



Application of a novel respirometric methodology to characterize mass transfer and activity of H₂S-oxidizing biofilms in biotrickling filter beds



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ABSTRACT

The elimination capacity of gaseous H₂S biofiltration can be limited either by mass transfer or bioreaction in the biofilm. Assessment of the biological activity of immobilized cells (biofilm) usually implies morphological and physiological changes during the adaptation of cells to respirometric devices operated as suspended cultures. In this study, respirometry of heterogeneous media is advised as a valuable technique for characterizing mass transport and biological activity of H₂S-oxidizing biofilms attached on two packing materials from operative biotrickling filters. Controlled flows of liquid and H₂S-containing air were recirculated through a closed heterogeneous respirometer allowing a more realistic estimation of the biofilm activity by the experimental evaluation of the oxygen uptake rate (OUR). Specific maximum OUR of 23.0 and 38.5 mmol O₂ (g biomass min)⁻¹ were obtained for Pall rings and polyurethane foam, respectively. A mathematical model for the determination of kinetic-related parameters such as the maximum H₂S elimination capacity and morphological properties of biofilm (i.e., thickness and fraction of wetted area of packing bed) was developed and calibrated. With the set of parameters obtained, the external oxygen mass transport to the wetted biofilm was found to limit the global H₂S biofiltration capacity, whereas the non-wetted biofilm was the dominant route for the gaseous O₂ and H₂S mass transfer to the biofilm. Oxygen diffusion rate was the limiting step in the case of very active biofilms.

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1. Introduction

Hydrogen sulfide (H₂S) is a volatile inorganic compound commonly found in waste gas streams (e.g., biogas from landfills and wastewater treatment plants), with a typical composition ranging from 0.0002% to 2.0% [1,2]. Biofilters (BF) and biotrickling filters (BTF) have been widely studied and applied by several research groups and companies to desulfurize polluted, odorous air or energetic gases such as biogas [3,4]. Therefore, the application of these technologies avoids the emission of harmful gases and odors, which

cause human hazard risks and also corrosion damages on cogeneration engines in case of recovering energy from H₂S containing waste gases.

Several parameters can be monitored and controlled during waste gas biofiltration, such as inlet and outlet gaseous pollutant concentrations or flow rates, which allow calculating the overall removal efficiency. However, biodegradation kinetics are usually difficult to determine [5]. Respirometry consists on the measurement and interpretation of the biological oxygen consumption rate under well-defined experimental conditions and is a typical tool to assess the degradation activity of microbial cultures [6,7]. The performance of this assay has been traditionally used with suspended cells [8,9], namely suspended culture respirometry (SCR), which implies biofilm destruction when it is applied to monitor biological activity of immobilized biomass. In SCR, the original physiology of cells, as well as the mass transport phenomena occurring in the

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Nomenclature

List of symbols

a	Packing specific surface area, $\text{m}^2 \text{m}^{-3}$
a_{g-b}	Gas–biofilm specific surface area, $\text{m}^2 \text{m}^{-3}$
a_{g-l}	Gas–liquid specific surface area, $\text{m}^2 \text{m}^{-3}$
a_{l-b}	Liquid–biofilm specific surface area, $\text{m}^2 \text{m}^{-3}$
$C_{g,i}^{\text{Bed}}$	Concentration of component i in the gas phase in the bed, g m^{-3}
$C_{g,i}^{\text{Free}}$	Concentration of component i in the gas phase in the free section, g m^{-3}
$C_{l,i}^{\text{Bed}}$	Concentrations of component i in the liquid phase in the bed, g m^{-3}
$C_{l,i}^{\text{Res}}$	Concentrations of component i in the liquid phase in the reservoir section, g m^{-3}
$D_{\text{eff},i}$	Diffusion coefficient of component i in the biofilm, $\text{m}^2 \text{h}^{-1}$
EC	Elimination capacity, $\text{g m}^{-3} \text{h}^{-1}$
He_i	Gas/liquid partition coefficient of component i in a air/aqueous system
K_B	The external mass transfer coefficient from external bulk phase to biofilm, m h^{-1}
$K_{L,a_{g-l}}$	Global mass transfer coefficient, h^{-1}
k_i	Saturation constant for component i , g m^{-3}
$K_{s,i}$	Half saturation constant for component i , g m^{-3}
N	Total number of layers that thickness biofilm was divided for the numerical resolution of the mathematical model
OUR	Oxygen uptake rate, $\text{g O}_2 \text{m}^{-3} \text{h}^{-1}$
OUR_{max}	Maximum oxygen uptake rate, $\text{g O}_2 \text{m}^{-3} \text{h}^{-1}$
OUR_{end}	Endogenous oxygen uptake rate, $\text{g O}_2 \text{m}^{-3} \text{h}^{-1}$
Q_g	Gaseous volumetric flow rate, $\text{m}^3 \text{h}^{-1}$
Q_l	Liquid volumetric flow rate, $\text{m}^3 \text{h}^{-1}$
$r_{b,i}$	Consumption rates of component i in the wetted biofilm, $\text{g m}^{-3} \text{h}^{-1}$
$r_{b-NW,i}$	Consumption rates of component i in the non-wetted biofilm, $\text{g m}^{-3} \text{h}^{-1}$
t	Time, h
t_{end}	Final time of respirometry assay, h
t_{max}	Time when maximum elimination capacity occurred, h
V_{bed}	Packed bed volume, m^3
V_{bio}	Biomass volume, m^3
V_g	Gaseous volume, m^3
V_l	Liquid volume, m^3
x	Thickness position in the biofilm, m
$Y_{\text{O}_2/\text{H}_2\text{S}}$	Yield coefficient, $\text{mol O}_2 \text{mol}^{-1} \text{H}_2\text{S}$

Subscripts

b	Section of biofilm wetted
$b-NW$	Section of biofilm non-wetted
i	Component i
max	Maximum

Superscripts

Bed	Packed bed of HR
Free	Gas free volume
Res	Reservoir liquid volume

Greek letters

α	Surface fraction of packing material wetted
β	Surface fraction of the packing material covered by biofilm

δ	Biofilm thickness, m
ϵ_l^{Bed}	Volume fraction occupied by the liquid in the packed bed, $\text{m}^3 \text{m}^{-3}$
ϵ_g^{Bed}	Volume fraction occupied by the gas in the packed bed, $\text{m}^3 \text{m}^{-3}$
ϵ_b^{Bed}	Volume fraction occupied by the biofilm in the packed bed, $\text{m}^3 \text{m}^{-3}$

biofilm, are not considered which drives to an overestimated biological activity [10]. A realistic assessment of the biodegradation activity measured from a sample of colonized packed bed would allow improving the strategies to adequately operate and control biofilters.

Some adapted respirometric methodologies to study biodegradation kinetics of immobilized biomass have been already proposed. In this sense, preliminary studies have been performed toward the application of heterogeneous respirometry (HR) to characterize H_2S -oxidizing biofilms [5,11]. However, in these methodologies the liquid and/or gas phases remained static [12,13], which do not simulate properly the dynamic nature of the flowing phases of a BF or BTF and, in consequence, altered the real biofilm conditions during tests. The effect of external mass transfer resistance on the H_2S elimination seems to be significant for the performance of BFs and BTFs, and especially in aerobic systems where the mass transport of gaseous oxygen to the biofilm could limit the global process [14]. Instead, the HR is a novel methodology based on the measurement of the biological activity of immobilized biomass with a minimum handling and damaging of the biofilm associated. HR also reproduces the dynamic conditions of the flowing phases. In addition, in the above mentioned studies only the pollutant concentration in the gas-phase has been monitored [15], while the oxygen concentration in the gas phase was not analyzed. The latter is a critical variable that defines the H_2S biological oxidation. Overall, the HR technique has not been extensively applied yet and requires further experimental and modeling research in order to be improved.

Thus, the aim of this work was to apply, assess and improve the HR methodology to characterize H_2S -oxidizing activity and mass transport phenomena of specialized biofilms grown on packed beds of desulfurizing BTFs. Complementary, a mathematical model is developed and calibrated to describe the process and the intrinsic oxygen uptake rates (OUR) induced by the oxidation of H_2S in the biofilm. The mathematical model considers both wetted and non-wetted biofilm surfaces of the packing material. Experimental data of oxygen profiles in the gas and the liquid phase together with the application of the mathematical model allowed estimating the maximum H_2S elimination capacity of the packing materials. Although no experimental data was available from inside the biofilm, the model was used to theoretically identify and assess the potential limiting steps in H_2S bio-oxidation.

2. Materials and methods

2.1. Heterogeneous respirometer setup

The experimental system consisted of a transparent PVC cylindrical BTF, with an internal diameter of 0.06 m and a height of 0.50 m. The packed bed height filled with random packing was 0.26 m (a working volume of 0.73 L). In Fig. 1, a schematic of the HR is presented. During respirometric assays the liquid phase was continuously recirculated at a flow rate of $2.25 \times 10^{-2} \text{m}^3 \text{h}^{-1}$ with a peristaltic pump (77200-12, Cole Parmer, USA) while the gas phase

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