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Model assessment of the prevailing fouling mechanisms in a submerged membrane anaerobic reactor treating low-strength wastewater

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ABSTRACT

Three models (blocking laws, combined and resistance-in-series) were applied to identify the prevailing fouling mechanisms in a submerged membrane in an up-flow anaerobic sludge blanket reactor treating municipal wastewater. Experimental runs were carried out at lab-scale with filtration periods of 4 and 10 min, followed by relaxation periods of one minute with and without nitrogen bubbling. In all conditions excepting one (IF4R), the blocking laws model showed a predominance of cake formation. With the combined model, cake formation coupled with intermediate, standard and complete fouling had the better fits in all conditions, excepting IF4 and IF4R. When sewage was fed, both models pointed at intermediate fouling in the absence of gas bubbling. The resistance-in-series model identified the positive effect of gas bubbling and a post-cake fouling behavior, not shown by the other two models. This modeling approach could be applied for achieving longer filtration runs in submerged UF membranes.

1. Introduction

In recent years, anaerobic membrane bioreactors (AnMBR) have been the focus of increased attention due to the improvements they bring to the well-known advantages of anaerobic processes: low energy requirement, biogas production and lower production of biomass. In this arrangement, membranes achieve total retention of biomass in the reactor, resulting in a filtered effluent, free of pathogenic microorganisms. However, their use is limited by membrane fouling, the same drawback of aerobic versions; on this regard, fouling appears to be more severe on anaerobic environments, due in part to the lower sludge filterability if compared to aerobic MBR (Spagni et al., 2010).

Membrane fouling is related to the formation of cake layer and pore blocking. In a new membrane, pore obstruction is dominant at the beginning of the filtration (standard blocking) due to the deposition of the soluble compounds, such as soluble microbial products, SMP. The internal clogging by soluble macromolecular species predominates at this early stage, being the main cause of irreversible fouling (the one that needs chemical cleaning to be removed) of UF membranes (Bae and Tak, 2005; Le-Clech et al., 2006). Then, cake layer formation occurs in the later stages of the filtration run, due to the deposition of sludge particles on the membrane surface. This situation can be described as a porous media with a complex system of interconnected inter-particle voids that acts as a barrier and prevents other species to infiltrate into the membrane pores (Meng et al., 2009).

Many studies have been carried out to understand the fouling mechanisms and identify control strategies, mostly on aerobic membrane bioreactors (MBR). The operational patterns have been based on constant TMP (Aslam et al., 2015; Drews et al., 2009; Hwang and Chen, 2007) and constant flux (Miller et al., 2014; Sioutopoulos and Karabelas, 2016; Suarez and Veza, 2000), the latter being more applied in submerged membrane bioreactors (Kovalsky et al., 2009; Le-Clech et al., 2006).

However, the application of models for the identification of fouling mechanisms in AnMBR for constant TMP operation is scarce (Charfi et al., 2012; Herrera-Robledo et al., 2011; Ho and Sung, 2009) and the available information for constant flux operation is also limited (Charfi et al., 2017; Ye et al., 2005).

In order to get a better understanding of the fouling phenomena and their predominant mechanisms, several models have been proposed.

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Table 1
Equations of the blocking laws model (BL) for two operating conditions.

Mechanism	Constant TMP[1]	Constant flux[2]	Fitted parameters
C	$K_bV = Q_o\,(1-e^{K_bt})$	$\frac{P}{P_o} = \frac{1}{1-K_bt}$	K_b (s^{-1})
S	$\frac{K_st}{2} = \frac{t}{V}-\frac{1}{Q_o}$	$\frac{P}{P_o} = \left(1-\frac{K_sJ_ot}{2}\right)^{-2}$	K_s (m^{-1})
CF	$K_cV = \frac{2t}{V}-\frac{2}{Q_o}$	$\frac{P}{P_o} = 1 + K_cJ_o^2t$	K_c (s/m^2),
I	$K_iV = \ln(1 + K_iQ_ot)$	$\frac{P}{P_o} = e^{K_iJ_ot}$	K_i (m^{-1})

[1] Hermia (1982).
[2] Hlavacek and Bouche (1993).

Blocking laws (BL) model, proposed by Hermans et al. (1936) and subsequently unified by Hermia (1982) is the most popular model used in MBR. This model describes four simple mechanisms of membrane fouling caused by particles: a) complete blocking (C), when each particle blocks a membrane pore without overlapping on other particles; b) standard blocking (S), when particles enter the membrane pores and are retained inside the filtering channels, resulting in a narrower water passage; c) cake formation (CF), when the particles accumulate on the membrane surface as a permeable layer of variable thickness that increases flow resistance; d) intermediate blocking (I), a combination of the former mechanisms. This model may be applied for evaluating the fouling mechanism of MBR operated at constant TMP. The model equations relate the filtered volume (V), the filtration time (t), and the initial flow rate (Q_o) with the corresponding constant (K_b for complete blocking, K_s for standard blocking, K_c for cake formation and K_i for intermediate blocking). Later, Hlavacek and Bouche (1993) extended the model to MBR operated with constant flux. In this case, the equations relate the operating (transmembrane) pressure (P), the filtration time (t) and the initial flux (J_o) with the corresponding constant (K_b, K_s, K_c and K_i). Table 1 presents the equations used in the BL model for both operating conditions.

The BL model cannot assess the relative importance of each mechanism at a particular time of a filtration run; furthermore, it is assumed that fouling is due only to one mechanism and that there is no detachment of particles from the membrane surface induced by air sparging, which is a common practice for submerged membranes. As a result, a single fouling model does not provide a good description of the filtration process in submerged MBR with air scouring. In fact, studies have shown a transition in fouling mechanism in such cases. Charfi et al. (2012) used the BL model for assessing the fouling mechanisms commonly found in AnMBR with ultrafiltration (UF) and microfiltration (MF) membranes. They used data from literature, gathering 32 short and long-term runs at constant TMP. In cases where the optimized flux expression was unable to describe the experimental data, they split the data in two sets corresponding to distinct phases. In the short-term experiments, fouling occurred during two separate phases, following either one or two fouling mechanisms. In the long-term runs, which included cleaning cycles, cake layer formation was identified as the predominant fouling mechanism in AnMBR.

Other models have been developed to solve some of the limitations of the classical BL model. Some consider the combination of fouling mechanisms in parallel (Bolton et al., 2006; Ho and Zydney, 2000) and others consider that the mechanisms ocurr in sequence (Hwang and Chen, 2007; Ye et al., 2005). Bolton et al. (2006) proposed five fouling behaviors combining the four fouling mechanisms of the BL model, testing their applicability for the sterile filtration of immunoglobulin (IgG) and the viral filtration of bovine serum albumin (BSA). Equations relating TMP pressure, filtration time and initial flux at constant flux operation were obtained with two fitted parameter depending on the combined mechanisms involved (K_c, K_s, K_b and K_i). The combined mechanisms (CM) model considers two fitted parameters and reduces to the single BL model equations in the absence of a second fouling mechanism. Table 2 summarizes the five combined fouling mechanisms at constant flux with the corresponding equations and characteristic parameters.

Table 2
Equations of the combined fouling model (CM) for constant flux operation.

Model	Equation	Fitted parameters
Cake-complete (CF-C)	$\frac{P}{P_o} = \frac{1}{(1-K_bt)}\left(1-\frac{K_cJ_o^2}{K_b}\ln(1-K_bt)\right)$	K_c ($s{\cdot}m^{-2}$) K_b (s^{-1})
Cake-intermediate (CF-I)	$\frac{P}{P_o} = \exp(K_iJ_ot)\left(1 + \frac{K_cJ_o}{K_i}(\exp(K_iJ_ot)-1)\right)$	K_c ($s{\cdot}m^{-2}$) K_i (m^{-1})
Complete-standard (C-S)	$\frac{P}{P_o} = \frac{1}{(1-K_bt)\left(1+\frac{K_cJ_o}{2K_b}\ln(1-K_bt)\right)^2}$	K_b (s^{-1}) K_s (m^{-1})
Intermediate-standard (I-S)	$\frac{P}{P_o} = \frac{\exp(K_iJ_ot)}{\left(1-\frac{K_s}{2K_i}(\exp(K_iJ_ot)-1)\right)^2}$	K_i (m^{-1}) K_s (m^{-1})
Cake-standard (CF-S)	$\frac{P}{P_o} = \left(\left(1-\frac{K_sJ_ot}{2}\right)^{-2} + K_cJ_o^2t\right)$	K_c ($s{\cdot}m^{-2}$) K_s (m^{-1})

Several authors have applied the CM model proposed by Bolton et al. (2006). Liu et al. (2008) used the model on a microfiltration membrane operated at constant flux for the treatment of synthetic surface water. The combined cake-complete and the cake-intermediate models demonstrated relative high consistency with experimental TMP data. The CM model provided the best fit to the experimental results, but not enough to predict the behavior of the membrane fouling. Wei et al. (2016) applied the CM model to fit their experimental data to understand the fouling mitigation mechanism of the soluble extracellular organic matter (EOM)-related membrane fouling by pre-ozonation. The combined cake-standard model had the best model fit at different ozone dosages. Based on the fitted parameters, the standard pore blocking played an important role in the EOM-related membrane fouling as well as the membrane fouling control. However, others authors have found that fitting their experimental data with the CM model did not provide better results than those obtained with the BL model (Bérubé et al., 2008).

As mentioned, although fouling in MBR has been studied extensively, there are limited information about the identification of AnMBR fouling mechanisms for constant flux operation. The aim of this work was to compare the fitness to experimental data of three models (BL, CM, and a resistance-in-series model) and by this means, to identify the prevailing fouling mechanisms in a UF membrane placed in the upper part of a UASB reactor treating municipal wastewater under different hydrodynamic conditions.

2. Material and methods

2.1. Experimental set-up

The experiments were performed using a lab-scale AnMBR under a constant flux mode. An UASB reactor (7.5 cm internal diameter and 0.95 m liquid depth, for a useful volume of 4.3 L) was operated at a hydraulic retention time (HRT) of 8 h with a PVDF submerged ultrafiltration tubular membrane with 100 kDa molecular weight cut-off (MWCO) (model VFU 100 manufactured by Memos, Germany). The mono tubular membrane (0.9 cm diameter, 30 cm length for a filtration surface of 0.0085 m^2) was placed in the third upper zone of the reactor (zone with the lowest suspended solids concentration). The reactor was inoculated with 1.1 L of granular sludge coming from a full-scale UASB reactor treating brewery wastewater (total suspended solids: 60 g L^{-1}; volatile solids – total solids ratio: 74%; specific methanogenic activity: 0.80 $gCOD_{CH4}{\cdot}gVS^{-1}\,d^{-1}$).

Two different sets of experiments of intermittent filtration (IF) were performed, using synthetic wastewater and real municipal wastewater at ambient temperature (18–24 °C). With synthetic municipal-like