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## Optimization of oxygen transfer through membrane diffusers for the biological sweetening of biogas

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### Abstract

Biological techniques for the removal of gaseous pollutants such as hydrogen sulfide have proved to be effective, environmentally friendly and economically viable. However, when high concentrations of hydrogen sulfide are treated the process is severely restricted by the oxygen deficit in the liquid phase. Thus, when the oxygen concentration is below the stoichiometric requirement, elemental sulfur is formed and accumulates in the packing material, increasing operating costs and, in extreme cases, requiring the bioreactor to be shut down. In this study the oxygen transfer efficiency provided by a membrane diffuser is evaluated under typical pressure and salinity conditions used for the biological treatment of hydrogen sulfide in biogas. The aim was to determine the optimal operating parameters for enhanced oxygen transfer. The addition of pure oxygen with a membrane diffuser to increase the oxygen transfer rate and the use of a non-aqueous phase to improve oxygen transfer in a bioscrubber system are also evaluated.

**Key-words:** oxygen transfer, biofiltration, membrane diffuser, non-aqueous phase, pressure

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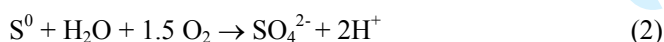
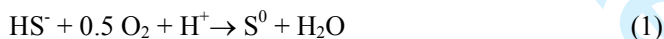
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## 1. Introduction

The recovery of energy-rich gases has grown in importance in recent years. European directive 1999/31/CE on landfill of waste stipulates that all landfill sites storing biodegradable waste must make provision for collecting and treating the biogas emitted for subsequent reuse as an energy source. Harnessing the biogas generated during anaerobic digestion processes at municipal landfills and wastewater treatment plants has therefore become a promising means of obtaining energy and reducing the emission of pollutant gases into the atmosphere. However, biogas contains hydrogen sulfide (H<sub>2</sub>S) at concentrations of 1000–4000 ppm<sub>v</sub> [1], which is sufficient to corrode processing equipment and, when released into the atmosphere, is an acid rain precursor. Cogeneration motors, used to generate electric and thermal energy, require H<sub>2</sub>S concentrations of less than 500 ppm<sub>v</sub>. Biological techniques have proved to be an effective and environmentally friendly solution for removing large quantities of hydrogen sulfide and are less costly than physicochemical techniques [2].

The most common systems for biological removal of hydrogen sulfide are **biotrickling filters and bioscrubbers** [3]. A bioscrubber consists of two operational components: a gas-liquid column (where contaminant transfer occurs) and an aeration tank. Instead, in a biotrickling filter, the pollutant is absorbed and removed in a packed column, being the liquid phase continuously recirculated from the bottom of the reactor [4].

**The biological oxidation of H<sub>2</sub>S occurs in two stages according to reactions 1 and 2. In the first step the hydrogen sulfide is oxidized into elemental sulfur, in the second stage the elemental sulfur is oxidized into sulfate. [5, 6]:**



When high concentrations of dissolved oxygen (DO) are present, hydrogen sulfide is oxidized to sulfate, which can be removed by purging the liquid phase. However, when DO levels are insufficient, microorganisms can only oxidize the H<sub>2</sub>S to elemental sulfur an insoluble solid that builds up in the packing material of the biofilter, increasing pressure drop, raising pumping costs and potentially requiring equipment to be shut down so that the packing material can be cleaned or replaced [7]. In standard processes, the contribution of oxygen to the biological process is achieved by injecting air together with the biogas. In this type of aeration system, oxygen transfer occurs throughout the biofilter. Oxygen has a low solubility in water of 8.37 mg l<sup>-1</sup> (air at 25 °C and 1 atm) [8], meaning that oxygen limitation is a common problem when treating large amounts of hydrogen sulfide. As such, one possible strategy for obtaining the biological oxygen demand required to oxidize hydrogen sulfide to sulfate is to optimize the air-to-water oxygen transfer.

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3 Despite its importance in the biological removal of H<sub>2</sub>S, optimization of oxygen transfer has  
4 received little attention in the literature. Despite its importance in the biological removal of H<sub>2</sub>S,  
5 optimization of oxygen transfer has received little attention in the literature. Alcantara *et al.*  
6 (2004) studied the relevance of dissolved oxygen in the liquid phase for a bioscrubber to remove  
7 hydrogen sulfide at different oxygen/sulfide ratios [9]. Authors observed that at flowrate ratios  
8 below 0.5 the hydrogen sulfide oxidation is incomplete producing mainly elemental sulfur. On  
9 the contrary, at flowrate ratios above 2 the oxidation is complete to sulfate. Similarly, Charnok  
10 *et al.* (2012) underlined the importance of oxygen transfer to achieve high hydrogen sulfide  
11 removal in biogas sweetening at extreme acidic conditions [10]. In this study the use of  
12 membrane diffusers to transfer oxygen directly into the liquid phase is proposed. As described  
13 above, biological oxidation of hydrogen sulfide generates sulfate that accumulates in the liquid  
14 phase. It is therefore of interest to examine the effect of sulfate on oxygen transfer. In addition,  
15 the biogas is often stored above atmospheric pressure, so it is also important to determine the  
16 possible effect of the working pressure in the system.

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18 Also the addition of a non-aqueous phase to optimize oxygen transfer in a bioscrubber system is  
19 proposed. The use of more than one phase has mainly been studied in two-phase partitioning  
20 bioreactors, where the presence of the non-aqueous phase has been found to increase the mass  
21 transfer rate [11]. Recently the use of solid inert polymers has been studied due their low cost,  
22 easy separation and recycling possibilities [12, 13].

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24 According to the stated above, main parameters that affect the gas-liquid oxygen transfer by  
25 means of membrane diffusers were tested under common biofiltration conditions. In addition  
26 the improvements that can be achieved through direct injection of oxygen and the use of a non-  
27 liquid phase (NLP) as mass transfer vector were also studied.

## 2. Materials and methods

28  
29 Oxygen transfer experiments were carried out using the equipment described in Rodriguez *et al.*  
30 (2012), which consists of a 200 l tank (114.4 cm height and 50 cm of inner diameter) and a 31  
31 cm fine bubble diffuser (Roediger ROEFLEX, Sacede) mounted near the tank bottom [14]. The  
32 variables measured were the liquid phase DO concentration and temperature (Cellox 325 probe,  
33 WTW), the supplied air flowrate (PT-313-0400 variable area flowmeter, Tecfluid), the tank  
34 pressure (ZSE40/ISE40 high-precision digital pressure switch, SMC) and the sulfate  
35 concentration (ICS-1000 ion chromatography system, DIONEX).

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37 Experiments were conducted at a constant temperature of 22±3 °C and at atmospheric pressure,  
38 except for those experiments in which the tank was pressurized, which were conducted at  
39 relative pressures in the range 0–0.5 bar. The reagents were nitrogen (99.99%, Abelló Linde),  
40 oxygen (99.95%, Abelló Linde) and sodium sulfate (99%, Scharlab). The transfer vector used  
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was Desmopan® (DP9370A, 3×3 mm cylinders, polyurethane of poly(oxytetramethylene)glycol and methyl diisocyanate, Bayer), as NLP.

For data acquisition a SCADA system in LabWindows™ was used. The DO sensor was connected to an inoLab740 meter (WTW) linked to the computer via a RS-232 interface.

The mass transfer coefficient was calculated from a mass balance model according to the standardized ASCE/EWRI 2-06 equation, 2007 [15], under dynamic conditions:

$$\frac{dC_L}{dt} = K_L a \cdot (C_S - C_L) \quad (3)$$

Equation (3) was solved by the fminsearch algorithm in Matlab using the Simplex search method at initial conditions  $t = 0$  and  $C_L = C_L(0)$ .

The mass transfer coefficient values obtained give a reference temperature of 20 °C, which can be corrected using Equation (4):

$$K_L a_{20} = K_L a \cdot \Theta^{(20-T)} \quad (4)$$

The amount of oxygen transferred to the liquid phase was determined using Equation (5), where SOTR is the standard oxygen transfer rate, expressed in Kg O<sub>2</sub> h<sup>-1</sup>:

$$SOTR = K_L a_{20} \cdot C_S \cdot V_T \quad (5)$$

Before each experiment, the liquid phase was deoxygenated with nitrogen to a concentration of 2–3 mg l<sup>-1</sup> DO. The coefficient measurement error was 10%, below the maximum allowed error of 15% established in ASCE/EWRI 2-06 [15].

As stated in the literature, sensor response times below 1/K<sub>L</sub>a can be disregarded when calculating the mass transfer coefficient [16]. Here, the sensor response time was determined using the method proposed by Vandu *et al.* (2004) [17], and a value of less than 1/K<sub>L</sub>a was obtained.

### 3. Results and discussion

#### 3.1. Effect of air flowrate

Taking as a reference the biofilter described by Tomàs *et al.* (2009) [18], the stoichiometric oxygen requirement for treating up to 1500 ppm<sub>v</sub> of H<sub>2</sub>S with a biogas flowrate of 40 m<sup>3</sup> h<sup>-1</sup> will be in the range 1–7 m<sup>3</sup> h<sup>-1</sup> of air (0.3–2.1 kg O<sub>2</sub> h<sup>-1</sup>). Consequently, experiments were conducted with supplied air flowrates from the diffuser in the range 1.2–6.7 m<sup>3</sup> h<sup>-1</sup> at atmospheric pressure. Figure 1 shows the air flowrates in the range 2.3–6.7 m<sup>3</sup> h<sup>-1</sup>, a linear relationship is observed between SOTR and the gas flowrate; when the air supply is increased, the oxygen transfer rates rises. These results are consistent with those obtained by Cachaza *et al.* (2008) [19] in a bubble column reactor, using a perforated plate to introduce air into the system.

Doubling the air flow from 1.2 to 2.34 m<sup>3</sup> h<sup>-1</sup> increased oxygen transfer by 59% (which graphically corresponds to a value of 0.026 kg O<sub>2</sub> m<sub>air</sub><sup>-3</sup>). When the supplied air flowrate is

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3 increased until  $6.7 \text{ m}^3 \text{ h}^{-1}$ , the oxygen transfer further improved by up to 118% ( $0.021 \text{ kg O}_2 \text{ m}_{\text{air}}^{-3}$ ).  
4 These results clearly indicate that the supplied air flowrate has a strong effect on the amount  
5 of oxygen transferred.  
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### 7 3.2. Addition of pure oxygen

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9 Figure 2 shows the SOTR obtained in experiments with addition of oxygen according to the  
10 supplied oxygen flowrate. Flows in the range  $0.24\text{--}0.48 \text{ m}^3 \text{ h}^{-1}$  were used, which supply an  
11 equivalent volume of oxygen to air flowrates of  $1.2\text{--}2.4 \text{ m}^3 \text{ h}^{-1}$ . As can be seen in Figure 2,  
12 lower oxygen transfer rates are achieved when pure oxygen is added to the system. If the same  
13 volumetric flowrates were used for pure oxygen and air, rather than an equivalent oxygen  
14 flowrate, oxygen transferred would be higher with the addition of pure oxygen. Using an  
15 equivalent oxygen flowrate ensures that the hydrodynamic conditions are comparable (same  
16 mass transfer coefficient) but the mass transferred is enhanced due to a higher driving force for  
17 oxygen pure in comparison to air.  
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20 At oxygen flowrates of  $0.24\text{--}0.48 \text{ m}^3 \text{ h}^{-1}$ , the total gas-liquid contact area is considerably  
21 smaller than that at the equivalent air flowrates (considering the bubbles size is kept constant at  
22 the flowrates range tested) and, what it is more important, the decrease of the turbulence at  
23 lower flowrates decrease significantly the oxygen transfer rate. In addition, when working with  
24 pure oxygen, the partial pressure of oxygen in the gas phase increases, raising the driving force  
25 of oxygen transfer. However, as can be seen in Figure 2, when pure oxygen is added at an  
26 equivalent oxygen flowrate to that achieved by the addition of air, the resulting oxygen transfer  
27 rate is higher. This is corroborated by discontinuous stirred-tank reactor experiments carried out  
28 by Pinelli *et al.* (2010) [20], who noted that the increase in  $k_L$  does not compensate for the  
29 reduction of the specific area, relating it to a decrease in gas hold-up. In addition, it is important  
30 to note that oxygen addition requires less energy consumption than the addition of air: the  
31 values of oxygen transfer as a function of energy supplied (measured as standard aeration  
32 efficiency, SAE, in  $\text{kg O}_2 \text{ h}^{-1} \text{ kW}^{-1}$ ) are 0.62 and  $3.31 \text{ kg O}_2 \text{ h}^{-1} \text{ kW}^{-1}$  for the addition of air and  
33 pure oxygen, respectively. However, the oxygen production entails an additional cost not  
34 accounted when air is used.  
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### 46 3.3. Effect of pressure

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48 Figure 3 shows the oxygen transfer rate at relative pressures inside the tank in the range  $0\text{--}0.5$   
49 barg, with an air flowrate of  $1.2 \text{ m}^3 \text{ h}^{-1}$  supplied by the diffuser. The pressure range used in these  
50 experiments reproduces the working pressure conditions of the biogas storage system for the  
51 Manresa and St. Joan de Vilatorrada (Barcelona, Spain) wastewater treatment plant [18].  
52 Contrary to the expected outcome, the oxygen transfer rate showed a slight decreasing trend as  
53 the relative pressure in the system increased (a total decrease in oxygen transfer of 8.3% was  
54 observed over the relative pressure range studied). However, considering the error associated  
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with SOTR measurements is 10%, and according to an ANOVA statistical test performed, this differences were not statistically different at 5% level of significance.

Bubble diameter should be smaller at higher working pressures. Smaller bubbles provide a larger total mass transfer area, meaning that the transfer rate should be higher in pressurized systems. Experiments were conducted to dismiss possible DO sensor errors caused by the increase in working pressure in the system (data not shown).

The literature is inconclusive regarding the effect of pressure increase on mass transfer coefficients. Han and Al-Dahhan (2007) [21] found that  $k_L$  remained constant or decreased slightly at higher pressures because the liquid properties varied only slightly in the pressure range tested (1.4–10.0 barg). Letzel and Stankiewicz (1999) [22], who used a pressure range of 1–13 barg in a bubble column, observed that  $k_L a$  increased with pressure. This outcome was explained by the fact that the higher pressure increases the gas density, reducing the stability of the bubbles and lowering the coalescence rate. By contrast, Yoshida and Arakawa (1968) [23] found that  $k_L a$  decreased at higher pressures, particularly with a low agitation rate, which was related to the surface renewal rate or degree of interfacial turbulence, arguing that the increase in pressure changes the surface tension and causes the gas-liquid interface to become rigid. The results presented in this study are in agreement with those reported by Heijnen *et al.* (1980) and Masutani and Stenstrom (1991) [24, 25].

### 3.4. Effect of sulfate concentration

Figure 4 shows the evolution of oxygen transfer at sulfate concentrations in the aqueous phase from 0 to 1000 mg l<sup>-1</sup>. The chosen range of concentrations was designed to ensure that bacterial activity is not inhibited, an effect that is observed at a sulfate concentration of 1900 mg l<sup>-1</sup>, according to Jin *et al.* (2005) [26] and 5000 mg l<sup>-1</sup> according to Ramirez *et al.* (2009) [27]. Experiments were conducted with air flowrates of 1.2, 1.8 and 2.4 m<sup>3</sup> h<sup>-1</sup>. Oxygen transfer increased with sulfate concentration in the aqueous phase, with a total increase in SOTR of 34% over the concentration range 0–1000 mg l<sup>-1</sup>.

Similarly, Jamnongwong *et al.* (2010) [28] studied the effect of salt content on mass transfer coefficients in a bubble column reactor equipped with a membrane diffuser. They concluded that the addition of salt increases the surface tension of the water—a phenomenon known as the Marangoni effect—to generate greater interfacial turbulence, raising the interface renewal rate and leading to increased oxygen transfer. Bubble column experiments conducted by Painmanakul and Hebrard (2008) [29] showed that higher salt concentrations reduce bubble coalescence, thereby increasing the mass transfer area and leading to a higher rate of oxygen transfer.

### 3.5. Effect of non-liquid phase addition

In biofiltration systems containing poorly water soluble compounds, if an immiscible second phase is added, the total oxygen transfer from the gas phase to the aqueous phase is the sum of the oxygen transferred from the gas to the non-aqueous phase and the oxygen transferred from the non-aqueous to the aqueous phase [30]. According to Muñoz *et al.* (2007) [31], the overall mass transfer in multiphase systems is controlled by transfer from the gas to the non-aqueous phase.

Figure 5 shows the oxygen transfer rate at air flows of 3.0, 4.2 and 6.6 m<sup>3</sup> h<sup>-1</sup> with NLP concentrations of 0, 1, 3, 5, 7 and 9% v/v. The oxygen transfer rate increased with NLP concentration at all three of the air flowrates tested, up to a concentration of 3% v/v. These results are consistent with those obtained by Quijano *et al.* (2010a) and (2010b) [32, 33], who experimented with the use of multiphase systems for removal of ethanol and toluene, respectively. However, when the NLP concentration was increased to above 3% v/v, oxygen transfer fell at the 3.0 and 4.2 m<sup>3</sup> h<sup>-1</sup> air flowrates. This abrupt drop in oxygen transfer at higher second-phase concentrations was also witnessed by Quijano *et al.* (2009) [34], who used silicone oil as a mass transfer vector at concentrations of 0–50% v/v. At silicone oil concentrations above 10%, the global mass transfer coefficient decreased.

In their study with bubble column using hollow glass spheres as NLP, Mena *et al.* (2011) observed a mass transfer increase until 2-3% v/v de NLP. For expandable polystyrene the results are quite different; in this case the mass transfer decreases with the addition of the non-liquid phase. In agreement with the results obtained in this study, the experiments of Albal *et al.* (1983) showed that for solid particles concentration less than 5% v/v the mass transfer increased and for concentrations higher than 5% the mass transfer decreased [36].

The reduction in mass transfer may be related to an increase in the apparent viscosity, since at higher viscosities the liquid turbulence decreases, leading to lower mass transfer rates [37].

At the highest air flowrate, oxygen transfer increased up to a NLP concentration of 5% v/v, which indicates that the optimal proportion depends on the air flowrate supplied, since increasing the air flowrate increases the liquid phase turbulence. The addition of a non-aqueous phase at a concentration of 3% v/v led to an average increase in oxygen transfer of 32%.

### 4. Conclusions

The experiments described above show that the typical working conditions of a biological desulfurization process are compatible with the addition of a fine bubble membrane diffuser and can in fact contribute to improving oxygen transfer rates. It was found that when pure oxygen is added instead of the equivalent oxygen volume in air, the rate of mass transfer is lowered due to the smaller available mass transfer area. Increasing the working pressure from 0 to 0.5 barg does not have a significant effect on oxygen transfer, but a 34% improvement in SOTR is observed



when sulfate concentration is increased from 0 to 1000 mg l<sup>-1</sup>. The addition of a 3% v/v non-aqueous phase increased oxygen transfer by an average of 32%. The addition of a transfer vector is a possible means of improving oxygen transfer, up to a certain concentration of the non-aqueous phase. This study demonstrates that oxygen transfer is largely insensitive to working pressure but responds strongly to the supplied air flowrate.

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### 6. Nomenclature

$a$  = Specific contact area, m<sup>2</sup> m<sup>-3</sup>

$C_L$  = Dissolved oxygen concentration at the liquid phase, kg m<sup>-3</sup>

$C_S$  = Dissolved oxygen concentration at saturation, kg m<sup>-3</sup>

DO = Dissolved oxygen

$K_L$  = Global mass transfer coefficient, s<sup>-1</sup>

$k_L$  = Liquid side mass transfer coefficient, m s<sup>-1</sup>

$K_{La_{20}}$  = Volumetric mass transfer coefficient corrected at 20 °C and pressure of 1 atm, s<sup>-1</sup>

NLP = Non-liquid phase

$Q_G$  = Gas flowrate, m<sup>3</sup> s<sup>-1</sup>

SAE = Standard Aeration Efficiency, kg O<sub>2</sub> kW<sup>-1</sup> h<sup>-1</sup>

SOTR = Standard Oxygen Transfer Rate, kg O<sub>2</sub> h<sup>-1</sup>

$t$  = experimental time, s

$T$  = Temperature, °C

$V_T$  = Volume of the tank, m<sup>3</sup>

$\Theta$  = empirical temperature correction factor, 1.024, dimensionless

$H$  = Henry coefficient, dimensionless

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3 Figure 1: SOTR with air flowrates in the range 1.4–6.7 m<sup>3</sup> h<sup>-1</sup>.

4 Figure 2: SOTR and flowrate for the addition of pure oxygen (Q<sub>O<sub>2</sub></sub>) and equivalent air (Q<sub>Air</sub>).

5  
6 Figure 3: SOTR and relative pressure inside the tank for an air flowrate of 1.2 m<sup>3</sup> h<sup>-1</sup>

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8 Figure 4: SOTR and sulfate concentration in the aqueous phase for air flows of 1.2, 1.8 and 2.4  
9 m<sup>3</sup> h<sup>-1</sup>

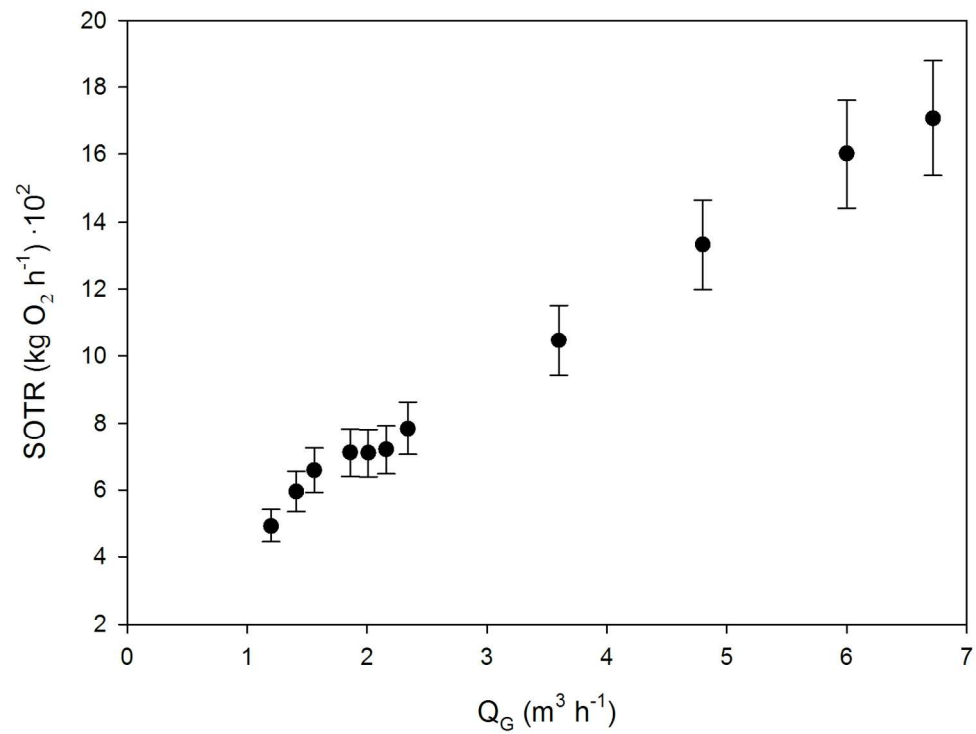
10 Figure 5: SOTR and NLP concentration, for air flowrates of 3.0, 4.2 and 6.6 m<sup>3</sup> h<sup>-1</sup>.  
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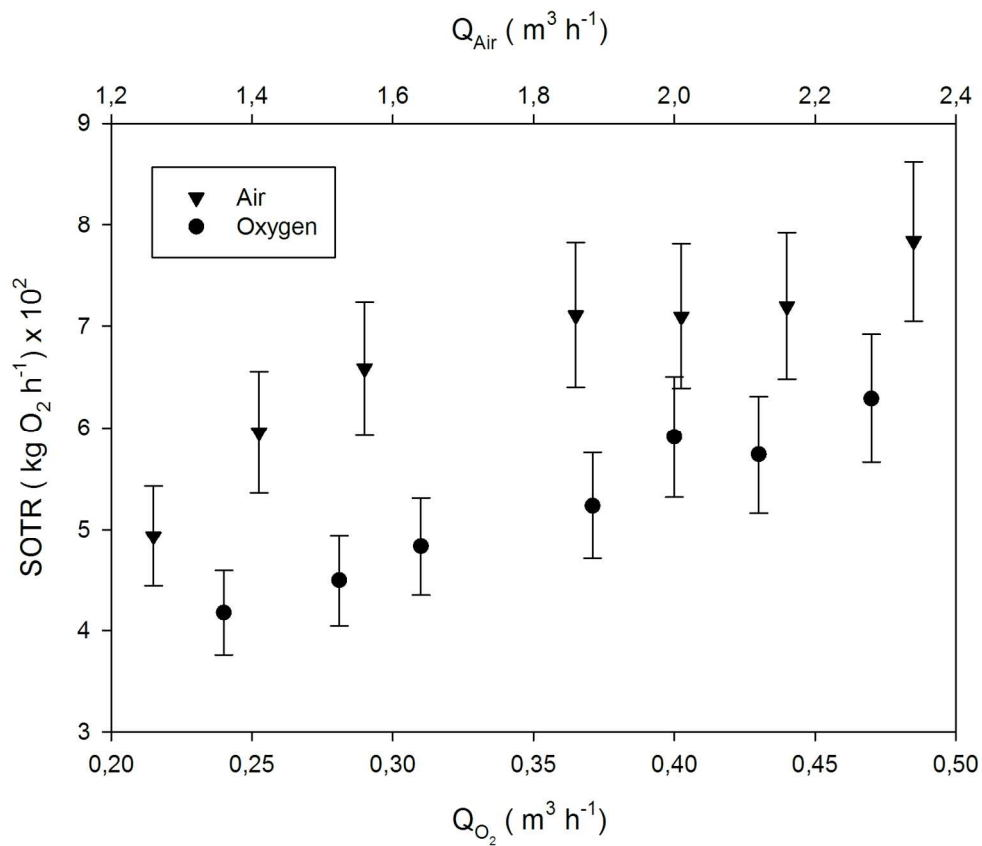
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3 **Text for the Table of Contents**  
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5 The effect of different operating variables on the oxygen mass transfer process has been tested  
6 in a pilot plant using a membrane diffuser as intensive gas-liquid contactor. Tests performed  
7 simulated those conditions found in the biofiltration of H<sub>2</sub>S at high loads for biogas sweetening.  
8 Results showed interesting effects on the mass transfer process when increasing the solution  
9 ions concentration and with the addition of a non-liquid phase (NLP).  
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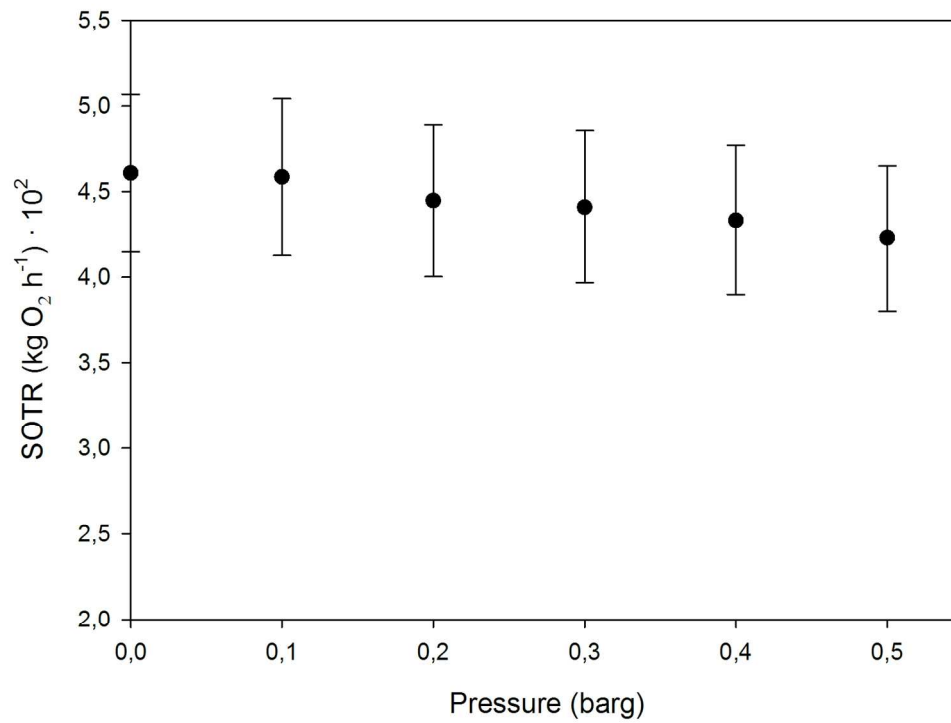
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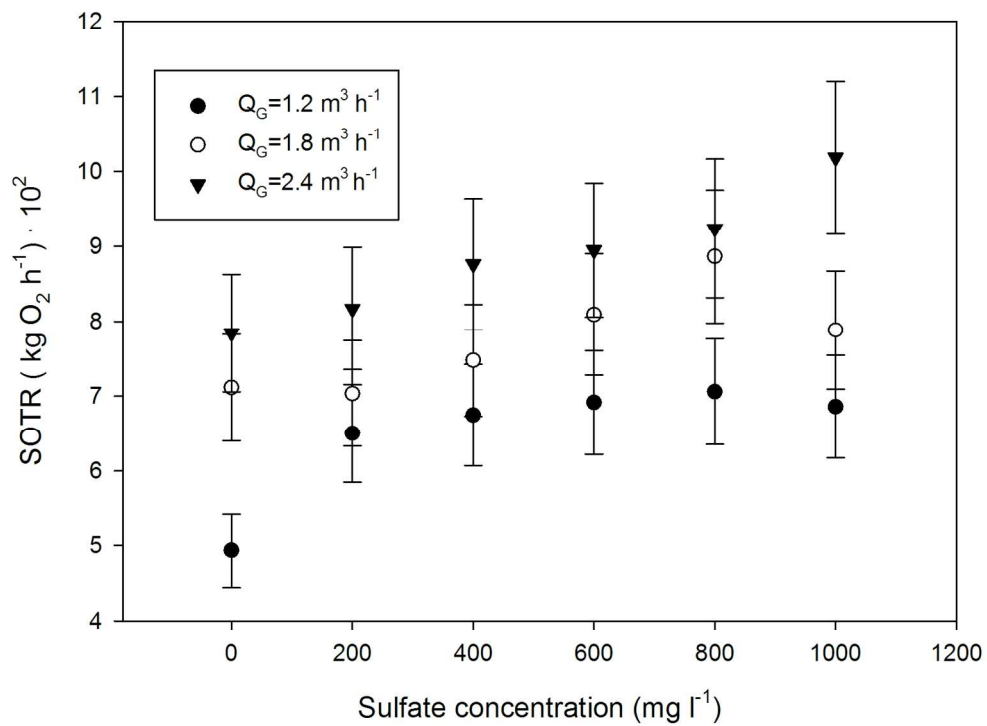


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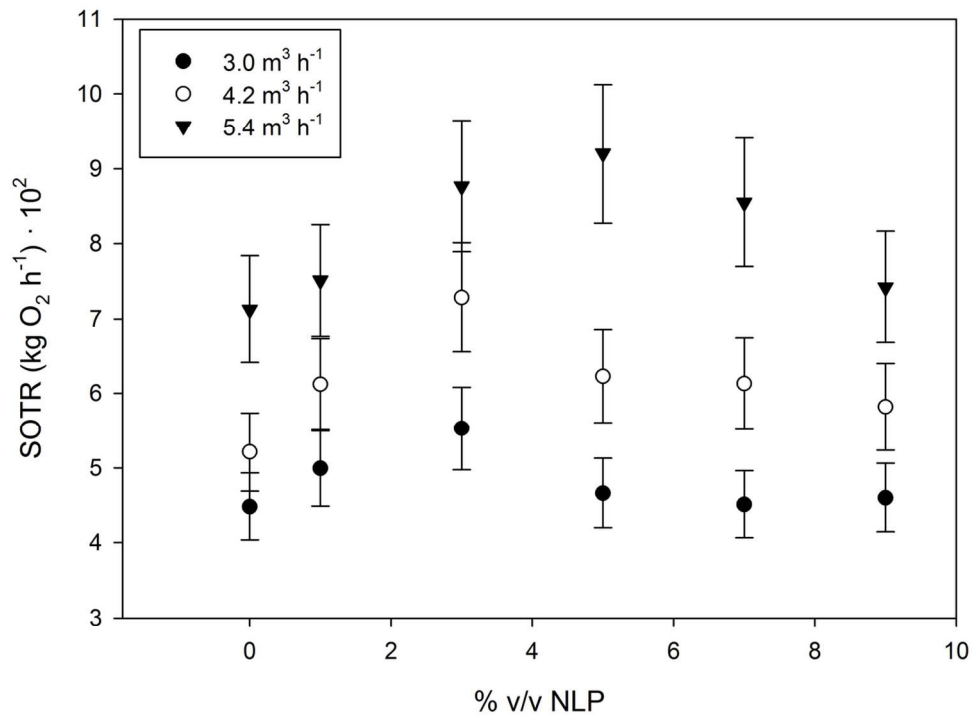


Figure 5  
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