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Improving mass transfer in gas–liquid dispersions by vibration excitement

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Abstract

The objective of our work is to demonstrate that significant enhancement of gas–liquid mass transfer in bubble columns can be achieved by subjecting the liquid phase to low-frequency vibrations. A special device, called a vibration exciter, is mounted at the bottom of the bubble column. The vibration is transmitted 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_La, were measured for a range of superficial gas velocities. Application of vibrations to the liquid phase leads to enhancement in ε and k_La values of up to 400%. A careful study of the influence of vibration frequency, vibration amplitude and column height reveals that subtle resonance phenomena are at play.

It is concluded that application of low-frequency vibration has the potential of improving the gas–liquid contacting in bubble columns.
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Keywords: Bubble column; Vibration excitement; Process intensification; Gas holdup; Mass transfer

1. Introduction

A bubble column reactor is a commonly used reactor type. Increase in the gas–liquid mass transfer coefficient often has a beneficial effect on the reactor performance. One common method for improving the mass transfer is by carrying out the reaction in a stirred vessel. The energy input from the stirrer device serves to break up the bubbles, leading to an increased contact area between gas and liquid. There are two important disadvantages of stirred vessels. The first relates to the large energy inputs required to achieve the desired improvement in interfacial transfer rates. The second disadvantage stems from the fact that stirring causes the gas bubbles to be re-circulated in the vessel. Bubble re-circulation is undesirable because the overall driving force for mass transfer is reduced. It is preferable to have piston flow of gas bubbles up through the liquid column, with little or no backmixing of the gas bubbles. Many aerobic fermentation processes (e.g. in beer manufacture) are carried out in stirred vessels. Some bacterial cells suffer death when the agitation is too severe. There is therefore a need to improve contacting without intense agitation.

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 & Davidson, 1966; Jameson, 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; Lemcoff & Jameson, 1975a, b). There is some indication from the patent literature (Kuesgen, Fieg, & Bartsch, 1999) of the use of vibration devices in high pressure reactors, presumably to intensify gas–liquid reactor operation. Pulsations and periodic operations (Gupta, Patel, & Ackerberg, 1982; Garcia-Anton, Perez-Herranz, & Guinon, 1997; Guinon, Perez-Herranz, Garcia-Anton, & Lacoste, 1995; Lemcoff & Jameson, 1975a, b; Gabarain, Castellari, Cechini, Tobolski & Havre, 1997) have been shown to improve mass transfer between solids and liquids.

The advantage of using low-frequency vibrations over mechanical agitation is that the plug flow character of the

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bubbles is maintained and no additional large-scale backmixing of the liquid phase is induced. Our objective in this paper is to undertake a systematic study of the influence of vibration frequency and amplitude on bubble size, gas hold-up and gas–liquid mass transfer in bubble columns and to show that significant improvement in gas–liquid contact can be achieved provided the operation conditions are properly tuned. A special vibration excitement device is used for this purpose.

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 4.0 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 of 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) twelve stainless steel capillaries of 0.9 mm i.d. (see inset on the right of Fig. 1). Stainless steel high-pressure pre-filters with pores of 2 μm are placed at the inlet of the capillaries resulting in a high pressure-drop restriction between the gas supply annulus and the capillaries, which in turn results in a constant gas flow. 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 0–25 mm. The vibrations follow a sinusoidal motion. The maximum acceleration under unloaded conditions is 700 m s^{-2}. 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. 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.

All the measurements have been carried out at room temperature with air as the gas phase and demineralised water as

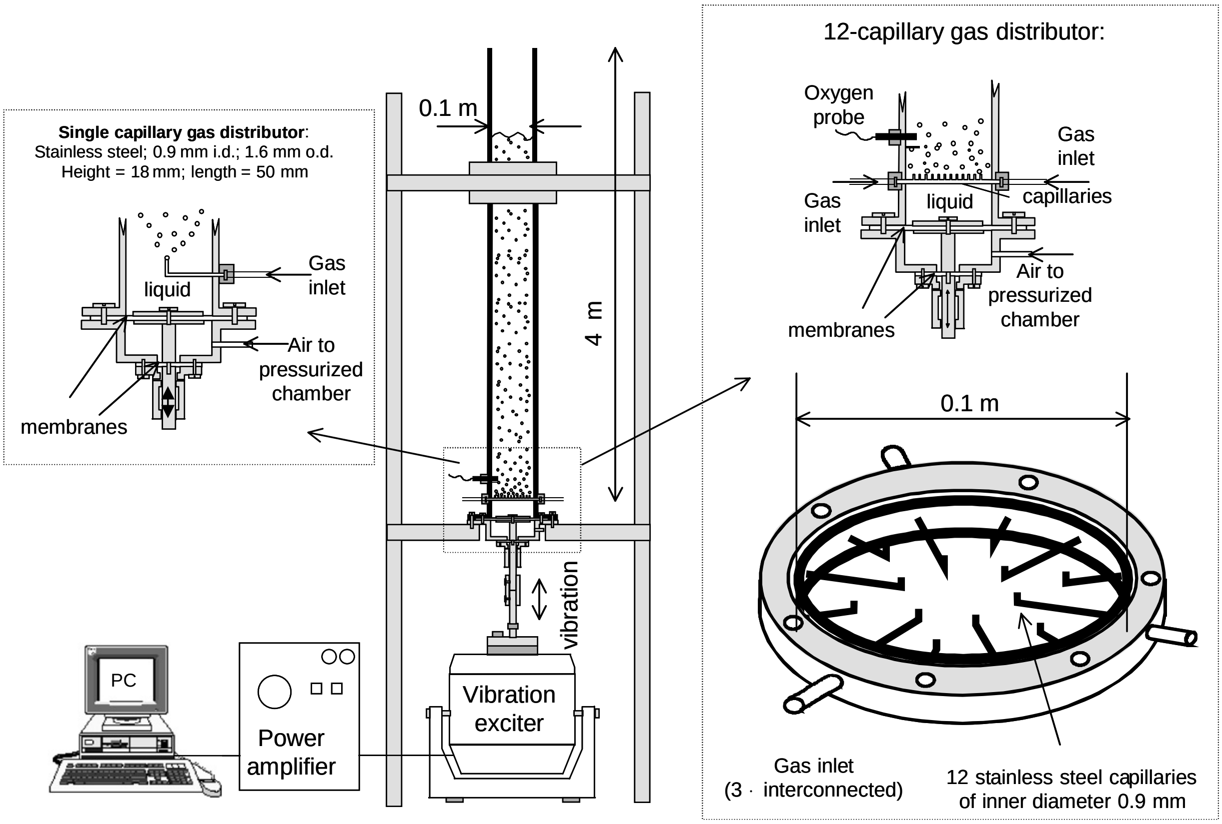


Fig. 1. Experimental set-up of the bubble column with vibration excitement device. Further details of the experimental set-up including photographs of the rig are found on our website: http://ct-cr4.chem.uva.nl/vibrationexciter.

the liquid phase. 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 (De Swart, van Vliet, & Krishna, 1996). For the set volumetric flow rate of the dispersed phase, the average air bubble diameter of the dispersion can be calculated. Additionally, we used a FASTCAM Ultima high-speed camera 40K (Roper Scientific MASD, Inc., San Diego, California, USA) at a frame rate of 2250 frames per second to examine the influence of vibrations on bubble formation in some more detail. The video recording using FASTCAM have been placed on our website: http://ct-cr4.chem.uva.nl/vibrationvideo.

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, liquid height, and the superficial gas velocity. The pressure at the top of the column is atmospheric. The liquid load in the bubble column acts on the vibration exciter and reduces the maximum acceleration to approximately 200 m s^{-2}.

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 (Letzel, Schouten, van den Bleek, & Krishna, 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.

Firstly, the oxygen was stripped from water almost completely by injection of pure nitrogen through the distributor; this took about 15 min. The oxygen concentration in the liquid phase is C_0. When all the oxygen has been stripped out, air was sparged into the column and the oxygen uptake into the liquid phase was monitored continuously by means of the oxygen sensor. The total measurement time was chosen large enough in order to reach the oxygen saturation concentration, C^*.

Assuming the liquid phase to be perfectly mixed, the dissolved-oxygen concentration is described by the relation

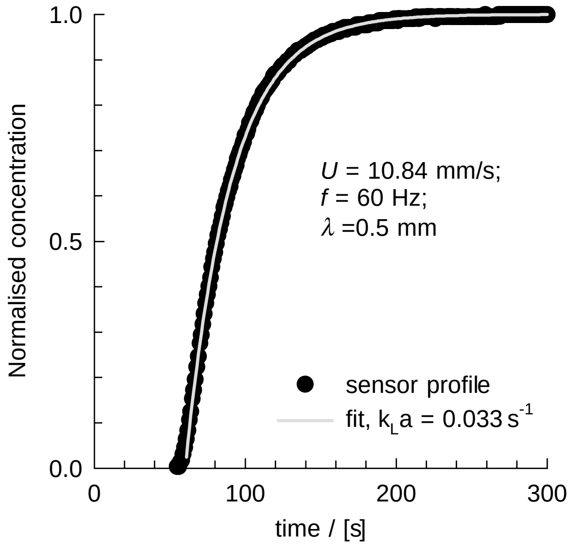


Fig. 2. Example of determination of mass transfer coefficient measurement together with a least-square fit of Eq. (4). $H_0 = 1.10$ m; $U = 10.84$ mm/s; $f = 60$ Hz and $\lambda = 0.5$ mm.

$$(1-\varepsilon)\frac{\mathrm{d}C}{\mathrm{d}t} = k_La(C^* - C), \tag{2}$$

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. Eq. (2) can be integrated with the following initial and final conditions:

$$t = 0: \quad C = C_0,$$

$$t \to \infty: \quad C = C^* \tag{3}$$

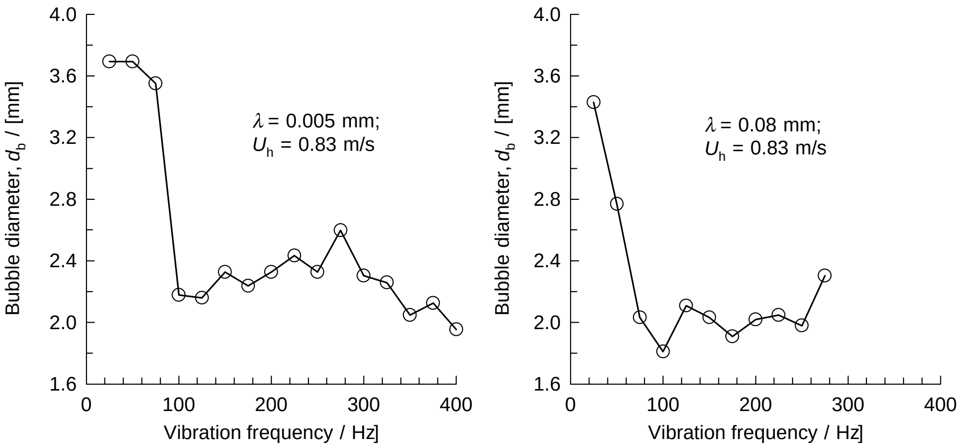
to obtain

$$\frac{C - C_0}{C^* - C_0} = 1 - \exp(-k_Lat). \tag{4}$$

The only unknown constant in Eq. (4) is k_La, which was determined using the regression solver routine in Microsoft Excel in which the mean square deviation between the experiments and the model predictions is minimised. Fig. 2 shows an example of the normalised output signal of the oxygen probe at a superficial gas velocity $U = 10.84$ mm s^{-1}, a vibration frequency $f = 60$ Hz and an amplitude $\lambda = 0.5$ mm. Also shown in Fig. 2 is the curve fitted with Eq. (4) taking $k_La = 0.033$ s^{-1}. We note that the model fits the experimental sensor profile very well.

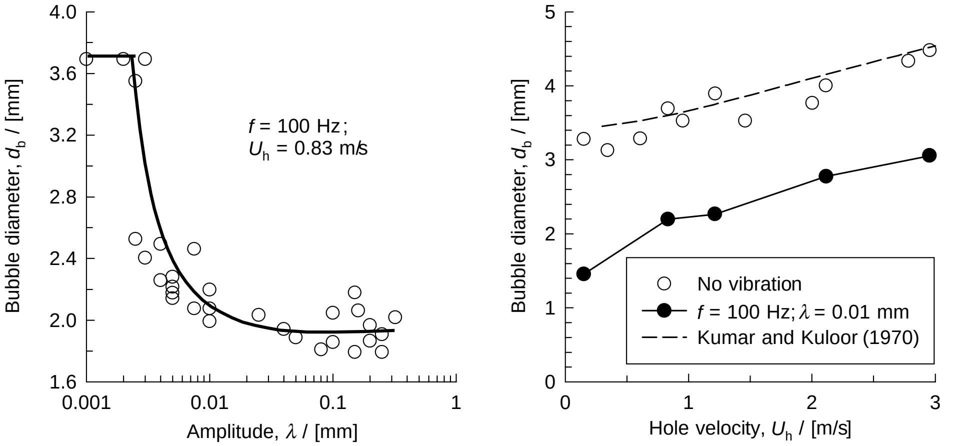
3. Bubble size measurements with single capillary distributor

In the first set of two experiments, the air flow rate through the single capillary was maintained at 5.3×10^{-7} m^3 s^{-1}



(a) Influence of varying frequency, λ = 0.005 mm

(b) Influence of varying frequency, λ = 0.08 mm



(c) Influence of varying amplitude

(d) Influence of varying hole velocity

Fig. 3. (a) Influence of variation in vibration frequency on bubble diameter at $\lambda = 0.005$ mm amplitude and hole velocity $U_h = 0.83$ m s^{-1}. (b) Influence of variation in vibration frequency on bubble diameter at $\lambda = 0.08$ mm amplitude and hole velocity $U_h = 0.83$ m s^{-1}. (c) Influence of vibration amplitude on the average bubble diameter at vibration frequency $f = 100$ Hz and hole velocity $U_h = 0.83$ m s^{-1}. (d) Influence of hole velocity U_h through capillary nozzle on the bubble diameter d_b in air–water operation. Measurements without vibration compared to data obtained with 100 Hz vibrations and amplitude $\lambda = 0.01$ mm. The dashed line represents the calculations of the bubble diameter using the model of Kumar and Kuloor (1970).

which corresponds to a hole velocity in the capillary of 0.83 m/s^{-1} and the amplitude λ was set at a constant value and the vibration frequency was varied in steps. The results are shown in Figs. 3(a) and (b) for $\lambda = 0.005$ and 0.08 mm, respectively. Let us consider the experiments at $\lambda = 0.005$ mm; see Fig. 3(a). There is a significant reduction in the bubble diameter from 3.7 mm (at 0 Hz) to 2.3 mm when the frequency f is set to 100 Hz. Increasing the frequency beyond 100 Hz does not reduce the bubble diameter further and it appears that there is an optimum at 100 Hz. In the next series of experiments, carried out with $\lambda = 0.08$ mm (Fig. 3(b)) we note that there is a continuous decrease in bubble size with increasing frequency in the 20–100 Hz range. Again we note that there is no significant bubble size decrease beyond a frequency of 100 Hz. Clearly both vibration frequency and vibration amplitude are important determinants of the bubble size. At a constant vibration frequency of 100 Hz, the influence of vibration amplitude, for a hole velocity $U_h = 0.83$ m s^{-1}, is shown in Fig. 3(c). We note a sharp decrease in the bubble diameter when the amplitude is increased to values above 0.0025 mm. Increase in the amplitude beyond 0.01 mm does not seem to have a further beneficial effect.

Fixing the vibration frequency at 100 Hz and the amplitude at 0.01 mm, we carried a series of experiments with variation in the air flow rate through the single capillary; see Fig. 3(d). For the no-vibrations situations, the measured bubble sizes agree reasonably well with predictions using the model of Kumar and Kuloor (1970). We note that the reduction of 40–50% of the value for no-vibrations case persists over a wide range of hole velocities when we apply liquid phase vibrations.

The physical explanation of the above observations is that application of vibrations to the liquid phase serves

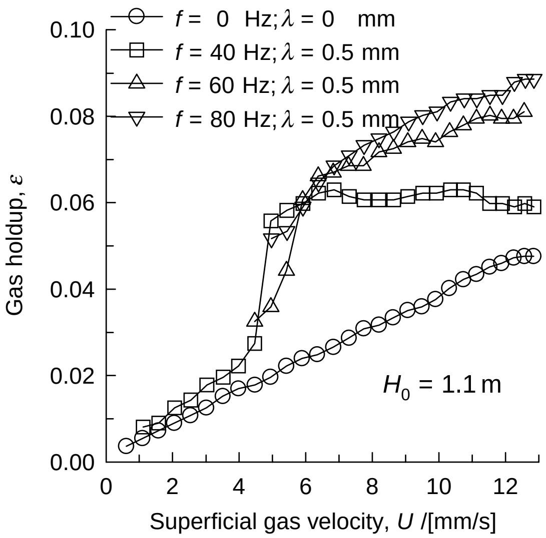


Fig. 4. Gas holdup, with and without vibrations, as function of the superficial gas velocity.

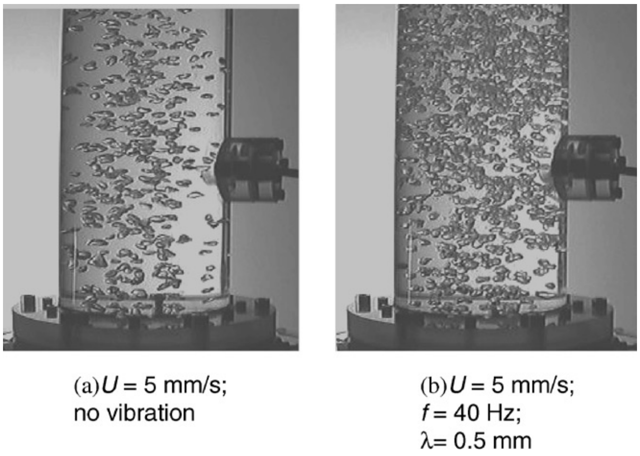


Fig. 5. Snapshots of the column operation with 12-capillary gas distributor operating at a superficial gas velocity $U = 5$ mm s^{-1} and liquid column height $H_0 = 1.10$ m. (a) Column operating without vibration excitement. (b) Operation with vibration $f = 40$ Hz and $\lambda = 0.5$ mm.

to overcome the surface tension forces and assist the break-up of the bubbles formed at the orifice (Grinis & Monin, 1999).

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. 4. Depending on the operating conditions, the improvement in the gas holdup is in the 100–300% range. For illustration, let us consider operation at $U = 5$ mm s^{-1}. For the no vibrations case, denoted here with the subscript 0, $\varepsilon_0 = 0.02$. When the vibrations are set at $f = 40$ Hz and $\lambda = 0.5$ mm, the gas holdup increases to a value $\varepsilon = 0.056$, representing a gas holdup enhancement $\varepsilon/\varepsilon_0 = 2.8$, i.e. an increase of 180%. Photographs of the corresponding gas–liquid dispersions are shown in Fig. 5. Visual inspection shows that application of vibrations leads to a larger number of smaller sized bubbles. The gas holdup data of Fig. 4 shows that increasing the vibration frequency from 40 to 60 Hz has a significant positive effect, especially for higher gas velocities U. Further increase in the vibration frequency to 80 Hz has only a minor added effect.

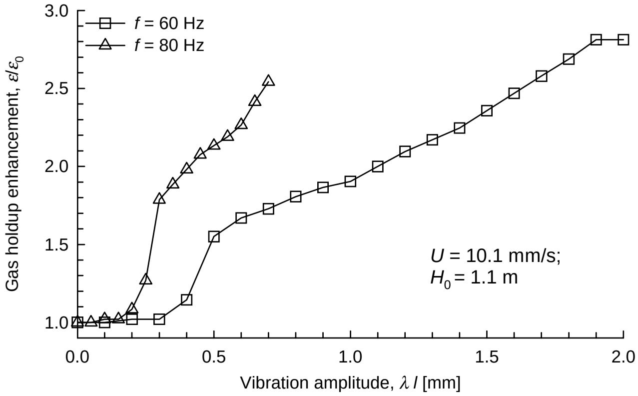
In order to gain further insights into the influence of vibrations on the enhancement of gas holdup $\varepsilon/\varepsilon_0$, we carried out a systematic study of the influence of (a) vibration amplitude, (b) vibration frequency and (c) clear liquid height; the results are shown in Fig. 6 for a constant superficial gas velocity $U = 10.1$ mm/s. Increase in vibration amplitude leads to an increase in $\varepsilon/\varepsilon_0$; see Fig. 6(a). The enhancement factor $\varepsilon/\varepsilon_0$ shows a steeper increase with λ when the vibration frequency is higher; compare the 80 and 60 Hz data in Fig. 6(a).

The influence of varying vibration frequency on $\varepsilon/\varepsilon_0$, at constant amplitude and clear liquid height, is much more intriguing and subtle; see Fig. 6(b). The sinusoidal waves generated by the vibration device are reflected at the top of the dispersion. For certain frequencies these reflected waves are synchronous, i.e. in phase, with the waves generated by the vibration exciter, thus leading to resonance. For this situation, we observe maxima in the gas holdup because the resonant (standing) waves tend to reduce the rise velocity of the bubble swarm. There is a set of characteristic frequencies for which the $\varepsilon/\varepsilon_0$, exhibits maxima. The dotted lines in Fig. 6(b) connect the series of first, second and third maxima. These maxima are off-set, approximately, by a frequency difference $\Delta f \approx 25$ Hz. From an energy input point of view it is desirable to operate the exciter at the conditions corresponding to the first maxima in Fig. 6(b).

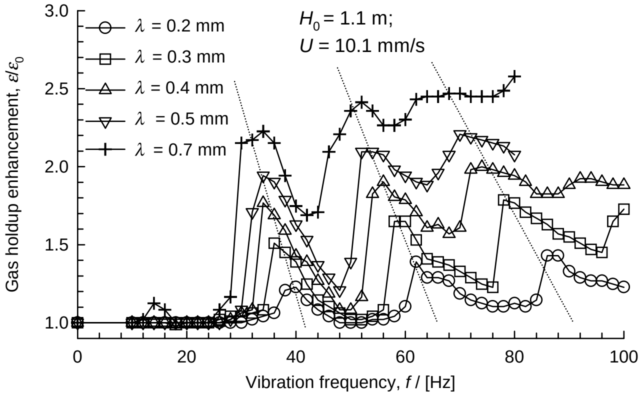
To emphasise further the resonance aspect, consider the influence of varying the clear liquid height for fixed values of vibration frequency and amplitude; see Fig. 6(c); we again observe multiple maxima in $\varepsilon/\varepsilon_0$ at "characteristic" values of the clear liquid height H_0. From Figs. 6(b) and (c) it appears that the maxima in $\varepsilon/\varepsilon_0$ are dictated both by the vibration frequency and clear liquid height H_0. From the set of experiments shown in Fig. 7 it is clear that the *product* of the vibration frequency and clear liquid height H_0 dictates the position of the holdup maxima.

From the gas holdup enhancement results presented in Figs. 6 and 7 we see that the gas holdup can be increased by more than factor 2 when the frequency and amplitude are properly "tuned".

(a) Influence of vibration amplitude



(b) Influence of vibration frequency



(c) Influence of varying clear liquid height

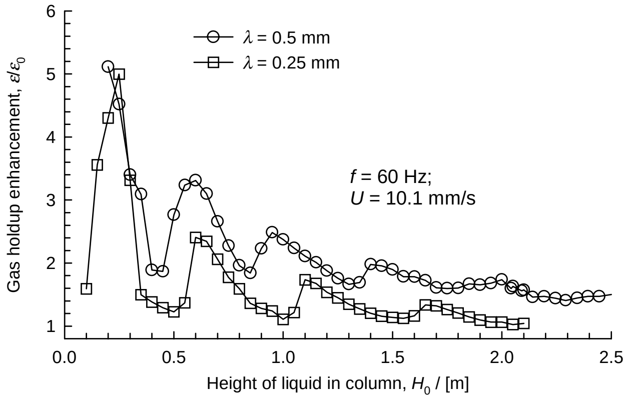


Fig. 6. (a) Enhancement in gas holdup with varying vibration amplitude. (b) Enhancement in gas holdup with varying vibration frequency. (c) Enhancement in gas holdup with varying clear liquid height.

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. 8(a). In consonance with the holdup measurements shown in Fig. 4, 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 bubble volume, k_La/ε, are plotted in Fig. 8(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_L, caused by increased turbulence at the gas–liquid interface.

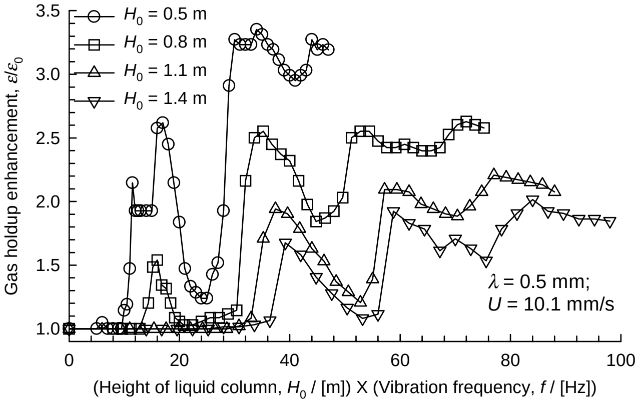


Fig. 7. Enhancement in gas holdup with the product of vibration frequency and clear liquid height.

6. Conclusions

The following major conclusions can be drawn from our study:

(a) Low-frequency vibrations, in the 40–100 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. 4.

(c) For any given superficial gas velocity the gas holdup increases with increasing amplitude; see Fig. 6(a).

(d) For a given value of superficial gas velocity U and vibration amplitude, the gas hold-up shows a series of maxima as a function of vibration frequency and clear liquid height H_0. This suggests that subtle resonance phenomena are in play; see Figs. 6 and 7.

(e) The increase in the k_La values is in consonance with the increase in the gas holdup values; see Fig. 8.

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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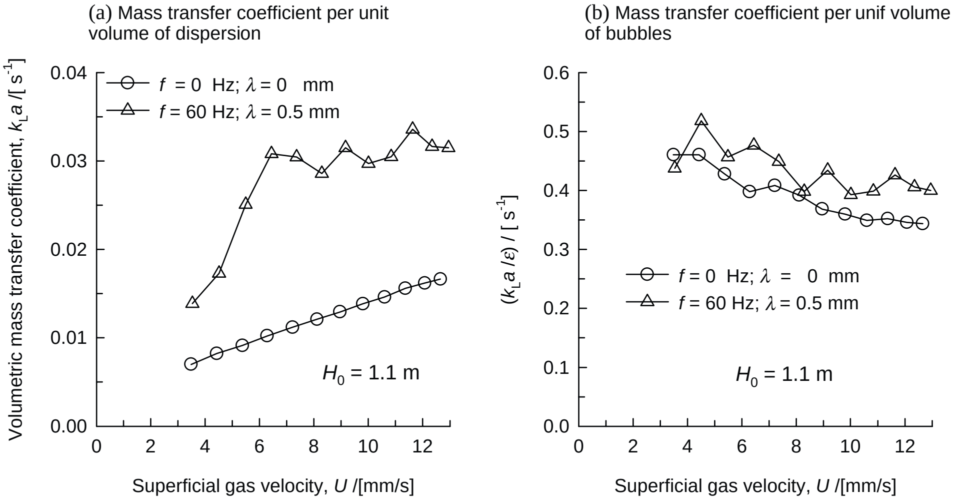

Fig. 8. (a) Volumetric mass transfer coefficient $k_L a$, with and without vibrations, as function of the superficial gas velocity, (b) $k_L a/\varepsilon$ as function of the superficial gas velocity.

development of novel concepts in reactive separations technology.

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