

Oxygen transfer in three-phase airlift and stirred tank reactors using silicone oil as transfer vector

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ABSTRACT

The use of organic liquids as vectors to enhance mass transfer has been applied since the 1970s. However, mass transfer in three-phase reactors is only partially understood. This paper aimed to characterize oxygen transfer in three-phase reactors containing air as gas, silicone oil as vector and water as aqueous phase. A mass transfer model that considers separately air/vector, vector/water and air/water oxygen transfers was developed. The model was used to describe oxygen transfer in airlift and stirred tank reactors containing from 0 to 50% of silicone oil. Under the experimental conditions, silicone oil had a positive effect on the overall oxygen transfer. In both reactor designs, the maximum overall oxygen transfer was observed with 10% silicone oil which was increased by 65 and 84% in the airlift and stirred reactor, respectively, compared to reactors operated without silicone oil. The overall transfer increase was mainly due to an enhanced air/water transfer. With 10% silicone oil, the air/water contribution to the overall oxygen transfer was 94.7 and 93.0% for the airlift and stirred reactor, respectively.

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1. Introduction

Transfer of oxygen, volatile organic compounds and other poorly water-soluble compounds from a gaseous phase to a liquid phase is a key issue in many biotechnological processes. To improve mass transfer, the use of vectors has been explored for more than 30 years [1]. Two types of vectors have been explored so far; (i) dynamic transporters such as hemoglobin to improve oxygen transfer [2] and (ii) passive transfer vectors. The use of passive transfer vectors is based on the addition of an immiscible liquid phase with high affinity for the poorly water-soluble compound. An additional gas/vector/water transfer pathway is then established (the terms “vector” and “water” refers to the organic and aqueous phases, respectively).

Several organic phases have been used as vectors, including perfluorocarbons, silicone oil and C₁₂ to C₁₆ alkanes [3]. According to Dumont and Delmas [4], although numerous works on three-phase bioreactors have been reported, no agreement on the positive or negative effect of vectors on mass transfer has been reached. Latter, Clarke and Correia [5] suggested, from a literature review, that the positive or negative effect of vectors on mass

transfer depends on the type and the percentage of vector used and the reactor design.

The mechanisms involved in gas/vector, vector/water and gas/water mass transfer have been only partially explained or quantified [6]. Most of the published works have focused on the effect of vector on the overall mass transfer coefficient or on biological processes. Relatively little attention has been given to the separate measurement of gas/vector, vector/water and gas/water mass transfer. For instance, the vector and the water phases are commonly assumed as a single homogeneous phase [7–12]. This impedes a separate estimation of the three partial mass transfers above mentioned.

Rols et al. [13] developed a method to determine partial oxygen transfer coefficient (k_La) in an air/dodecane/water system. This method included a non-dispersed vector separated from the water by a PTFE membrane, which is quite different to the well mixed conditions prevailing in three-phase reactors. Yeom and Daugulis [14] measured the partial transfer of benzene in a two stage bioreactor system where absorption was done in a column separated from the bioreactor, which is an uncommon configuration. To our knowledge, these are the sole reports on partial mass transfers estimation.

The aim of this work was to measure separately the gas/vector, vector/water and gas/water oxygen transfer rates in three-phase

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reactors. For that purpose, a model based on the oxygen mass balances was developed. This model was used to characterize mass transfer in lab-scale airlift and stirred tank reactors containing from 0 to 50% of silicone oil.

2. Mathematical model

The experimental system used in this work was air and silicone oil dispersed in an aqueous sulfite solution. Silicone oil was selected as vector. It has been reported as a non-toxic and non-biodegradable organic phase [3,15,16]. The sulfite solution simulates oxygen uptake as occurring in biological processes. In this system, two assumptions can be made; (i) oxygen is transferred from air to water, directly or through vector and (ii) oxygen uptake occurs only in water. The last assumption has been made previously by several authors who observed bacterial adhesion onto the vector but no bacterial presence into it [3]. According to these assumptions, three different oxygen transfers can take place; gas/water, gas/vector and vector/water (Fig. 1). In this work mass transfer was quantified by oxygen transfer rates, expressed as mass of oxygen transferred per unit of reactor volume and per unit of time. Under this way, transfer rates are comparable to each other.

Two transfer rates were defined for each of the three oxygen transfer pathways; the actual transfer rate (OTR_{ij}) and the maximum transfer rate also called oxygen transfer capacity (OTC_{ij}). Subscripts i and j refer to the origin and the target phases; g , v and w for gas, vector and water respectively. OTC_{ij} are the OTR_{ij} observed when the maximal gradient concentration is reached between the i th and j th phases. Thus OTR_{ij} can be expressed as a fraction of OTC_{ij} as follows;

$$OTR_{gv} = OTC_{gv} \cdot \left(1 - \frac{C_v}{C_v^*}\right) \quad (1)$$

$$OTR_{vw} = OTC_{vw} \cdot \left(1 - \frac{C_w}{C_w^*}\right) \left(\frac{C_v}{C_v^*}\right) \quad (2)$$

$$OTR_{gw} = OTC_{gw} \cdot \left(1 - \frac{C_w}{C_w^*}\right) \quad (3)$$

In these equations, C_i and C_i^* represent the actual and the saturation concentration of oxygen in the i th phase, respectively. In the presence of sodium sulfite the oxygen mass balance in vector and water can be written as follows;

$$\frac{dC_v}{dt} = [OTR_{gv} - OTR_{vw}] \frac{V_r}{V_v} \quad (4)$$

$$\frac{dC_w}{dt} = [OTR_{gw} + OTR_{vw} - q] \frac{V_r}{V_w} \quad (5)$$

In Eqs. (4) and (5), q is the oxygen consumption rate due to sulfite oxidation and V_r , V_w and V_v , are the volume of the vector,

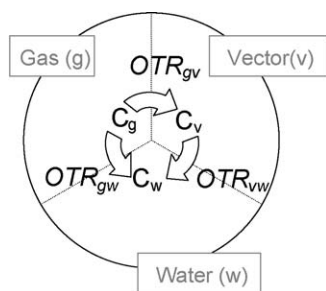


Fig. 1. Mass transfer model.

the water phase and the reactor, respectively. In Eqs. (4) and (5), OTR_{ij} can be substituted by Eqs. (1)–(3). Additionally, the presence of sulfite ensures that C_w equal zero making the value of OTR_{gw} equal to OTC_{gw} and giving Eqs. (6) and (7). Eq. (8) gives the sulfite oxidation rate, where $Y_{SO_3^{2-}/O_2}$ is the sulfite/oxygen stoichiometric yield.

$$\frac{dC_v}{dt} = \left[OTC_{gv} \left(1 - \frac{C_v}{C_v^*}\right) - OTR_{vw} \left(\frac{C_v}{C_v^*}\right)\right] \frac{V_r}{V_v} \quad (6)$$

$$\frac{dC_w}{dt} = \left[OTC_{gw} + OTR_{vw} \left(\frac{C_v}{C_v^*}\right) - q\right] \frac{V_r}{V_w} \quad (7)$$

$$\frac{dSO_3^{2-}}{dt} = -(Y_{SO_3^{2-}/O_2} \cdot q) \frac{V_r}{V_w} \quad (8)$$

3. Materials and methods

3.1. Setup and operating conditions

Two lab-scale reactors were used; an airlift (ALR) and a stirred tank (STR) reactor. The ALR, designed according to literature [17], was a glass bubble reactor (0.12 m inner diameter, 0.57 m height, 6.4 L total volume, 5 L working volume) with a concentric tube (0.075 m inner diameter, 0.35 m height) placed at 0.05 m from the bottom of the reactor. A porous plate (0.05 m diameter) was used for air supply. The stirred tank reactor was a glass reactor (0.133 m inner diameter, 0.55 m height, 7.6 L nominal volume, 5 L working volume), fitted with a double Rushton turbine operated at 300 rpm. Air was supplied through a perforated ring (orifice diameter of 1×10^{-3} m). In both reactors, the air flow-rate was maintained at $8.34 \times 10^{-5} \text{ m}^3 \text{ s}^{-1}$ and controlled by a variable area flow-meter (Cole Parmer, Mexico) previously calibrated with a mass flow controller (GFC171S, Aalborg, Mexico). The experiments were performed at ambient temperature (23 ± 2 °C). The reactors were operated with 0–50% v/v, silicone oil (polydimethylsiloxane 200R, Dow Corning, Mexico). This silicone oil is characterized, at 25 °C, by a dynamic viscosity of 0.019 Pa s, a specific gravity of 0.95 and a surface tension of 20.6 mN m^{-1} [18].

3.2. Mass transfer experiments

Mass transfer was characterized in the reactors with sulfite solution and silicone oil as liquid phases. A sodium sulfite concentration matching the ionic strength of typical culture medium (8.0×10^{-2} M), corresponding to a sodium sulfite concentration of 2.6×10^{-2} M was chosen. This allowed the measurement of mass transfer under similar conditions as real culture medium. Before mass transfer measurements, the reactors containing the sodium sulfite concentration, water and vector volumes were operated until saturation of vector and water was reached. This typically lasted about 10–20 min. A final concentration of 2.5×10^{-4} M of cobalt sulfate was then injected in the reactor to start the sulfite oxidation reaction (Eq. (9)).



The system rapidly reached a pseudo steady-state during which the oxygen concentration in the vector (C_v) was constant and the sulfite concentration decreased linearly. This pseudo steady-state lasted until sulfite concentration became limiting and the system returned back to saturation. During the mass transfer experiments, the oxygen concentration in water and vector was measured as well as the sulfite concentration in water. Eqs. (4) and (5) were fitted to experimental data to estimate OTC_{gw} , OTC_{gv} and OTC_{vw} . Then, the corresponding values of OTC_{gw} , OTR_{gv} and OTR_{vw} were obtained from Eqs. (1)–(3).

3.3. Measurement techniques

The overall gas holdup was measured in both reactors according to the volumetric method described by Vandu et al. [19]. Silicone oil and water viscosity was measured at 23 °C with a RV2 viscosimeter (Haake, Mexico). Silicone oil and water surface tension were measured with a 2141 tensiometer (Analyte, Mexico). During experiments, sulfite concentration was measured in water samples taken from the bottom of the reactor, by iodometric back-titration method according to Zhao et al. [20]. Dissolved oxygen concentration in water was measured at the bottom of the reactors, using a polarographic oxygen probe, connected to a DO Meter (HI 2400, Hannah Instrument, Mexico). The oxygen concentration in the vector was measured after vector sampling. For that purpose, aeration and mixing of the reactor was shortly stopped. The vector started to separate immediately from water and a sample of approximately 5 mL of the vector was taken from the top of the reactor and was used to measure dissolved oxygen concentration. After sampling, aeration and mixing were started again. For data interpretation the time scale was corrected to discard times during which the aeration and mixing were

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