Modeling of Mass-transfer in Bubbly Flows Encompassing Different Mechanisms

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Modeling of Mass-transfer in Bubbly Flows
Encompassing Different Mechanisms

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Abstract

Models proposed to describe the liquid side mass transfer coefficient in absorption prosesses differ widely in such basic questions as on which of the local flow variables they are based. Comparison of different alternatives with experimental data taken from the literature suggests that there are two basic mechanisms, a laminar and a turbulent one, each of which dominates under suitable conditions. A dimensionless number that allows to identify the corresponding regimes is suggested together with a preliminary model encompassing both. New experiments will be needed to come to a final conclusion.

Keywords: mass-transfer, penetration model, dispersed gas liquid multiphase flow, Euler Euler two fluid model, closure relations, CFD simulation

1 INTRODUCTION

Mass-transfer from gas bubbles to the surrounding liquid or vice versa is an important consideration in chemical engineering. Models of this process that can be used in full CFD simulations or simplified treatments of industrial equipment have been proposed for a long time. However even quite basic questions apparently have not yet found definitive answers.

This is evidenced for example by looking at some recent works performing Euler-Euler simulations of bubble- and airlift-columns. Talvy et al. (2007), Wiemann and Mewes (2005), and Cockx et al. (2001) use correlations for the mass transfer coefficient involving the mean bubble motion relative to the liquid. Dhanasekharan et al. (2005) use a correlation based on liquid turbulence. Wang and Wang (2007) provide a comparison of models of both types. Krishna and van Baten (2003) simply take the mass-transfer coefficient as constant. Similarly for aerated stirred tanks Fayolle et al. (2007) use a correlation based on bubble size and relative velocity. Gimbun et al. (2009) compare this to a correlation based on liquid turbulence.

Obviously it is not even clear on which parameters the mass transfer coefficient depends, let alone what form the dependence should have. Broadly there are two types of models which assume different mechanisms, namely (i) laminar or mean flow and (ii) turbulent eddies, to govern the mass-transfer.

An attempt to delineate regimes where each of the two mechanisms is dominant has been made by Alves et al. (2006) on the basis of data for vertical bubbly pipe flow from Vasconselos et al. (2002). However, their main result is not expressed in a way that is applicable as a general estimate as will be explained below. Moreover, the selection of models to obtain quantitative estimates for both mechanisms may not be adequate for the flow configuration considered and no attempt at a unified model comprising both regimes is made.

While the present contribution still falls short to provide definitive answers, some progress can be made concerning these issues which may be helpful to direct further research on the topic in particular new experiments. To this end some of the available modeling options are summarized in section 2 and discussed in comparison with a selection of experimental data in section 3. Following the discussion in section 4, a proposal for a preliminary unified model is made from which estimates on the transition between both regimes can be drawn.

2 MASS-TRANSFER MODELS

In the following we assume that the resistance to mass-transfer is dominated by the liquid side so that only the liquid mass-transfer coefficient k_L needs to be considered. This assumption applies e.g. during absorption from gas bubbles which are saturated with the transferred species. Predictions of k_L are frequently made based on the penetration model. While this simple conceptual model may not provide a high quantitative accuracy it seems well suited to discuss the general questions posed above.

The penetration model Higbie (1935) considers one-dimensional time-dependent diffusion of the transferred component from the interface with a bulk concentration imposed at infinity. These approximations are suitable for a thin concentration boundary layer at a fluid interface. The mass flux at the interface is evaluated from Fick's law and averaged over a time-interval up to the so-called contact time τ_c. After this time the surface is supposed to be renewed, i.e. to be brought into contact with liquid at the bulk concentration. Depending on the mechanism by which this renewal occurs, different expressions are used for the contact time to be discussed shortly. The expression obtained for the mass-transfer coefficient is

$$k_L \propto \frac{2}{\sqrt{\pi}} \left(D_A \tau_c^{-1} \right)^{1/2} , \tag{1}$$

where D_A denotes the diffusion coefficient of the transferred component A in the liquid. There are more refined versions of the model (Dankwerts 1951) which do not take all elements of the surface to have the same contact time but instead allow a distribution of contact times. A few examples of contact time distributions considered in Danckwerts et al. (1963) give the same functional dependence of k_L on τ_c, the latter now being interpreted as the average contact time. Only the prefactor in Eq. (1) is modified in these examples. Admitting that some adjustment of the prefactor may be necessary in the end to obtain agreement with measured values for k_L we keep the value used in Eq. (1) for the time being and focus on the parameters on which contact time is supposed to depend.

Three expressions are frequently used for τ_c. The first one was proposed by Higbie (1935) assuming laminar flow around the bubble in which fluid elements enter the interface at the front stagnation point and leave it at the rear one. This results in the expression

$$\tau_c^{-1} = \frac{u_{rel}}{d_B} , \tag{2}$$

where d_B is the bubble size and u_{rel} its velocity relative to the liquid.

The two other expressions assume turbulent flow but take eddies of different size to dominate the interface renewal. Fortescue and Pearson (1967) proposed that this process is governed by the largest turbulent eddies, i.e. those in the production range of the turbulent spectrum. This gives the equivalent expressions

$$\tau_c^{-1} \propto \frac{\varepsilon}{\kappa} \propto \frac{\Lambda}{\sqrt{\kappa}} \propto \frac{\sqrt{2}\ \varepsilon^{1/3}}{\Lambda^{2/3}} , \tag{3}$$

where κ, ε and Λ denote the turbulent kinetic energy, dissipation and integral length scale.

In contrast, Lamont and Scott (1970) took the smallest eddies, i.e. those in the dissipation range, as the relevant ones. Then the expression

$$\tau_c^{-1} \propto \left(\frac{\varepsilon}{\nu} \right)^{1/2} \tag{4}$$

is obtained, where ε is again the turbulent dissipation and ν the viscosity of the liquid.

The original works (Fortescue and Pearson, 1967, Lamont and Scott, 1970) contain additional prefactors in Eq. (1) which come from assumptions on the local velocity field within a turbulent eddy. Since this introduces additional hypotheses which are hard to verify we discard them here in line with the consideration above.

3 COMPARISON WITH EXPERIMENTAL DATA

Qualitative evidence from experimental work shows that both laminar and turbulent mechanisms are relevant under appropriate conditions. Numerous investigations are available on mass-transfer from single bubbles rising in a quiescent liquid as summarized by Clift et al. (1978). Since turbulence is absent, only the laminar mechanism is possible and results are in reasonable agreement with Eq. (2) substituted in (1).

Conversely in turbulent flows where either u_{rel} is zero or d_B tends to infinity, laminar flow cannot contribute to the mass-transfer and only the turbulent mechanism is operative. The first condition is realized for example in horizontal bubbly flow (Lamont and Scott, 1970), the latter one at a free surface (Fortescue and Pearson, 1967). An open question remains whether large or small eddies are the relevant ones for the mass-transfer.
A quantitative comparison is made in the following on the basis of available data for bubbly flows in ducts. To this end the turbulent dissipation is taken as being equal to the total power input (Lamont and Scott, 1970, Alves et al., 2006). Together with the Blasius correlation for the friction factor (Pope, 2000) which applies to fully turbulent flows at $Re_H \geq 4000$ and low gas fraction, this gives

$$\varepsilon = \frac{2\ J_L^3}{D_H} f = 0.16\ Re_H^{2.75} \frac{\nu^3}{D_H^4}. \tag{5}$$

Here J_L is the liquid superficial velocity, f the Fanning friction factor, D_H the hydraulic diameter of the duct and $Re_H = J_L\, D_H / \nu$ the duct Reynolds number. For the integral turbulent length scale the correlation of Lauffer (1954) can be used (Hosokawa and Tomiyama, 2004), i.e.

$$\Lambda = 0.2\ D_H. \tag{6}$$

This gives the following expression for the mass-transfer coefficient according to the large eddy model

$$k_L \approx 1.7 \frac{(\nu\ D_A)^{1/2}}{D_H} \left(\frac{J_L D_H}{\nu} \right)^{0.46}, \tag{7}$$

while according to the small eddy model

$$k_L \approx 0.71 \frac{(\nu\ D_A)^{1/2}}{D_H} \left(\frac{J_L D_H}{\nu} \right)^{0.69}. \tag{8}$$

A corresponding expression for the laminar model is

$$k_L \approx 1.13 \frac{(\nu\ D_A)^{1/2}}{d_B} \left(\frac{u_{rel} d_B}{\nu} \right)^{1/2}. \tag{9}$$

where $Re_B = u_{rel}\, d_B / \nu$ is identified as the bubble Reynolds number.
A presentation of Eqs. (7) - (9) has been chosen from which both the dependence on the variables measured in experiments and the appropriate non-dimensionalized forms are easily inferred. Note that Eqs. (7) - (8) and Eq. (9) suggest different relevant length scales, which would complicate the comparison of turbulent and laminar models based on a dimensionless form of the equations.
Lamont and Scott (1970) have shown data for both horizontal and vertical flow in round pipes with diameters D_H of 8 and 16 mm using CO2 bubbles in water. The pipe Reynolds number Re_H is varied in the range from 2000 to 20000 which includes both developed turbulence and the transition region. The purity of the water is not specified. Information on the bubble sizes can be found in the quoted reference (Lamont 1966). From that work it is seen that the bubbles shrink by ~30% during the absorption. The dependence on the bubble size on the flow rate is smaller than this variation. Rough estimates for the two pipe diameters and orientations are listed in Table 1. A discussion of the applicability of the small eddy

model proposed is given by Lamont and Scott (1970), but an explicit comparison between data and models is lacking from the paper. Such a comparison is shown in the following.

	hor. 8mm ∅	**hor. 16mm ∅**	**vert. 8mm ∅**	**vert. 16mm ∅**
d_B [mm]	4.5	8.5	4.5	7.0

Table 1: Estimated bubble sizes from Lamont (1966).

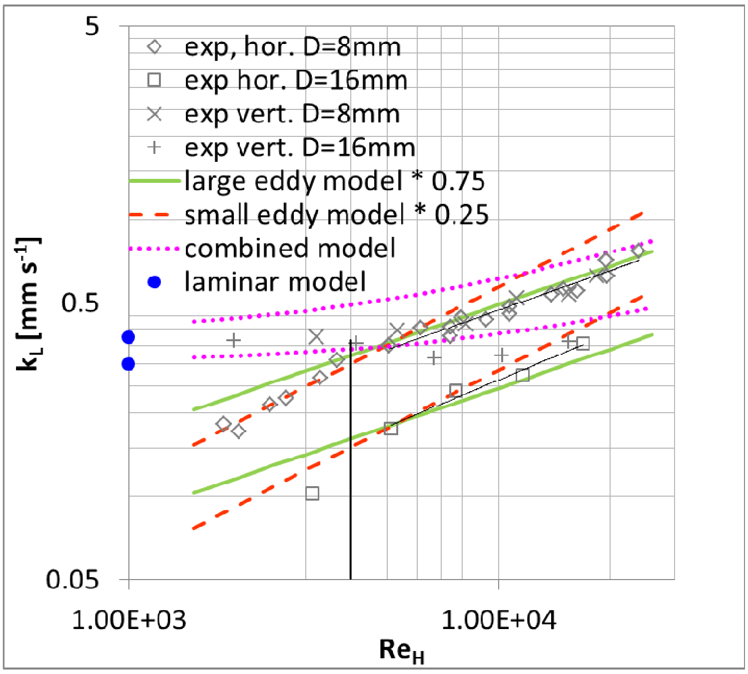

Figure 1: Data of Lamont and Scott (1970) compared with predictions of the penetration model using different expressions for the contact time.

The symbols in Fig. 1 display the measured values of k_L as function of Re_H which combines the varied parameters J_L and D_H in the experiments of Lamont and Scott (1970). It is seen that values for horizontal (◇ and □) and vertical (× and +) flow coincide with each other at large values of the liquid flux. Upon decreasing Re_H, k_L at first also decreases, but for vertical flow it tends to a constant when Re_H falls below a certain value while for horizontal flow it continues to decrease. The thin black lines show power law approximations to the data for values of $Re_H \geq 4000$, for which the model predictions Eqs. (7) and (8) are applicable. For D_H = 8 mm (◇ and ×) the fitted exponent is 0.47 while for D_H = 16 mm (□ and +) it is 0.56. Predictions of the large eddy model according to Eq. (7) are shown by the thick lines in Fig. 1 those of the small eddy model according to Eq. (8) by the dashed lines. The exponent 0.46 predicted for the large eddy model is not too far from the exponent fitted to the data while the predicted exponent of 0.69 for the small eddy model is significantly larger. To keep the figure clear, prefactors have been applied to both the large and small eddy models as indicated in the legend. Without such prefactors the large eddy model matches the data within ~33% while the small eddy model is by a factor of ~4 off. The values approached by the data for the vertical flows at lower values of Re_H are not too far from those predicted by the laminar

model Eq. (9) (solid dots) for the respective values of the bubble sizes taken from Table 1. Note that these values really correspond to Re_H tending to zero.
It may be concluded that at high Reynolds number mass transfer is governed by the turbulent mechanism. For the vertical orientation a crossover to the laminar mechanism occurs as Re_H is decreased. For the horizontal orientation such a crossover is not seen, but effects of not fully developed turbulence should be taken into account. The large eddy model offers a reasonable description of the turbulent mechanism even when no additional prefactor is introduced. The small eddy model in contrast predicts a wrong dependence on Re_H and without additional prefactor also a wrong magnitude of k_L. The laminar model is consistent with the data considering that there is some uncertainty associated with the values of the bubble size in the experiment.
Alves et al. (2006) have presented data for vertical downward flow in a slightly diverging square duct. By matching the liquid velocity with the terminal rise velocity of the bubble this facilitates long observation of individual bubbles. Air bubbles in clean water are used, the hydrodynamic diameter of the duct at the measurement position is $D_H \approx 10$ mm and the bubble terminal velocity is $u_{rel} \approx 0.25$ m/s. These conditions amount to a value of $Re_H = 2500$ falling in the transition region between laminar and turbulent flow. Bubble sizes vary between 1 and 6 mm or equivalently the bubble Reynolds number is $Re_B = u_{rel}\, d_B / \nu = 250 \ldots 1500$ corresponding to the ellipsoidal bubble shape regime. Alves et al. (2006) have compared these data with the laminar and small eddy models but not with the large eddy model. This latter comparison will be included in the following.

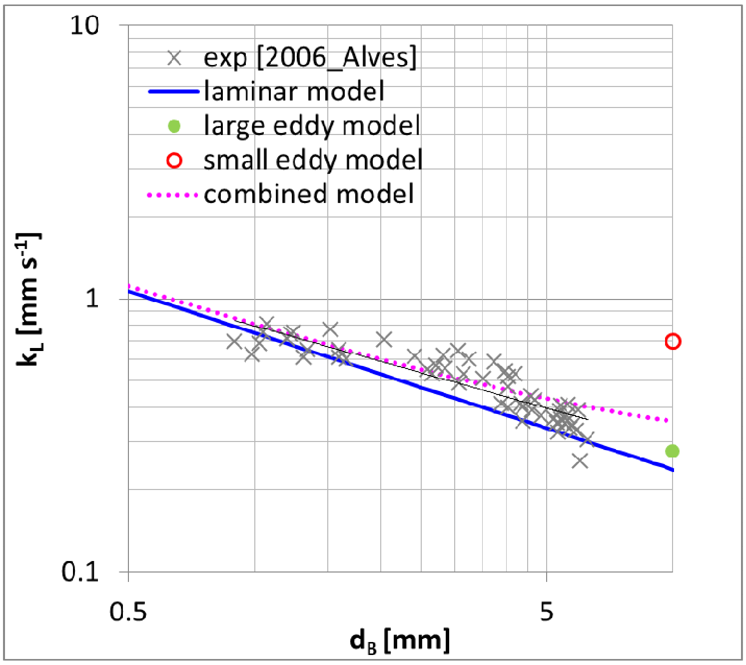

Figure 2: Data of Alves et al. (2006) compared with predictions of the penetration model using different expressions for the contact time.

The symbols (×) in Fig. 2 display the measured values of k_L as function of d_B which is the varied parameter in the experiments of Alves et al. (2006). The thin line gives a power law fit to the data as $k_L \propto d_B^{-0.43}$. The fitted exponent is not too far from –0.5 as predicted by the laminar model, Eq. (9), which is shown by the thick line. Some systematic deviation from power law behaviour is evident which may result from deviations from a spherical bubble shape. The solid dot gives the prediction for k_L by the large eddy model, Eq. (7), which really

corresponds to d_B tending to infinity. This prediction is about 30% smaller than the measured values for the largest bubbles. The corresponding prediction by the small eddy model, Eq. (8), shown by the open dot, is about 3 times larger unless an additional prefactor is introduced. This corresponds with the measured values for the smallest bubbles.
Since the data for k_L show a significant variation with d_B it may be concluded that the laminar mechanism for interface renewal here is the dominant one. The turbulent mechanism based on the large eddies is not completely negligible but less effective than the laminar one which is consistent with the data. In contrast, renewal based on the small turbulent eddies would be more effective than due to laminar flow contradicting the data unless additional assumptions are introduced.
To round off the picture it is worthwhile to mention some data for other systems.
Fortescue and Pearson (1967)have investigated absorption in decaying grid-generated turbulence in open channel flow. The data support their large eddy model. A comparison to the small eddy model has been made by Lamont and Scott (1970), where no good agreement was obtained for the scaling with Re_H.
Prasher (1973) has investigated absorption in turbulent open channel flow. They presented $k_L(\varepsilon)$ which by Eq. (3) in (1) gives $k_L \propto \varepsilon^{0.167}$ for the large eddy model and by Eq. (4) in (1) gives $k_L \propto \varepsilon^{0.25}$ for the small eddy model. Their finding is an exponent of 0.213 which is hardly closer to the latter than the former. The data show a rather large scatter which makes it difficult to distinguish between the two values for the exponent which are quite close.
Prasher and Wills (1973) show a similar analysis for an areated stirred tank where however the exponent was fixed to 0.25. A power law fit to the data results in $k_L \propto \varepsilon^{0.141}$ which clearly favors the large eddy model. The difference has probably gone unnoticed due to the again rather large scatter in the data.
Kress and Keyes (1973) investigated desorption in bubbly pipe flow and find a behavior of the mass transfer coefficient that does not match any of the models discussed here. This may be due to the fact that during desorption significant concentration gradients are present within the gas phase and the assumption that mass transfer be governed by the liquid side resistance in invalid.

4 DISCUSSION AND CONCLUSIONS

We have considered the question on which variables the mass transfer from bubbles to the surrounding liquid depends during absorption. The penetration model $k_L \propto (2/\sqrt{\pi})\ \sqrt{(D_A/\tau_c)}$ provides a useful framework to formulate different hypotheses based on assumptions on the nature of the flow around the bubbles, namely laminar or turbulent and in the latter case the size of the dominant eddies. Predictions resulting from the different assumptions have been compared with data available from the literature.
Qualitative reasoning shows that both laminar and turbulent mechanisms do exist under appropriate conditions. Quantitatively the penetration model with $\tau_c^{-1} = u_{rel} / d_B$ provides a reasonable first approximation for the laminar case. Deviations with respect to the data are most likely due to the assumption of a spherical bubble shape implicit in the use of the spherical equivalent diameter, while in reality the bubbles are of ellipsoidal shape. For the turbulent case the large eddy model $\tau_c^{-1} \propto \sqrt{2}\ (\varepsilon/\Lambda^2)^{1/3}$ provides a better match for the dependence of k_L on the duct Reynolds number Re_H than the small eddy model with $\tau_c^{-1} \propto \sqrt{(\varepsilon/\nu)}$. In addition, the large eddy model immediately gives the right magnitude of k_L while for the small eddy model the introduction of an additional prefactor is unavoidable.
To motivate the small eddy model, it is often simply argued that having the smallest time-scale, according to Eq. (1) these would give the highest transfer and thus would dominate.

However, this argument does not consider the conditions for an efficient surface renewal which requires that fresh liquid be brought to the surface *from a large distance*. But fast transport over a large distance is obtained for high velocities, so the dominant eddies really are those with the highest velocity and these correspond to the largest ones. It is conceivable that for a more precise description a weighted integral of contributions due to eddies of all sizes will have to be considered. Further investigation of this issue would require data on k_L with little statistical noise for a wide range of Re_H in a system where the laminar mechanism is supressed.

Considering the existence of two mechanisms dominating the mass transfer in different regimes the next question is how both can be combined into a unified model that contains both regimes and describes the transition between them. This transition should be given in terms of an appropriate dimensionless number rather than by an absolute value of the turbulent dissipation as stated in [2006_Alves]. The analysis given so far suggests the ratio of inverse contact times as given by Eqs. (2) and (3) for this purpose, i.e. if $u_{rel} / d_B \gg \sqrt{2}\,(\varepsilon / \Lambda^2)^{1/3}$ the laminar mechanism is dominant while if $u_{rel} / d_B \ll \sqrt{2}\,(\varepsilon / \Lambda^2)^{1/3}$ the turbulent mechanism is dominant. Concerning a model for the transition region one may try to use the sum of renewal rates τ_c^{-1} for both mechanisms in Eq. (1) which amounts to assuming that they both act in parallel. This is shown as the dotted lines labeled "combined model" in Figs. 1 and 2. For the data in Fig. 2 the match is quite good, but for the data in Fig. 1 the transition region is far too broad. In this latter case is appears more appropriate to use the maximum of both renewal rates in Eq. (1) so that only the more efficient one is active. This would also be in reasonable accord with the data in Fig. 2. We note that these two possibilities are the extremes of a general approach to combining two different asymptotic regimes discussed by Churchill and Usagi (1972). For the present case this approach can be written as

$$k_L^{tot} = \left(\left(k_L^{laminar} \right)^n + \left(k_L^{turbulent} \right)^n \right)^{1/n} , \tag{10}$$

which gives the first variant for $n = 2$ and the second for $n \to \infty$. Other values give a transition of varying abruptness. Further investigation of this issue would require data with controlled variation of both Re_H and Re_B independently and over large ranges.

The need for new experiments to provide definitive answers to the questions considered has been repeatedly stated. Such experiments should ideally be done on single bubbles where the interfacial area can be well characterized and the transferred mass be determined with high precision by modern high-speed imaging techniques. To facilitate establishment of a reliable model including both laminar and turbulent mechanisms as large range of bubble sizes, d_B = 1 … 10mm and a large range of duct Reynolds numbers Re_H = 2000 … 20000 should be covered simultaneously.

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6 NOMENCLATURE

Notation	Unit	Denomination
d_B	m	bulk bubble diameter
D_A	$m^2\ s^{-1}$	diffusion coefficient of transferred species
D_H	m	equivalent hydrodynamic diameter of duct
Eo	-	Eötvös Number
f	-	Fanning friction factor
J_L	$m\ s^{-1}$	liquid volumetric flux = superficial velocity
k_L	$m\ s^{-1}$	mass transfer coefficient
Mo	-	Morton Number
Re	-	Reynolds number
Sc	-	Schmidt number
Sh	-	Sherwood number
u_{rel}	$m\ s^{-1}$	bubble relative velocity
ε	$m^2\ s^{-3}$	turbulent dissipation rate
κ	$m^2\ s^{-2}$	turbulent kinetic energy
Λ	m	turbulent integral length scale
ν	$m^2\ s^{-1}$	kinematic viscosity
τ_c^{-1}	s^{-1}	inverse contact time = renewal rate

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