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SHORT COMMUNICATION

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Utilisation of bubble resonance phenomena to improve gas–liquid contact

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Abstract In several branches of science and technology a gaseous phase is dispersed into a liquid in the form of bubbles, a gaseous component then dissolves into the liquid and subsequently undergoes chemical reaction. The overall process performance can be improved substantially when the area of gas–liquid contact is increased. By subjecting the liquid phase to low frequency vibrations, the bubbles are shown to suffer significant breakage, induced by resonance. When the vibration is properly tuned, the interfacial area is found to increase by a factor of 1.8–2.4, depending on the properties of the liquid. Resonance-induced bubble breakage phenomena have a great potential for improving the rates of chemical processes involving fast reactions, with minimal energy input.

Introduction

Aeration is the pumping or sparging of bubbles into liquid, often with mechanical stirring to break up and distribute the bubbles; see Fig. 1a, b. Aeration typically supplies the gas needed for a chemical reaction in a liquid, or the oxygen demanded by organisms in bio-engineering. Aeration of fluids is used in rectification, absorption, waste-water treatment, and biotechnological processes. In all such processes, a component needs to be transferred from the gas bubbles to the surrounding liquid. In the ozone treatment of water, for example, ozone is dispersed as gas bubbles in water. The ozone dissolves in water and reacts chemically with organic pollutants. The chemical reaction is fast and the overall process is limited by the efficiency of transfer of ozone from the gas bubbles to the liquid phase. A significant improvement in the water purification efficiency, leading to vastly improved water quality and smaller equipment costs, can be realised if the physical mass transfer process can be enhanced. One common method for improving this mass transfer efficiency is by carrying out the reaction in a stirred vessel (see Fig. 1b). 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 (see Fig. 1b). 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

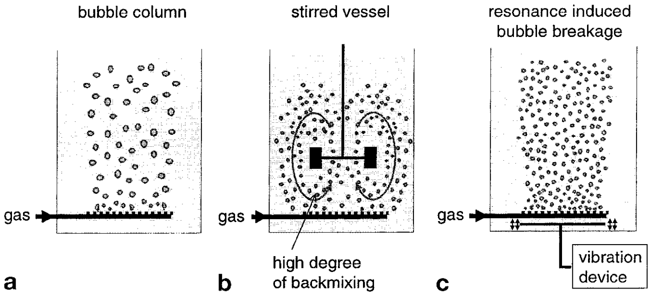

Fig. 1 Typical aeration device, a bubble column reactor (**a**); Stirred gas-liquid dispersion (**b**); Resonance-induced breakage caused by low-frequency vibrations (**c**)

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column, with little or no backmixing of the gas bubbles.

Edible oils require to be partially hydrogenated in order to modify their melting behaviour and their taste stability. For example, the hydrogenation of trioleoylglycerol (Ol_3) to tristearoylglycerol (St_3) occurs according to the scheme: $Ol_3 \rightarrow StOl_2 \rightarrow St_2\,Ol \rightarrow St_3$ where $StOl_2$ and St_2Ol represent the partially hydrogenated products (Krishna and Sie 1994). With respect to product properties like melting behaviour, selective hydrogenation giving the maximum concentration of intermediates $StOl_2$ and St_2Ol is required. Industrially, the hydrogenation is usually carried out in a bubble column reactor in which fine catalyst particles are dispersed in the liquid phase into which hydrogen is dispersed in the form of gas bubbles. The selectivity and conversion achieved in this process can be improved if the mass transfer from the gas bubbles to the oil phase can be enhanced, while maintaining piston flow of gas bubbles through the slurry of oil and catalyst.

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 contact without intense agitation.

The research reported in this communication provides a novel method for improving the contact efficiency of bubbly dispersions in liquids which has none of the disadvantages associated with stirred vessels. The approach we take relies on interactions between low-frequency sound waves and gas bubbles in liquids.

It is well known that bubbles produce an acoustic signal on formation and deformation (Minnaert 1933; Strasberg 1953, 1956; Leighton 1994). Much of the sound we associate with running water is made by bubbles. The sound of a rushing stream, a waterfall, or liquid pouring into a glass is due to bubbles being formed (Minnaert 1933; Manasseh 1997). Why do bubbles make sound? As a bubble is formed or broken up, it suffers a severe distortion to its surface. As the surface recoils, the gas within the bubble gets compressed. Minnaert first analysed the sounds of bubbles produced in various liquids and concluded that most of the sound was produced by radial pulsation of the bubble. Like any natural system with inertia and stiffness, the gas and surrounding liquid vibrate in response to this shock. In effect, the bubbles 'ring' like bells. Bubbles moving freely through liquid may also emit sound, probably as they are impacted by turbulent eddies. Bubbles have a natural frequency that depends on their size. Big bubbles emit low-frequency sounds while small bubbles emit high-frequency sounds – like big bells and small bells. This simple fact, first quantified in 1933 by Minnaert, is the key to bubble acoustics. As the bubbles get larger, the pitch of the note gets lower. It was suggested by Leighton and Walton (1987) that the sound spectrum produced by bubbles in the environment could be used to calculate the bubble size spectrum. The sound that bubbles emit yields a measurement tool that is currently being exploited within the chemicals, minerals-processing and waste-water industries (Manasseh 1997).

Our approach to improving bubble–liquid contact is to exploit bubble acoustics in reverse, i.e. we subject the bubble to low-frequency vibrations and rely on resonance phenomena to induce bubble breakage (see Fig. 1c). When the liquid is made to vibrate at the characteristic bubble frequency, the bubble will resonate and suffer breakage because of the resulting violent oscillations. The inspiration for our work is drawn from the work of Smith (1935) who concluded that bubbles should show a high degree of resonance since the damping coefficient was small. He also suggested that the intense local strains in the vicinity of the pulsating bubble might account for the destructive effects of sound on vegetation and other living matter.

The novelty of the approach presented in this communication lies in the fact that we utilise the resonance phenomena not to characterise bubble size, as is invariably seen in the published literature on bubble acoustics (Leighton 1994), but to induce bubble breakage and thereby increase the gas–liquid interface area.

Influence of low-frequency vibrations on bubble size

In order to demonstrate resonance-induced bubble breakage, we carried out a series of experiments in a cylindrical polyacrylate column of 730 mm height and 100 mm i.d. A vibration device (which was simply a commercially available loudspeaker) was mounted at

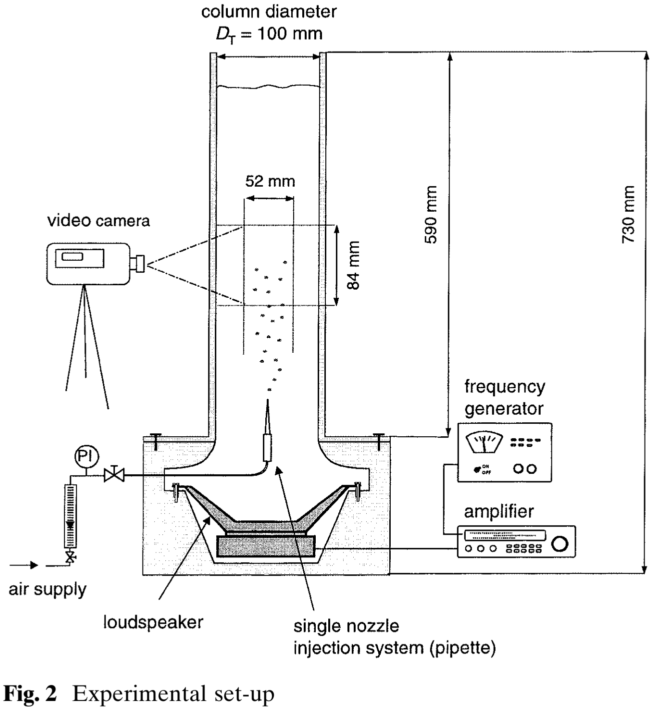

Fig. 2 Experimental set-up

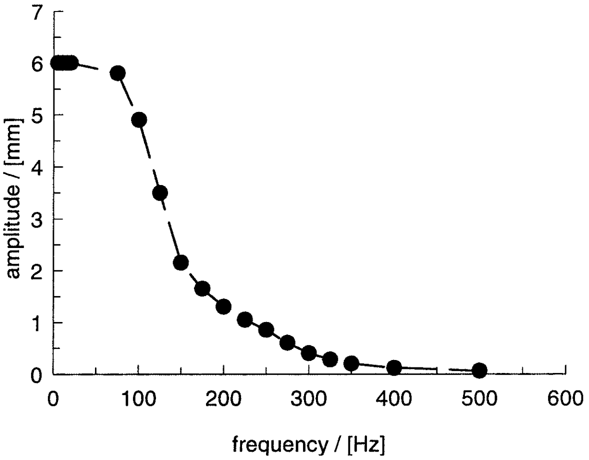

Fig. 3 Amplitude vs frequency characteristics of the vibration device (loudspeaker)

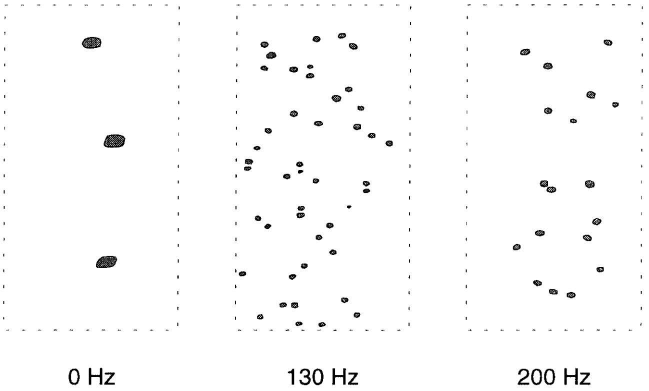

Fig. 4 Typical video snapshots taken at three different vibration frequencies for the air–water system. The actual video images (placed on our website: http://ct-cr4.chem.uva.nl/sonication) have been retraced. The *dotted contour lines* cover the area indicated in Fig. 2. The air flow rate in these experiments was maintained at 0.17 ml/s

the bottom of the column (Fig. 2). The outer diameter of the loudspeaker was 0.18 m and the vibrating diaphragm had a diameter of 0.12 m. The vibration device was connected to a frequency generator. The amplitude vs. frequency characteristics of the speaker are shown in Fig. 3. The column was filled with demineralised water, tellus oil or paraffin oil; the physical properties of the systems are specified in Table 1. Air was introduced into the column through a single nozzle with an opening of 0.4 mm. The volumetric gas flow was accurately measured and maintained constant throughout the series of experiments. Bubbles were successively formed through this orifice, placed in the centre of the column. The loudspeaker was made to vibrate at various frequencies in the range 0–400 Hz. At each vibration frequency, video recordings of the bubble dispersion were made at 25 frames per second for a period of 5 s. Frame-by-frame analysis of the video images, gave accurate information on the number of bubbles passing through the observation window (sketched in Fig. 2) in the time interval of the observations (5 s).

Let us first consider the results with injection of air through a single nozzle into demineralised water at a flow of 0.17 ml/s. Typical snapshots of the bubble dispersions taken at three different vibration frequencies are shown in Fig. 4. The corresponding video recordings can be viewed by logging on to our website: http://ct-cr4.chem.uva.nl/sonication/. We see from Fig. 4 that at 130 Hz the bubble breakage is such as to increase the number of bubbles by one order of magnitude. Quantitative information on the number of bubbles within the observation window are given in Fig. 5a. At 0 Hz (no-vibration), seven bubbles issue from the orifice per second. When the vibration is tuned to 100 and 130 Hz there is a dramatic increase to 107 and 112 bubbles per second, respectively. At intermediate vibration frequencies there is no significant effect. The sharp peaks at 100 and 130 Hz testify to the fact that this is a resonance effect and not just an influence of vibration. There is a smaller peak at 200 Hz. Beyond 200 Hz there is no improvement over the no-vibration case. From a knowledge of the volumetric flow of the gas, the average bubble diameter can be calculated and the results are presented in Fig. 5b. We see, for example, that at 130 Hz the average bubble diameter is reduced from 3.5 mm to 1.5 mm. The increase in the interfacial area relative to the no-vibration case is shown in Fig. 5c. Vibrations, when properly tuned, cause the interfacial area to increase to 240% of the value corresponding to the 0 Hz (i.e. no-vibration) case.

Table 1 Physical properties of the liquid phase (at atmospheric conditions)

Liquid	Physical property		
	Density ρ_L/(kg/m^3)	Dynamic viscosity η_L/(Pa × s)	Surface tension σ/(N/m)
Water	998	0.001	0.07275
Tellus oil	862	0.075	0.028
Paraffin oil	795	0.0029	0.028

It must be remarked here that calculations using the Minnaert (1933) formula for the resonance frequency of a bubble of 3.5 mm yields a value of 1900 Hz. In our experiments we observed breakage at frequencies in the 100–200 Hz range, much lower than the resonance frequency calculated from the Minnaert formula. One reason for this discrepancy is that the Minnaert formula is valid for a single spherical bubble, whereas for air–water systems the bubbles of 3.5 mm assume an ellipsoidal shape (see Fig. 4). Furthermore, bubble oscillations are non-linear and there are complex bubble–bubble interactions which distort the resonance characteristics. The fact that bubble breakage is observed only for a narrow range of frequencies between 100 and 200 Hz is testimony to the fact that resonance-induced breakage is at work.

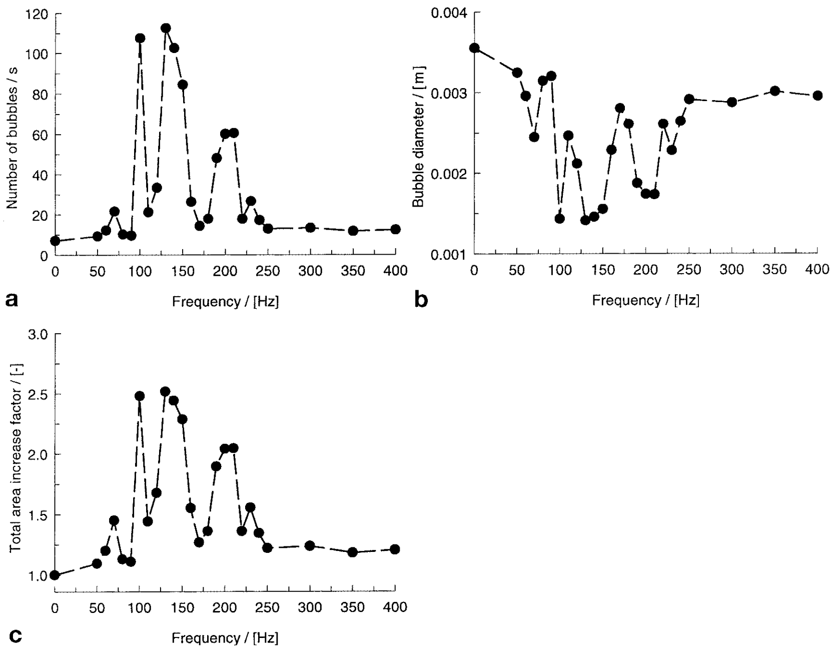

Fig. 5 Influence of vibration frequency on the number of bubbles per second within observation window (**a**), average diameter of the bubbles (**b**) and increase in the surface area of bubble with respect to the no-vibration case (**c**)

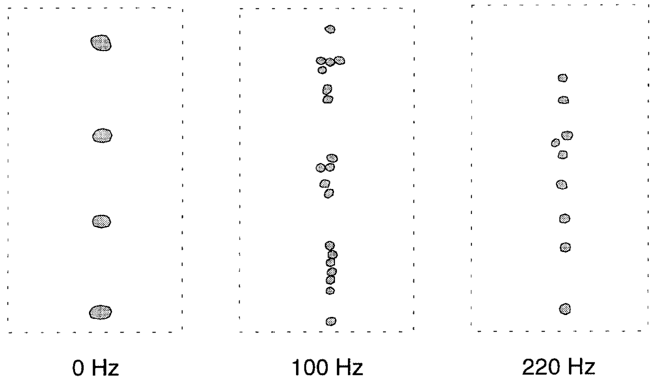

Fig. 6 Typical video snapshots taken at three different vibration frequencies for the air–tellus oil system. The actual video images (placed on our website: http://ct-cr4.chem.uva.nl/sonication/) have been retraced. The *dotted contour lines* cover the area indicated in Fig. 2. The air flow rate in these experiments was maintained at 0.18 ml/s

The resonance-induced bubble breakage phenomenon also manifests itself for other liquids. Typical snapshots of the bubble dispersions with tellus oil and paraffin oil are shown, respectively, in Figs. 6 and 7 (the actual video recordings can be viewed our website: http://ct-cr4.chem.uva.nl/sonication/). For the highly viscous tellus oil, the bubble size decreases from 3.7 mm (no-vibration) to 2.1 mm at 100 Hz. The corresponding increase in the gas–liquid interfacial area is by a factor of 1.8 (Fig. 8a). For paraffin oil, the bubble size decreases from 3 mm (no-vibration) to 1.3 mm at 200 Hz. The corresponding increase in the interfacial area is by a factor of 2.3 (see Fig. 8b).

Concluding remarks

We have shown that low-frequency vibrations, in the 100–200 Hz range, has the effect of inducing a significant amount of bubble breakage. The area increases, determined from video imaging techniques, are about 80% to 140%, depending on the properties of the liquid.

The energy input required for vibrating the liquid at such frequencies is estimated to be 0.5 kW/m^3. This is an order of magnitude lower than the energy requirements of stirred vessels. In contrast to a stirred vessel,

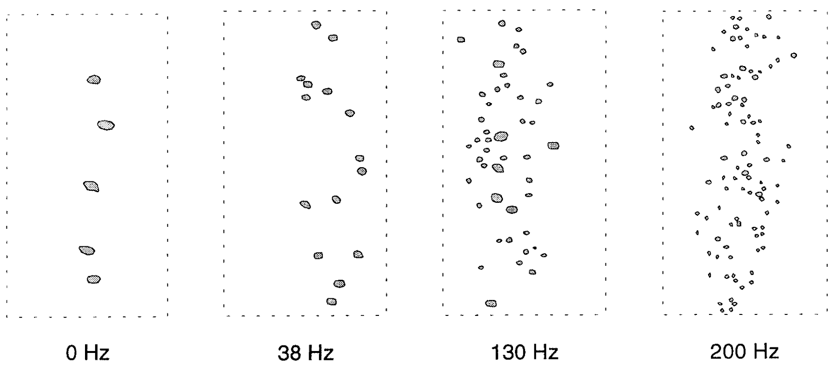

Fig. 7 Typical video snapshots taken at three different vibration frequencies for the air–paraffin oil system. The actual video images (placed on our website: http://ct-cr4.chem.uva.nl/sonication/) have been retraced. The *dotted contour lines* cover the area indicated in Fig. 2. The air flow rate in these experiments was maintained at 0.18 ml/s

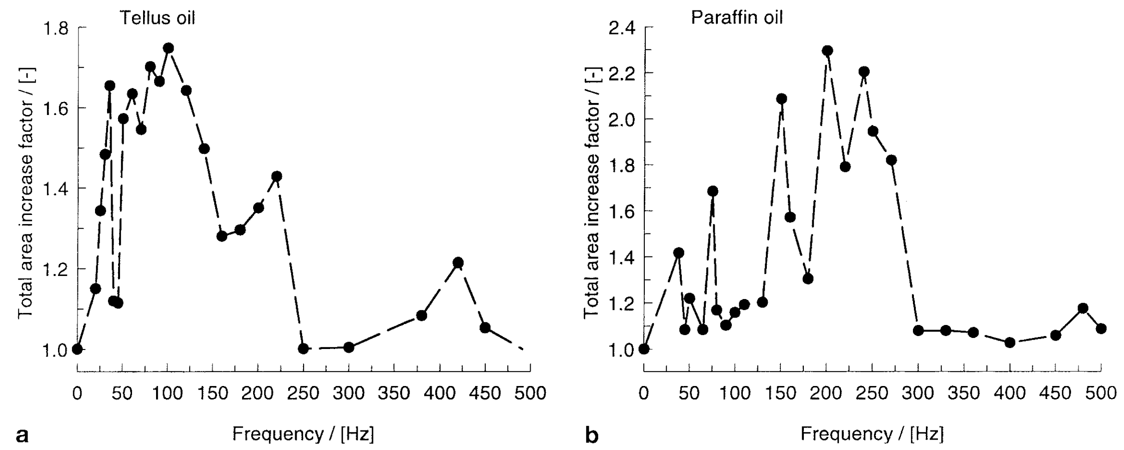

Fig. 8 Influence of vibration frequency on the increase in the surface area of bubble with respect to the no-vibration case with Tellus oil (**a**) or paraffin oil (**b**) as liquid phase

the bubbles maintain their piston flow character as they flow up the column. Incorporation of a vibration device at the bottom of an industrial reactor is a relatively simple matter and it is anticipated that the benefits to conversion in, say, a water ozonation plant could be of great significance. Also, in view of the modest energy inputs, when compared to a stirred vessel, the increased costs associated with the power required for low-frequency vibrations may be well justified.

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