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Long-term use of the critical flux for fouling control in membrane bioreactors treating different industrial effluents: bench and pilot scale

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ABSTRACT

This paper presents an evaluation, in the long term, of the critical flux test as a tool to monitor and control fouling in membrane bioreactors (MBRs). The critical flux was assessed in this work on bench- and pilot-scale MBRs treating different kinds of effluents. The results showed that the critical flux test is important for the indication of the optimal operational flux and serves as a tool to compare propensity to fouling in several systems, independent of the characteristics of the feed or the operational conditions. The results also show that the improvement in the quality of the sludge or the improvement in the hydrodynamic conditions is an effective example of alternatives to increase the critical flux. As an example, these improvements can be achieved by dosing powder activated carbon, improving specific membrane permeability or increasing aeration rate.

Keywords: MBR; Critical flux; Fouling control

1. Introduction

Membrane bioreactors (MBRs) have been extensively employed for industrial wastewater treatment because they are more effective in removing pollutants than conventional processes. However, intrinsic phenomena of the membrane separation process, such as concentration polarization and fouling, are limiting factors to the use of this technology because they are responsible for the permeate flux reduction and for the increase in the transmembrane pressure (TMP) in permeation systems.

Membrane fouling can be managed by operating the system below the so-called *critical flux* (J_C). The critical flux is a quantitative parameter for the filterability of different membranes and different activated sludge mixtures. Field et al. [1] were the first researchers to conceptualize critical flux. According to these authors, the critical flux is the flux below which no decline in the permeate flux with the operating time is observed and above which fouling occurs. The critical flow can also be explained when there is a balance between the convection rate of these materials towards the membrane by the permeate drag flow and the back transport velocity of material from the membrane to the solution bulk due to shear and Brownian

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diffusion [2]. Thus, the critical flux value depends on the hydrodynamic conditions of the process, the membrane characteristics, operating conditions, feed characteristics, and sludge properties [3].

Field et al. [1] also outlined two forms of critical flux: strong and weak. The more rigid form (strong) is the one indicating the critical flux as being the point at which, for the same TMP, the solution flux becomes lower than the pure solvent flux. The second form (weak) considers that there will always occur some fouling at the beginning of permeation, primarily due to the static adsorption of solutes at the membrane, and that, for this reason, the solution flux will always be inferior to the pure solvent flux. In this case, the critical flux is considered the point at which the flux curve vs. pressure becomes non-linear.

However, according to Le Clech et al. [4], the strong form of critical flux is rarely observed in multi-dispersed and complex systems like the MBRs. The authors also showed that in MBRs, even for permeate fluxes as low as $2\,\mathrm{L\,h^{-1}\,m^{-2}}$, there is still a slight increase in the operating pressure over time for the maintenance of a constant permeate flux. Therefore, both forms of critical flux, strong and weak, have suffered relaxations when used in MBRs [5]. While, before, a flux below which no fouling could be observed was believed to exist; today the existence of subcritical fouling has already been proven [6].

Thus, instead of talking of a flux below which there is no fouling, a more recent understanding considers a flux below which the fouling rate is low and constant and above which it is intense, compromising the sustainability of the operation. Field and Pearce [7] named this flux value threshold flux.

There are different methodologies that can be applied to measure critical flux. They consist, basically, in a direct observation of the deposition of particles on the membrane surface, in a mass balance between the concentration of compounds at the entry and the exit of a membrane cell, and in the evaluation of the filtration profile, where the critical flux can be determined based on the relation between pressure and flux. Of these methods, the only one with practical applicability to pilot- or real-scale MBRs is the based on the relation between pressure and flux [8].

The simplest method to determine the critical flux through observation of the filtration profile consists in the fixation of the flux or pressure and on the measurement of the respective variable. The critical flux is defined as the point at which the linear relation between the two variables can no longer be observed. However, this method does not present high accuracy [8].

According to Bacchin et al. [5], the methods that work with a constant pressure present the advantage of reading of the flux in a stationary state because the occurrence of fouling causes a flux reduction, which consequently reduces the fouling rate and stabilizes the system. On the other hand, the methods that fix the flux and monitor the pressure allow the determination of the fouling rate, which is essential in order to evaluate the sustainability of the system.

The flux-step and TMP-step methods correlate with the imposition of a flux value and pressure reading corresponding to a given time segment, or in the opposite case, the imposition of a pressure value and reading of the flux, respectively. After the given time segment, the fixed variable suffers an increase and changes in the other variable are once again observed. It should be stressed that the time of maintaining each stage and the magnitude of the increase influence the critical flux value obtained [4,8]. Flux-step methods are to be preferred to TMP-step methods because the deposition rate of material on the membrane is better controlled, as the convective flux to the membrane is steady [4].

When defining the critical flux point, some authors arbitrarily determined a pressure variation considered critical (e.g. $\mathrm{d}P/\mathrm{d}t > 0.1\,\mathrm{mbar\,min^{-1}}$ for Le Clech et al. [4]). According to Bacchin et al. [5], this definition bares advantages in the sense that the choice of the critical flux point becomes clear and can be applied by several researchers.

Some authors investigated the variation in the aeration frequency close to the membrane surface, aiming to improve the hydrodynamics and consequently, at a reduction in fouling [9]. Psoch and Schiewer [10] investigated the use of backflushing together with the aeration frequency as a technique for the reduction in fouling in long-term MBR operations. Wu et al. [11] evaluated several different parameters like sludge concentration, aeration flow, and membrane properties for a better understanding of the critical flux values obtained. Studies in bench scale conducted by Navaratna and Jegatheesan [12] indicate that the intermittent operation of the MBR with relaxation of the membrane is efficient for fouling control and in accordance with the mathematical model applied, the critical flux decreases exponentially with the presence of the SMP (soluble microbial products) and variation in the sludge concentration, and linearly with the Extracellular polymeric substances. In the same study, the authors confirm that the determination of the critical flux by use of the flux-step method is more adequately applied in long-term operations than the short-term methods.

In literature, most articles about the use of critical flux for fouling control focus on concepts on the topic, methodologies, and trials. This paper presents an evaluation, in the long term, of critical flux test as a tool for monitoring and control of fouling in MBRs treating different effluents, as well as on the evaluation of the application of flux improvement techniques with the aim to elevate the critical flux. The critical flux trial methods adopted were flux step and TMP step [4], because they are easily applicable to large-scale units, apart from the shorter time required for the analysis, and other advantages mentioned before.

In this context, the aim of this work is to evaluate the long-term use of the critical flux for fouling control in MBRs treating different industrial effluents in bench and pilot scale.

2. Methodology

In this work, the long-term use of critical flux for fouling monitor in MBRs treating bleach pulp mill and dairy effluents in bench scale and refinery effluents in pilot scale was evaluated. At first, the effluents were characterized and used as feeding for the MBR, where the critical flux was periodically determined for every treatment system.

2.1. Effluents samples

In this article, petroleum refinery, bleach pulp mill, and dairy effluents were used as feeding line for treatment in the examined MBRs.

The petroleum refinery effluent used in the study came from Refinaria Gabriel Passos (REGAP Refinery) in Betim, Minas Gerais, Brazil. REGAP is a petroleum refinery owned by Petrobras, which produces paint thinner, asphalt, coke, sulfur, gasoline, LPG, diesel, and aviation kerosene. The effluent was sent to the pilot-scale units after a pretreatment in the oil–water separator, flotation, sand filter, and a hydrogen peroxide dosage for sulfide concentration control.

The bleach pulp mill effluent used for the experiments was collected in a bleached eucalyptus pulp (short-fiber) industry located in Brazil. The pulp production process used is the kraft process and the bleaching process is the D_{HT}(EP) D_1P (acid extraction with chlorine dioxide and alkaline extraction with hydrogen peroxide). The effluent was collected during the second stage of bleaching (alkaline extraction with hydrogen peroxide) and previously submitted to microfiltration for removal/recovery of lost fibers during the process.

The dairy effluent here evaluated came from a dairy industry, situated in the state of Minas Gerais, Brazil, which produces UHT milk, yoghurt, Minas cheese, cream cheese and Petit-suisse. The effluent was collected at the company's effluent treatment station after the stages of sieving and flotation with compressed air.

2.2. Physicochemical characterization of effluent

The effluents were characterized according to the parameters of chemical oxygen demand (COD) (5220 B), BOD (5210 B), pH (4500-H^+ B), alkalinity, color (spectrophotometer Hach DR2800), total solids (2540 B), total nitrogen (4500-N C) and ammonia (4500-NH_3 C), phosphorus (4500-P B), phenol index (5530 C), chlorides (4500-Cl), and total organic carbon (TOC) (SHIMADZU TOC-VCPN). The analyses were conducted in accordance with the Standard Methods for the Examination of Water and Wastewater [13].

2.3. MBR units and operational condition

The bench unit used for the treatment of effluents from the pulp and dairy industry consisted of two tanks for feed and permeate storage (13 and 4 L, respectively) and an aerobic bioreactor operated with a submerged microfiltration hollow fiber membrane module. Fig. 1 presents a chart of the submerged MBR system used. The total biological tank volume was 13 L. The system possesses five process currents: an MBR feeding line containing the effluent to be treated, a compressed air line for bioreactor aeration, a biologically degraded and microfiltered effluent line, a vacuum line, and a permeated backwash line. Table 1 shows the characteristics of the membrane modules and operational conditions of the MBR for each effluent treated.

The bench MBRs were operated using a backwash with duration of 15 s for each 15 min of permeation. The pH of the reaction liquid was maintained between 7–7.5 and the biological suspension temperature was kept between 25–30˚C. In the MBR fed with dairy effluent, the distributed aeration was applied on the base of the module. For chemical cleaning of the membrane, the modules were immersed in an ultrasonic bath with a 200-ppm hypochlorite solution. The cleaning time was 20 min, after which the module was rinsed with running water to remove the cleaning agent. The hydraulic permeability of the membrane in water after the cleaning was measured to evaluate the efficiency of the membrane cleaning.

To investigate the critical flux in pilot scale, two submerged MBR configurations were assessed for the treatment of a petroleum refinery effluent. The first MBR has flat-sheet microfiltration membranes (Kubota)

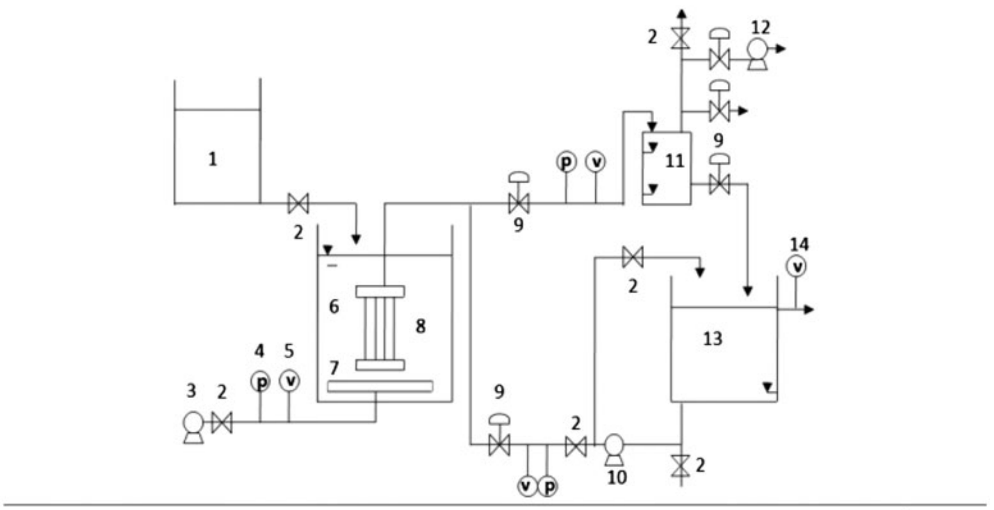

Fig. 1. Chart of MBR system.

Table 1
Characteristics of bench scale MBR and operational conditions for dairy and bleach pulp mill effluents

		Dairy		
Parameters	Units	Stage 1	Stage 2	Pulp
Volume of the biological reactor	m^3	0.0042	0.0042	0.0047
Aeration flow	$Nm^3\,h^{-1}$	0.5 (0.0*)	0.5 (3.5*)	0.5
Hydraulic retention time	H	6	6–8	6
Solids retention time	D	25–80	60	Infinite
MLVSS	$g\,L^{-1}$	7.0	22.0	3.6
Organic load	$kg\,COD\,d^{-1}$	10	16	36
Feed to micro-organism rate	d^{-1}	1.3	0.8	1.8
Permeate flow	$L\,h^{-1}$	0.8	0.55–0.75	0.5
Membrane material	–	Polyetherimide	Polyetherimide	Polyetherimide
Average pore diameter	μm	0.5	0.5	0.5
Membrane area	m^2	0.044	0.2	0.02
Membrane configuration	–	MF/Hollow fiber	MF/Hollow fiber	MF/Hollow fiber

*Air stream specifically used in the membrane module.

(MBR1), and the second MBR has hollow fiber ultrafiltration membranes (Zenon) (MBR2). Fig. 2 shows a chart of the experimental apparatus.

MBR1 has a membrane module submerged in the biological tank ($V = 8\,m^3$), while MBR2 has a membrane module submerged in an external membrane tank, i.e. MBR1's biological tank, which means both MBRs share the same biological tank. In order to maintain similar solid concentrations or liquid characteristics in both MBRs, the sludge from MBR1 was pumped into MBR2 at a flow rate of 2.5 times the permeation flow rate, and the sludge from MBR2 was pumped into MBR1 in order to keep the useful volume of MBR2 constant.

The driving force for permeation in MBR1 was the hydrostatic pressure of the water column. The unit had an aeration system to ensure oxygen was provided to the biological process and to ensure fouling control by the shear stress caused by the ascending (tangential) flow of air bubbles. MBR2 was operating

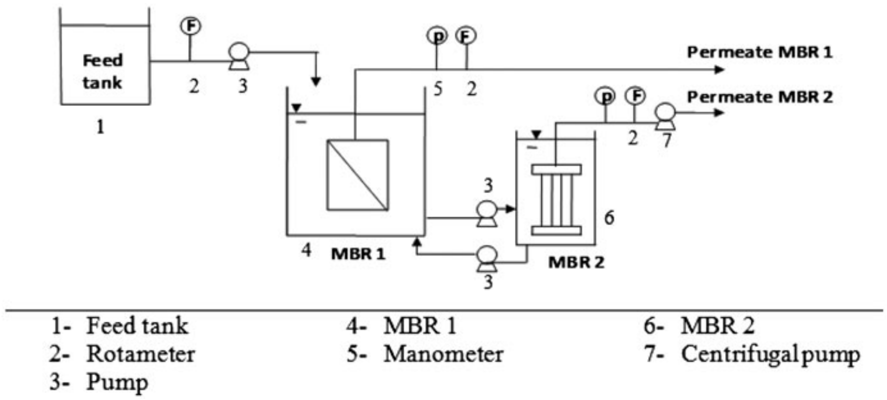

Fig. 2. Chart of MBR pilot-scale system, MBR flat plate (MBR1), and hollow fiber (MBR2).

in low vacuum (10–50 kPa) induced by a centrifugal permeate pump. The unit also had an aeration system with two tubes with 3 and 10 mm holes in the lower extremity of the module. Table 2 shows the characteristics of both MBRs as well as their operational conditions.

To maintain the permeate flux, since the MBRs were operated with constant flux, one minute of relaxation was performed after every nine minutes of permeation on MBR1, and 15 s of backwash after every 15 min of operation on MBR2. Regarding membrane cleaning, MBR1 was initially supposed to undergo a maintenance cleaning once a week with a 500-mg L^{-1} sodium percarbonate solution for 2 h and, at second stage, it was submitted to a recovery cleaning with a 5,000-mg L^{-1} sodium hypochlorite solution when permeability reached 100 $L\,h^{-1}\,m^{-2}\,bar^{-1}$. MBR2 underwent weekly maintenance cleanings of the membranes with a 200-mg L^{-1} sodium hypochlorite solution for 20 min and recovery cleanings with a 1,000-mg L^{-1} sodium hypochlorite solution for 24 h and a citric acid solution with pH lower than 2 for 20 min when the pressure to keep a constant flow was higher than 0.4 bar.

The performance of all MBRs was assessed through a periodic monitoring of the following process variables: permeate flow, applied pressure, TOC or COD, ammonia, phosphorus, total solids concentration, and critical flux.

2.4. Critical flux test procedure

The critical flux values were obtained from the flux-step or TMP-step method. To determine the critical flux in bench or pilot scale, the membrane module was previously, chemically, cleaned and immersed in the biological reactor. At the bench-scale units, the critical flux was determined by keeping the pressure

Table 2
Characteristics of MBR pilot scale and operational conditions for effluents of petroleum refinery

Parameters	Units	MBR1	MBR2
Volume of the biological reactor	m^3	8	8
Aeration flow	$Nm^3\,h^{-1}$	45	25
Hydraulic retention time	h	5.6	5.6
Solids retention time	d	40	40
MLVSS	$g\,L^{-1}$	8	8
Organic load	$kg\,COD\,d^{-1}$	13	13
Feed to micro-organism rate	d^{-1}	0.2	0.2
Permeate flow	$m^3\,h^{-1}$	0.5	1.2
Membrane material	–	PVDF	PES
Average pore diameter	μm	0.4	0.04
Membrane area	m^2	70	60
Membrane configuration	–	MF/Flat plate	UF/Hollow fiber

constant and monitoring the corresponding permeate flux (TMP step). For pilot-scale units, the critical flux was determined by keeping the permeate flux constant and monitoring the operating pressure (flux step). This difference in the methodology for pilot-scale and bench-scale units was necessary due to limitations on the equipment used in the bench unit, which did not allow for highly sensible pressure measurements and the possibility to maintain a strictly constant flux.

For the TMP-step method, for each pressure value, the filtration time was 18 min, after which the applied pressure was increased by 0.05 bar. The critical flux corresponded to the value at which a decrease in permeate flux was observed during the 18 min of permeation at constant pressure or constant flow. For the flux-step method, the filtration time for each constant permeate flux was also 18 min, after which it was increased by $2\,L\,h^{-1}\,m^{-2}$.

3. Results and discussion

3.1. Effluents characterization and the MBRs' removal efficiencies

The results of the physical-chemical characterization of effluents from the petroleum refinery, dairy, and cellulose industries are described in Table 3.

The organic matter content—in terms of COD, BOD, and TOC—in the pretreated refinery effluent is within the range documented in literature for this type of wastewater [14,15]. COD/BOD ratio values (average of 2.2) suggest that the effluent was suitable for biological treatment.

Table 3
Physical-chemical characteristics of effluents from the petroleum refinery, dairy, and cellulose industries

Parameters	Unit	Effluents		
		Refinery	Dairy	Pulp
COD	$mg\,L^{-1}$	610	3,963	1,455
BOD	$mg\,L^{-1}$	276	1,954	741
TOC	$mg\,L^{-1}$	205	953	205
TS	$g\,L^{-1}$	7.9	4.1	5.9
TFS	$g\,L^{-1}$	0.8	1.8	3.7
TVS	$g\,L^{-1}$	7.1	2.2	1.7
Ammonia	$mg\,L^{-1}$	30.4	10	1
Phosphorus	$mg\,L^{-1}$	1	10	29
Chloride	$mg\,L^{-1}$	293	–	–
Alkalinity	$mg\,L^{-1}$	282.8	819	–
pH	–	8.5	7.7	11

The bleach pulp mill effluent showed a high concentration of organic matter for COD and BOD. The COD values obtained are higher than the typical values found in literature ranging from 500 to 1,500 $mg\,L^{-1}$ of COD and 200 to 800 $mg\,L^{-1}$ of BOD [16,17]. Although the concentration of organic matter in the effluent depends on the type of bleaching and dosage of reagents used, the higher values obtained can be attributed to the partial closing of the water circuit within the bleaching process and/or to the low fiber recovery during the process. Low concentration of nutrients, ammonia, and phosphorus were also observed, indicating the possible need to adjust the dosage of nutrients in case of biological treatment.

Among the three effluents evaluated in this study, the one coming from the dairy industry presents the highest concentration of organic matter. Most of this organic matter is due to the presence of suspended solids, which can be observed by looking at the considerable difference between the average values of total COD (3,963 $mg\,L^{-1}$) and soluble COD (2,590 $mg\,L^{-1}$). The average BOD/COD relation of 0.5, besides being somewhat lower than the values mentioned in literature [18,19], is still high and indicates an elevated biodegradability of the effluent. The relation between organic matter and nutrients expressed in terms of BOD/Nitrogen/Phosphorus of 100:7:1 also appears to be adequate for biological treatment. According to Jordão and Pessoa [19], the optimal relation between nutrients for an aerobic biological system is 100:5:1.

Table 4 presents the results of the removal efficiency of pollutants from the MBRs in bench and pilot scale. It can be observed that the application of the MBR technology is effective for the removal of organic matter in terms of COD as well as BOD. The larger removal of organic matter from the MBR treating dairy effluents can be justified by the effluent's elevated biodegradability and by the elevated concentration of biomass in the reactor (Table 1). A high removal of nutrients can also be observed for dairy effluents. Higher ages of the sludge usually used in MBRs contribute to the occurrence of nitrification in these systems, because nitrifying bacteria, responsible for the conversion of ammonia into nitrate, are notoriously slow-growing micro-organisms [20]. This explains the significant removal of ammonia by the system. On the other hand, the high removal of phosphorus can be related to the precipitation of phosphates with calcium and sodium cations to be found in high concentration in the effluent in question, and to the assimilation of the micro-organisms [21]. The low color removal during the biological treatment of bleaching pulp mill effluent in the MBR is in agreement with the literature, which suggests

Table 4
Quality of the permeate and efficiency of the removal of the main physical-chemical parameters from the MBRs used for treatment of effluents from petroleum refinery, cellulose industry, and dairy industry

	Refinery		Dairy		Pulp	
	Permeate	% Removal	Permeate	% Removal	Permeate	% Removal
COD (mg L^{-1})	88	86	58	98	80	95
BOD (mg L^{-1})	4.5	98	6	99	–	–
Apparent color (uC)	60	8	27	99	63	36
Ammonia (mg L^{-1})	1.4	95	2.0	96	C*	–
Phosphorous (mg L^{-1})	C*	–	2.0	89	C*	–
Total solids (mg L^{-1})	–	–	1,647	46	2,220	80

Note: C* correction of nutrients for biological process.

that the coloration of bleaching effluents normally remains unaltered or could even increase its concentration in the treatment due to the formation of new chromophores resulting from the partial oxidation of organic matter [22].

3.2. *Critical flux in MBR treating petroleum refinery effluent: an example in pilot scale*

In accordance with what has already been presented, the treatment of the petroleum refinery effluent was relieved by two MBR configurations, given that, for each one of them, the usage of the critical flux was studied as a tool to monitor and control fouling. In Fig. 3, the performance of the (a) MBR 1 and (b) MBR 2 in terms of monitoring the operating pressure, the permeation flux (which was kept approximately constant the whole time aiming at preserving the hydraulic retention time, HRT), and the critical flux is presented.

The critical flux tests were carried out with a frequency of approximately 7 d. In the Fig. 3, the value obtained on a specific day has been extended until the performance of the next test in order to make the visualization of the data and the interpretation of the results easier. It is nevertheless known that the variation of the critical flux can be instantaneous and that this representation does not correspond exactly to reality, being a purely illustrative nature.

A variation in critical flux is observed during the time of observation, which may be associated with the variation of feed characteristics, and thus of the quality of the biological sludge, as discussed earlier, which is confirmed by studies described in literature [4]. Operation of the unit at an operational flux higher than the critical flux results in a high fouling rate, as expected. However, it is observed that, even operating the MBR at an operational flux lower than the critical

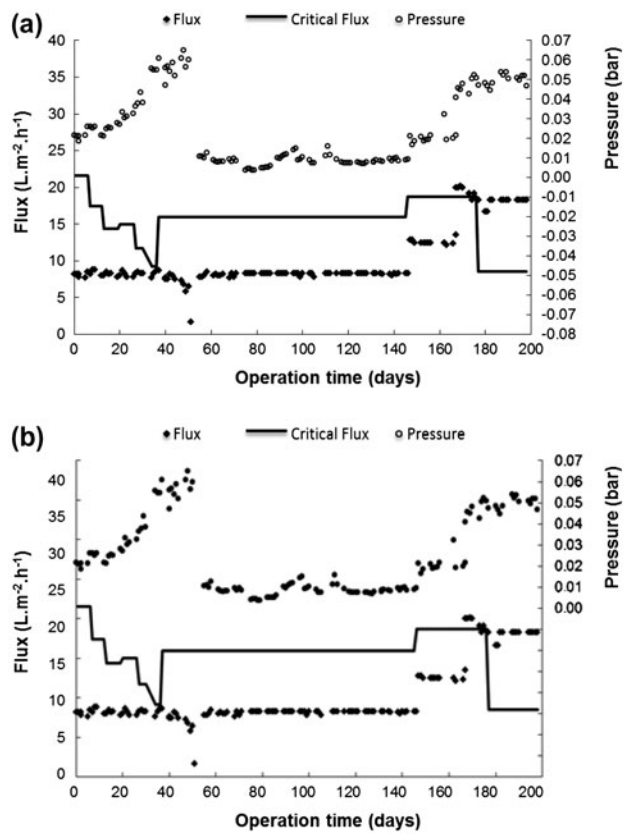

Fig. 3. Operational pressure and flux and critical flux throughout the (a) MBR1 and (b) MBR2 operation.

flux, there is still membrane fouling, which is consistent with the results obtained by Pollice et al. [6] and Le Clech et al. [4], who observed the occurrence of subcritical fouling in complex systems such as MBRs. Although one may note that the larger the difference between critical flux and permeate flux, the lower the fouling rates, it is important to note that the critical flux measurements were performed punctually once a week and after cleaning the membrane. Since the critical flux depends on sludge characteristics, which change constantly, and membrane characteristics, operation under the critical flux level cannot be ensured during the entire time of evaluation.

3.3. Critical flux in MBR treating bleach pulp mill effluent: an example in batch scale

Fig. 4 shows the performance of the MBR treating bleach pulp mill effluent in terms of monitoring the operating pressure, of the permeation flux (which was kept approximately constant the whole time aiming at preserving the HRT), and of the critical flux.

When the employed operational flux was greater than the critical flux, the applied pressure increased with the time of operation and returned to the initial value only after chemical cleaning. Conversely, when the operational flux employed was below the critical flux, an operation with stationary applied pressure was observed. The permeate flux was kept constant during operating time and the increase between days 33 and 37 was intentionally applied to confirm the earlier observation.

3.4. Critical flux in MBR dairy effluent: an example in batch scale

In Fig. 5, the performance of the MBR treating dairy effluent in terms of monitoring the operating pressure, the permeation flux (which was kept approximately constant the whole time in order to preserve the HRT), and the critical flux is shown.

The MBR used for treating dairy effluent was operating in two different stages: during the first stage (days 1–77), no aeration between the fibers of the membrane was applied. In this case, the only inlet of

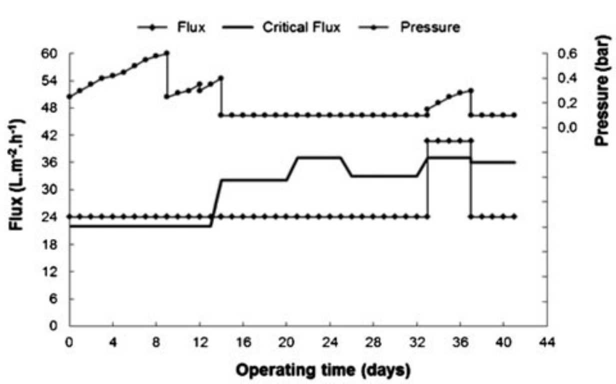

Fig. 4. Operational pressure and flux and critical flux throughout MBR treating bleach pulp mill effluent operation.

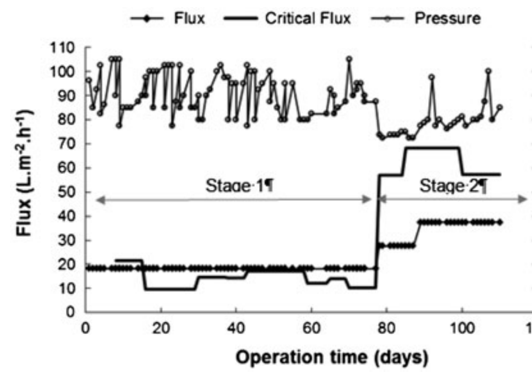

Fig. 5. Operational pressure and flux and critical flux throughout MBR treating dairy effluent operation.

air into the system was directed at the diffusers located at the bottom of the aerobic biological tank. During the second stage (days 78–110), an aeration system was placed at the base of the MF module as well, aiming at the promotion of aeration and shearing between the fibers.

High values of critical flux can be observed during the second operational stage, which justifies the stability observed during this stage, proven by the low increase in pressure, even though the MBR was operated with fluxes as high as 27.5 and 37.5 $L\,h^{-1}\,m^{-2}$. It was, however, observed that the results of the critical flux during the earlier stage were by far inferior. Results oscillated between 21.5 and 9.6 $L\,h^{-1}\,m^{-2}$. Therefore, the intense fouling observed through the rapid increase in pressure in stage 1 can be justified by the fact that the operational flux established, of 18 $L\,h^{-1}\,m^{-2}$, was higher than almost all critical flux measured.

The relation between critical flux, operational flux, and fouling can clearly be seen. When the operational flux is lower than the critical flux, the system is able to operate in a stable manner without the need of a constant increase in operating pressure. When, however, an operational flux superior to the critical flux is established, the operation becomes almost unfeasible, with rapid and more intense fouling. It can be noted that the period of lower fouling during the first stage, observed between days 9 and 16, corresponds to the only moment when the operational flux was found to be below the critical flux.

3.5. Implications for practice

It is believed that the critical flux test has the importance of being indicative of the flux above which fouling becomes truly severe and of serving as a tool to compare propensity to fouling in several systems, independently of the characteristics of the feed or the operation conditions. It, therefore, helps in the selection of the MBR's operational flux, considering that an operation of the MBRs at a flux lower than the critical flux can lead to little or no fouling [2,4].

Fig. 6(a) shows the relation between the fouling rate measured during the operation of the MBR (relation between the difference in pressure after the operation (P_2), fouled membranes, and after chemical cleaning (P_1), clean membrane, per time interval $\Delta t = t_2 - t_1$) and the difference between the critical flux (J_c) and operational permeation flux (J_p) for different effluents when operated under subcritical conditions. Fig. 6(b) presents a chart illustrating the calculation method for fouling rates.

The results showed for all evaluated effluents that the larger the difference between J_c and J_p, the smaller the observed fouling rate; this was already expected. The manner in which these results were evaluated allowed for a balancing of this tendency with an acceptable adjustment. The main importance of this quantification is to subsidize the establishment of the MBR project flux, in real scale, aiming to improve the technical and economical viability, and also to serve

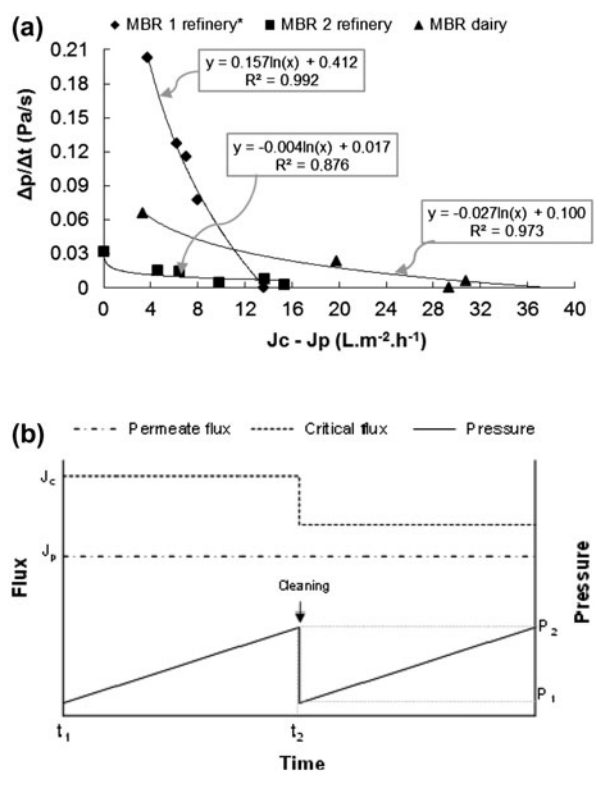

Fig. 6. Quantification of the relation between the fouling rate between chemical cleanings and the difference between critical flux and operational flux for different effluents. *Unit of pressure values for MBR1 = 100 Pa.

as a diagnostic tool for the demands for fouling control measurements for systems that have already been implemented and are operating, which do not allow significant variations of the permeate flux. It should be mentioned that the evaluation of this relation for the MBR treating the cellulose effluent was not possible, because no fouling was observed during the operation under subcritical conditions.

Therefore, the importance of controlling the operational flux in order to keep it lower than the critical flux becomes obvious. However, in most applications, the operational flux must be kept constant, because it is necessary to treat all generated effluents. Thus, after the implementation of the unit, the best way to keep the operational flux lower than the critical flux is to increase the critical flux, and not to reduce the operational flux. One way to increase the critical flux is to improve the quality of the sludge or to improve the hydrodynamic conditions of the process, which can be accomplished, for example, by dosing powder activated carbon or other specific membrane permeability improvers, and by elevating the aeration and shear rate.

The membrane permeability improver, a commercial modified cationic polymer, was used to increase the critical flux in the MBR treating refinery effluent. The mechanism of such products is based on coagulation/flocculation of the sludge, increasing the size of the flocs and retaining SMPs and EPSs in them, thus reducing their concentration in the medium and increasing membrane filterability. The use of permeability improvers was assessed by dosing such compounds in pilot-scale units. The biological tank received a certain dosage of a modified cationic polymer (commercial permeability improver) to recover sludge quality. The optimal dosage was determined by tests with variable concentrations of the commercial flux improver, in a methodology similar to that described by Koseoglu et al. [23]. The optimal polymer concentration was the one that resulted in better sludge filterability and higher SMP and EPS removal from the medium.

On the 12th day of operation (Fig. 3(b)) of the MBR2 for petroleum refinery effluent treatment, the critical flux decreased from 19 to 15 $L\,h^{-1}\,m^{-2}$, a value below the operational flux due to an alteration in the feed composition that stressed the biomass. On the 54th day of operation, a chemical cleaning was performed on the membrane with the purpose to recover permeability; however, no significant increase in permeability was observed. After the flux improver was added, on the 57th day of operation, an increase in critical flux was observed. The flux improver concentration used was 250 $mg\,L^{-1}$ based on results of an optimal dosage test. After the dosage, the membrane permeability remained stable, and no stressful situation affecting the biomass was detected. The system was monitored during a period of 30 d, due to the fact that part of the sludge was accidentally lost on the 87th day of operation and during this time (57–87th day of operation) the operational permeate flux was lower than the critical flux. During this period, the permeability improver proved to be an efficient way of controlling emergency situations of filterability loss caused by stress to the biomass.

In the MBR treating bleach pulp mill effluent, the PAC dosage was assessed to verify the efficiency in improving the critical flux. It is observed that the addition of PAC to the MBR on the 13th day of operation (Fig. 4) resulted in the increase in the critical flux value in a way that the operational flux was below the critical flux, resulting in an operation with stationary applied pressure.

To prove that the applied pressure in the stationary regime was the result of an operation with a flux lower than the critical flux, from the 32nd to 36th day of MBR operation, the operational flux was increased to be higher than the critical flux. This showed that, in this period, the applied pressure increased progressively to maintain a constant operational flux. After the cleaning, the applied pressure reduced to the initial value, indicating the non-occurrence of irreversible fouling. When the operational flux returns to a value lower than the critical flux, the applied pressure again operates in a stationary regime.

The increase in critical flux after the addition of PAC to the MBR could be attributed to physical and/or physical-chemical factors. The last ones are due to adsorption of SMP and EPS in PAC, thereby reducing their concentration in the medium and consequently, reducing membrane fouling. However, it can also be associated with the physical abrasive effect of carbon, which helps in the removal of the cake layer deposited on the membrane surface.

In the MBR treating dairy effluents, a high aeration rate was used to increase the critical flux. Aeration induces a cross-flow movement of water along the fibers, thereby increasing shear and particle back transport, and reducing the accumulation of foulants. Kim and DiGiano [24] observed a linear increase in the critical flux with the aeration rate, which suggests that the particle back-transport rate increases linearly with the aeration rate to offset the increase in the convective transport toward the membrane. The elevated values of critical flux during the second stage of operation shown in Fig. 5 are due to the application of aeration along the fibers, capable of controlling the material deposition rate on the membrane surface. The elevated critical flux justifies the stability observed during this stage, even when the MBR operates with fluxes as high as 27.5–37.5 $L\,h^{-1}\,m^{-2}$.

These results support the importance of controlling the operational flux to keep it lower than the critical flux. However, no single and precise agreed protocol exists for critical flux measurements, making a comparison of reported data difficult. Variables include step duration, step height, initial state of the membrane (new/backwashed/cleaned), feed characteristics, and system hydraulics.

It can thus be observed a necessity to standardize the methodology for critical flux and the necessity for the same to be precise and at the same time objective for a large-scale application. If the critical flux test was conducted in pilot scale, in order to foresee the critical flux in real scale, it would have to have a membrane module with a length closest possible to the real application and an equivalent aeration rate because these factors have a significant influence on the critical flux measurement [24]. Another important aspect is the frequency with which the critical flux is monitored. The critical flux is influenced by the characteristics of feed and sludge, and therefore fluctuates in time. In this case, the smaller the time interval between measurements, the better the inference from the data and consequently, the better the fouling control.

4. Conclusion

The results obtained during this study showed that the critical flux determination on batch- or pilot-scale units can be used as a parameter for monitoring the fouling in MBR units. It can, furthermore, be concluded that one way to increase the critical flux is to improve the quality of the sludge or to improve the hydrodynamic conditions process. In this study, powder activated carbon or a specific membrane permeability improver dose and a high aeration rate are presented as appropriate strategies to increase the critical flux.

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