Intensification of Slurry Bubble Columns by Vibration Excitement

Jürg Ellenberger and Rajamani Krishna*

Department of Chemical Engineering, University of Amsterdam, Nieuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands

We show that application of low-frequency vibrations, in the 30 to 60 Hz range, to the liquid phase of an air - water - silica catalyst slurry bubble column causes significant enhancement of both gas holdup and volumetric mass transfer coefficient over a wide range of superficial gas velocities. The increase in the gas holdup is attributed mainly to a significant reduction in the rise velocity of the bubble swarm due to the generation of standing waves in the column. Furthermore, application of vibrations to the liquid phase serves to stabilize the homogenous bubbly flow regime and delay the onset of the churn-turbulent flow regime.

On montre que l'application de vibrations de faibles fréquences (entre 30 et 60 Hz) à la phase liquide d'une colonne à bulles à suspensions de catalyseur air-eau-silice, permet une amélioration significative à la fois du coefficient de rétention de gaz et du coefficient du transfert de matière volumétrique pour une vaste gamme de vitesses de gaz superficielles. L'augmentation de la rétention de gaz est imputée principalement à une réduction importante de la vitesse de montée de l'essaim de bulles qui est due à la création de vagues stationnaires dans la colonne. En outre, l'application des vibrations à la phase liquide sert à stabiliser le régime d'écoulement à bulles homogène et retarde l'apparition du régime d'écoulement agité-pistonnant.

Keywords: slurry bubble column, vibration excitement, process intensification, gas holdup, mass transfer, standing waves.

A bubble column reactor is commonly used in the process industries for carrying out a variety of liquid phase reactions (Deckwer, 1992). There are two regimes of operation for a bubble column. At low superficial gas velocities, U, we have homogeneous bubbly flow in which the dispersion consists of bubbles that are roughly uniform in size. Homogeneous bubbly flow is sustainable up to a superficial gas velocity $U = U_{trans}$, called the transition gas velocity. When U is increased to values beyond U_{trans}, we enter the heterogeneous or churn-turbulent flow regime. In this churn-turbulent flow regime we have a wide distribution of bubble sizes, ranging from 3 mm to 50 mm depending on the system properties (De Swart *et al.*, 1996). The wide distribution of bubble sizes causes a wide gas phase residence time distribution; this is often detrimental to reactor conversion and selectivity.

Earlier work has shown that smaller bubble sizes and improved mass transfer can be achieved by application of low-frequency vibrations in gas-liquid bubble columns (Ellenberger and Krishna, 2002; Krishna and Ellenberger, 2002a, 2002b, 2003; Krishna et al., 2000). The major objective of the present work is to extend this work to *slurry* bubble columns, operating with slurries of varying solid contents, to show that similar improvements are achievable.

Experimental Set-up and Procedures

The experimental set-up consists of a bubble column, a vibration exciter, a power amplifier, a vibration controller and a personal computer. A schematic diagram of the experimental set-up is given in Figure 1, and is the same as in our earlier publications (Ellenberger and Krishna, 2002; Krishna and Ellenberger, 2002a, 2003). Further details of the experimental set-up, including photographs of the rig may be found on our website: *http://ct-cr4.chem.uva.nl/vibrationexciter.*

With the 12-capillary gas inlet device, two physical quantities, the gas holdup and the volumetric mass transfer coefficient k_La, have been measured for varying conditions of vibration frequency, vibration amplitude, and the superficial gas velocity in the column. All the measurements have been carried out at room temperature with air as the gas phase and demineralised water as the liquid phase, containing varying concentration of solids. The solid phase used consisted of porous silica particles whose properties were determined to be: skeleton density = 2100 kg/m^3; pore volume = 1.05 mL/g; particle size distribution, d_p: 10% < 27 μm; 50% < 38 μm; 90% < 47 μm. The solids concentration ε_s, is expressed as the volume fraction of solids in gas free slurry. The pore volume of the particles (liquid filled during operation) is counted as being part of the solid phase. The pressure at the top of the column is atmospheric.

The gas holdup ε is measured by visually recording the dispersion height H above the gas distributor. The volumetric mass transfer coefficient k_La is measured by means of the dynamic oxygen absorption method, described in earlier work (Ellenberger and Krishna, 2002;

Author to whom correspondence may be addressed. E-mail address: krishna@ science.uva.nl

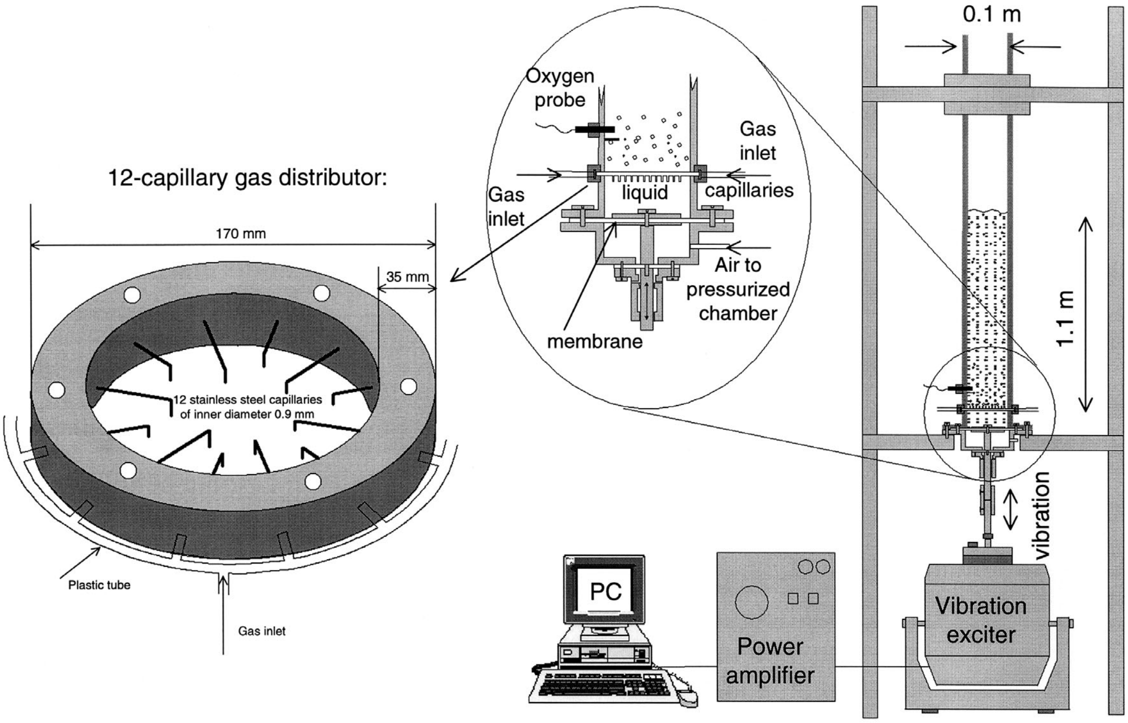


Figure 1. Experimental set-up of the bubble column with vibration excitement device. Further details of the experimental set-up, including photographs of the rig are to be found on our website: *http://ct-cr4.chem.uva.nl/vibrationexciter.*

Krishna and Ellenberger, 2002a), and the reader is referred to these papers for details of the experimental procedure. Assuming the liquid phase to be perfectly mixed, the dissolved-oxygen concentration is described by the relation:

$$\varepsilon_L \frac{dC}{dt} = k_L a\left(C^* - C\right) \tag{1}$$

where a is the interfacial area per unit volume of dispersion, k_La is the volumetric mass transfer coefficient, C is the dissolved-oxygen concentration in the liquid bulk and t represents the time. Equation (2) can be integrated with the following initial condition: $t = 0$, $C = 0$, to obtain:

$$\frac{C}{C^*} = 1 - \exp\left(-k_L a \varepsilon_L t\right) \tag{2}$$

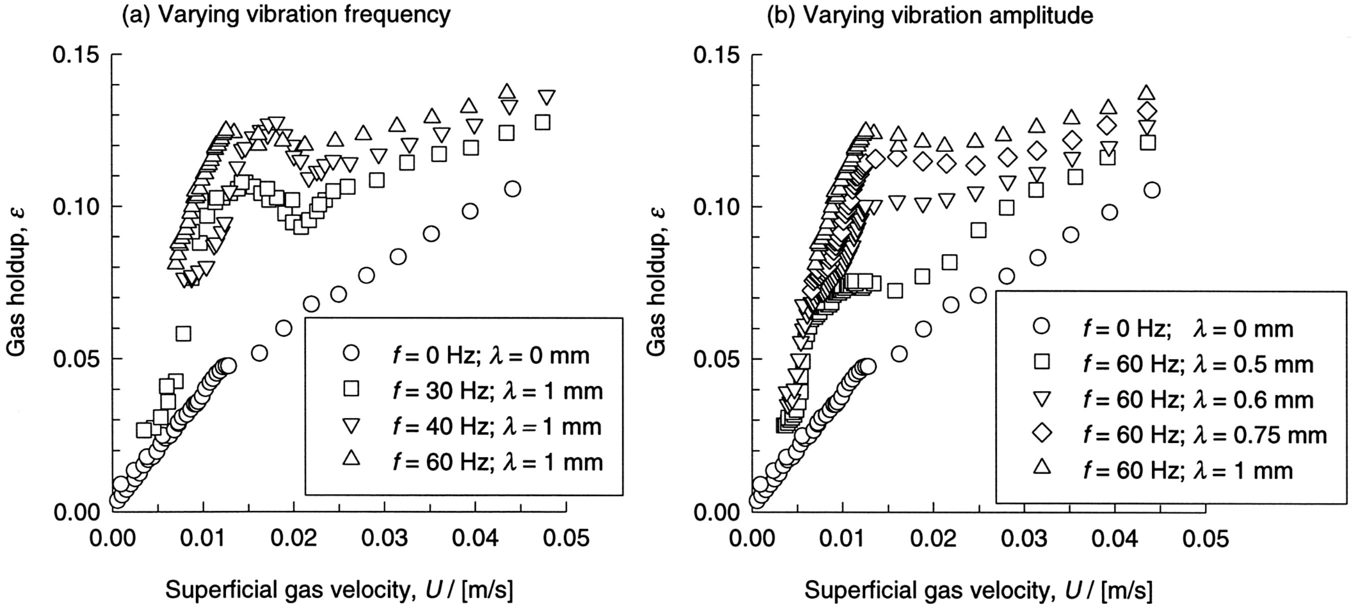


Figure 2. Gas holdup, with and without vibrations. (a) Influence of vibration frequency, (b) Influence of vibration amplitude.

The only unknown constant in Equation (4) is k_La, which was determined by fitting the measured dynamic response.

Gas Holdup in Air-water System

Before considering the measurements with slurries, let us consider gas holdup with the air-water system. For a range of superficial gas velocities, U, the gas holdup was measured for the no-vibrations case, along with three cases in which the vibration frequencies were set at 30, 40 and 60 Hz at a constant amplitude λ = 1 mm and clear liquid height H_0 = 1.10 m; the results are shown in Figure 2 (a). Depending on the operating conditions, the improvement in the gas holdup is in the 100 – 300% range. The higher the vibration frequency, the higher is the gas holdup. A similar picture emerges when the vibration frequency is held constant at 60 Hz and the vibration amplitude λ is set at values of 0.5, 0.6, 0.75 and 1 mm; see Figure 2 (b). The higher the vibration amplitude, the higher is the gas holdup.

In order to gain further insight into the reasons behind the increase of gas holdup due to application of the vibrations to the liquid phase, we adopt the drift-flux analysis of Wallis (1969) for the holdup experiments. The Wallis drift-flux is defined as $V_{slip}\varepsilon(1-\varepsilon)$, where V_{slip} is the slip velocity between the gas and liquid phases. For a bubble column with no net liquid flow, $V_{slip} = U/\varepsilon$. The Richardson and Zaki (1954) expression for the slip velocity is given by:

$$V_{slip} = V_{b0}(1-\varepsilon)^{n-1} \tag{3}$$

where V_{b0} is the single-bubble rise velocity and n is the Richardson and Zaki index. The factor $(1-\varepsilon)^{n-1}$ in Equation (2) describes the hindering effect of the rising bubble swarm. In Figure 3, we plot $V_{slip}\varepsilon(1-\varepsilon) \equiv U(1-\varepsilon)$ against ε for the set of experiments shown in Figure 2. Also shown by continuous lines in Figure 3 are calculations using Equation (2), indicated by the continuous lines drawn taking the Richardson and Zaki index n = 2 (a typical value for air-water systems) and a fitted value for V_{b0} for the data points with

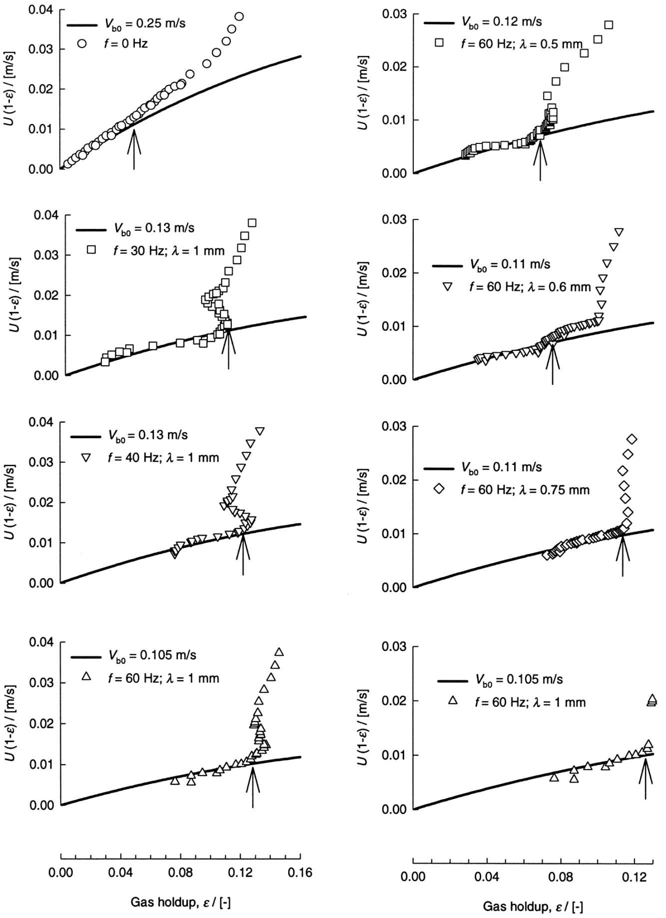


Figure 3. Wallis plot for the set of holdup measurements shown in Figure 2.

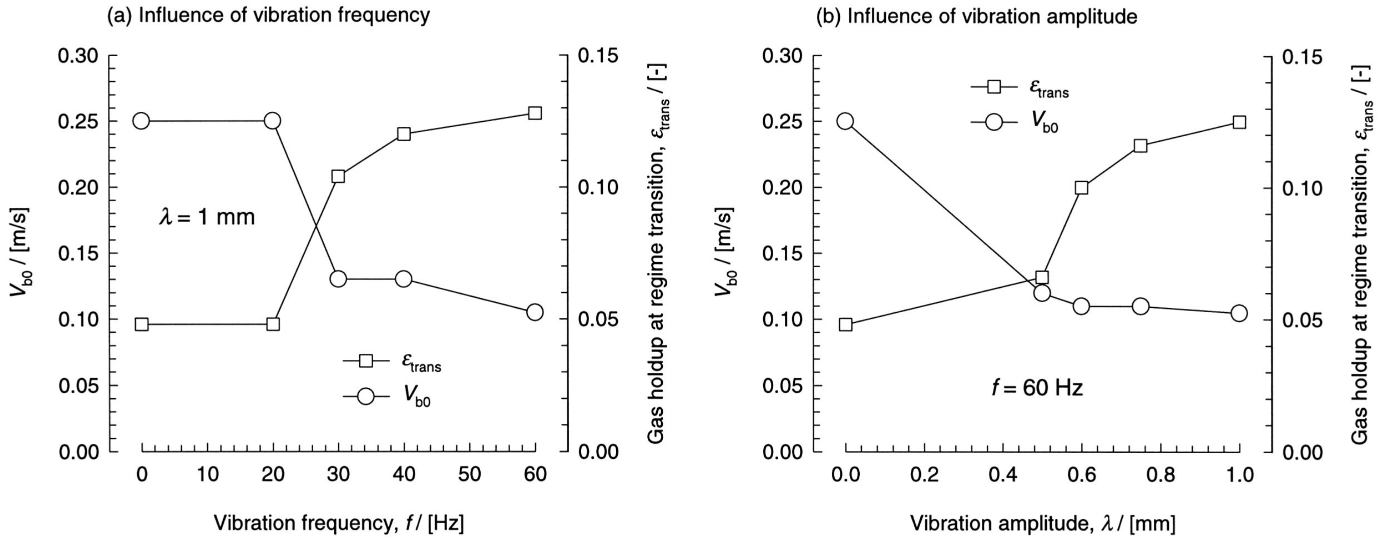
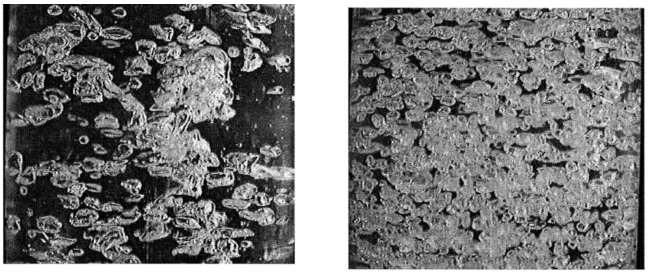


Figure 4. Variation of ε_{trans} and V_{b0} with the vibration amplitude frequency and amplitude.



(a) $U = 7.4$ mm/s;
no vibration

(b) $U = 7.4$ mm/s;
$f = 60$ Hz;
$\lambda = 0.5$ mm

Figure 5. Snapshots of the column operation with 12-capillary gas distributor operating at a superficial gas velocity $U = 7.4$ mm/s and liquid column height $H_0 = 1.10$ m. (a) Column operating without vibration excitement, (b) Operation with vibration $f = 60$ Hz and $\lambda = 0.5$ mm.

low gas holdups. The point of departure of the experimental data points from Equation (2) signifies the regime transition point (holdup ε_{trans}, superficial gas velocity U_{trans}). The regime transition holdup ε_{trans} is plotted in Figure 4 as a function of the (a) vibration frequency and (b) amplitude. It is clear that application of vibrations tends to delay the transition to the heterogeneous flow regime. A snapshot of the column operation at $U = 7.4$ mm/s, without application of vibration, is shown in Figure 5 (a). The operation is clearly in the heterogeneous flow regime, as is evidenced by the presence of a few large bubbles. The corresponding snapshot for column operation with a vibration frequency $f = 60$ Hz and amplitude $\lambda = 0.5$ mm is shown in Figure 5 (b). The bubbles are significantly smaller in size and the dispersion is uniform.

The fitted value of V_{b0}, the single-bubble rise velocity, is plotted as a function of vibration (a) frequency, and (b) amplitude in Figure 4. For the no-vibrations case, the value of $V_{b0} = 0.25$ m/s which is a typical value for air-water systems with bubbles in the 3-7 mm size range. The value of V_{b0} reduces to about

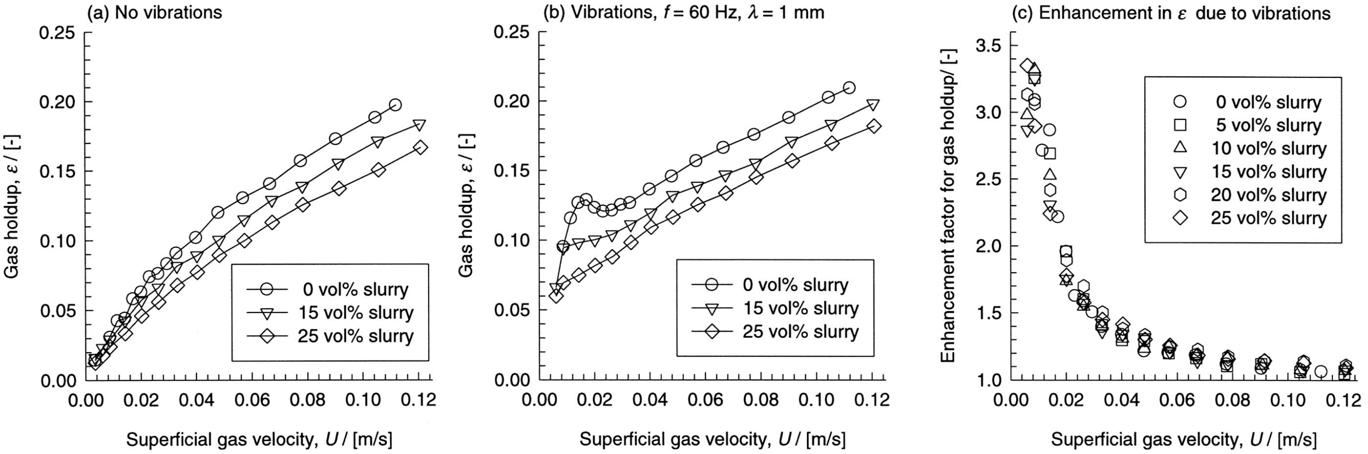


Figure 6. Influence of vibrations on gas holdup with slurries of varying concentration.

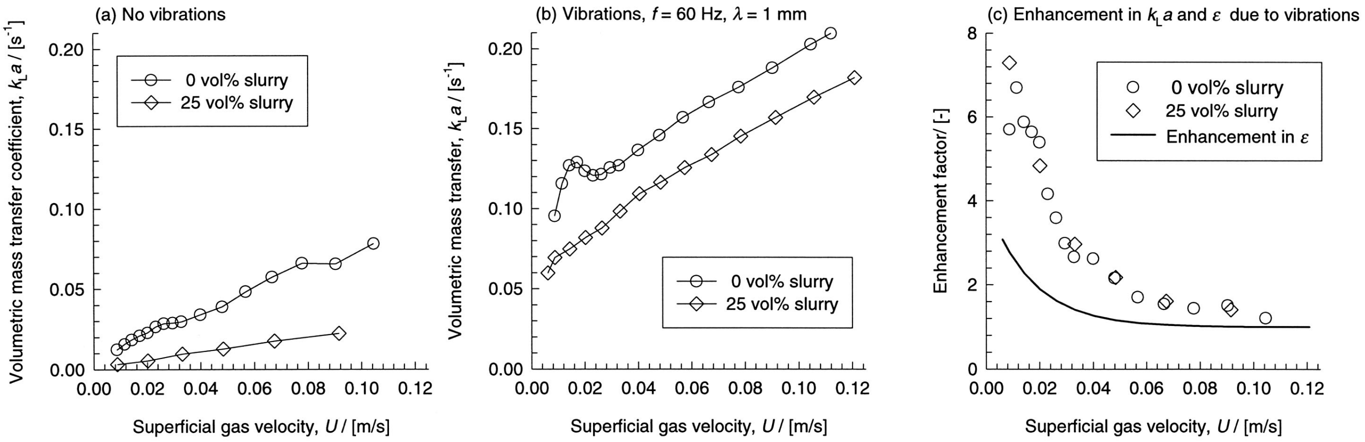


Figure 7. Influence of vibrations on k_La with slurries of varying concentration.

0.11 m/s on application of vibration. This significant reduction in the rise velocity of the bubble cannot be attributed to the reduction in the bubble size. For air-water systems, the single bubble rise velocity is practically independent of the bubble size for d_b values in the 3 – 7 mm size range (Clift et al., 1978; Krishna et al., 1999). Therfore, the conclusion is that the vibrations to the liquid phase have the effect of reducing the rise velocity of the bubble swarm due to the creation of standing waves.

Gas Holdup and Mass Transfer in Slurries

From the foregoing campaign we conclude the maximum improvement in gas holdup is obtained when the vibration amplitude is set at $\lambda = 1$ mm and the vibration frequency is adjusted to $f = 60$ Hz; with these vibration parameters we measured the gas holdup in water slurries containing varying concentration of solids (up to 25 vol%); the results are presented in Figure 6. Let us first consider the no-vibrations case (see Figure 6 (a)): increasing slurry concentration tends to decrease the gas holdup. This is due to the enchanced coalescence caused by the presence of the silica particles. With vibrations ($\lambda = 1$ mm; $f = 60$ Hz), a similar trend is observed with increasing slurry concentrations but the holdup values are significantly higher than for the corresponding no-vibrations case. The enhancement factor for gas holdup due to vibrations, defined as the ratio of the gas holdup with and without vibrations, is shown in Figure 6 (c) for a range of slurry concentrations. It is interesting to note that enhancement factor is practically independent of the slurry concentration. We note also that the enhancement is highest at low superficial gas velocities and decreases sharply with increasing values of U.

We carried out a series of mass transfer experiments for a range of superficial gas velocities for two situations: (i) no vibrations, and (ii) vibrations $f = 60$ Hz and $\lambda = 1$ mm. The k_La values for these two sets, for 0 and 25 vol% slurries, are shown in Figure 7 (a) and (b). The enhancement in k_La due to vibrations is plotted in Figure 7 (c). It is interesting to note that the enhancement in k_La is significantly higher than the corresponding enhancement in the gas holdup. This result suggests that the k_La improvement on application of vibrations is a consequence of significantly smaller bubble sizes (see also Figure 5).

Conclusions

The following major conclusions can be drawn from our study:

a. Low-frequency vibrations, in the 30 – 60 Hz range, are capable of causing a significant increase in the gas holdup; see Figure 2.
b. Application of vibrations tends to delay the transition to the churn-turbulent flow regime; see Figure 3.
c. The increase in the gas holdup is largely to be attributed to the reduction in the rise velocity of the bubble swarm, due to the generation of standing waves
d. For the experiments with varying slurry concentrations, the enhancement in gas holdup and k_La values are practically independent of slurry concentration. The enhancement decreases strongly with increasing superficial gas velocity.
e. It is concluded that application of low-frequency vibrations has the potential of improving the gas-liquid contacting in bubble columns.

Acknowledgement

This research was supported by a grant from the Netherlands Foundation for Scientific Research (NWO) for development of novel concepts in reactive separations technology.

Nomenclature

a	interfacial area per unit volume of dispersion, (m^2 m^{-3})
C	oxygen concentration in the liquid phase, arbitrary units
C^*	saturation concentration of oxygen in liquid, arbitrary units
d_b	diameter of bubble, (m)
d_p	diameter of particle, (m)
f	vibration frequency, (Hz)
H	height of dispersion, (m)
k_L	mass transfer coefficient in liquid phase, (m/s)
t	time, (s)
U	superficial gas velocity, (m s^{-1})
U_{trans}	superficial gas velocity at regime transition, (m s^{-1})
V_{b0}	single bubble rise velocity, (m s^{-1})

Greek Symbols

ε	total gas hold-up
ε_L	liquid hold-up
λ	vibration amplitude, (m)

Subscripts

0	referring to initial conditions
trans	referring to regime transition point
G	referring to gas
L	referring to liquid

References

Clift, R., J.R. Grace and M.E. Weber, "Bubbles, Drops and Particles", Academic Press, San Diego, CA (1978).

De Swart, J.W.A., R.E. van Vliet and R. Krishna, "Size, Structure and Dynamics of ''Large'' Bubbles in a Two-dimensional Slurry Bubble Column", Chem. Eng. Sci. **51**, 4619–4629 (1996).

Deckwer, W.D., "Bubble Column Reactors", John Wiley, New York, NY (1992).

Ellenberger, J. and R. Krishna, "Improving Mass Transfer in Gas-liquid Dispersions by Vibration Excitement", Chem. Eng. Sci. **57**, 4809–4815 (2002).

Krishna, R. and J. Ellenberger, "Improving Gas–liquid Contacting in Bubble Columns by Vibration Excitement", Int. J. Multiphase Flow **28**, 1223–1234 (2002a).

Krishna, R. and J. Ellenberger, "Improving Gas-liquid Mass Transfer in Bubble Columns by Applying Low-frequency Vibrations", Chem. Eng. Technol. **25**, 159–162 (2002b).

Krishna, R. and J. Ellenberger, "Influence of Low-frequency Vibrations on Bubble and Drop Sizes Formed at a Single Orifice", Chem. Eng. Process. **42**, 15–21 (2003).

Krishna, R., J. Ellenberger, M.I. Urseanu and F.J. Keil, "Utilisation of Bubble Resonance Phenomena to Improve Gas–liquid Contact", Naturwissenschaften **87**, 455–459 (2000).

Krishna, R., M.I. Urseanu, J.M. van Baten and J. Ellenberger, "Wall Effects on the Rise of Single Gas Bubbles in Liquids", Int. Commun. Heat Mass Transf. **26**, 781–790 (1999).

Richardson, J.F. and W.N. Zaki, "Sedimentation and Fluidization. I", Trans. Inst. Chem. Engrs. (London) **32**, 35–53 (1954).

Wallis, G.B., "One-dimensional Two-phase Flow", McGraw-Hill, New York, NY (1969).

Manuscript received September, 2002; revised manuscript received February 10, 2003; accepted for publication February 25, 2003.