



Brief communication

Improving gas–liquid contacting in bubble columns by vibration excitement

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1. Introduction

A bubble column reactor is a commonly used reactor type; see Fig. 1(a). 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; see Fig. 1(b). 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 (cf. Fig. 1(c)), at frequencies of the order of 100 Hz, can (a) influence bubble rise (Jameson and Davidson, 1966; Jameson, 1966), (b) reduce the bubble size (Grinis and Monin, 1999; Krishna et al., 2000) and (c) improve gas–liquid mass transfer (Baird, 1963; Bartsch, 1995; Harbaum and Houghton, 1960). There is some indication from the patent literature (Kuesgen

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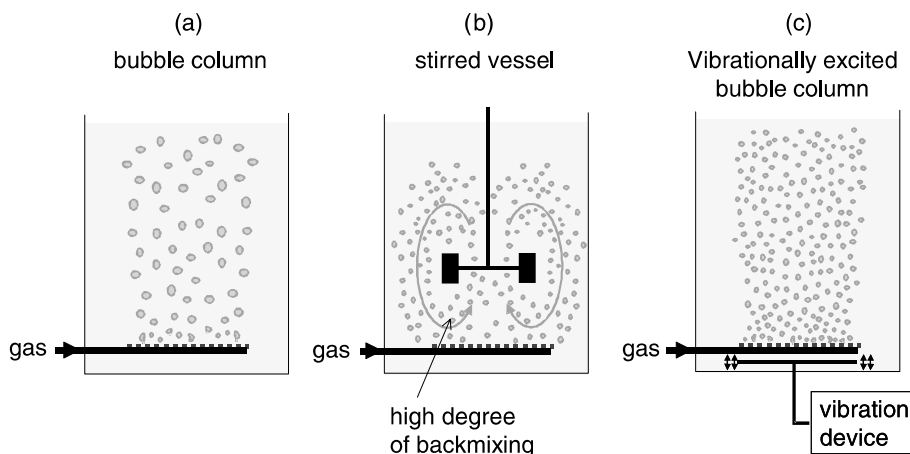


Fig. 1. (a) Typical aeration device, a bubble column reactor. (b) Stirred gas–liquid dispersion. (c) Liquid phase subjected to vibrations.

et al., 1999) of the use of vibration devices in high-pressure reactors, presumably to intensify gas–liquid reactor operation.

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. Our objective in this paper is to undertake a systematic study of the influence of vibration frequency and amplitude on gas hold-up and gas–liquid mass transfer in bubble columns. A special vibration excitement device is used for this purpose.

2. Experimental setup and procedures

The experimental setup consists of a bubble column, a vibration exciter, a power amplifier, a vibration controller and a personal computer. A schematic diagram of the experimental setup is given in Fig. 2. 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. 2. At a distance of 0.1 m above the membrane, air is fed to the bubble column through 12 stainless steel capillaries of 0.9 mm inner diameter. A schematic drawing of the gas distributor is shown in inset to Fig. 2. 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 and 25 mm. The vibrations follow a sinusoidal motion. The

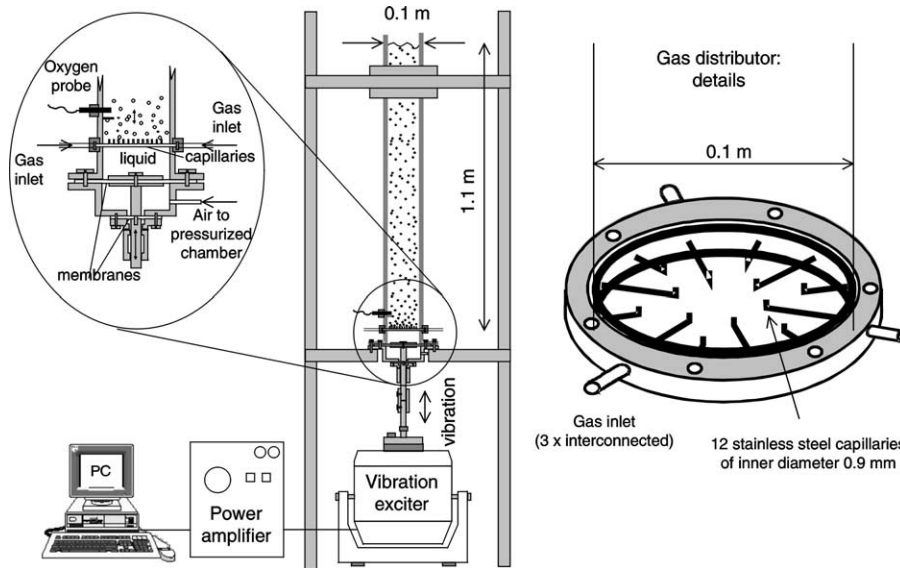


Fig. 2. Experimental setup of the bubble column with vibration excitation device. Further details of the experimental setup including photographs of the rig are to be found on our website: <http://ct-cr4.chem.uva.nl/vibrationexciter>.

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 setup including photographs of the rig are to be found on our website: <http://ct-cr4.chem.uva.nl/vibrationexciter>.

Two physical quantities, the gas hold-up and the volumetric mass transfer coefficient $k_L a$, 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. 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 hold-up ε is measured by visually recording the dispersion height H above the gas distributor; the gas hold-up is then calculated from

$$\varepsilon = 1 - \frac{H_0}{H} \quad (1)$$

where H_0 is the height of the ungasged liquid in the column. In all the experiments reported in this paper the value of H_0 was held constant at 1.1 m .

The volumetric mass transfer coefficient $k_L a$ is measured by means of the dynamic oxygen absorption method, described in earlier work from our group (Letzel et al., 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:

$$\frac{dC}{dt} = k_L a (C^* - C) \quad (2)$$

where a is the interfacial area per unit volume of dispersion, $k_L a$ 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:

$$\begin{aligned} t = 0 : \quad C &= C_0 \\ t \rightarrow \infty : \quad C &= C^* \end{aligned} \quad (3)$$

to obtain

$$\frac{C - C_0}{C^* - C_0} = 1 - \exp(-k_L a t) \quad (4)$$

The only unknown constant in Eq. (4) is $k_L a$, 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. 3 shows an example of the normalised output signal of the

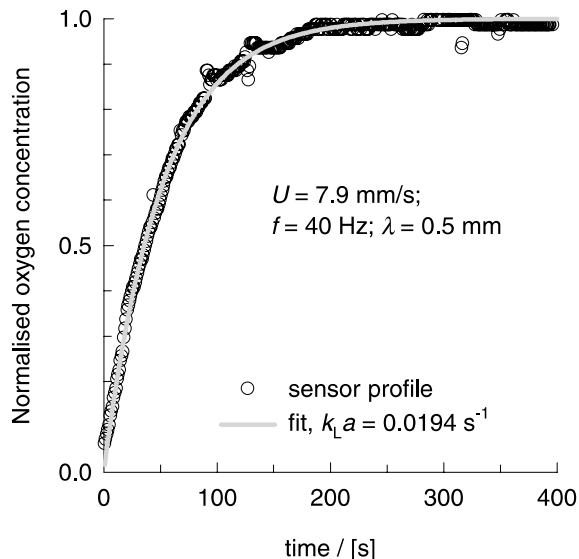


Fig. 3. Example of a mass transfer measurement together with a least-square fit of Eq. (4). $H_0 = 1.10 \text{ m}$; $U = 7.9 \text{ mm/s}$; $f = 40 \text{ Hz}$ and $\lambda = 0.5 \text{ mm}$.

oxygen probe at a superficial gas velocity $U = 7.9$ mm/s, a vibration frequency $f = 40$ Hz and an amplitude $\lambda = 0.05$ mm. Also shown in Fig. 3 is the curve fitted with Eq. (4) taking $k_L a = 0.0194$ s⁻¹. We note that the model fits the experimental sensor profile very well.

3. Experimental results and discussion

3.1. Gas hold-up

In order to illustrate the efficacy of liquid vibrations we show photographs of the column operation, focusing on the top of the dispersion layer, in Fig. 4. When there is no gas injected into the system, the clear height is determined visually to be $H_0 = 1.10$ m; see Fig. 4(a). Now consider air flow at a superficial velocity $U = 6$ mm/s. The height of dispersion increases to a value $H = 1.128$ m, giving a gas hold-up value (using Eq. (1)) $\varepsilon = 0.0248$; see Fig. 4(b). When the liquid phase is vibrated at $f = 40$ Hz and an amplitude $\lambda = 0.5$ mm, the height of dispersion increases to a value $H = 1.169$ m, giving a gas hold-up value (using Eq. (1)) $\varepsilon = 0.059$; see Fig. 4(c). With vibration the hold-up is seen to be a factor 2.4 times the no-vibrations case. Visual inspection reveals that the bubbles are significantly smaller in size; this is also evidenced in the photographs shown in Figs. 4(b) and (c).

We carried out a series of measurements with varying superficial gas velocity, U , vibration frequency f and amplitude λ with the objective of determining the best set of parameters which leads to the maximum increase in the gas hold-up.

Consider first a set of three experiments at three different superficial gas velocities $U = 5.8, 10.1$ and 13.5 mm/s wherein f is varied over in the range 0–120 Hz for a set of values of the amplitude λ ; the results are shown in Figs. 5(a), (b) and (c) respectively. For any superficial gas velocity we note that the gas hold-up increases significantly with vibration excitement, in comparison to the no-vibrations case. The waves generated by the vibration device are reflected at the top of the dispersion. For certain frequencies these reflected waves are synchronous with the waves generated by the vibration exciter, thus leading to resonance. For this situation, we have maxima in the gas

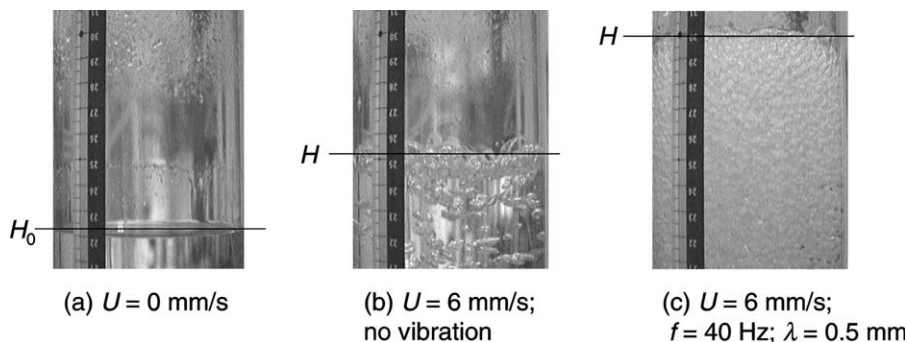


Fig. 4. Snapshots of the column operation taken near the top of the dispersion: (a) no gas flow through the column; the height of liquid column is H_0 ; (b) column operating at $U = 6$ mm/s without vibration excitement. The height of the dispersion is H ; (c) column operating at $U = 6$ mm/s and with vibration $f = 40$ Hz and $\lambda = 0.5$ mm. The height of the dispersion is H .

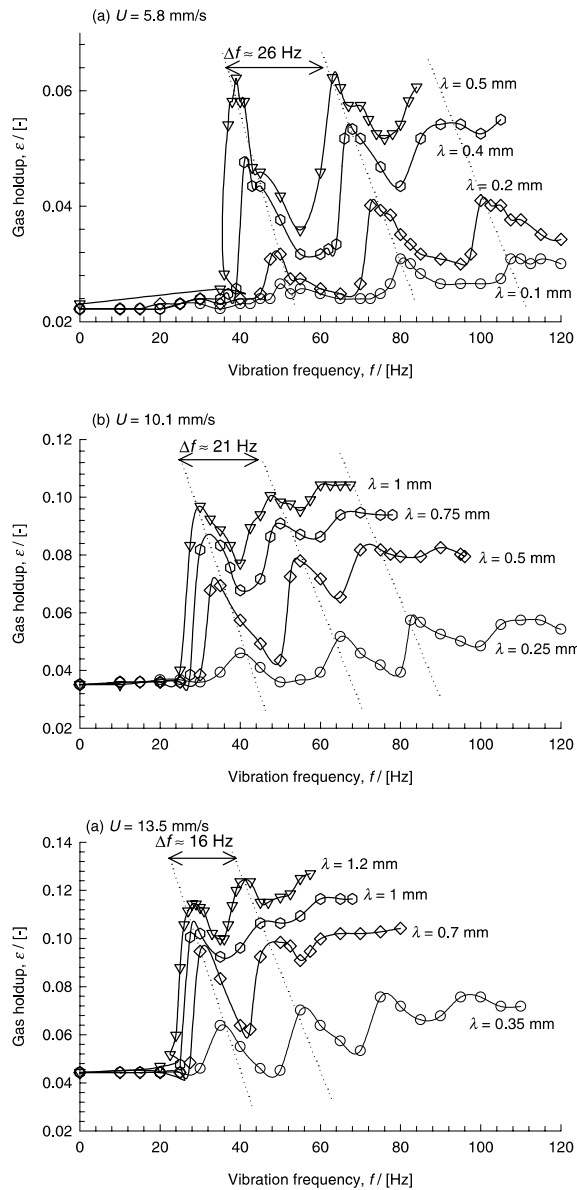


Fig. 5. Gas hold-up as function of the vibration frequency and amplitude for a clear liquid height $H_0 = 1.10$ m: (a) $U = 5.8$ mm/s; (b) $U = 10.1$ mm/s; (c) $U = 13.5$ mm/s. The dotted lines trace the maxima in the gas hold-ups.

hold-up because the resonating waves tend to cause maximum bubble breakage. We also note that for a given superficial gas velocity and set amplitude, there is a number of characteristic frequencies for which the gas hold-up exhibits maxima. Visual inspection also shows a substantial decrease of the bubble diameter at these characteristic frequencies. The dotted lines in Fig. 5 connect the series of first, second and third maxima. These maxima are off-set by a nearly constant frequency difference Δf . For operation at $U = 5.8$ mm/s; the maxima are off-set by a value

$\Delta f \approx 26$ Hz; see Fig. 5(a). The same picture emerges for operation at $U = 10.1$ mm/s. However, the frequency off-set separating the hold-up maxima has a reduced value, $\Delta f \approx 21$ Hz; see Fig. 5(b). For operation at $U = 13.5$ mm/s, the frequency off-set further reduces to $\Delta f \approx 16$ Hz; see Fig. 5(c). The off-set frequency shows a linear decline with the superficial gas velocity, suggesting that subtle resonance phenomena are at play here; see Fig. 6(a).

From Fig. 5 we also observe that for any given superficial gas velocity and set vibration frequency, the gas hold-up increases with increasing vibration amplitude. With increasing amplitude, the characteristic frequencies (corresponding to hold-up maxima) shift to lower values. There is however a maximum in the amplitude which can be applied at a given value of U ; increase of the amplitude beyond this maximum values leads to an unstable, splashing gas–liquid dispersion for

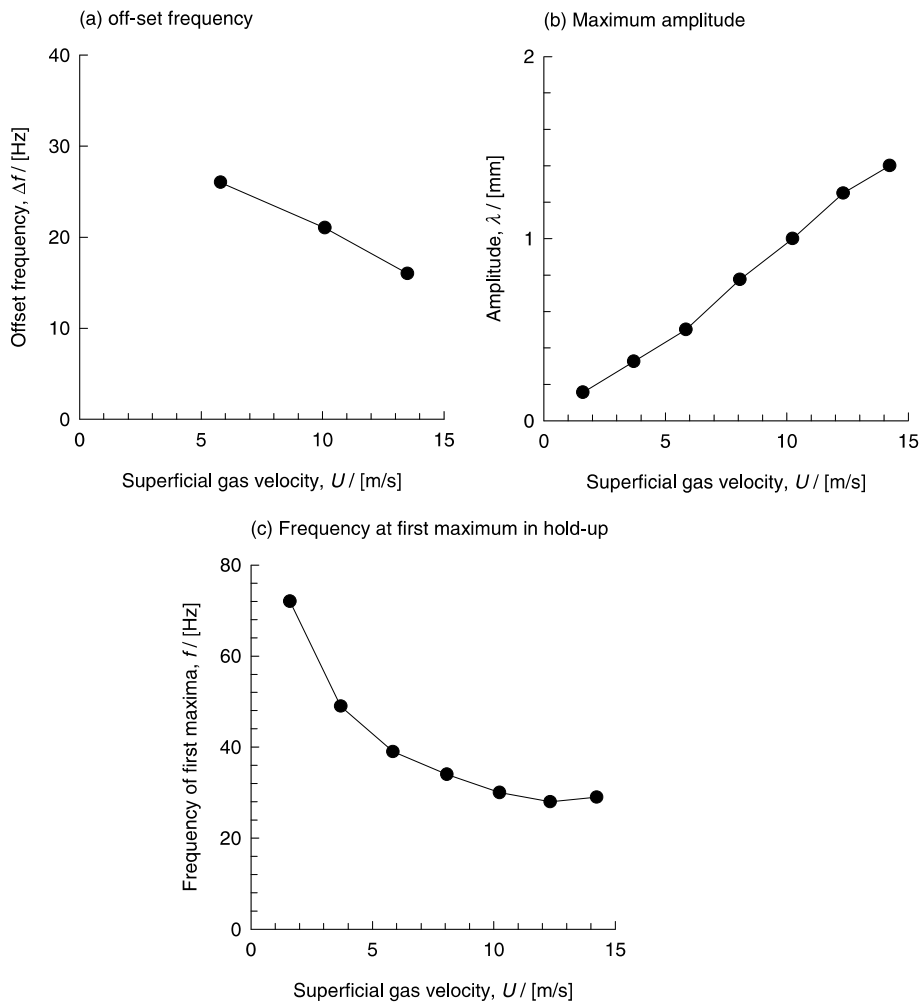


Fig. 6. (a) Off-set frequency as a function of the superficial gas velocity U . (b) Maximum amplitude of vibration for stable operation as a function of U . (c) “Optimum” frequency corresponding to the first maximum in the gas hold-up for operation at the maximum vibration amplitude.

which the measurement of the gas hold-up becomes impossible. The maximum amplitude for different superficial gas velocities is plotted in Fig. 6(b) and seems to be an almost linear function of the gas velocity.

With the amplitude set at the maximum allowable value for stable operation (values given in Fig. 6(b)), the frequencies corresponding to the first hold-up maxima are plotted in Fig. 6(c). We note that these “first-characteristic frequencies” decrease with increasing superficial gas velocity and tends to reach a plateau value of 30 Hz.

From an energy input point of view it is desirable to operate the exciter at the conditions corresponding to the conditions specified in Figs. 6(b) and (c).

A second set of experiments has been carried out for superficial gas velocities ranging from 2 to 14 mm/s. For each of these velocities, the gas hold-up is measured at 0 Hz (no vibration) and at the maximum amplitude, along with the “first-characteristic frequencies”, specified in Figs. 6(b) and (c). From the hold-up results presented in Fig. 7 we see that the gas hold-up can be increased by more than factor 2 when the frequency and amplitude are properly “tuned”.

3.1.1. Volumetric mass transfer coefficients

Consider operation of the column at $U = 10.1$ mm/s. Keeping the amplitude constant at $\lambda = 0.5$ mm and varying the frequency we performed mass transfer measurements; these results are shown in Fig. 8(a). It is interesting to note that the $k_L a$ values also show maxima at the same frequencies for which the gas hold-up exhibit maxima. This result is to be expected as a higher gas hold-up would imply better gas–liquid contact and therefore better mass transfer. In Fig. 8(b) we have plotted the ratios of the gas hold-up, and volumetric mass transfer coefficient, at any given frequency with the values for the no-vibrations case. The results show that the improvement in the $k_L a$, as compared to the no-vibrations case, is equal to the improvement in the gas hold-up for

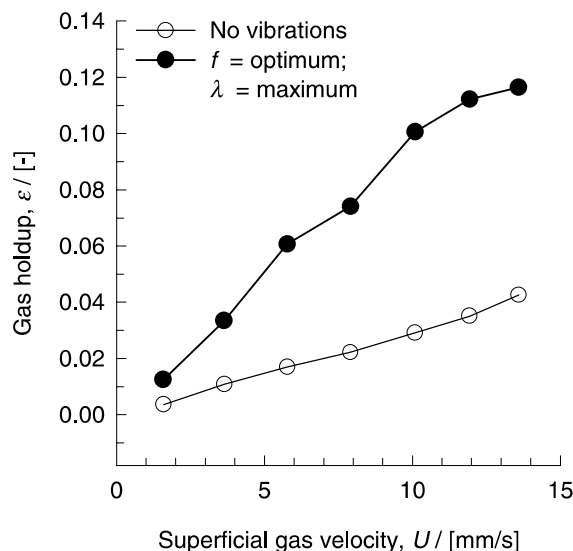


Fig. 7. Gas hold-up, with and without vibrations, as function of the superficial gas velocity. The vibration amplitude and frequency used correspond to those specified in Figs. 6(b) and (c).

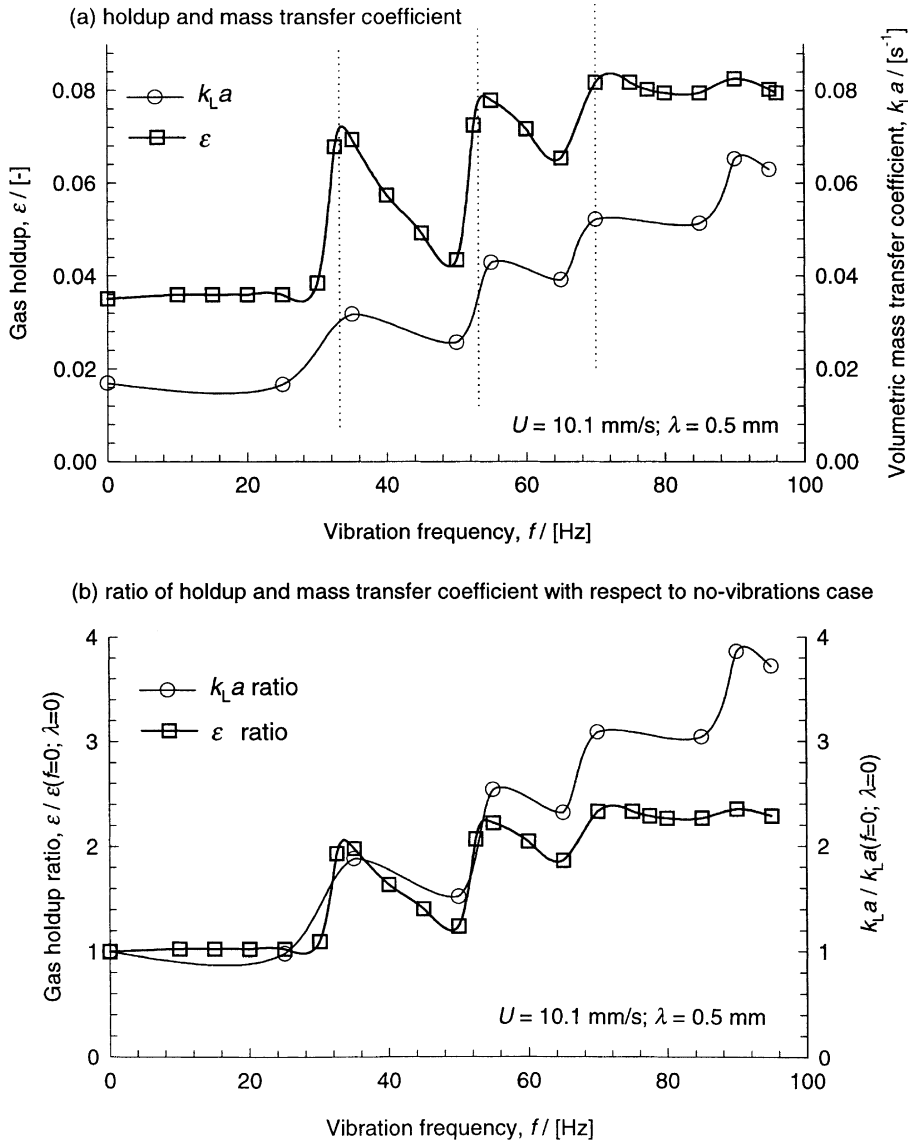


Fig. 8. (a) Volumetric mass transfer coefficient and gas hold-up as function of the frequency f at a superficial gas velocity $U = 10.1 \text{ mm/s}$ and an amplitude $\lambda = 0.5 \text{ mm}$. (b) Ratios of $k_L a$ and gas hold-up ϵ with respect to values for the no-vibrations case as a function of f .

frequency values below about 40 Hz. However, above 60 Hz, the improvement in the $k_L a$ is about 1.5–2 times the improvement in the gas hold-up. This result tends to suggest that at higher vibration frequencies the interface between the gas bubbles and the liquid becomes more turbulent and results in a higher value for k_L . The improvement in the volumetric mass transfer coefficient is not only due to a reduction in bubble size, which results in an increase in the specific interfacial area a , but also due to an enhancement in the k_L .

We carried out a series of mass transfer experiments for a range of superficial velocities in the range 2–14 mm/s for two situations: (i) no vibrations, and (ii) vibrations at the “optimum” frequencies corresponding to the values in Fig. 6(c) and maximum amplitudes according to Fig. 6(b). The $k_L a$ values for these two sets are shown in Fig. 9(a). In consonance with the hold-up measurements shown in Fig. 7, we find that the $k_L a$ values are improved by a factor of 2 due to

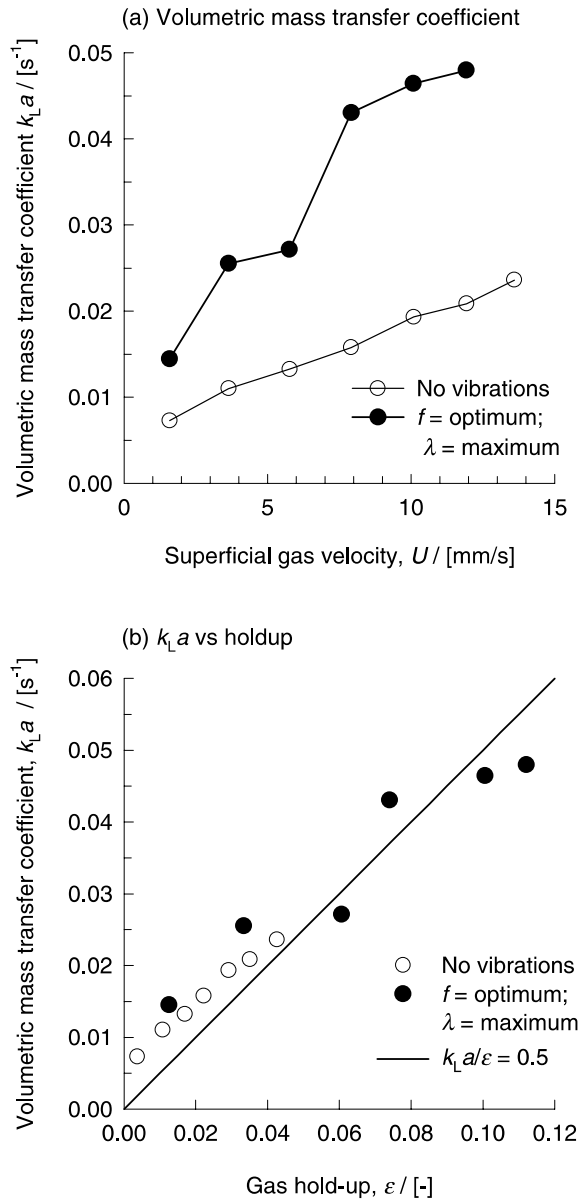


Fig. 9. (a) Volumetric mass transfer coefficient $k_L a$, with and without vibrations, as function of the superficial gas velocity. The vibration amplitude and frequency used correspond to those specified in Figs. 6(b) and (c). (b) $k_L a$, as function of the gas hold-up ϵ .

vibration excitement. Since the frequencies correspond to those obtained for the first hold-up maxima, we would expect on the basis of the results shown in Fig. 8(b) that the improvement in $k_L a$ to be equal to the improvement in the gas hold-up. In order to demonstrate this we plot the $k_L a$ against the gas hold-up in Fig. 9(b). The values of $k_L a$ appear to be linearly dependent on the gas hold-up, either with or without vibrations. The value of $k_L a/\varepsilon$ is seen to approximately equal to 0.5, which is the value also obtained by Letzel et al. (1999) for bubble column operation.

4. Conclusions

In this paper we have demonstrated that significant improvement in the gas hold-up and volumetric mass transfer coefficient can be realised by vibration excitement.

The following major conclusions can be drawn from our study:

- (a) 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. This suggests that subtle resonance phenomena are in play; see Fig. 5.
- (b) The vibration frequency corresponding to the first hold-up maximum appears to decrease with increasing U and reaches a plateau value of 30 Hz; see Fig. 6(c).
- (c) For any given superficial gas velocity the gas hold-up increases with increasing amplitude but beyond a certain maximum value, given in Fig. 6(b), the operation of the column becomes unstable.
- (d) Compared to the no-vibrations case the gas hold-up increases by a factor of about 2, provided the vibration frequency and amplitude are properly “tuned” according to the recipe specified in Figs. 6(b) and (c); see Fig. 7.
- (e) Compared to the no-vibrations case the volumetric mass transfer coefficient $k_L a$ increases by a factor of about 2, when operating at the frequency corresponding to the first maximum in the gas hold-up; see Fig. 9(a). For operation at higher vibration frequencies, the improvement in $k_L a$ increases to a value of 4.
- (f) The increase in the $k_L a$ values is in consonance with the increase in the gas hold-up values; see Fig. 9(b).

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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