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Shaken, not stirred, bubble column reactors: Enhancement of mass transfer by vibration excitement

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Abstract

In this paper we discuss the advantages of a bubble column reactor, wherein the liquid phase is subjected to low-frequency vibrations. A special device, called a vibration exciter, mounted at the bottom of the bubble column, transmits the vibration to the liquid phase by means of a piston. Both the amplitude of the vibration, and its frequency can be adjusted quite accurately. We show that application of low-frequency vibrations, in the 40–120 Hz range, to the liquid phase of an air–water bubble column causes significantly smaller bubbles to be generated at the nozzle. In experiments with a single capillary nozzle the bubble size is reduced by 40–50% depending on the gas flow rate. Using a 12-capillary nozzle arrangement, the gas holdup, ε, and the volumetric mass transfer coefficient, $k_L a$, were measured for a range of superficial gas velocities. Application of vibrations to the liquid phase leads to enhancement in ε and $k_L a$ values by a factor of two or more. 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 homogeneous bubbly flow regime and delay the onset of the churn-turbulent flow regime.

It is concluded that application of low-frequency vibration has the potential of improving the gas–liquid contacting in bubble columns.

Keywords: Bubble column; Vibration excitement; Process intensification; Gas holdup; Mass transfer; Standing waves

1. Introduction

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 $U = U_{\text{trans}}$, 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 to 50 mm depending on the system properties (De Swart, van Vliet, & Krishna, 1996). The wide distribution of bubble sizes causes a wide gas phase residence time distribution; this is often detrimental to reactor conversion and selectivity.

Uniformity of bubble sizes in gas–liquid reactors can be achieved by means of mechanical agitation or *stirring*. However, the stirring action of the agitator causes intense backmixing of both liquid and gas phases. In some biotechnological applications the high shear, experienced near the agitator, is undesirable. The major objective of the present communication is to demonstrate the advantages of a *shaken* or vibrated bubble column reactor in which the liquid phase is subjected to low-frequency vibrations in the 40–120 Hz range. A special vibration excitement device is used for this purpose. The advantage of using low-frequency vibrations over mechanical agitation is that the plug flow character of the bubbles is maintained and no additional large-scale backmixing of the liquid phase is induced.

There is some evidence in the published literature to show that the application of vibrations to the liquid phase, at frequencies of the order of 100 Hz, can (a) influence bubble rise in gas–liquid dispersions (Jameson, 1966; Jameson & Davidson, 1966), (b) reduce the bubble size (Grinis & Monin, 1999; Krishna, Ellenberger, Urseanu, & Keil, 2000) and (c) improve gas–liquid mass transfer (Baird, 1963; Bartsch, 1995; Harbaum & Houghton, 1960; Krishna & Ellenberger, 2002). Vibrations have found to have a

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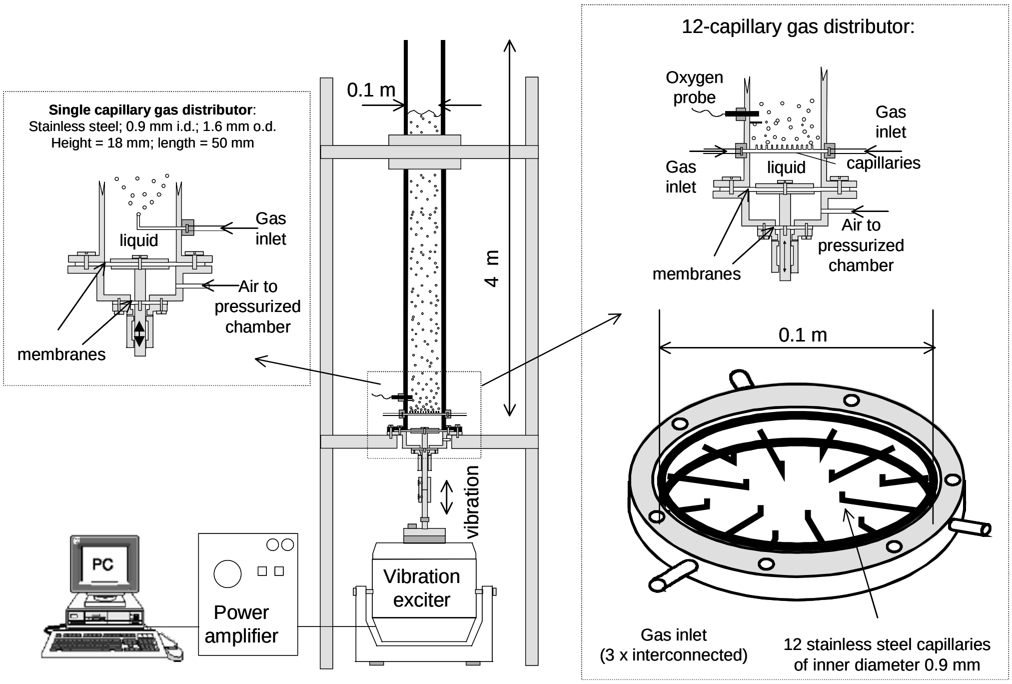


Fig. 1. Experimental set-up of the bubble column with vibration excitement device. Further details of the experimental set-up including photographs of the rig are to be found on our website: http://ct-cr4.chem.uva.nl/vibrationexciter.

beneficial effect in gas–solids fluidization (Herrera, Levy, & Ochs, 2002; Noda, Mawatari, & Uchida, 1998). Our present work extends the published works in bubble columns to high superficial gas velocities in order to show that application of vibrations helps to stabilize the homogeneous bubble flow regime and results in a more uniform dispersion of smaller sized bubbles.

2. 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 Fig. 1. The bubble column, made of polyacrylate, has an inner diameter of 0.10 m and a height of 2.0 m. The bottom of the column is sealed by a silicon rubber membrane of 0.4 mm thickness and clamped between two metal disks of 0.096 m in diameter; see inset to Fig. 1. At a distance of 0.1 m above the membrane, air is fed to the bubble column through two types of gas distributors: (a) a single capillary of 0.9 mm i.d. (see inset on left of Fig. 1), and (b) 12 stainless steel capillaries of 0.9 mm i.d. (see inset on the right of Fig. 1). The gas flow is controlled by means of a calibrated flowmeter (*Brooks*). In order to hold the membrane at constant vertical position after filling the column with the liquid phase, a chamber for pressure compensation is mounted below the membrane. The membrane is connected to an air-cooled vibration exciter (*TIRAvib 5220*, Germany). The amplifier of this vibration-exciter is controlled by the *SignalCalc* 550 *Vibration-controller* in a PC environment. The frequency range is 10–5000 Hz. Depending on the frequency the amplitude can be varied between 0 and 25 mm. The vibrations follow a sinusoidal motion. For mass transfer measurements an oxygen electrode (*Yellow Springs Incorporated*) is placed in the bubble column at a distance of 0.1 m above the gas distributor in such a way that the gas bubbles did not hit the electrode; see upper right inset to Fig. 1. 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.

With the single capillary gas inlet device, bubble sizes were measured for a range of vibration frequencies, and inlet gas velocities. At each vibration frequency, video recordings, using a Panasonic DSP colour CCD camera, of the air–water dispersion were made at 25 frames per second for a period of 5 s. Frame-by-frame analysis of the video images, gives accurate information on the number of bubbles passing through the observation window in the time interval of the observation (5 s). The video imaging technique is the same as that described in an earlier publication (Krishna et al., 2000). For the set volumetric flow rate of the dispersed phase, the average air bubble diameter of the dispersion can be calculated.

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

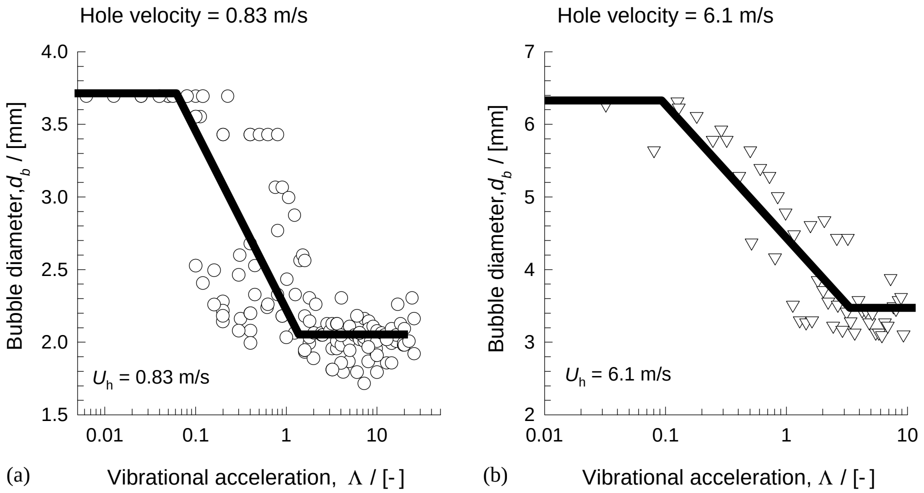


Fig. 2. Influence of vibrational acceleration $\Lambda = \lambda(2\pi f)^2/g$ on the bubble diameter for hole velocity (a) $U_h = 0.83$ m/s and (b) $U_h = 6.1$ m/s.

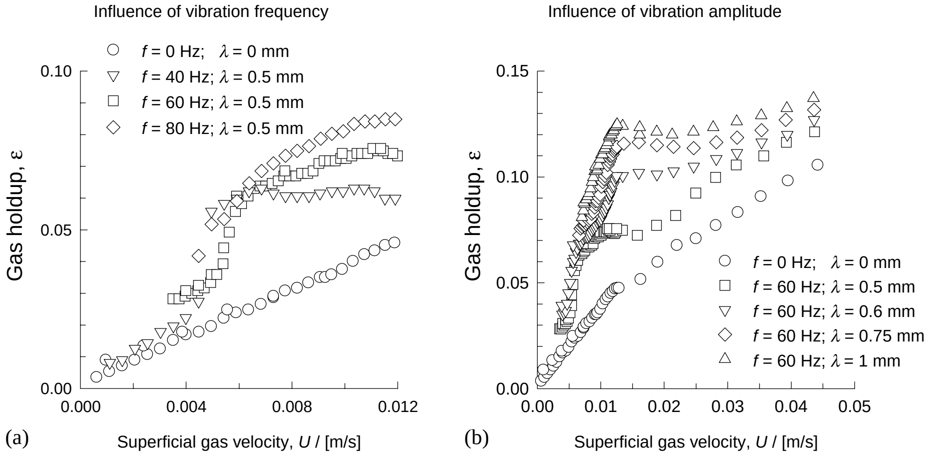


Fig. 3. Gas holdup, with and without vibrations. (a) Influence of vibration frequency, (b) influence of vibration amplitude.

demineralized water as the liquid phase. The pressure at the top of the column is atmospheric. All experiments reported here have been carried out with the liquid in batch mode.

The gas holdup ε is measured by visually recording the dispersion height H above the gas distributor; the gas holdup is then calculated from

$$\varepsilon = 1 - \frac{H_0}{H}, \tag{1}$$

where H_0 is the height of the ungassed liquid in the column.

The volumetric mass transfer coefficient k_La is measured by means of the dynamic oxygen absorption method, described in earlier work from our group (Krishna & Ellenberger, 2002; Letzel, Schouten, Krishna, & van den Bleek, 1999). The dissolved oxygen concentration is measured by means of an oxygen electrode, which is connected to a microammeter. The signal of the oxygen electrode is directly proportional to the oxygen concentration C in the liquid phase. By fitting the transient oxygen uptake profile, the volumetric mass transfer coefficient k_La can be determined.

3. Bubble size measurements with single capillary distributor

In the first set of experiments, the air flow rate through the single capillary was maintained at 5.3×10^{-7} m^3/s which corresponds to a hole velocity in the capillary, $U_h = 0.83$ m/s and bubble sizes were measured for a range of frequencies, $f = 20$–160 Hz, and vibration amplitudes, $\lambda = 0.0025$–0.32 mm. The measured bubble sizes are plotted in Fig. 2(a) as a function of the vibrational acceleration which is a dimensionless quantity defined by $\Lambda = \lambda(2\pi f)^2/g$ where $g = 9.81$ m/s^2 is the gravitational acceleration. Below a value $\Lambda = 0.1$, no reduction in bubble size is observed and the value of $d_b = 3.4$ mm corresponds to the no-vibrations case. When Λ is increased beyond 0.1, the bubble sizes tend to be reduced with increasing Λ. For $\Lambda > 1$, no further significant bubble size reduction is observed. At a hole velocity of $U_h = 0.83$ m/s the maximum reduction in bubble size is from 3.4 to 2 mm, i.e. 40%. A similar picture emerges at higher hole velocities. Measurements of bubble sizes for $U_h = 6.1$ m/s are shown in Fig. 2(b). We note that the

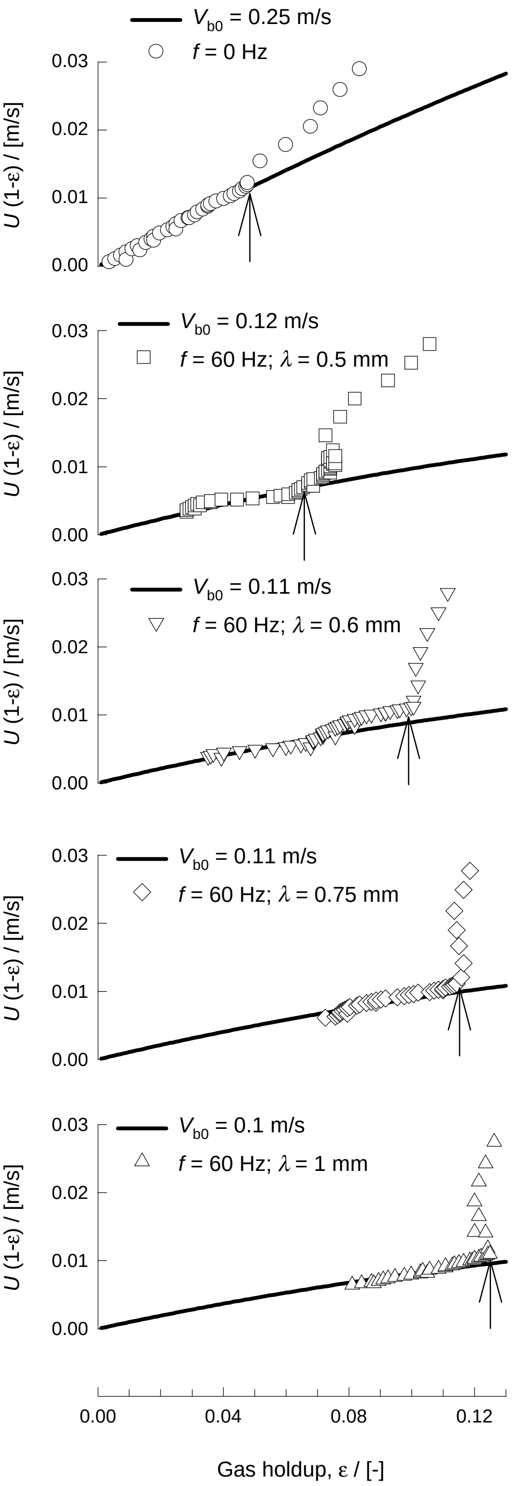


Fig. 4. Wallis plot for the set of holdup measurements shown in Fig. 3(b).

bubble sizes reduce by about 50% from a value of 6.4 mm to about 3 mm for $\Lambda > 1$. The physical explanation of the above observations is that application of vibrations to the liquid phase serves to overcome the surface tension forces and assist the break-up of the bubbles formed at the orifice (Grinis & Monin, 1999).

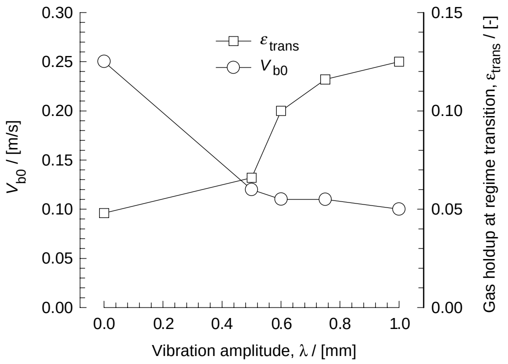


Fig. 5. Variation of $\varepsilon_{\text{trans}}$ and V_{b0} with the vibration amplitude λ.

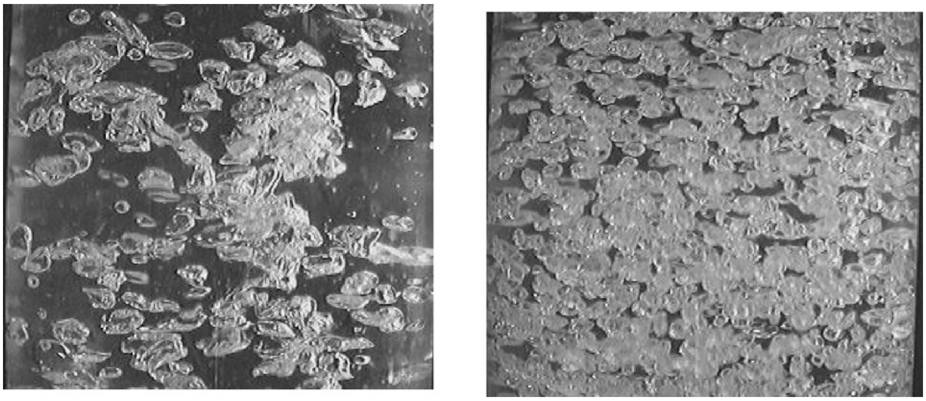
4. Gas holdup measurements with 12-capillary distributor

We now consider the influence of vibrations on the gas holdup in the 0.1 m diameter column using the 12-capillary gas distributor device. 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 40, 60 and 80 Hz at a constant amplitude $\lambda = 0.5$ mm and clear liquid height $H_0 = 1.10$ m; the results are shown in Fig. 3(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 Fig. 3(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 $V_{\text{slip}}\,\varepsilon(1-\varepsilon)$ where V_{slip} is the slip velocity between the gas and liquid phases. For a bubble column with no net liquid flow, $V_{\text{slip}} = U/\varepsilon$. The Richardson and Zaki (1954) expression for the slip velocity is given by

$$V_{\text{slip}} = V_{b0}(1-\varepsilon)^{n-1}, \tag{2}$$

where V_{b0} is the single-bubble rise velocity and n is the Richardson and Zaki index. The factor $(1-\varepsilon)^{n-1}$ in Eq. (2) describes the hindering effect of the rising bubble swarm. In Fig. 4, we plot $V_{\text{slip}}\varepsilon(1-\varepsilon) \equiv U(1-\varepsilon)$ against ε for the set of experiments shown in Fig. 3(b) with varying vibration amplitudes. Also shown by continuous lines in Fig. 4 are calculations using Eq. (2), shown 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 low gas holdups, below the regime transition point. The point of departure of the experimental data points from Eq. (2) signifies the regime transition point (holdup $\varepsilon_{\text{trans}}$, superficial gas velocity U_{trans}). The regime transition holdup $\varepsilon_{\text{trans}}$ is plotted in Fig. 5 as a function of



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

(b) U = 7.4 mm/s; f = 60 Hz; λ= 0.5 mm

Fig. 6. 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.

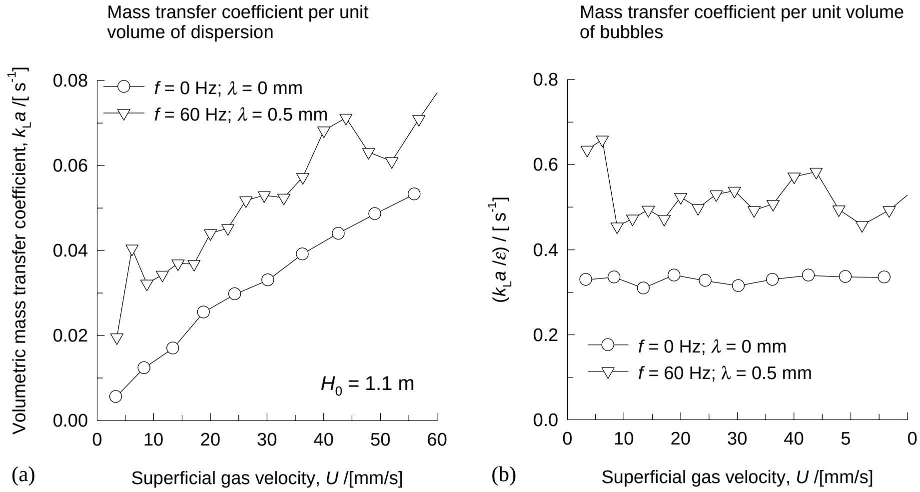


Fig. 7. (a) Volumetric mass transfer coefficient $k_L a$, with and without vibrations, as function of the superficial gas velocity. (b) $k_L a/\varepsilon$ as function of the superficial gas velocity.

the vibration 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 Fig. 6(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 Fig. 6(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 against λ in Fig. 5. 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 ≈ 0.11 m/s on application of vibration. This significant reduction in the rise velocity of the bubble size cannot be attributed to the reduction in the bubble size (evidenced in the single capillary experiments in Fig. 2). 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, Grace, & Weber, 1978; Krishna, Urseanu, van Baten, & Ellenberger, 1999). The inescapable conclusion, therefore, 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.

5. Mass transfer measurements with 12-capillary distributor

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 $\lambda = 0.5$ mm. The k_La values for these two sets are shown in Fig. 7(a). In consonance with the hold-up measurements shown in Fig. 3, we find that the k_La values are improved by about factor of 2 due to vibration excitement. The corresponding values of the volumetric mass transfer coefficient per unit volume of the dispersed bubbles, k_La/ε, are plotted in Fig. 7(b). The values of k_La/ε with vibrations are higher than the corresponding no-vibrations case. This result suggests that the k_La improvement on application of vibrations is a consequence not only of the increased interfacial area a, but also due to a higher value of k, caused by increased turbulence at the gas–liquid interface.

6. Conclusions

The following major conclusions can be drawn from our study:

(a) Low-frequency vibrations, in the 40–120 Hz range, are capable of causing a 40–50% reduction in the bubble sizes formed at a single capillary.
(b) Compared to the no-vibrations case the gas holdup increases by a factor of about 2; see Fig. 3.
(c) Application of vibrations tends to delay the transition to the churn-turbulent flow regime; see Fig. 5.
(d) 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
(e) The increase in the k_La values is in consonance with the increase in the gas holdup values; see Fig. 7.

It is concluded that application of low-frequency vibrations has the potential of improving the gas–liquid contacting in bubble columns.

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