



Interfacial gas–liquid transfer area in alkane–aqueous dispersions and its impact on the overall volumetric oxygen transfer coefficient

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ABSTRACT

K_La and gas–liquid interfacial area per unit volume were quantified in 2.5–20% n -C_{10–13} aqueous dispersions agitated at 600–1200 rpm. The interfacial area was quantified using high speed photography and image analysis. Interfacial area correlated strongly with K_La over all agitation rates and alkane concentrations, suggesting that the interfacial area was the major factor defining K_La in these dispersions.

The bubble diameter and gas hold up were identified as key parameters defining an optimal K_La at 5% alkane at agitation rates of 800–1200 rpm, through their impact on the interfacial area. Decreased bubble diameter and increased gas hold up with alkane addition from 2.5% to 5% resulted in increased interfacial area. However, a decreased gas hold up on alkane addition above 5% resulted in a correspondingly decreased interfacial area, despite the continued decline in bubble diameter. The decrease in bubble diameter and the decrease in gas hold up have in part been attributed to the reduction of surface tension from 26.0 to 17.7 and the doubling of viscosity to 2.0 mN/m respectively over the range of alkane concentrations examined.

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1. Introduction

The growing interest in hydrocarbon-based bioprocesses and the key role of the overall volumetric oxygen coefficient (K_La) in the operation, design and scale up of these processes highlights the need for an understanding of the factors which underpin the behaviour of K_La in response to process conditions in these systems.

In a hydrocarbon–aqueous system, oxygen transfer has been postulated to take place via one or more of several possible alternate transfer paths between the organic, aqueous and gaseous phases, and interphasic K_La proposed. It has been suggested that the most likely transfer path will depend on whether the hydrocarbon adsorbs onto the gas bubble or remains freely dispersed, and, if the former, whether it is adsorbed as beads (non-spreading) or as a continuous film (spreading) around the surface. A simplified approach is used here, neglecting the different paths the oxygen may take to transfer from the gas bubbles to the aqueous phase.

In aqueous systems, the behaviour of K_La is largely attributable to fluid turbulence which impacts on the gas–liquid interfacial area per unit volume available for transfer and the resistance to transfer.

In the case of hydrocarbon–aqueous systems, the K_La behaviour is likely to be additionally influenced by the fluid properties. A combined influence of fluid turbulence and fluid properties on K_La is supported by the impact of hydrocarbon on K_La behaviour demonstrated in numerous studies of agitated hydrocarbon–aqueous systems. Enhanced K_La has been reported in stirred tank reactors (STRs) with dodecane [1] and perfluorodecalin [2] and in airlift reactors and STRs with dodecane [3]. K_La behaviour, however, varies markedly on hydrocarbon addition under different conditions of agitation and hydrocarbon type and concentration. A recent review [1] has identified 3 distinctly different K_La behavioural trends in dispersions comprising 20 different hydrocarbons at concentrations from 0% to 33%, and has characterised these behavioural trends according to the hydrocarbon moiety and concentration, operating conditions and geometric constraints. Type 1 behaviour is characterised by a peak K_La associated with a specific system-dependent hydrocarbon concentration, type 2 behaviour by increased K_La with increasing hydrocarbon and type 3 behaviour by a constant or decreased K_La on hydrocarbon addition.

While the effect of turbulence and fluid properties on K_La in hydrocarbon-based systems has been observed, the causative factors underlying this behaviour have yet to be quantified. In this paper, correlation of K_La with measured interfacial area over a wide range of agitation rates and alkane concentrations in alkane–aqueous dispersions enabled the identification and elucidation of the parameters characterising K_La behaviour in

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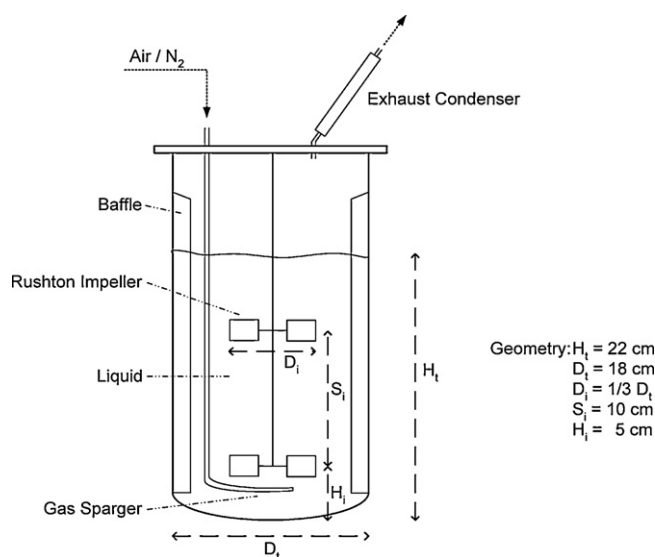


Fig. 1. Bioreactor system geometry.

hydrocarbon-based systems, and provided insight into the underlying mechanisms which dictate this behaviour.

2. Materials and methods

2.1. Experimental system

Experiments were performed in an aerated 7.5 l baffled bioreactor (New Brunswick Bio Flo 110, USA) with a 5 l working volume and system geometry as shown in Fig. 1. Agitation was provided by two identical, six-flat-bladed Rushton turbines. Aeration at 0.8 vvm (volume air per volume liquid per minute), was provided by means of a ring sparger situated directly below the lower Rushton turbine. Temperature was maintained at 22 ± 0.1 °C by means of a heating jacket and coiling coils.

The experiments were conducted in an immiscible alkane–aqueous system at discrete alkane concentrations of 2.5%, 5%, 10% and 20% (v/v), each at agitation rates of 600 rpm, 800 rpm, 1000 rpm and 1200 rpm. The alkane cut, n -C_{10–13}, which comprised 10% n -C₁₀, 30% n -C₁₁, 30% n -C₁₂ and 30% n -C₁₃ alkane, was used throughout (SASOL, South Africa). Deionised water was used in all the experiments.

2.2. Parameter measurement and calculation

2.2.1. Determination of the overall volumetric oxygen transfer coefficient and the oxygen transfer rate

The dissolved oxygen concentration (DO) in the liquid was measured by means of a submerged polarographic oxygen probe (Toledo/Ingold® InPro® 6800 Series) with a Teflon membrane. $K_L a$ was determined from the DO vs. time profile using the pressure step procedure, taking the response lag of the oxygen probe into account, as previously described [4].

The maximum oxygen transfer rate (OTR) was calculated from the $K_L a$ and oxygen solubility ($C_{\text{saturated}}$) according to Eq. (1).

$$\text{OTR} = K_L a C_{\text{saturated}} \quad (1)$$

where $C_{\text{saturated}}$ (in mg l⁻¹) was calculated according to the volume average model [5] using Henry's Law for oxygen solubility in water and the literature values for oxygen solubility in pure alkanes [1].

2.2.2. Determination of bubble diameter, gas hold up and interfacial area

Bubble characteristics were determined using high speed photography and image analysis software. Images (1024 × 1018 pixels) were recorded at 30 frames per second at a resolution of approximately 1 Mb with a Kodak MegaPlus ES 1.0 camera (Redlake MASD, USA) connected to a Matrox Meteor-II/1394 board. Both the frame grabber and the camera were operated by Matrox Inspector 3.1 [6]. Two sets of 300 image frames were captured with the second set to confirm that steady-state had been reached. Approximately 600 bubble images were acquired at random at each process condition examined. This number of images was considered sufficient since 500–800 bubbles have been shown to be a statistically representative number for adequate quantification of the bubble size [7,8].

To facilitate image processing, computational image analysis software was developed using Matlab® programming. The original image was converted to a form from which the required bubble descriptors could be extracted, as follows. A binary gradient mask of the image was created to show lines of high contrast depicting the bubble outlines. The gaps in these lines were closed by subsequent dilation of the image and the interior holes filled. Finally, segmented bubbles were obtained by image erosion from which the projected area (A_p) of each bubble was calculated, based on the number of pixels within a specified area.

The equivalent diameter (D_{eq}), i.e. the diameter of a circle with the same projected area as the bubble, was obtained from the projected area according to Eq. (2) and used to determine the Sauter mean bubble diameter (D_{32}) according to Eq. (3).

$$D_{\text{eq}} = 2 \sqrt{\frac{A_p}{\pi}} \quad (2)$$

$$D_{32} = \frac{\sum_{i=1}^n n D_{\text{eq}}^3}{\sum_{i=1}^n n D_{\text{eq}}^2} \quad (3)$$

The Sauter mean diameter was used as the preferred bubble size descriptor as it is the most commonly used in image analysis [9–20].

The gas hold up was measured using the dispersions height technique [16,20,21]. Here, the liquid level in the bioreactor was measured with no aeration (V_0) relative to that during aeration at 0.8 vvm (V). The volume gas hold up (ε_G) was calculated according to Eq. (4).

$$\varepsilon_G = \frac{V - V_0}{V} \quad (4)$$

The interfacial transfer area (a) was calculated from the bubble Sauter mean diameter and the gas hold up assuming spherical bubbles, according to Eq. (5).

$$a = \frac{6\varepsilon_G}{D_{32}} \quad (5)$$

2.2.3. Determination of surface tension and viscosity

Viscosity was determined using a rheometer (Paar Physica MCR 501, Anton Paar, Austria) with a double gap measuring system and Rheoplus® software. Surface tension was determined using a tensiometer (CSC Du Noüy). This measured the force required for a submerged platinum ring to break through the interface as it was raised out of the liquid, from which the surface tension was calculated. Prior to measurement of the surface tension and viscosity, the alkane–aqueous dispersions were homogenised at 6 bar (gauge) in order to obtain an emulsion that would be stable during the period of measurement.

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