Investigating the effects of hydrodynamics and mixing on mass transfer through the free-surface in stirred tank bioreactors

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HIGHLIGHTS

- A stirred tank aerated through the free-surface is globally and locally characterized.
- PIV and PLIF are used to assess flow, mixing and mass transfer dynamics.
- Large-scale flow patterns highly influence mass transfer through the free-surface.
- Mixing efficiency is described based on time stabilization and spatial uniformity.
- Vertical gradients of dissolved gas concentration are present and persistent.

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ABSTRACT

In stirred-tank bioreactors, flow structures of various length and time scales are implied in scalar transport phenomena, such as gas species transfer through the liquid free-surface and their homogenization in the bulk. A proper understanding of the underlying mechanisms, *i.e.* hydrodynamics, mixing and mass transfer, and of their interactions is required to design and develop reliable and efficient production-scale bioprocesses. The objective of the present work is to experimentally investigate the coupling between gas-liquid mass transfer of oxygen with mixing efficiency and circulation patterns inside an arbitrarily chosen stirred-tank configuration aerated through the liquid free-surface, a baffled 20 L-vessel agitated by two Rushton turbines. Based on global parameter values, the most appropriate rotating speed, $N = 300$ rpm, is selected in order to further study local hydrodynamic quantities using Particle Image Velocimetry (PIV), as well as mixing and mass transfer dynamics using Planar Laser-Induced Fluorescence (PLIF). The results obtained with these local experimental methods are analyzed in detail. Their averages are first successfully compared to global data. Statistical analysis of their spatial distributions show that large-scale flow patterns significantly influence mass transfer through the free-surface of the stirred tank. Even if global measurements show that global characteristic times for mixing and mass transfer differ by two orders of magnitude, local experimental characterization shows persistent vertical gradients of dissolved gas concentrations. So the dissolved gas concentration is not as perfectly uniform as one might expect.

1. Introduction

Stirred-tank reactors are commonly used in various chemical and biological processes because of their flexibility and good performance due to the wide range of available operating conditions.

Besides, biological processes often require gas-liquid contact for acceptable product formation. In some applications, aeration through the liquid free-surface is enough to meet the gas-liquid mass transfer demand. This is typically the case when either gas requirements/reactor volume are relatively low, or when bubbles are undesirable. Even when dealing with sparged reactors, aeration through the liquid free-surface is a factor to be considered when scaling up/down processes as its contribution to the overall mass transfer is linked to the evolution of the surface area to volume ratio (Godoy-Silva et al., 2009).

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In most stirred-tank reactors, hydrodynamics consists of turbulent flow structures of various length and time scales whose relative contributions to the interfacial transfer rate and to the mixing of scalar quantities such as dissolved gas concentration depend on the equipment-scale. Therefore, the design of a reliable and efficient industrial bioreactor requires a better identification of all characteristic scales relative to fluid dynamics, mixing and mass transfer. The dependence of these phenomena and of their interconnections regarding the equipment scale and geometry may be then properly understood. In practice, the influence of the vessel size on transport structures is usually not considered when scaling-up, leading then to suboptimal operations (Nienow, 1997; Wernersson and Trägårdh, 1999; Mathpati et al., 2009). Traditional scale-up criteria based on the conservation of global quantities such as specific dissipated power P/V, mixing time t_M or overall mass transfer coefficient $K_L a$ should be used with caution as they do not necessarily account for the heterogeneous spatial distributions of the flow variables and for their impact on process performance.

Consequently, the scale-up of a bioprocess from lab to industrial scale often results in a decrease of productivity compared to the lab-scale (Enfors et al., 2001). Large gradients (substrate, nutrients, dissolved oxygen or carbon dioxide, pH) appear when the bioreactor volume increases and can lead to modifications of the biological responses in terms of physiology or/and metabolism compared with lab-scale culture (Larsson et al., 1996; Delvigne et al., 2006; Garcia-Ochoa and Gomez, 2009; Morchain et al., 2014). Understanding and modeling these complex interactions between biological reaction and hydrodynamics are known to be key problems when dealing with bioprocesses, but can be expected only if hydrodynamics and physical (non biological) phenomena responsible for gradients are fully understood.

From fluid dynamics theory, it is well known that a turbulent flow can be divided into a macroscopic convection velocity field (large-scale motions, circulation) and a microscopic eddy-like velocity field which is, finally, dominated by diffusion (small-scale motions, turbulence). From this mechanistic representation, the mixing process is often divided into several simpler stages from the *macro*-scale to the *micro*-scale, *i.e.* from the scale of the whole vessel to finer scales associated to turbulent properties (Bałdyga and Pohorecki, 1995).

Gas-liquid mass transfer is also usually described by considering mechanisms occurring at these two scales (Martín et al., 2008). The exchange of species basically occurs through thin boundary layers at the gas-liquid interface by molecular diffusion processes. However, for poorly soluble gases, the transport of species is most often controlled by resistance on the liquid side which is dependent on flow structures and dynamics at and near the interface. Indeed, macro-scale motions determine the fluid paths across the reactor as well as their contribution to liquid free-surface aeration. They also lead to periodic and local deformation of the liquid free-surface. Besides, these large-scale structures are directly affected by the reactor design, and thus also by its size. On the other hand, micro-scale motions are responsible for the rippled liquid free-surface and for the concentration gradients surrounding it and within the bulk. Turbulence is known to be of key importance for the gas transfer process. Many researchers have tried to explain the contribution of each flow scale on mass transfer process using conceptual models (*e.g.* Danckwerts, 1951; Fortescue and Pearson, 1967; Lamont and Scott, 1970; McCready et al., 1986; Banerjee et al., 2004). However, one of the main bottlenecks in the development and validation of detailed gas-liquid mass transfer models is the scarcity of reliable experimental information, relative to phenomena occurring near the interface. In parallel to the evolution of conceptual models, investigation techniques have thus also been improved. Even if computer simulations have provided estimates of required parameters for simple flow conditions at low turbulent Reynolds numbers, these simulations remain, up to now, too costly to be used for high Reynolds numbers.

Despite some research advances, the physical mechanisms of coupling between turbulence and gas-liquid mass transfer still remain unclear, such as the characteristic scales of eddies involved in the transfer or the effects of surface chemistry. Studies on mass transfer process across the air-water interface driven by grid-stirred turbulence provide some insights into the transfer mechanisms along with flow and scalar structures near the liquid free-surface (Chu and Jirka, 1992; Herlina and Jirka, 2004; Tsumori and Sugihara, 2007; Janzen et al., 2010; Variano and Cowen, 2013; Herlina and Wissink, 2014). These works indicate that the larger and smaller eddies are respectively able to distort and penetrate the boundary layer. In particular, surface-renewal motions observed in vertical planes have a much larger length-scale than the Kolmogorov scale η; they are rather within the range of the Taylor macro- and micro-scales, Λ and λ.

The aim of the present work is thus to develop an experimental approach to characterize an arbitrarily chosen standard agitation configuration in terms of global and local quantities in order to better understand the coupling between gas-liquid mass transfer, mixing efficiency and circulation patterns.

For this purpose, a stirred-tank reactor aerated through the liquid free-surface is first characterized globally. The specific dissipated power, the mixing time and the global mass transfer coefficient are measured for different rotation speeds. The most appropriate agitation condition is selected on the basis of the values of these traditional scale-up criteria.

Then, local detailed information relative to a large volume fraction of the stirred vessel is collected to further study the system performance in terms of hydrodynamics, mixing and mass transfer. This requires accurate measurements of both flow quantities and concentration fields. To measure flow quantity field, Particle Image Velocimetry (PIV) is particularly appropriate, capable of measuring the flow field in an entire plane of the system with a few operations, yielding also enough information to obtain additional flow features such as turbulence and its characteristic length-scales (Bugay, 1998; Escudié and Liné, 2003; Khan et al., 2004; Micheletti et al., 2004; Gabriele et al., 2009). To measure the concentration fields, Planar Laser-Induced Fluorescence (PLIF) has proven to be a suitable measurement technique. It has already been used to assess mixing characteristics (Houcine et al., 1996; Guillard et al., 2000; Fall et al., 2001; Busciglio et al., 2014) in stirred-tank reactors, or to characterize mass transfer through an air-water interface, mainly using oxygen (Woodrow and Duke, 2001; Herlina and Jirka, 2004; Falkenroth et al., 2007; Janzen et al., 2010; Jimenez et al., 2013, 2014) but also carbon dioxide (Tsumori and Sugihara, 2007; Variano and Cowen, 2013). Besides, both techniques are considered as proper tools for numerical CFD model validation (Coroneo et al., 2011; Delafosse et al., 2014). The results obtained with these local experimental methods are analyzed in detail. Their averages are compared to global data. Statistical analysis of their spatial distributions gives a further insight on the effects of hydrodynamics and mixing on mass transfer through the free-surface in stirred-tank bioreactors.

2. Equipment and methods

2.1. Stirred-tank configuration

The investigated stirred-tank reactor, presented in Fig. 1(a), has already been used in previous works (Delafosse et al., 2014). This standard lab-scale set-up is a flat-bottom cylindrical vessel of