

Effect of solids on homogeneous–heterogeneous flow regime transition in bubble columns

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Abstract

Experiments were conducted to study the effect of the presence of the solid phase on the homogeneous–heterogeneous flow regime transition in a bubble column 0.14 m diameter. Air, distilled water and calcium alginate beads (2.1 mm, 1023 kg/m³) at concentrations $c = 0$ –30% (vol.) were the phases. The basic data were the voidage–gas flow rate dependences. The critical point, where the homogeneous regime loses stability and the transition begins, was evaluated by the drift flux model. The critical values of voidage and gas flow rate were the quantitative measures of the homogeneous regime stability. These were plotted against the solid phase concentration. It was found, that both the voidage and the critical values increased with the solid content at low solid loading, approx. $c = 0$ –3%, and decreased at higher loading, $c > 3\%$. The homogeneous regime was thus first stabilized and then destabilized. To explain this dual effect, possible physical mechanisms of the solid phase influence on the uniform bubble bed were discussed.

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1. Introduction

In bubble column reactors there are two principal flow regimes: homogeneous and heterogeneous (see Fig. 1) (Deckwer, 1992; Kastanek et al., 1993; Molerus, 1993). The *homogeneous* regime is produced by plates with small and closely spaced orifices at low gas flow rates. The bubbles generated at the plate are small, almost spherical and monodisperse, and rise roughly vertically with small vertical and horizontal fluctuations. Coalescence and break-up are negligible and no large-scale liquid circulations occur in the bed. The long-time radial profiles of voidage and liquid velocity are flat. The *heterogeneous* regime is produced either by plates with small and closely spaced orifices at high gas flow rates, or by plates with large orifices at any

gas flow rate (pure heterogeneous regime). This regime is characterized by a wide bubble size distribution, due to generation of large and highly non-uniform bubbles. Bubble coalescence is promoted and macro-scale circulations of the liquid phase are present. The long-time radial profiles are roughly parabolic with a maximum at the centre.

The homogeneous–heterogeneous regime transition is a gradual process of increasing the number and size of coherent structures (circulations) in the bubble bed. The transition is intermittent in space and time and both regimes co-exist in the bubble column. The two flow regimes can be identified from the character of the experimental voidage e –gas flow rate q graph (see Fig. 2). The homogeneous voidage increases progressively with gas flow rate (convex graph) while heterogeneous voidage follows a rational function (concave graph) (e.g. Zahradnik et al., 1997; Ruzicka et al., 2001a).

Bubble column reactors have different behaviour in the homogeneous and heterogeneous regimes, since the rate of

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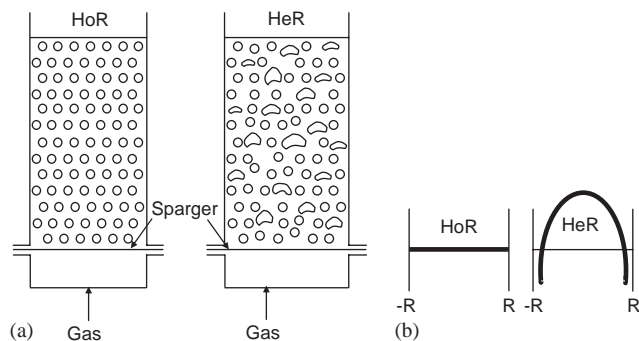


Fig. 1. Definition sketch of homogeneous and heterogeneous flow regimes in bubble columns: (a) flow pattern, and (b) long-term radial profiles of voidage and gas phase velocity.

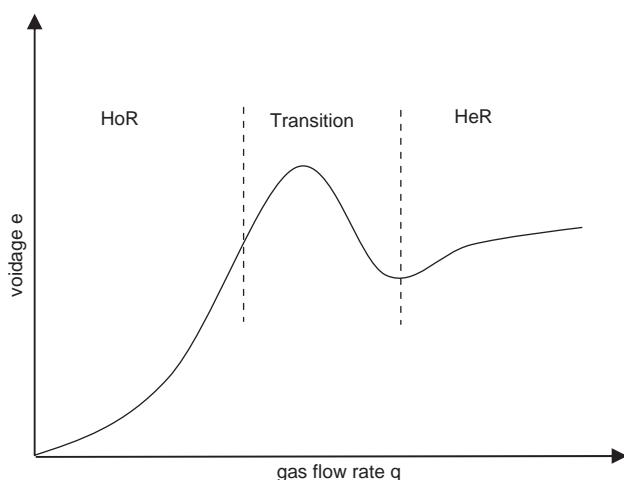


Fig. 2. Typical graph of (voidage e)–(gas flow rate q) dependence $e=e(q)$. Homogeneous regime loses stability in inflection point where regime transition begins.

transport processes depends on the hydrodynamics. Therefore, for rational reactor design and operation, it is of crucial importance to know the range of parameters over which the respective regime prevails. This naturally leads to the stability issue and the regime transition condition. In our previous studies focused on the regime transition, we suggested two stability theories: one kinematic, based on the concept of the Darwinian drift of bubbles (Ruzicka et al., 2001a), and the second, more elaborated, dynamic, based on the analogy with the Rayleigh–Benard instability in thermal convection (Ruzicka and Thomas, 2003). The latter yields a stability criterion for the homogeneous regime in terms of the following parameters: column dimensions, effective viscosity of bubbly mixture, hydrodynamic diffusivity of bubbles. The latter two have a clear physical meaning, but their concepts are not yet well developed, especially at intermediate Re . We also performed experiments to validate particular aspects of the stability criterion, namely, the effect of column dimensions (Ruzicka et al., 2001b) and liquid viscosity (Ruzicka et al., 2003, 2004). In this study, we consider the effect of the

presence of the solid phase, which is not explicitly involved in the above theories.

The behaviour of the gas–liquid–solid systems has been studied for a long time (e.g. Shah, 1979). These systems can be considered as bubbly flows with the presence of solids, or, as liquid–solid fluidized beds with presence of gas bubbles. Therefore, various research communities dealing with bubble columns (Pandit and Joshi, 1984), airlift reactors (Lu et al., 1995), bubbly flows (Douek et al., 1997), flotation columns (Ityokumbul et al., 1995), pulp slurry columns (Xie et al., 2003) and fluidized beds (Muroyama and Fan, 1985) are interested in this complicated three-phase system. Because they operate the equipments under different conditions, the results are not always comparable. One obvious difference is the liquid throughput, which is typically zero in bubble columns, often nonzero in flotation and always nonzero in fluidized beds and bubbly flows. Another difference is in the solid particles, regarding their size, shape, material and surface properties. Big wettable beads likely produce different effects from those by fine hydrophobic particles of a catalyst, or by flexible and sticky fibres in pulp suspensions in paper industry.

Despite the intense research, our knowledge about the possible effects of solids on gas–liquid systems is far from satisfactory. Even less are understood the physical mechanisms underlying the known macroscopic effects. Often, the results are ambiguous or even contradictory. Partly because of comparing results obtained under aforementioned different operating conditions, partly due to complex nature of the solid influence, where many aspects have to be taken into account. The presence of solids affect the gas–liquid mixture in many different ways: bubble formation (Yoo et al., 1997; Luo et al., 1998; Fan et al., 1999), bubble rise (Bly and Worden, 1992; Luo et al., 1997a; Fan et al., 1999), axial (Gandhi et al., 1999) and radial (Warsito et al., 1997) profiles, mixing and dispersion (Smith and Ruether, 1985; Matsumoto et al., 1989), mass transfer (Koide et al., 1984; Quicker et al., 1984; Pandit and Joshi, 1986; Charinpanitkul et al., 1993), and voidage and flow regimes. Unfortunately, it seems that there is no authoritative review available covering in detail this broad area where the reader could be referred to. However, some particular aspects have been reviewed (Pandit and Joshi, 1984; Fan and Tsuchiya, 1990; Fan et al., 1999, see also the proceedings from the GLS Congresses). References are given below, having a relevance for the present study: effect of solids on voidage and flow regimes in bubble columns.

Most of the published work report that the voidage (gas holdup) generally decreases with increasing solid concentration (e.g. Kara et al., 1982; Kelkar et al., 1984; Koide et al., 1984; Banisi et al., 1995 a,b; Lu et al., 1995; Reese et al., 1996; Swart et al., 1996; Jianping and Shonglin, 1998; Fan et al., 1999; Krishna et al., 1999; Zon et al., 2002). Equivalently, the mean bubble speed must increase with solids. This is usually attributed to an increase in bubble coalescence caused by the solids, which results in bigger and faster

bubbles (e.g. Kato et al., 1972; Kara et al., 1982; Lu et al., 1995; Jianping and Shonglin, 1998; Krishna et al., 1999; Zon et al., 2002; Su and Heindel, 2003). An apparent shift in the bubble population from small to large bubbles is documented (Swart et al., 1996). Further, reduction of bubble breakup (Gandhi et al., 1999; Su and Heindel, 2003) and increase of mixture viscosity (Kara et al., 1982; Luo et al., 1997a; Tsuchiya et al., 1997; Jianping and Shonglin, 1998; Fan et al., 1999) are suggested as alternative probable reasons. Another possibility can be the reduction of the space available for the g–l mixture in presence of solids (Lu et al., 1995). Effects of hydrodynamic interactions between bubbles and solids are considered too (Fan and Tsuchiya, 1990). Relative importance of several possible mechanisms (coalescence, mixture density and viscosity, radial profiles, wake effects) causing the decrease of voidage in a particular flotation system has been evaluated (Banisi et al., 1995b).

On the other hand, an interesting dual effect of solids on gas holdup has also been observed (Kara et al., 1982; Pandit and Joshi, 1984; Sada et al., 1986 a,b; Bukur et al., 1990; Khare and Joshi, 1990; Jamialahmadi and Muller-Steinhagen, 1991; Garcia-Ochoa et al., 1997; Xie et al., 2003, see also Table 1 in Banisi et al., 1995a), indicating the presence of two counteracting physical mechanisms. Besides the above mentioned drop in voidage with solids, Kara et al. (1982) also find a subtle opposite effect: “... In fact, systems with 10- μm particles indicated slightly higher gas holdups than obtained from an air–water system. Although the reason for this behaviour is unknown at the present time, it was thought that different wettability ... caused that increase...”. In their review, Pandit and Joshi (1984) state that the solids reduce the gas holdup, but some data in their Fig. 2 indicate that the opposite may also be true. With fine 7 μm wettable solids, Sada et al. (1986a) find a maximum in the (voidage e)–(solid content c) dependence about $c \approx 5\%$, where the unexpected increase is explained by suppression of coalescence by presence of solids in the liquid film between bubbles. A similar result is found also in their sequel paper (Sada et al., 1986b). Next, Bukur et al. (1990) find the dual effect (maximum between $c = 20\%$ and 30%) of small solids (iron oxide, silica) in the batch-mode but not in the continuous mode of column operation, assuming that coalescence suppression prevails in the former and nonuniformities due to inflowing liquid in the latter modes. Khare and Joshi (1990) prove unequivocally the dual effect, with a pronounced maximum at about $c = 0.6\%$ of fine alumina particles. Jamialahmadi and Muller-Steinhagen (1991) report an important difference between wettable (Styrocel) and non-wettable (Nylon, Diakon) particles ($\sim 1\text{ mm}$): the former increase the holdup (suppress coalescence by slowing the drainage) while the latter decrease the holdup (enhance the drainage). Zon et al. (2002) confirm that hydrophobic particles reduce the holdup. On the other hand, Kelkar et al. (1984) report that the wettability enhances the coalescence, and, Banisi et al. (1995a) find that no apparent difference exists between

hydrophilic and hydrophobic particles. The dual effect is observed also for pulp slurry (Xie et al., 2003). Occasionally, under certain conditions, roughly negligible effect of solids on voidage is reported too (Ityokumbul et al., 1995).

Closely related is the effect of the particle size on gas holdup. Usually, a decrease is reported (e.g. Kato et al., 1972; Kara et al., 1982; Jamialahmadi and Muller-Steinhagen, 1991; Lu et al., 1995). Sometimes, an increase is detected (Banisi et al., 1995a). A negligible effect can be found too (Kelkar et al., 1984; Matsumoto et al., 1989; Fan et al., 1999). Pandit and Joshi (1984) resolve this ambiguity by showing that the graph $e = e(d_p)$ has a peculiar structure of four qualitatively different regions: (i) small particles ($Re < 2$) increase the holdup, (ii) medium particles ($2 \leq Re \leq 300$) decrease, (iii) large particles ($Re > 500$) increase it again, and then (iv) the graph saturates at a roughly constant level (see also Pandit and Joshi, 1986). They interpret this peculiarity in terms of bubble size and bubble speed. Garcia-Ochoa et al. (1997) confirm the existence of the first two regions also for heavy polydisperse pyrite particles. Khare and Joshi (1990) study in detail the first two regions and prove that the dual effect of solids on voidage exists in the first region, where small particles suppress the coalescence. Regarding the complicated relations between holdup and solids size and content, i.e., the character of the function $e = e(d_p, c)$, Banisi et al. (1995a) suggest a consensus: small amount of fine particles (suppressing coalescence) and large amount of big particles (break up of large bubbles) tend to increase the holdup (reduce mean bubble speed). Otherwise a decrease can be expected (e.g. much of small particles, medium particles at moderate content, little of big particles).

In spite of all the efforts aimed at the gas holdup studies, the information about the effect of solids on the flow regimes is very scarce. Often, no attempt is made to specify the prevailing flow regime during the experiment. Sometimes, the type of the regime is assessed. For instance, Kara et al. (1982) and Clark (1990) plot the lines corresponding to the homogeneous and heterogeneous regimes in the plane (drift flux j)–(voidage e) and find that their experimental data fall between these two, i.e., the transition regime prevails. In a similar way, Luo et al. (1997b) report stabilization of the homogeneous regime by increased pressure in a three-phase bubble column. Koide et al. (1984) demarcate the respective regimes in the e – q plane by lines of recommended correlations and find their data in the transition and heterogeneous regimes. Kelkar et al. (1984) estimate the heterogeneous regime based on large values of the distribution parameter in the drift flux model. Further, the regime type is assessed visually from the shape of the $e(q)$ graph. Based on this rather subjective method, Jamialahmadi and Muller-Steinhagen (1991) find delayed/advanced transition for wettable/nonwetable solids, i.e., different kinds of particles produce different effects. Reese et al. (1996) find advanced transition for pulp slurry and Krishna et al. (1997) that for fine silica particles. Based on a substantial voidage

drop caused by fine glass powder, Clark (1990) assumes that the original transition regime shifts to the heterogeneous one due to enhanced bubble coalescence.

There are also few studies where the *transition point* is determined. Krishna et al. (1999) use the homogeneous drift-flux model for the critical point (beginning of the transition) and demonstrated by two experimental points that the presence of solids (14% of fine silica in ethanol) dramatically reduces the critical values of gas flow and voidage, hence destabilizes the uniform regime. Su and Heindel (2003) use the heterogeneous drift-flux model for the critical point (end of the transition) to find the same effect of pulp slurry (Rayon fibre).

It follows that the knowledge regarding the flow regimes and their transitions in three-phase systems is indeed far from satisfactory. We lack both data and their interpretation in terms of underlying physical processes. The purpose of this study is to contribute to this subject and examine the effect of solid particles on homogeneous regime stability. For that, two kinds of experiments are done: the basic regime transition study (macro-scale) and an auxiliary visualization study (micro-scale). The results show that the homogeneous regime is stabilized by low solid load, but destabilized by high solid load. The discussion offers some clues towards the explanation of this dual effect.

2. Experiments and data treatment

2.1. Measurements and errors

The measurements were performed in a cylindrical plexiglas bubble column of 0.14 m diameter. The column was equipped with a 3 mm brass perforated plate with 0.5 mm orifices, 10 mm pitch, and relative free area 0.2%. This plate ensures generation of the homogeneous, transition and heterogeneous bubbling regimes. Such a plate is a typical gas distributor for production of the uniform bubbly layers for stability studies, and is also suitable for applications. Typical bubble size in the homogeneous regime was 4–5 mm, with the following features: terminal speed $u_0 \approx 0.2$ m/s, $Re \approx 10^3$, $We \approx 2.7$, $EO \approx 3.3$, $Mo \approx 1.9 \times 10^{-6}$. Compressed filtered air from laboratory lines was the gas phase. Distilled water was the liquid phase. Calcium alginate beads, roughly spherical particles, with equivalent diameter $d_p = 2.1$ mm and density $\rho_p = 1023$ kg/m³ were the solid phase. The choice of the phases corresponds to our interest in three-phase airlift reactors with immobilized biomass. The solids are well-defined completely wettable objects with reasonable rigidity that do not form agglomerates and are big enough not to affect the surface properties of the gas–liquid interface. The following nine values of solid loading were used: 0 (water), 1, 3, 5, 10, 15, 20, 25 and 30 vol%. The clear liquid height was $H = 0.4$ m for all experiments (no liquid throughput). The dependence of the voidage e on the gas flow rate q was measured three times and then averaged.

The gas flow rate varied in the range $q = 0$ –0.1 m/s, which covers the homogeneous regime and part of the transition regime. The gas flow was read from a rotameter. The 33 measuring points covered densely the range from $q = 0.0144$ to 0.0722 m/s, where the transition point were located, with the step of 1.8 mm/s. The gas holdup was determined from the bed expansion. The estimated error of the results is less than 5%.

The claimed 5% is the upper limit for voidage in the range measured (homogeneous regime + part of transitional regime). When the layer is uniform, the surface is stable and horizontal and the interface can be located with precision of 1 mm (resolution of the ruler). For layers with $H \sim 40$ –55 cm (voidage 0–30%) this gives an error 0.25–0.18% in H for the homogeneous range up to the critical point, which causes a comparable error in measuring e , being a function of H . Going further into the transition regime, the surface starts to wave and the uncertainty increases. Its position was determined as the mean over certain number of periods of the oscillations, providing enough data to obtain the deviation within the claimed 5% range. These data, however, are well beyond the transition point and are shown only for depicting the trend of the e – q dependence for larger q . We consider that reading the gas flow from the rotameters was precise.

As for the critical values, the simultaneous application of several methods gives the uncertainty in its determination within one experimental data point. So the error in q_c is the discrete step size in q , i.e., 0.0018 m/s, which amounts to 4.5–5% with the typical values of q_c being 0.035–0.040 m/s (Fig. 6b). Since the data points were connected with a continuous line, the actual precision in determination of q_c is much better and these 4.5–5% represent the upper limit. The error of e_c comprises (i) that of measuring e and (ii) that of determining the transition point on the e -coordinate. The former contribution is the above mentioned less than 1%. The latter relates to the difference between the neighbouring data points. Considering that the e changes from 0 to 0.3 within about 30 measuring points (Fig. 3), the mean step in e is 0.01, which amounts to 5% with the typical values of e_c being 0.2 (Fig. 6a). Since the data points were connected with a continuous line, the actual precision in determination of e_c is much better and the 5% represents the upper limit.

2.2. Evaluation of critical point

The dependence $e = e(q)$ for each solid content c was the primary data. The critical point could be found as the *inflexion point* of the data graph, but its direct determination in this way is difficult and inaccurate.

Therefore, the data were re-plotted according to the *drift-flux model* by Wallis (1969) and the point was determined from the deviation of the data from the theoretical line of the uniform regime. This is a standard procedure. The theoretical

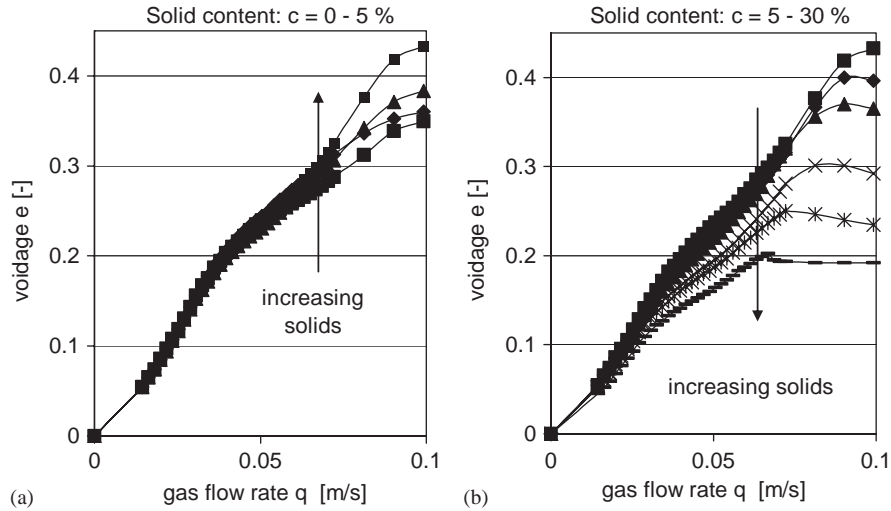


Fig. 3. Primary data: voidage e vs. gas flow rate q : (a) low solid content c increases voidage $c = 0$ vol% (water) (■), 1 vol% (◆), 3 vol% (▲), 5 vol% (■) and (b) higher solid content c decreases voidage. $c = 5$ vol% (■), 10 vol% (◆), 15 vol% (▲), 20 vol% (×), 25 vol% (*) and 30 vol% (—).

line $j = j(e)$ is defined as

$$j_t = e(1 - e)u \quad (1)$$

which must be closed by a formula for the hindered bubble speed u , which is the mean slip speed in case of no liquid flux through the column. Instead of many common empirical relations (e.g. by Richardson–Zaki), we prefer our equation based on the concept of Darwinian drift (Ruzicka et al., 2001a),

$$u = u_0(1 - ae/(1 - e)), \quad (2)$$

where the two parameters have a clear physical meaning: u_0 —bubble terminal speed, a —drift coefficient. For each data line $e(q)$, they are obtained by linearization of Eq. (2), using the basic relation

$$e = q/u \quad (3)$$

which is the mass conservation of the gas phase. The experimental drift flux is obtained from Eq. (1) using Eq. (3),

$$j_e = (1 - e)q. \quad (4)$$

The transition *begins* where Eq. (4) separates from Eq. (1): it is the critical point $[q_c, e_c]$, the instability threshold. The numerical values of q_c and e_c are the quantitative measures of the homogeneous regime stability. The evaluation procedure is an iterative process. First, the homogeneous data range is assessed. Then, these data are used for the linearization. These two steps are repeated, till the correlation coefficient of the linearization is sufficiently close to unity.

Additionally, the regime transition was also found using the *slip-speed concept*, where, at the critical point, the slip speed data given by Eq. (3), $u = q/e$, based on the measured q and e , depart from the model line (2). The homogeneous regime ends where the hindrance disappears. The result is the average over these two methods. Since these two

methods are equivalent, only using different co-ordinates, the results—when evaluated correctly—were close together. This was the test of correctness.

2.3. Criteria for transition

There are not many stability criteria available for the homogeneous regime in g–l system. Even less is known about the criteria designed for g–l–s systems. Those we are aware of are discussed below.

(i) The simplest and sometimes most effective is the *rule of thumb*, that the transition in water–air system under normal conditions begins somewhere around 0.03 m/s and is completed around 0.1 m/s, say. Thus the q_c is expected to be within this interval, depending on its definition: some authors take the critical point at the maximum of $e(q)$ graph, others at the beginning or the end of the transition. We prefer the beginning of the transition range where the instability occurs first (see Fig. 2). For instance, in Krishna's simple and flexible model designed mostly for practical purposes, it is estimated $q_c \approx 0.09$ m/s (Krishna et al., 1991). Empirical criteria of this kind usually stems from long-term experience and are reliable, but not much precise.

(ii) Other kind of criteria are basically *empirical* or *semi-empirical correlations* for the criticals. They are based on experimental data, should be both reliable and precise. Their basic weakness is that they lack the universal character. They usually refer to the particular situations under which the data were collected, and reflect particular effects of only certain parameters. Unfortunately, there are not many of them available at present. One such is by Wilkinson et al. (1992), who suggested a correlation for critical voidage,

$$e_c = 0.5 \exp(-193 \rho_g^{-0.61} \mu^{0.5} \sigma^{0.11}), \quad (5)$$

based on data collected from the literature as well as on their own experiments. The critical gas flow rate q_c is given by e_c/u_s , where u_s is the speed of so-called ‘small bubbles’ that are responsible for the prevailing part of the uniform voidage,

$$u_s = 2.25(\sigma/\mu)(\sigma^3\rho/g\mu^4)^{-0.273}(\rho_g/\rho)^{0.03}. \quad (6)$$

Here, ρ_g is the gas density, μ the liquid viscosity, σ the surface tension, ρ the liquid density, and g the gravity. Another example is the criterion due to Reilly et al. (1994), stemming from an assumption of a specific form of the relation between the gas momentum flux and the voidage. It is based on data obtained in 0.15 m diameter column with water and non-aqueous liquids in the form:

$$e_c = 0.59B^{1.5}(\rho_g^{0.96}\sigma^{0.12}/\rho), \quad (7)$$

$$q_c = (\sigma^{0.12}/2.84\rho_g^{0.04})e_c(1 - e_c), \quad (8)$$

where B is an empirical parameter ≈ 4 , depending on the kind of the liquid.

(iii) The third kind of criteria is based on an underlying *theoretical concept*. Their reliability relies on reliable closures used in the analysis. Depending on their predictive value, they belong to two classes. A posteriori criteria are used for evaluation of the critical point from data already measured. Two common examples are the slip speed concept and the drift-flux model, used also for our data. The former is based on the empirical fact that, in uniform bed, the bubble speed decreases with bubble concentration (hindrance), the latter on the mass conservation of the phases. Both rely heavily on robust closures for the slip speed. A priori criteria are more ambitious and take the form of relations for the criticals. They belong to two qualitatively different groups.

The first group is based on strictly one-dimensional (1D) models of the flow. These have been developed by the nuclear/mechanical engineering community for externally driven g–l flows, flow regimes and their stability, in long and narrow pipes of cooling circuits in nuclear power plants where the liquid speed is large (bubbly flows). These models were adopted/developed by chemical engineers to investigate fluidized beds and bubble columns. These are several studies devoted to 1D bubble columns (e.g. Hoefsloot and Krishna, 1993; Mineev et al., 1999; Joshi et al., 2001; Leon-Becerril and Line, 2001), and further will surely appear due to the relative simplicity of the concept and ease at evaluating the effects of particular forces (e.g. added mass, drag, lift, etc.). The common feature of these models is that the first instability mode is the planar wave mode, where regions of low and large voidage appear periodically along the (infinite) pipe due to the positive feed-back effect produced by the hindrance (the more bubbles the lower their speed). These models are generally not suitable for bubble columns due to completely different conditions: internally (buoyancy) driven flow, short and wide containers, low liquid speed. The effect of the horizontal extent of the column and the presence of the boundaries on all sides prevent us

from treating the bubble columns as a infinitely long 1D systems with flat radial profiles.

The second group is based on two-dimensional (2D) models of the flow. We are currently aware about two of them. First, Shnip et al. (1992) performed linear stability analysis of relatively simple governing equations for gas–liquid flow and obtained an implicit stability criterion for the homogeneous regime in the form:

$$\frac{2g}{\Delta P J' u_0} < \left(\frac{\pi}{D}\right) \frac{\sinh(\pi A)}{\cosh(\pi A) - 1}. \quad (9)$$

Here, ΔP relates to the pressure drop across the plate and J' equals $u + e(\partial u/\partial e)$; both must be obtained from some closure relations, usually empirical. D is the column diameter and A the column aspect ratio H/D . To obtain the critical values, Eq. (9) must be solved for e_c , upon substituting some $u(e)$, e.g. that by Richardson–Zaki. Note that the effect of viscosity is absent in Eq. (9) and can enter only via the closure for u . Second, Ruzicka and Thomas (2003) undertook a different approach, based on the analogy between the buoyancy-driven instability of uniform dispersed layers and the Rayleigh–Benard instability in thermal convection. In both these cases, the original homogeneous state is broken by onset of large-scale circulations when increasing the energy input into the system. The Rayleigh number is the order parameter. This generic physical concept yields the following explicit stability criterion:

$$e < e_c = \frac{\mu^* \kappa}{\rho g} \left(\frac{k_1}{H^3} + \frac{k_2}{H^{3-c} D^c} \right). \quad (10)$$

Here, μ^* is the effective dynamic viscosity of the bubbly mixture and κ is the hydrodynamic diffusivity of bubbles, that must be closed. k_1 , k_2 and c are empirical parameters that depend on the columns size (Ruzicka et al., 2001b). For the bubble column used in this study ($D = 0.14$ m, $H = 0.4$ m), Eq. (10) reads

$$e_c = 2.11 \times 10^5 \mu^* \kappa \quad (11)$$

which predicts a linear increase of the stability with the viscosity and diffusivity. Estimating these two, $\mu^* \sim \mu \approx 10^{-3}$ Pa s and $\kappa \sim (\text{bubble size}) \times (\text{bubble speed}) \approx 0.005 \text{ m} \times 0.2 \text{ m/s} = 10^{-3} \text{ m}^2/\text{s}$, we have for tap water a constant value $e_c = 0.211$.

All the above concepts relate to g–l systems and do not contain the effect of solids explicitly. However, it can be involved indirectly, through the dependence of certain quantities on the solid content. These can be either the constitutive properties of the multiphase system (density, viscosity, diffusivity, etc.), or closure relations for pressure drop, slip speed, etc. Thus, the g–l criteria can in principle be used for the data obtained in this study.

There are also studies devoted to flow regime identification in true three-phase g–l–s systems, mainly liquid-fluidized beds. The main difference from bubble columns is the essentially non-zero liquid speed, because the liquid

passes through the system. Since the flow maps are usually plotted in the co-ordinates ‘liquid speed’ and ‘gas speed’, only the ‘gas speed’-axis is applicable to bubble columns. For instance, Zhang et al. (1997) classify up to seven different flow regimes of the three-phase flow and present correlations for the boundary lines separating them in the parameter plane. Most of these regimes do not occur in a typical bubble column (e.g. slug, bridging, annular). For the lowest value of the liquid speed considered, 10^{-3} m/s, their map predicts transition from discrete bubbly flow to slug flow at gas speed ≈ 3 cm/s, which is not observed in bubble columns. The authors also warn the reader: “*Note that this map . . . is applicable only for the size of column and type of distributor studied. In addition, the map is strictly only applicable at the height of the measurement probe . . .*”. With their column width 0.0826 m, column height 2 m, 2-mm orifices, and probe position 0.65 m, their flow maps are completely irrelevant for our data.

Nowadays, we can do nothing but agree with Krishna et al. (1993): “*There is a need to better understand flow regime transitions and the development of a unified theory of multiphase flow regime transitions will be useful and enlightening*”. The convective theory of stability of dispersed layers based on the generic physical concept (Ruzicka and Thomas, 2003) is one step towards this distant goal. Also, regarding the g–l–s systems, the statement by Fan et al. (1999) “*The studies of the regime transition in three-phase fluidized beds and slurry bubble columns are scarce*” is still valid. The present study contributes to filling in this gap.

2.4. Visualization experiments

Auxiliary visualization experiments were done in order to investigate the three phase systems in more detail, namely the pattern of the bubble–particle interactions, to obtain arguments to support some possible mechanisms responsible for the observed trends.

The measurements were performed in a cylindrical plexiglas bubble column of 7 cm diameter and 0.84 m high. At the bottom, the column was equipped with one 0.3 mm inner diameter needle for generation of bubbles of similar size like those in the main experiments. Compressed air from laboratory lines passing through a microvalve was the gas phase. The liquid phase was tap water. The same alginate beads as before were used. In some experiments, a narrow glass tube (6 or 14 mm diameter) was placed into the column to facilitate frequent and intense contact between bubbles and particles.

Two cameras were used for the visualization. First, commercial analogue Panasonic S-VHS-C camera, connected to cassette recorder and monitor. Second, high-speed digital system Kodak EKTAPRO, with speed up to 10 000 fps. The images were downloaded from the fast memory unit through a SCSI interface to PC. The pictures were taken from two different places in the column: at the bottom and 0.255 m above

it. Different situations were studied: behaviour of bubbles in the gas–liquid system, and the effect of particle–bubble interactions in the gas–liquid–solid system.

3. Results

3.1. Primary data: voidage–gas flow rate

The plot of the basic $e(q)$ graphs is shown in Fig. 3. For low solid content, $c \leq 5\%$, the data presented in Fig. 3a witness a significant increase of the voidage with increasing the solid load. On the other hand, at larger content $c \geq 5\%$, the voidage displays a substantial reduction in Fig. 3b. This dual effect of the particles on the gas holdup is interesting, since it indicates the presence of two competing mechanisms, one stabilizing and the other destabilizing the uniform three-phase system. This result also reconciles the contradictory findings reported in the literature. The quantitative aspect of the change in voidage is documented in Fig. 4a. The corresponding variations in the mean speed of the gas phase is shown in Fig. 4b. Note that the data in Fig. 4 belong to the transition regime, where the bubble speed is enhanced by the liquid circulations.

3.2. Secondary data: drift-flux plot

A demonstration of the determination of the critical point based on the drift-flux model is presented in Fig. 5. The data are shown in the co-ordinates voidage e –drift flux j , according to Eqs. (1) and (4). It is clearly seen, where the data separate from the uniformity line.

3.3. Main result: stability

The critical values of voidage e_c and gas flow rate q_c are plotted versus the solid content c in Fig. 6. The picture unequivocally demonstrates the ambiguous effect of the solid particles on the stability of the homogeneous flow regime. The stabilization occurs at low solid load, $c \leq 3\%$, and the destabilization at higher load, $c > 3\%$. Qualitatively, the behaviour of the voidage in Fig. 3 and the critical voidage in Fig. 6 is similar: both have a maximum with respect to the solid content. Quantitatively, there is a little discrepancy: the maximum is at $c = 3\%$ in Fig. 6 while at $c = 5\%$ in Fig. 3. This suggests that the shapes of the $e(q)$ graphs are not universal in the following sense: there are exceptions to the expected rule: the larger the voidage, the larger the critical voidage. The absolute values of the criticals in Fig. 6 may seem rather low, $q_c \sim 0.035$ – 0.04 m/s and $e_c \sim 0.13$ – 0.2 , say. This is because they refer to the beginning of the transition process.

The stabilization effect amounts to 11% of increase in q_c and 13% of increase in e_c , relative to the g–l air/water system. The increase of e_c between $c = 0\%$ and 1% can be

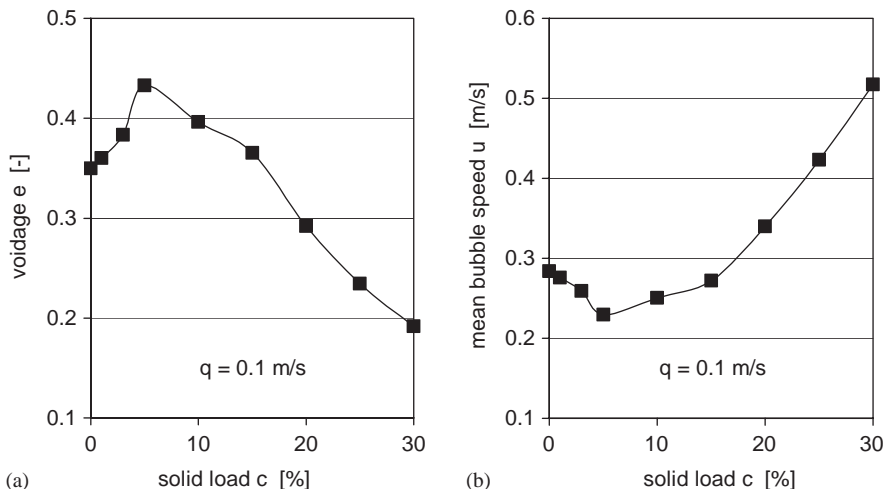


Fig. 4. Primary data: effect of solid content c on (a) voidage e and (b) mean bubble speed u .

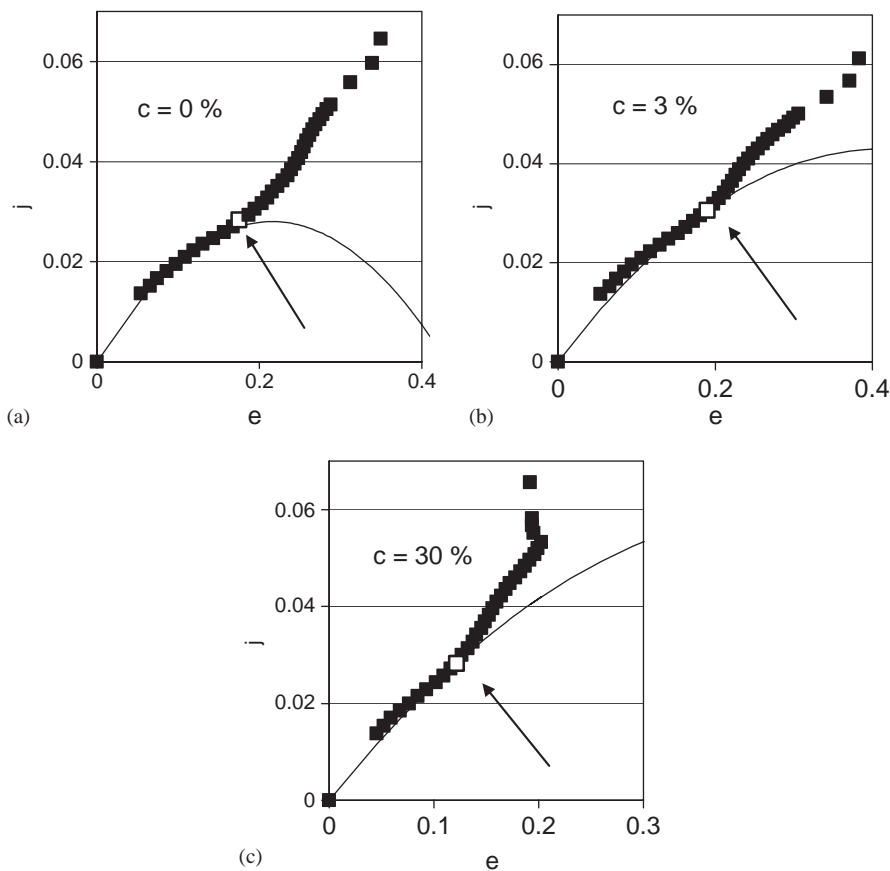


Fig. 5. Secondary data: drift-flux plot of drift flux j (m/s) vs. voidage e . Model—smooth line of j_T by Eq. (1). Data—marks of j_c calculated by Eq. (4). Blank data point—critical point. Different solid loads: (a) $c = 0\%$, (b) $c = 3\%$, and (c) $c = 30\%$.

described by a modified Eq. (11),

$$e_c = 0.175 \times 10^5 \mu^* \kappa (1 + 2.23 f), \tag{12}$$

where the original critical voidage 0.211 of g–l system with tap water was replaced by a somewhat lower value 0.175 for distilled water, with $f = c/100$ being the solid volume fraction. Eq. (12) should not be con-

sidered a reliable correlation; it only demonstrates how to incorporate the stabilizing effect of solids into the stability criterion (10) and indicate its magnitude. The destabilization trend in Fig. 6a can well be fitted with a straightline:

$$e_c = 0.21 - 0.25 f \quad (R_{xy} = 0.987). \tag{13}$$

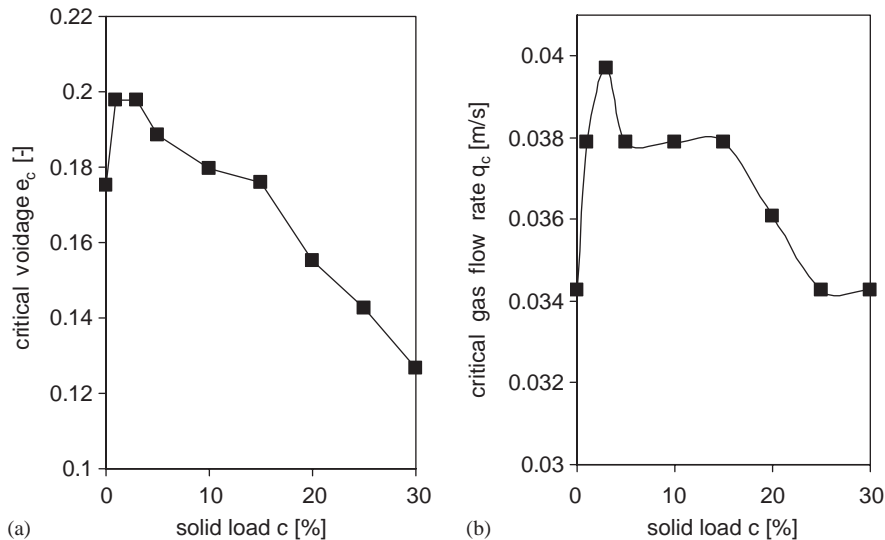


Fig. 6. Main result: homogeneous regime stability measured by critical values of (a) voidage e_c and (b) gas flow rate q_c .

4. Discussion

The purpose of this discussion is to provide some suggestions regarding the explanation of the observed dual effect of the solids: first increase and then decrease of both the voidage e and the critical voidage e_c , hence increase and decrease of the homogeneous regime stability. Correspondingly, by Eq. (3), the presence of solids first reduces and then enhances the mean bubble rise speed.

Strictly speaking, the suspended solids should be regarded as a new phase, and the original set of equations for g–l system should be expanded to g–l–s system. The difference between the solutions of the respective sets of governing equations is exactly the ‘effect of solids’, hard to predict. Therefore, we resort to making a list of particular effects known from literature, suggesting possible ways how can the presence of the solid phase affect the behaviour of bubble bed. First, the corresponding physical mechanisms is explain, then the magnitude of the effect in case of our experimental data is assessed. In quantitative evaluations, the following relations between the voidage and the quantities that can be directly affected by the solids can be used: $e \sim 1/u$ by Eq. (3), $u \sim u_0$ by Eq. (2), $u_0 \sim ((\rho - \rho_g)d/\rho C)^{0.5} \approx (d/C)^{0.5}$, since $\rho \gg \rho_g$, so that at an error of order $O(10^{-3})$ the bubble speed does not explicitly depend on liquid density, $C \sim 1/Re$, $Re = \rho d u_0 / \mu$ (consequently, $e \sim \mu^{0.5}$). Since the possible effects depend on the solid load, they are evaluated at the point where the stability diagrams in Fig. 6 change their trends, i.e., at the characteristic value $c \approx 3\%$, the corresponding solid volume fraction f being ≈ 0.03 .

(i) The first effect is the steric effect, consisting in the simple fact that the solids occupy certain space of the column. Consequently, the bubble concentration is different whether based on g–l or g–l–s volume. At any given q , the effective bubble concentration e^* based on the g–l volume is by a

factor $1/(1-f)$ larger than the common voidage e used here and based on the g–l–s volume. Thus also the true critical value e_c^* is reached sooner, at lower gas input, hence destabilization. This effect can be particularly strong at large f , i.e., at high solid loads.

In our case, when the solid content is $c \approx 3\%$, this destabilizing effect is weak, $1/(1-f) \approx 1.03$, i.e., about 3%. However, at large c of 20–30%, this effect can contribute to the instability (descending branch in Fig. 6).

(ii) The second effect is the density effect. Although the density itself should not affect the single bubble rise, the influence of solids can be estimated in terms of effective (mixture, apparent) density: $\rho^* = (1-f)\rho + f\rho_p$. The concept of effective density, hence buoyancy, applies only when the size d of a body (here a bubble) immersed in a dispersion is much larger than the size d_p of the dispersed particles, the quantitative criterion for the body and the particles of like shapes being: $d > d_p/f^{1/3}$.

In our case, with almost neutrally buoyant dispersed particles ($\rho_p = 1023 \text{ kg/m}^3$), the solid–liquid density difference is small as $O(10^{-2})$, so that an effect of the same magnitude is expected due to the effective density—if applicable. Evaluation of the criterion for applicability of the concept of effective density with $d \approx 0.004 \text{ m}$, $d_p \approx 0.002 \text{ m}$, and both the characteristic solid load $f \approx 0.03$ and maximum solid load $f = 0.3$, we obtain $0.004 > 0.0065$ and $0.004 > 0.003$, respectively. This means that the concept is inappropriate at low solid load and only very weakly applies at large solid load (the l.h.s. is not ‘much larger’ than the r.h.s.). We conclude that the possible density effect is very minute.

(iii) The third effect is the viscosity effect. It relates to one particular change of the liquid flow field caused by the presence of solids. Each particle in the flow presents a new boundary surface with the no-slip condition, where the liquid velocity must accommodate to zero. Therefore,

additional velocity gradients arise and the viscous dissipation increases. This is reflected by the effective (mixture, apparent) viscosity μ^* of a suspension, which is larger than that of a pure fluid and increases with the particle content. Consequently, the free rise speed of a buoyant body is reduced; not because of higher friction at the surface—it experiences the pure fluid, but because of higher capacity of the flow for absorbing the energy released by the body motion. Therefore, the concept of effective viscosity applies generally, whenever the dispersed particles are presented, regardless of their relations to the immersed body, as for the shapes, sizes, etc. With bubbles, the reduction of bubble rise speed results in larger voidage at the same gas input ($e \sim 1/u_0$), hence stabilization. On the other hand, the bubble coalescence is promoted in viscous media, bigger and fast bubbles are formed, which results in lower voidage, hence destabilization. Thus, the viscosity plays a dual role in the stability of uniform bubble bed. There are many studies devoted to elaborating formulas for μ^* (e.g. Thomas, 1965; Barnea and Mizrahi, 1973; Tsuchiya et al., 1997; Cheng and Law, 2003). A common form is a power series $\mu^*/\mu = 1 + b_1 f + b_2 f^2 + \dots$, with the coefficients of $O(10^0)$. The resulting figure can be modified by the fact that the effective viscosity increases with particle size (for d_p larger than $\sim 10^1 \mu\text{m}$), particle density and particle anisometry (e.g. Clarke, 1967).

In our case, we had relatively large particles, but spherical and almost neutrally buoyant. At low solid load, with typical $f \approx 0.03 \sim O(10^{-2})$, the viscosity effect is of the same leading-order $O(10^{-2})$. Using the standard value $b_1 = 2.5$ it gives $\mu^*/\mu = 1.075$. Since $e \sim \mu^{0.5}$, the effect voidage is $e^*/e \approx 1.037$, i.e., about 3–4%. This effect (reduction of bubble speed) can contribute to the increase of stability in Fig. 6. At high solid load, with $f \approx 0.2\text{--}0.3$, this effect is of $O(10^{-1})$ in viscosity, giving $\mu^*/\mu \approx 1.75$. This may not be enough to promote a massive coalescence (coalescence was not actually observed), so that another effect must be responsible for the decrease of stability in Fig. 6.

(iv) The fourth effect concerns the physical chemistry of surfaces. Depending on the interfacial properties of the g–l–s system (hydro-philicity/phobicity, wettability, etc.), particles tend to increase or reduce their concentration near the g–l interface. The deposition at the bubble surface affects the original slip boundary condition. Stabilization of the surface then causes higher drag, hence lower rise speed. Also, bubble shape oscillations can be affected, and the result in terms of bubble speed is difficult to assess. The concentration differences along the interface can serve as a driving force for various processes and complicated electrokinetic phenomena can be encountered. Changes in the interfacial properties affect the tendency to coalescence and breakup. These effects will be strong in case of small particles, much smaller than bubbles.

In our case, the particles are big (comparable with bubble size) and completely wettable. Therefore, no interface effects are expected.

(v) The fifth effect concerns the bubble size at detachment, when formed in a suspension. In systems with a small effect of particle inertia, the influence of solids is negligible (Yoo et al., 1997), which is also our case. On the other hand, in the opposite case, the bubble size generally increases due to additional downward forces exerted by settling solids on the growing bubble (Luo et al., 1998). At low gas flow (lower than necessary for complete suspension, e.g. Roy et al., 1964; Pandit and Joshi, 1984), the solids settle on the plate and the bubbles coalesce there (Ityokumbul et al., 1995).

In our case, with neutrally buoyant particles $\rho_p \approx \rho$, we did not observe intensive particle settling. We conclude that this effect can be neglected.

(vi) The sixth effect relates to bubble rise velocity in suspension. The contribution of effective viscosity is treated in (iii). Here, the effect of direct bubble–particle interactions is considered. Generally, the presence of particles reduce the bubble speed (Fan and Tsuchiya, 1990; Luo et al., 1997a), the reason being the hydrodynamic forces and mutual collisions. Both delay the bubble motion. One aspect of the retardation is the hindrance effect from particles to bubbles. It can be expressed in form of a series, $u_0^*/u_0 = 1 + B_1 f + B_2 f^2 + \dots$, with the coefficients of $O(10^0)$ (e.g. Bly and Worden, 1992). Taking $B_1 \approx 5$ and $f \approx 0.03$, one obtains 15% effect. Another effect is the lateral bubble motion induced and/or enhanced by collisions with the particles. The buoyant potential energy of a bubble is partitioned into more degrees of freedom to the detriment of the vertical velocity component. It results in a net reduction of the mean rise speed.

In our case, this effect is documented by the auxiliary visualization experiments focused on a simple situation. Fig. 7 shows a typical collision event, where a bubble is deflected from its original trajectory after the contact with a particle. These events were frequent in the column and can contribute to the increase of stability in Fig. 6. Preliminary estimates indicate that the speed reduction could be 5–15%. Consequently, the hydrodynamic g–s interactions at low c can be important in stabilizing the bubble bed by reducing the vertical component of the bubble speed.

(vii) The seventh effect relates to bubble coalescence in suspension. This is usually considered to be the reason for the destabilizing effect of the solid phase. The properties of the solids are very important here. Depending on their size, density and surface properties (wettability), they can both suppress and promote the coalescence, the detail mechanism of which has not been fully understood yet.

In our case, during the visualization experiments we observed an increase in number of coalescing events with increasing solid content. Fig. 8 shows a typical situation where the rise of two bubbles is hindered by a small cloud of solids so that they remain in contact for a time long enough to complete the coalescence process. We did not succeed in assessing this effect quantitatively.

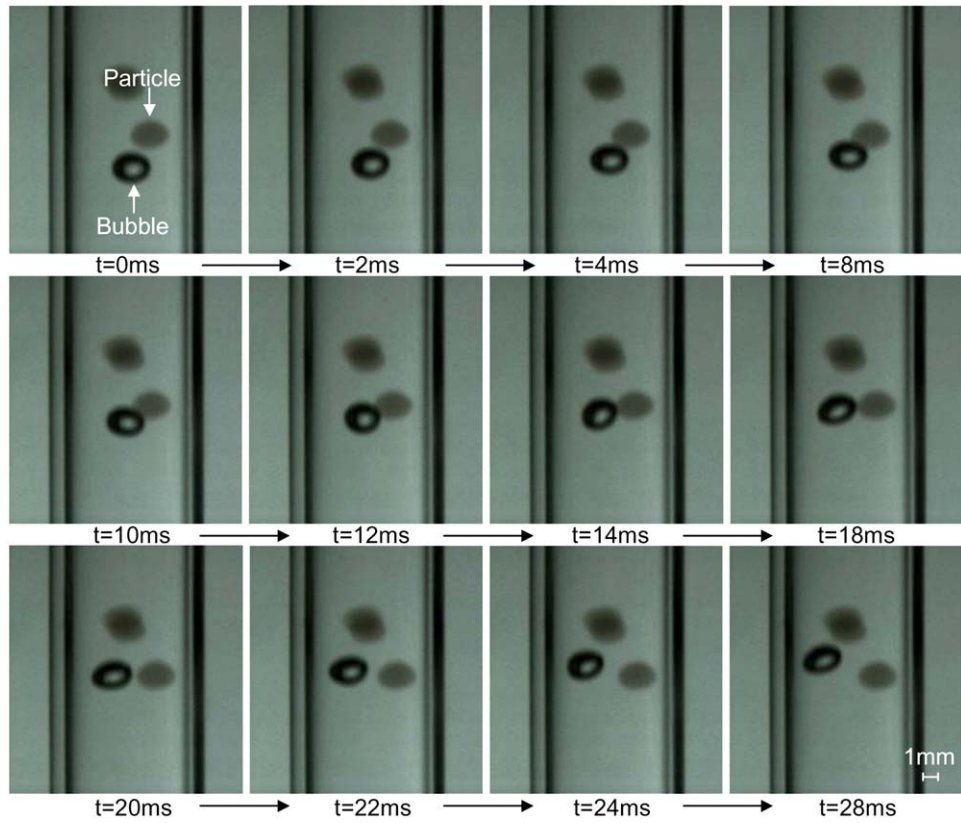


Fig. 7. Auxiliary visualization experiment. Demonstration of bubble deflection from vertical direction after collision with a solid particle.

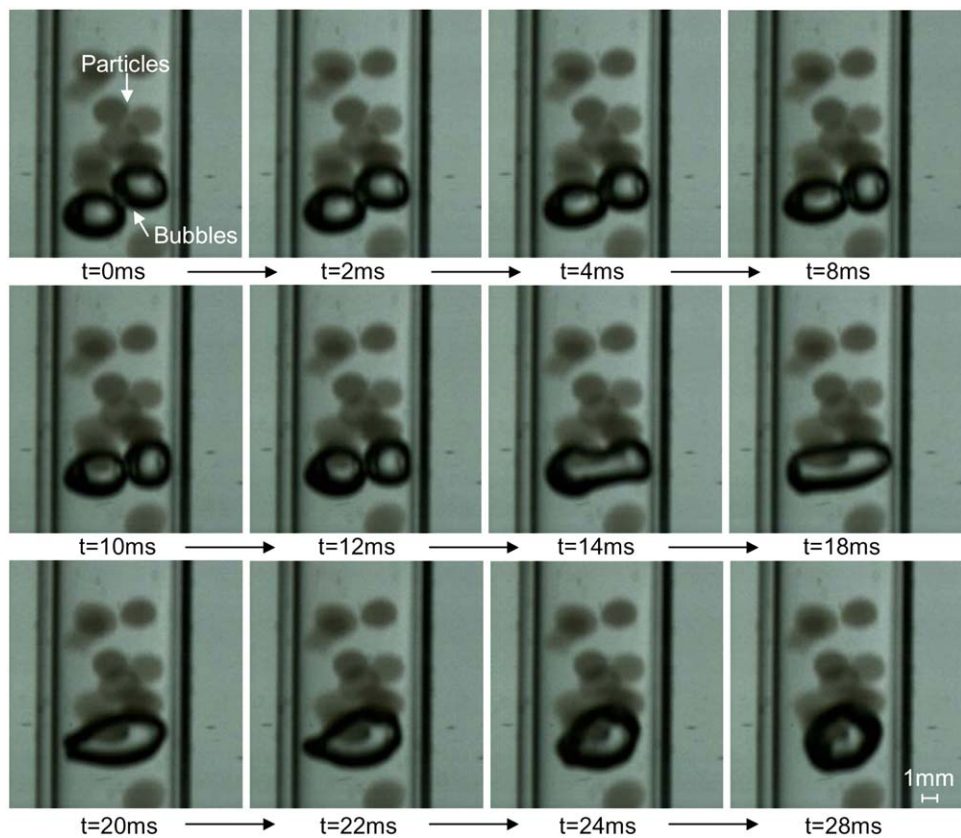


Fig. 8. Auxiliary visualization experiment. Demonstration of bubble coalescence induced by collision with a swarm of solid particles.

(viii) The last possible effect mentioned here relates to spatial inhomogeneities in distribution of solid particles. The homogeneity of the three-phase bed can be broken by nonuniformities originated in any of the two dispersed phases. When pronounced radial profiles develop in the solid phase, the flow regime transition can occur even if the bubbles are distributed uniformly. On the other hand, a statistically uniform distribution of solids can act against the clustering tendency of the gas phase, hence stabilize the bed. Thus, interactions between two phenomena should be considered: (1) fluidization (sedimentation) of solids by liquid and (2) generation of bubbly layer, both uniform at low gas input and solid load. The mechanism of breakage of the uniformity in both cases is believed to be the advection of randomly formed buoyant clusters that introduces the large-scale motions, circulations. The clustering tendency of the dispersed phases finds its long-term expression in the nonuniform spatial profiles.

In our case, with particles and bubbles of comparable size, we can presume a comparable tendency to formation of clusters as a result of action of hydrodynamic forces. Since the g–l density difference $\sim O(10^2)$ is much larger than the s–l difference $\sim O(10^{-3})$, the clusters of solids can generate only very small destabilizing buoyant energy, when compared with clusters of bubbles. Therefore, we assume that the nonuniformity starts in the gas phase first.

Note that qualitatively same dual effect on the stability of the homogeneous flow regime exerted by the presence of solids has been found also for another important parameter—the liquid viscosity: small viscosity stabilizes uniform bubble bed while large viscosity destabilizes the bed, the underlying physical mechanism being currently under study (Ruzicka et al., 2004).

5. Conclusions

The effect of solid particles on homogeneous regime stability and regime transition in a three-phase bubble column was investigated experimentally. The stability was expressed by the critical values of gas holdup and gas flow rate. The experiments showed that for low solid loading (0–3%) the homogeneous regime is stabilized, while for higher solid loadings (> 3%) destabilization occurs. Several possible physical mechanisms underlying this dual effect were discussed. The visualization indicated the importance of hydrodynamic bubble–particle interactions.

Notation

a	coefficient of Darwinian drift, dimensionless
c	solid content in bubble column, vol%
C	bubble drag coefficient, dimensionless
d	bubble diameter, m
d_p	solid particle diameter, m

D	column diameter, m
e	voidage, gas holdup, dimensionless
f	solid content in bubble column $f = c/100$, dimensionless
g	gravity, m/s^2
H	column height (clear water height), m
j	drift flux, $\text{m}^3/\text{m}^2 \text{ s}$
q	gas flow rate, m/s
Re	Reynolds number of a single bubble, $Re = \rho d u_0 / \mu$
u	mean bubble rise speed (mean velocity of gas phase in column), m/s
u_0	terminal bubble speed, m/s

Greek letters

κ	hydrodynamic bubble diffusivity, m^2/s
μ	liquid dynamic viscosity, Pa s
ρ	liquid density, kg/m^3
ρ_g	gas density, kg/m^3
ρ_p	solid particle density, kg/m^3
σ	interfacial tension (liquid surface tension), N/m , J/m^2

Subscript and superscript

c	critical value (threshold of homogeneous regime instability)
*	'effective' value

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