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Chemical Engineering and Processing 44 (2005) 121–130
Chemical Engineering and Processing
www.elsevier.com/locate/cep

Mechanism of mass transfer from bubbles in dispersions Part II: Mass transfer coefficients in stirred gas–liquid reactor and bubble column

V. Linek*, M. Kordač, T. Moucha

Department of Chemical Engineering, Prague Institute of Chemical Technology, Technická 5, 166 28 Prague 6, Czech Republic

Received 15 December 2003; accepted 14 May 2004
Available online 2 July 2004

Abstract

Experimental data on the average mass transfer liquid film coefficient (k_L) in an aerated tank stirred by Rushton turbine and in bubble column are presented. Liquid media were used as 0.8 M sodium sulphite solution, pure or with the addition of Sokrat 44 (copolymer of acrylonitrile and acrylic acid) or short-fiber carboxymethylcellulose (CMC) for the Newtonian and long-fiber CMC for the non-Newtonian viscosity enhancement and ocenol (*cis*-9-octadecen-1-ol) or polyethylenglycol (PEG) 1000 for surface tension change. Volumetric mass transfer coefficient (k_La) and specific interfacial area (a) were measured by the Danckwerts' plot method. Coefficients k_L measured by pure oxygen absorption in pure sulphite solution and Newtonian viscous liquids are well fitted by the "eddy" model in the form of $k_L = 0.448\,(ev/\rho)^{0.25}\,(D/v)^{0.5}$ with a mean deviation of 20%. Surface-active agents (ocenol and PEG) and non-Newtonian additive (long-fiber CMC) reduced k_L value significantly but their effect was not described satisfactorily neither by surface tension nor by surface pressure. It is shown that the decisive quantity to correlate k_L in the stirred tank and bubble column is power dissipated in the liquid phase rather than the bubble diameter and the slip velocity. Absorption of air did not yield correct k_L data, which did not depend on or slightly decreased with increasing power. This is due to the application of an improper gas phase mixing model for absorption data evaluation.

Keywords: Gas–liquid contactor; Bubble column; Agitated vessel; Mass transfer coefficient; Viscosity; Surfactants

1. Introduction

Mass transfer from swarm of bubbles into turbulent liquid controls the rate of many chemical and biochemical processes. It is assumed that the mechanism of mass transport in liquid phase is due to a renewal of the liquid at the bubble surface. Models of the process differ in the scale of flow, which is responsible for the renewal.

The first group of models ("eddy" models) assumes that the liquid renewal is due to small-scale eddies of the turbulent field. These models are based on idealized eddy structures of turbulence in the bubble vicinity. Lamont and Scott [1] have assumed that the small scales of turbulent motion, which extend from the smallest viscous motions to the inertial ones, affect the mass transfer. In any case, these motions are much smaller in scale than the gas bubbles. As a result, the size of the gas bubble is not a very critical parameter for the estimation of k_L. They deduced [1] the following relation for k_L:

$$k_L = c_1 \left(\frac{ev}{\rho}\right)^{0.25} \left(\frac{D}{v}\right)^{0.5} \quad (1)$$

Different authors predict different values of the constant c_1: 0.301 [2], 0.4 [1], 0.531 [3], 0.592 [4] and 1.13 [5].

The second group of models ("slip velocity" models) assumes a gross mean flow of fluid relative to the bubble (slip velocity) and a bubble surface mobility control of this renewal rather than the small-scale eddies of the turbulent field. The model proposed by Calderbank and Moo-Young [6] belongs to this group. They [6] have divided bubbles by size into two categories: the "small" bubbles ($d < 1$ mm), which behave always as a rigid sphere, and the "large" ones ($d > 2.5$ mm), which always have a completely mobile

* Corresponding author. Tel. +420 2 2435 3298; fax: +420 2 3333 7335.
E-mail address: linekv@vscht.cz (V. Linek).

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doi:10.1016/j.cep.2004.05.009

surface. An expression for the calculation of k_L for the "large" bubbles has been deduced from dimensionless analysis of mass transfer from rising particle in gravitational field.

$$k_L = 0.42\left(\frac{\Delta\rho\nu g}{\rho}\right)^{1/3}\left(\frac{D}{\nu}\right)^{1/2} \quad (2)$$

For the "small" bubbles, an expression for the calculation of k_L has been obtained by the equation proposed by Frössling [7] for dissolution of a rigid sphere.

$$k_L = 0.31\left(\frac{\Delta\rho\nu g}{\rho}\right)^{1/3}\left(\frac{D}{\nu}\right)^{2/3} \quad (3)$$

The transition between these two bubble categories was unclear and strongly dependent on the presence of surfactants. An expression for calculation of the mass transfer coefficients in this transition region was not given.

Recently, Vasconcelos et al. [8] and Alves et al. [9] proposed another variant of the "slip velocity" model. Their model is based on experimentally observed phenomena of an abrupt change of the dissolution rate of free-floating bubbles held stationary in a downward water flow. Starting from these experiments, they have interpreted the mass transport from bubbles in terms of bubble contamination kinetics, using a stagnant cap model, according to which bubbles suddenly change from a mobile interface to a rigid condition when surface tension gradients, caused by surfactant accumulation, balance out shear stress. The fresh bubble entering the dispersion has clean, completely mobile interface. For the bubbles with completely mobile surface, k_L is given by the equation, which follows from the well-known penetration model with exposition time equal to d/v_{sl}:

$$k_L^{mobil} = 1.13\sqrt{\frac{v_{sl}}{d}}D^{1/2} \quad (4)$$

After some time (τ^{mobile}), enough contaminant for transition to rigidity is accumulated on the surface. For the rigid bubbles, an expression for the calculation of k_L follows from Frössling [7] equation.

$$k_L^{rigid} = 0.6\sqrt{\frac{v_{sl}}{d}}D^{2/3}\upsilon^{-1/6} \quad (5)$$

Gas–liquid slip velocities v_{sl} are assumed to be close to single-bubble terminal velocities in still water, v_t, on which a correction for turbulence is introduced, $v_{sl} = 0.65v_t$ [9]. Assuming monosized bubble dispersion and considering only two possible values of k_L (k_L^{mobil} and k_L^{rigid}, depending on surface mobility), they deduced the following relation for the average mass transfer coefficient:

$$k_L = \frac{k_L^{mobil}\tau^{mobile} + k_L^{rigid}(\tau_R - \tau^{mobile})}{\tau_R} \quad (6)$$

where τ_R is the average bubble residence time.

$$\tau_R = \frac{V_L\varepsilon}{q(1-\varepsilon)} \quad (7)$$

The time τ^{mobile} depends on bubble diameter and surfactant concentration.

$$\tau^{mobile} = k\frac{d^{1/2}\ln(d/h_{trans})}{c_{surf}} \quad (8)$$

where d is an average bubble diameter, k is an empirical constant related with surfactant properties, c_{surf} is the concentration of surfactant and h_{trans} is the bubble clean segment height at the transition point from mobile to rigid.

The main difference between the "eddy" and the "slip velocity" models is in the influence of turbulence intensity on mass transfer coefficient: the "eddy" models predict an increase while the "slip velocity" models a decrease of k_L with increasing turbulence intensity, i.e. with increasing power dissipated in the liquid. This is due to the fact that k_L from rigid bubbles is cca four times lower than from the ones with a mobile surface. The small bubbles, which either have lower k_L (according to Calderbank's conception) or become rigid more quickly (according to Alves' conception, τ^{mobile} diminishes with diminishing bubble diameter d, see Eq. (8)), are generated in large amount just at higher turbulence intensities. As a result, this leads to the decrease of average mass transfer coefficient with increasing power dissipated in the liquid. Literature data on k_L in stirred tanks supporting both these models can be found.

In dispersions with no mechanical agitation (bubble columns, air-lifts), the only source of energy is the expansion of the entering gas which gives the relation $e = \rho g v_s$. Reith [10], Dillon and Harris [11], Kawase et al. [5] and Vázquez et al. [12] revealed strong dependence of liquid-side mass transfer coefficient on gas flow rate in bubble columns: $k_L \sim v_s^{0.25/0.5}$. Slight dependence or independence of k_L on v_s was found by Schumpe and Deckwer [13], Bouaifi et al. [14], Wongsuchoto et al. [15] and Vasconcelos et al. [8]. The values of k_L corresponded to values predicted by Calderbank's model for "large" bubbles. Values which correspond also to "small" bubbles were found by Vázquez et al. [12] and Vasconcelos et al. [8] only, who worked with solutions of surfactants. Vasconcelos et al. [8] interpreted their results in terms of bubble contamination kinetics.

In dispersions with mechanical agitation, following authors found an increase of k_L with increasing energy dissipation e: Prasher and Wills [4] ($k_L \sim e^{0.25}$); Figueiredo and Calderbank [16] ($k_L \sim e^{0.33}$); Bouaifi and Roustan [17] ($k_L \sim e^{0.22}$); Panja and Rao [18] ($k_L \sim e^{0.15}$); and Linek et al. [19] ($k_L \sim e^{0.14}$). Power dissipated in the liquid by an agitator is proportional to the third power of impeller rate ($e \sim f^3$). Thus, we can add the results of authors that presented k_L as a function of impeller rate f: Koetsier and Thoenes [20] ($k_L \sim f^{0.9} \sim e^{0.3}$) and Yoshida and Miura [21] ($k_L \sim f^{0.6} \sim e^{0.2}$). Robinson and Wilke [22] measured k_L that did not depend on power dissipated and Hassan and Robinson [23] obtained coefficients that decreased with increasing power dissipated. It was shown [32] that the reported independence on or the decrease of k_L with increasing power dissipated was a result of misinterpretation of k_La data.