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Dynamic characterization of external and internal mass transport in heterotrophic biofilms from microsensors measurements

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ABSTRACT

Knowledge of mass transport mechanisms in biofilm-based technologies such as biofilters is essential to improve bioreactors performance by preventing mass transport limitation. External and internal mass transport in biofilms was characterized in heterotrophic biofilms grown on a flat plate bioreactor. Mass transport resistance through the liquid-biofilm interphase and diffusion within biofilms were quantified by in situ measurements using microsensors with a high spatial resolution (<50 μm). Experimental conditions were selected using a mathematical procedure based on the Fisher Information Matrix to increase the reliability of experimental data and minimize confidence intervals of estimated mass transport coefficients. The sensitivity of external and internal mass transport resistances to flow conditions within the range of typical fluid velocities over biofilms (Reynolds numbers between 0.5 and 7) was assessed. Estimated external mass transfer coefficients at different liquid phase flow velocities showed discrepancies with studies considering laminar conditions in the diffusive boundary layer near the liquid-biofilm interphase. The correlation of effective diffusivity with flow velocities showed that the heterogeneous structure of biofilms defines the transport mechanisms inside biofilms. Internal mass transport was driven by diffusion through cell clusters and aggregates at Re below 2.8. Conversely, mass transport was driven by advection within pores, voids and water channels at Re above 5.6. Between both flow velocities, mass transport occurred by a combination of advection and diffusion. Effective diffusivities estimated at different biofilm densities showed a linear increase of mass transport resistance due to a porosity decrease up to biofilm densities of 50 g VSS$\cdot$L^{-1}. Mass transport was strongly limited at higher biofilm densities. Internal mass transport results were used to propose an empirical correlation to assess the effective diffusivity within biofilms considering the influence of hydrodynamics and biofilm density.

1. Introduction

Biofilters and biotrickling filters are the most important biofilm-based reactors for air pollution control. However, the limited knowledge about biofilms performance due to large technical limitations in biofilms monitoring, mainly due to their reduced size (ranging from few microns to few millimeters), has led to often assume large model simplifications in biofiltration modeling (Beyenal and Lewandowski, 2005). Characterization of mass transport and biological reactions within biofilms is essential to improve biofilms models and therefore, our ability to design and operate biofilm reactors.

Two main phenomena are considered when modeling biofilms performance (Bishop et al., 1995; Picioreanu et al., 2000a): (1) pollutants and substrates transport at the liquid-biofilm interphase (external mass transport) and within biofilms (internal mass

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Nomenclature

a	specific mass transfer area (m^2 m^{-3})
$A_{transfer}$	interfacial area of the monitored system (m^2)
$C_{DO,b}$	DO concentration inside the biofilm (mg L^{-1})
$C_{DO,B}$	DO concentration on biofilm surface (mg L^{-1})
$C_{DO,B\ (t\ =\ 0)}$	initial DO concentration on biofilm surface (mg L^{-1})
$C_{DO,L}$	DO concentration inside the liquid boundary layer (mg L^{-1})
$C_{DO,L\ (i)}$	DO concentration in the liquid phase at the inlet of a FPB section (mg L^{-1})
$C_{DO,L\ (inlet)}$	DO concentration in the liquid phase at the inlet of the FPB (mg L^{-1})
$C_{DO,L\ (o)}$	DO concentration in the liquid phase at the outlet of a FPB section (mg L^{-1})
$C_{DO,L\ (t\ =\ 0)}$	initial DO concentration in the liquid boundary layer (mg L^{-1})
D_b	effective diffusivity within biofilms (m^2 s^{-1})
D_i	the effective diffusivity within the boundary layer (m^2 s^{-1})
D_r	dimensionless effective diffusivity (−)
F	objective function in optimization procedure for parameters estimation
k_L	external mass transfer coefficient (m s^{-1})
L_c	boundary layer thickness (m)
n	number of experimental measurements
Q_L	liquid flow rate (m^3 s^{-1})
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number
t	time (s)
t_{exp}	monitoring time (s)
V_L	liquid phase volume (m)
$\nu_{sampling}$	sampling frequency (s^{-1})
X_b	biofilm density (g VSS·L^{-1})
$y_{\theta,i}$	simulated oxygen concentration in optimization procedure (mg L^{-1})
$y_{exp,I}$	experimentally measured oxygen concentration in optimization procedure (mg L^{-1})
z	biofilm depth (m)
$z_{profile}$	depth of DO profile (μm)
$\Delta z_{profile}$	distance between monitoring points (μm)

transport) and (2) the biodegradation of pollutants in the biofilm. Literature is abundant in assessing biokinetic parameters, which are usually adapted from suspended cultures studies for further biofilm modeling (Bonilla-Blancas et al., 2015; Hille et al., 2009; Mannucci et al., 2012) despite this approach is somehow questionable due to physiological differences between attached and suspended growth systems (Yurt et al., 2003). Oppositely, literature is scarce in the characterization of external and internal mass transport in biofilm modeling. External mass transport resistance is often modeled through mass transfer coefficients (Horn and Hempel, 1997) usually calculated as a function of hydrodynamic conditions in the reactor (Picioreanu et al., 2000b). Different works have studied external mass transport resistance in the liquid boundary layer from experimental measurements with microsensors (Horn and Hempel, 1995; Wäsche et al., 2002; Zhang and Bishop, 1995). However, these measurements only provided limited information of external mass transport resistance since did not considered diffusive and convective phenomena within the boundary layer as suggested by different authors (Beyenal and Lewandowski, 2000; Wäsche et al., 2002). On the other hand, internal mass transport in biofilm systems is commonly addressed by diffusive mechanisms following Fick's law (Stewart, 2003).

Effective diffusivities within biofilms are highly influenced by biofilms structure and heterogeneity. Some works have considered the effect of biofilms heterogeneity in effective diffusivity correlations through the use of different structural macroscopic parameters such as biofilm density, porosity and tortuosity. Although the use of these correlations for effective diffusivity is sometimes used in biofiltration models, its reliability is under suspect. Most of these researches were based on theoretical models (Zhang and Bishop, 1994) or important theoretical assumptions (Hinson and Kocher, 1996). Oppositely, several works have shown the high performance of microsensors measurements for the quantification of effective diffusivity within biofilms (Beuling et al., 1999; Fu et al., 1994; Hille et al., 2009; Ning et al., 2012). However, the reliability of these studies is compromised since empirical correlations for effective diffusivity estimation within biofilms were developed combining experimental estimates inside biofilms showing dry mass densities between 10 and 65 g L^{-1}, and sludge granules, with dry mass densities above 100 g L^{-1}.

The confidence intervals of mass transport parameters are not commonly determined. However, the assessment of confidence intervals is as important as the estimation of the parameter. A mathematical method based on the Fisher Information Matrix (FIM) is a proven procedure that accurately provides confidence intervals of estimated parameters. FIM method is based on the calculation of the covariance inverse matrix and is directly associated to the uncertainty of estimated parameters and the quality and quantity of experimental data. In addition, the FIM method can be adapted for optimal experiment design (Dochain and Vanrolleghem, 2001), thus the reliability of mass transport parameters can be enhanced due to the selection of optimal experimental conditions during the design of the experiments.

The goal of this work was to take advantage of the high resolution of microsensors measurements (10 μm) to experimentally quantify external and internal mass transport resistance. Oxygenation profiles were recorded using a dissolved oxygen (DO) microsensor specially designed for biofilms profiling (Moya et al., 2014). Oxygenation data were used to develop methodologies for mass transport parameters estimation to further investigate the influence of hydrodynamic conditions and biofilm density on external and internal mass transport. Reliability of estimated parameters was increased by selecting the optimal experimental conditions, evaluating the parameter sensitivities and the quality of estimated parameters (confidence intervals).

2. Materials and methods

2.1. Development of a heterotrophic biofilm

Mass transport phenomena were studied in a heterotrophic biofilm grown in a flat plate bioreactor (FPB), shown in Fig. 1. The reactor consisted of an open channel, manufactured in methacrylate (PMMA), 20 cm in length, 3.5 cm in width, and 1.3 cm in depth. The wall effect in the open channel was assessed in modeling studies (Prades et al., 2015) concluding that this effect could be neglected in laminar flow conditions. The reactor setup included two tanks of 40 mL (depicted in Fig. 1), at the inlet and the outlet of