Although the influence of TMP on the permeate quality was not statistically significant, a recovery of 53% of the effluent was possible at 0.4 MPa. This technology incorporating membranes shows promise for the production of effluent that can be reused for different applications. The obtained permeates could be reused for irrigation, cooling and external cleaning (washing cars, floors and some external surfaces), construction, and flushing, and the retentates could be recycled back into the anaerobic bioreactor. Such reuse of permeates could provide economic benefits by reducing water consumption.

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#### References

- [1] B. Farizoglu, S. Uzuner, The investigation of dairy industry wastewater treatment in a biological high performance membrane system, *Biochem. Eng. J.*, 57 (2011) 46–54.
- [2] BREF 2006, Integrated pollution prevention and control, Reference Document on Best Available Techniques in the Food, Drink and Milk Industries, August 16, 2006.
- [3] F. Carta-Escobar, J. Pereda-Marin, P. Alvarez-Mateos, R. Romero-Guzman, M.M. Duran-Barrantes, F. Barriga-Mateos, Aerobic purification of dairy wastewater in continuous regime Part I: Analysis of the biodegradation process in two reactor configurations, *Biochem. Eng. J.*, 21 (2004) 183–191.
- [4] M.C. Cammarota, D.M.G. Freire, A review on hydrolytic enzymes in the treatment of wastewater with high oil and grease content, *Bioresour. Technol.*, 97 (2006) 2195–2210.
- [5] H. Lin, W. Peng, M. Zhang, J. Chen, H. Hong, Y. Zhang, A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives, *Desalination*, 314 (2013) 169–188.
- [6] D.T. Sponza, H. Atalay, Simultaneous phosphorus, nitrogen and dinitrotoluene removals in batch anaerobic/anoxic/aerobic sequential, *Process Biochem.*, 40 (2005) 25–34.
- [7] J.P. Kushwaha, V.C. Srivastava, I.D. Mall, An overview of various technologies for the treatment of dairy waste waters, *Crit. Rev. Food Sci. Nutr.*, 51 (2011) 442–452.
- [8] D. Dionisi, M. Majone, V. Tandoi, M. Beccari, Sequencing batch reactor: influence of periodic operation on performance of activated sludges in biological wastewater treatment, *Ind. Eng. Chem. Res.*, 40 (2001) 5110–5119.
- [9] J.P. Kushwaha, V.C. Srivastava, I.D. Mall, Sequential batch reactor for dairy wastewater treatment: Parametric optimization; kinetics and waste sludge disposal, *J. Environ. Chem. Eng.*, 1 (2013) 1036–1043.
- [10] C. Meunier, O. Henriot, B. Schoonbroodt, J.M. Boeur, J. Mahillon, P. Henry, Influence of feeding pattern and hydraulic selection pressure to control filamentous bulking in biological treatment of dairy waste waters, *Bioresour. Technol.*, 221 (2016) 300–309.
- [11] A.E. Ghaly, A comparative study of anaerobic digestion of acid cheese whey and dairy manure in a two-stage reactor, *Bioresour. Technol.*, 58 (1996) 61–72.
- [12] E. Castelló, C.G. Santos, T. Iglesias, G. Paulino, J. Wenzel, L. Borzacconi, C. Etchebehere, Feasibility of biohydrogen production from cheese whey using a UASB reactor: links between microbial community and reactor performance, *Int. J. Hydrog. Energy*, 34 (2009) 5674–5682.
- [13] B. Sarkar, P.P. Chakrabarti, A. Vijaykumar, V. Kale, Wastewater treatment in dairy industries – possibility of reuse, *Desalination*, 195 (2006) 141–152.
- [14] J. Wojdalski, B. Drózd, J. Piechocki, M. Gaworski, Z. Zander, J. Marjanowski, Determinants of water consumption in the dairy industry, *Polish J. Chem. Technol.*, 15 (2013) 61–72.
- [15] B. Balanec, G. Gesan-Guiziu, B. Chaufer, M. Rabiller-Baudry, G. Daufin, Treatment of dairy process waters by membrane operations for water reuse and milk constituents concentration, *Desalination*, 147 (2002) 89–94.
- [16] D. Bixio, C. Thoeye, J. De Koning, D. Joksimovic, D. Savic, T. Wintgens, T. Melin, Wastewater reuse in Europe, *Desalination*, 187 (2006) 89–101.
- [17] A. Chollangi, M.M. Hossain, Separation of proteins and lactose from dairy wastewater, *Chem. Eng. Process.*, 46 (2007) 398–404.
- [18] F. Aghili, A.A. Ghoreysi, A. Rahimpour, M. Rahimnejad, Enhanced treatment of pretreated sour whey by PAC adsorption/membrane process, *Chem. Eng. Process.*, 99 (2016) 80–85.
- [19] J. Luo, L. Ding, B. Qi, M.Y. Jaffrin, Y. Wan, A two-stage ultrafiltration and nanofiltration process for recycling dairy wastewater, *Bioresour. Technol.*, 102 (2011) 7437–7442.
- [20] S. Judd, *The MBR Book: Principles and applications of membrane bioreactors in water and wastewater treatment*, Elsevier Ltd., Oxford, Oxfordshire, 2006.
- [21] M. Qadir, D. Wichelns, L. Raschid-Sally, P.G. McCornick, P. Drechsel, A. Bahri, P.S. Minhas, The challenges of wastewater irrigation in developing countries, *Agric. Water Manage.*, 97 (2010) 561–568.
- [22] S.M.K. Sadr, D.P. Saroj, S. Kouchaki, A.A. Ilmohade, S.K. Ouki, A group decision-making tool for the application of membrane technologies in different water reuse scenarios, *J. Environ. Manage.*, 156 (2015) 97–108.
- [23] B. Van der Bruggen, M. Manttari, M. Nyström, Drawbacks of applying nanofiltration and how to avoid them: a review, *Sep. Purif. Technol.*, 63 (2008) 251–263.
- [24] S.S. Madaeni, Y. Mansourpanah, Chemical cleaning of reverse osmosis membranes fouled by whey, *Desalination*, 161 (2004) 13–24.
- [25] S.J. Lee, M. Dilaver, P.K. Park, J.H. Kim, Comparative analysis of fouling characteristics of ceramic and polymeric microfiltration membranes using filtration models, *J. Membr. Sci.*, 432 (2013) 97–105.
- [26] R.W. Baker, *Membrane Technology and Applications*, second ed., John Wiley and Sons, 2000.
- [27] A. Lateef, M.N. Chaudhry, S. Ilyas, Biological treatment of dairy wastewater using activated sludge, *Science Asia*, 39 (2013) 179–185.
- [28] R.V. Kumar, L. Goswami, K. Pakshirajan, G. Pugazhenthii, Dairy wastewater treatment using a novel low cost tubular ceramic membrane and membrane fouling mechanism using pore blocking models, *J. Water Process Eng.*, 13 (2016) 168–175.
- [29] M. Zielińska, M. Galik, Use of ceramic membranes in a membrane filtration supported by coagulation for the treatment of dairy wastewater, *Water Air Soil Pollut.*, 228 (2017) 173.
- [30] APHA Standard Methods for the Examination of Water and Wastewater, 18<sup>th</sup> ed., APHA, AWWA and WEF, Washington, 1992.
- [31] M. Zielińska, K. Bernat, D. Kulikowska, A. Cydzik-Kwiatkowska, I. Wojnowska-Baryła, Activated sludge activity in the treatment of anaerobic sludge digester supernatant, *Environ. Protect. Eng.*, 38 (2012) 17–27.
- [32] L.H. Andrade, F.D.S. Mendes, J.C. Espindola, M.C.S. Amaral, Nanofiltration as tertiary treatment for the reuse of dairy wastewater treated by membrane bioreactor, *Sep. Purif. Technol.*, 126 (2014) 21–29.
- [33] A. Mohseni-Bandpi, H. Bazari, Biological treatment of dairy wastewater by sequencing batch reactor, *Iranian J. Env. Health Sci. Eng.*, 1 (2004) 65–69.



- [34] A. Tawfik, M. Sobhey, M. Badawy, Treatment of a combined dairy and domestic wastewater in an up-flow anaerobic sludge blanket (UASB) reactor followed by activated sludge (AS system), *Desalination*, 227 (2008) 167–177.
- [35] C.N. Laabs, G.L. Amy, M. Jekel, Understanding the size and character of fouling-causing substances from effluent organic matter (EfOM) in low – pressure membrane filtration, *Environ. Sci. Technol.*, 40 (2006) 4495–4499.
- [36] I.S. Kim, N. Jang, The effect of calcium on the membrane bio-fouling in the membrane bioreactor (MBR), *Water Res.*, 40 (2006) 2756–2764.
- [37] H.S. You, C.P. Huang, J.R. Pan, S.C. Chang, behavior of membrane scaling during cross flow filtration in the anaerobic MBR system, *Sep. Purif. Technol.*, 41 (2006) 1265–1278.
- [38] A. Aouni, C. Fersi, B. Cuartas-Urbe, A. Bes-Pia, M.I. Alcaina-Miranda, M. Dhahbi, Study of membrane fouling using synthetic model solutions in UF and NF processes, *Chem. Eng. J.*, 175 (2011) 192–200.
- [39] X. Sun, C. Wang, Y. Li, W. Wang, J. Wei, Treatment of phenolic wastewater by combined UF and NF/RO processes, *Desalination*, 355 (2015) 68–74.
- [40] A.L. Lim, R. Bai, Membrane fouling and cleaning in micro-filtration of activated sludge wastewater, *J. Membr. Sci.*, 216 (2003) 279–290.
- [41] S. Muthukumar, D.A. Nguyen, K. Baskaran, Performance evaluation of different ultrafiltration membranes for the reclamation and reuse of secondary effluent, *Desalination*, 279 (2011) 383–389.
- [42] A. Bick, L. Gillerman, Y. Manor, G. Oron, Economic assessment of an integrated membrane system for secondary effluent polishing for unrestricted reuse, *Water*, 4 (2012) 219–236.
- [43] L.H. Andrade, F.D.S. Mendes, J.C. Espindola, M.C.S. Amaral, Reuse of dairy wastewater treated by membrane bioreactor and nanofiltration: Technical and economic feasibility, *Brazilian J. Chem. Eng.*, 32 (2015) 735–747.
- [44] S. Oota, T. Murakami, K. Takemura, K. Noto, Evaluation of MBR effluent characteristics for reuse purposes, *Water Sci. Technol.*, 51 (2005) 441–446.
- [45] D. Falsanisi, L. Liberti, M. Notarnicola, Ultrafiltration (UF) pilot plant for municipal wastewater reuse in agriculture: impact of the operation mode on process performance, *Water*, 2 (2010) 872–885.
- [46] S.L. Kim, J. Paul Chen, Y.P. Ting, Study on feed pretreatment for membrane filtration of secondary effluent, *Sep. Purif. Technol.*, 29 (2002) 171–179.