

## RESEARCH NOTES

## Influence of Gas Density on the Stability of Homogeneous Flow in Bubble Columns

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Stability analysis is used to show that an important parameter determining the stability of homogeneous bubbly flows in a bubble column is the Richardson-Zaki exponent  $n$  in the bubble swarm velocity relationship  $U_{\text{swarm}} = v_{\infty}(1 - \epsilon)^n$ . Analysis of bubble swarm velocity shows that with increasing gas density the value of this exponent decreases; physically this means that increasing gas density results in reduced interaction between neighboring bubbles and, consequently, reduced chance of propagation of instabilities. This provides a rationalization of the experimental observation that the influence of increased gas density,  $\rho_G$ , is to delay the transition from homogeneous bubbly flow to churn-turbulent flow.

Industrial gas-liquid bubble column reactors are often operated at high pressures but have become the subject of experimental studies only relatively recently (Clark, 1990; Hikita et al., 1980; Idogawa et al., 1986; Oyevaar, 1989; Öztürk et al., 1987; Reilly et al., 1986; Wilkinson, 1991). These studies show that the influence of increased gas density is to significantly increase the gas holdup. In an initial attempt to explain the influence of increased gas density on bubble column hydrodynamics, Krishna et al. (1991) suggested a simple mechanistic model pictured in Figure 1. The essence of their model is as follows:

(i) The gas holdup varied linearly with the gas velocity till the regime transition point  $U_{\text{trans}}$  is reached.

(ii) At a superficial gas velocity,  $U$ , greater than  $U_{\text{trans}}$  the gas holdup is a sum of two contributions: (a) the small bubble holdup, which is constant and equal to the total gas holdup at the velocity  $U_{\text{trans}}$ , and (b) the holdup of large fast-rising bubbles.

(iii) The influence of increased gas density is to delay the transition from homogeneous to heterogeneous, or churn-turbulent, flow regime; see Figure 2. Put another way, increased gas density tends to stabilize the homogeneous bubbly flow regime.

While this simple model of Krishna et al. (1991) was successful in correlating gas holdup data over an extremely wide range of conditions, their key result given in Figure 2 represents an entirely empirical observation. The express aim of the present paper is seek a theoretical rationalization for the observed stabilizing influence of increased gas density on the homogeneous flow regime. The stability analysis of Biesheuvel and Gorissen (1990) is used as a starting point.

## Stability Analysis of Biesheuvel and Gorissen (1990)

Biesheuvel and Gorissen (1990) have presented a theoretical analysis of stability of homogeneous bubbly

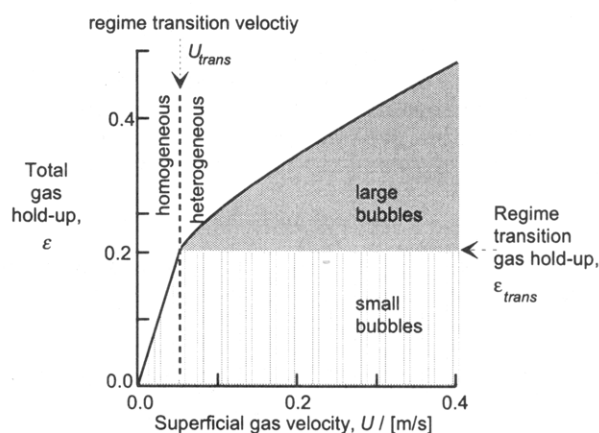


Figure 1. Model for gas holdup in bubble columns in the homogeneous and heterogeneous flow regimes. After Krishna et al. (1991).

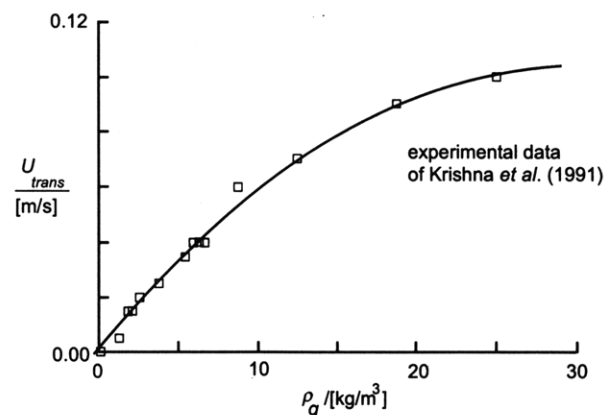


Figure 2. Dependence of the regime transition velocity  $U_{\text{trans}}$  on the gas density  $\rho_G$ . Data from Krishna et al. (1991) for 0.16-m-diameter bubble column with water as the liquid phase and system pressures ranging from 0.1 to 2 MPa with a variety of gases used as dispersed phase (nitrogen, carbon dioxide, argon, helium, and sulfur hexafluoride).

flows subject to void fraction disturbances. They develop the following criterion for instability of homogeneous bubbly flow:

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$$-\epsilon^2 \frac{\partial v_0}{\partial \epsilon} \frac{\partial}{\partial \epsilon} \left[ \left( \rho_G + \frac{1}{2} \rho_L m_0 \right) v_0 \right] \geq - \left[ \frac{\partial p_e}{\partial \epsilon} + \frac{12 \pi a \mu_L}{(4/3) \pi a^3} \frac{1}{(1-\epsilon)^p} \delta_e \right] \quad (1)$$

where we retain the nomenclature of Biesheuvel and Gorissen (1990) and take the opportunity of pointing out a sign error in the first right member of their final result, eq 61. Replacing  $\geq$  in eq 1 with an  $=$  allows calculation of the maximum, stable, gas holdup  $\epsilon_{\text{trans}}$  for homogeneous bubbly flow. The physical significance of the various parameters in eq 1 are given below.

$v_0$  is the velocity of the bubble swarm in the zero volume flux frame; this velocity is related to the single, isolated, bubble rise velocity,  $v_\infty$ , by

$$v_0 = v_\infty (1 - \epsilon)^p \quad (2)$$

In the laboratory fixed reference frame the bubble swarm velocity is (Wallis, 1969)

$$U_{\text{swarm}} \equiv \frac{U}{\epsilon} = \frac{v_0}{1 - \epsilon} = v_\infty (1 - \epsilon)^n \quad (3)$$

where the Richardson-Zaki exponent  $n = p - 1$ .

For a bubble of radius  $a$  the effective mass of the bubble is  $(4/3) \pi a^3 (\rho_G + (1/2) \rho_L m_0)$ , where  $(4/3) \pi a^3 ((1/2) \rho_L m_0)$  is the added mass contribution and  $m_0 = (1 + 2\epsilon)/(1 - \epsilon)$  is the correction to this contribution due to gas holdup (Biesheuvel and Spoelstra, 1989). The term  $\delta_e$ , the effective diffusivity of the bubble swarm, is taken to be proportional to the bubble radius and the mean velocity fluctuation

$$\delta_e = \alpha a [\Delta v^2]^{1/2} \quad (4)$$

The constant of proportionality  $\alpha$  is taken to be unity in the Biesheuvel and Gorissen analysis and the mean velocity fluctuations calculated from

$$\Delta v^2 = \frac{\epsilon}{\epsilon_{\text{cp}}} \left( 1 - \frac{\epsilon}{\epsilon_{\text{cp}}} \right) v_0^2 \quad (5)$$

where the correction factor  $(\epsilon/\epsilon_{\text{cp}})(1 - (\epsilon/\epsilon_{\text{cp}}))$  meets requirements of vanishing of the velocity fluctuations in the limit  $\epsilon \rightarrow 0$  and in the limit of closest packing of the spherical bubbles  $\epsilon \rightarrow \epsilon_{\text{cp}}$ .

The term  $p_e$  represents the kinetic contribution to the so-called "effective" pressure

$$p_e = \epsilon (\rho_G + (1/2) \rho_L m_0) \Delta v^2 \quad (6)$$

which is the product of the effective density and the mean velocity fluctuation.

The stability criterion (1) can be understood as follows. A displacement  $dz$  of the bubble would require a force equal to

$$-\epsilon^2 \frac{\partial v_0}{\partial \epsilon} \frac{\partial}{\partial \epsilon} \left[ \left( \rho_G + \frac{1}{2} \rho_L m_0 \right) v_0 \right] \frac{d\epsilon}{dz}$$

this is a negative force which indicates that the bubbles are attracted to regions of larger void fraction. This force is opposed by the force arising from random motion of the bubbles

$$\left[ \frac{\partial p_e}{\partial \epsilon} + \frac{12 \pi a \mu_L}{(4/3) \pi a^3} \frac{1}{(1-\epsilon)^p} \delta_e \right] \frac{d\epsilon}{dz}$$

tending to equalize bubble voidage fluctuations. If the force which tends to drive the bubbles in the direction of larger void fraction is larger than the opposing force causing

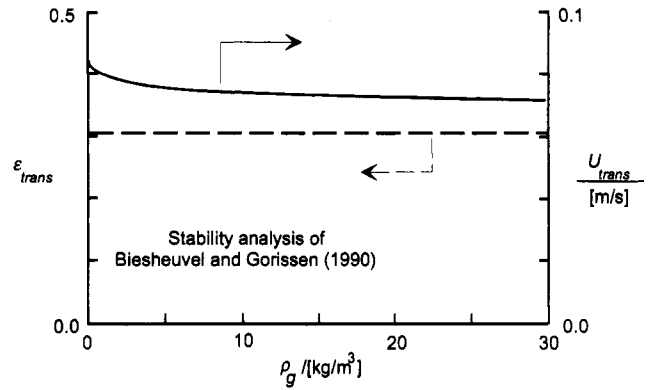


Figure 3. Dependence of  $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$  on the gas density. Theoretical predictions using the original stability analysis of Biesheuvel and Gorissen (1990).

voidage equalization, the homogeneous bubbly regime is unstable and transition takes place to the churn-turbulent regime.

The Biesheuvel-Gorissen criterion (1) for instability involves the following system parameters:  $\rho_G$ ,  $\rho_L$ ,  $a$ ,  $\mu_L$ ,  $v_\infty$ , and the exponent  $p$ . In their analysis of the air-water system at atmospheric pressure conditions, Biesheuvel and Gorissen (1990) take  $p = 2$ , i.e.,  $n = 1$ .

For the set of experiments portrayed in Figure 2, the liquid-phase properties are  $\mu_L = 0.001$  Pa s,  $\rho_L = 1000$  kg  $\text{m}^{-3}$ , and  $\sigma = 0.072$  N  $\text{m}^{-1}$ . The bubble diameter,  $d = 2a$ , can be estimated from the correlation of Wilkinson (1991)

$$\frac{d^2 (\rho_L - \rho_G) g}{\sigma} = 8.8 \left( \frac{U_{\text{ML}}}{\sigma} \right)^{-0.04} \left( \frac{\sigma^3 \rho_L}{g \mu_L^4} \right)^{-0.12} \left( \frac{\rho_L}{\rho_G} \right)^{0.22} \quad (7)$$

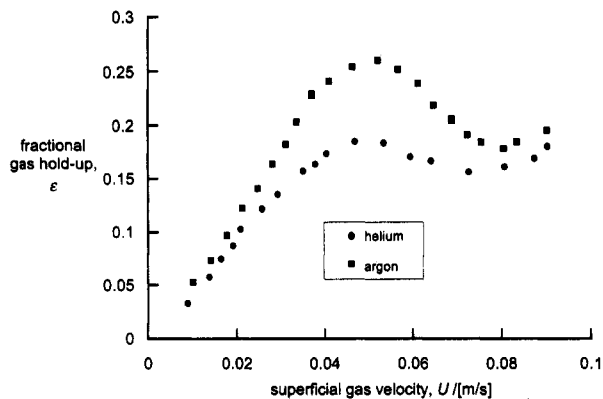
The single bubble rise velocity can be estimated using the small bubble velocity correlation of Wilkinson (1991), valid in the homogeneous flow regime

$$\frac{v_\infty \mu_L}{\rho} = 2.25 \left( \frac{\sigma^3 \rho_L}{g \mu_L^4} \right)^{-0.273} \left( \frac{\rho_L}{\rho_G} \right)^{0.03} \quad (8)$$

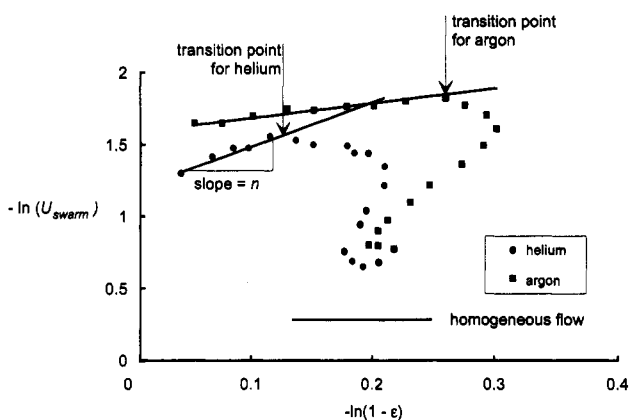
With the above parameter estimates the gas holdup at the regime transition point,  $\epsilon_{\text{trans}}$ , was determined using the equality sign in eq 1; calculations for a range of gas densities are shown in Figure 3. Also shown in Figure 3 are calculations for the superficial gas velocity at the regime transition point  $U_{\text{trans}}$ . The results in Figure 3 show that  $\epsilon_{\text{trans}}$  is practically constant and independent of the gas density.  $U_{\text{trans}}$ , on the other hand, decreases, though slightly, with increasing gas density. Clearly, the analysis of Biesheuvel and Gorissen (1990) is unable to provide an explanation of the experimental results shown in Figure 2.

### Modified Stability Analysis

An important assumption made in the analysis of Biesheuvel and Gorissen (1990) is with regard to the value of the Richardson-Zaki exponent  $n = p - 1 = 1$ . Batchelor's (1988) analysis of the analogous situation of the stability of a homogeneously fluidized gas-solid bed shows that the Richardson-Zaki exponent is an important parameter determining stability. Intuitively one can understand the importance of the exponent  $n$  on the stability of the system; this parameter determines the interaction between the particles (equivalent to bubbles). If  $n$  is large, the particles "feel" each other much more than with small  $n$ . Further, examination of experimental data on the Richardson-Zaki exponent  $n$  for gas-solid fluidized beds shows that the exponent decreases with increasing gas density (cf. Ri-



**Figure 4.** Gas holdup  $\epsilon$  as a function of superficial gas velocity  $U$  for helium-water and argon-water systems. Measurements by Koetsier et al. (1976) in a 0.05-m-diameter column with sintered plate gas distributor.

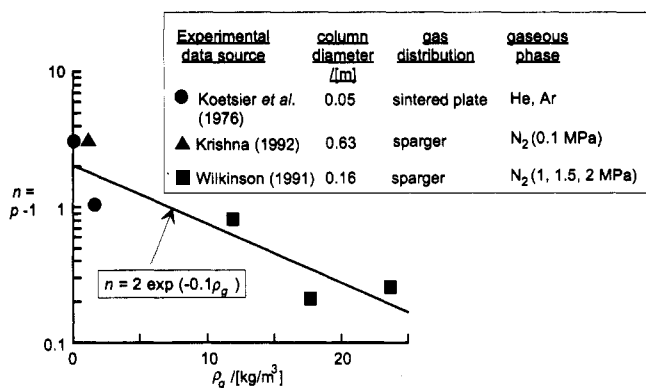


**Figure 5.** Log-log plot of the swarm velocity  $U_{\text{swarm}} (\equiv U/\epsilon)$  versus  $(1 - \epsilon)$  for helium-water and argon-water systems. Data calculated from Figure 4.

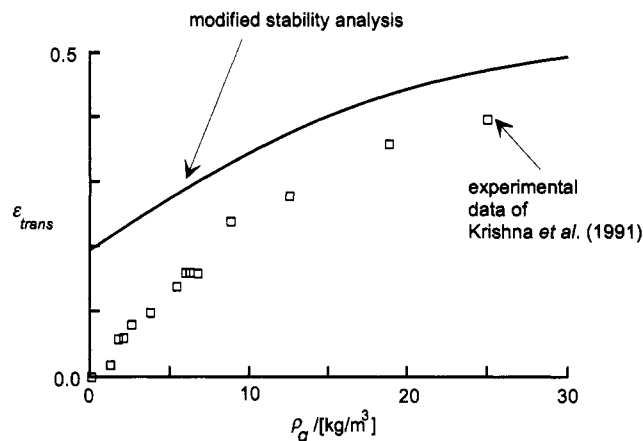
Richardson and da S. Jeronimo, 1979). This would mean that increasing gas density should make the regime of homogeneous fluidization more stable; this is in agreement with the experimental observations showing that the minimum bubbling velocity  $U_{\text{mb}} (\equiv U_{\text{trans}})$  increases with increasing system pressure (cf. Rowe et al., 1982). If the analogy with gas-solid fluidized beds is accepted, we at least have a qualitative explanation for the experimental observations in Figure 2. It remains to verify this analogy.

In order to determine the value of the exponent  $n \equiv p - 1$  for bubble columns, we analyzed available experimental data for gas holdup in bubble columns in the homogeneous flow regime. Six sets of experimental data were used in the ensuing analysis from three different sources, (i) helium-water and argon-water data of Koetsier et al. (1976), (ii) nitrogen (at 0.1 MPa)-water data of Krishna (1992), and (iii) nitrogen (at 1, 1.5, and 2 MPa)-water data of Wilkinson (1991), in order to establish the value of the exponent  $n$  as a function of the gas density  $\rho_g$ .

To illustrate our procedure, we reproduce in Figure 4 the gas holdup results of Koetsier et al. (1976) for the systems helium-water and argon-water. The data were used to make a log-log plot of the bubble swarm velocity  $U_{\text{swarm}}$  versus  $(1 - \epsilon)$ ; this plot is shown in Figure 5. The slope of the initial linear portion of the curve yields the value of the exponent  $n$ . The six sets of experimental data were analyzed in a similar manner to obtain the exponent  $n$  for each data. A plot of  $n$  versus the gas density  $\rho_g$  for the six data sets is shown in Figure 6; the data could be correlated as



**Figure 6.** Correlation of the Richardson-Zaki exponent  $n$  as a function of the gas density. Data for bubble columns from three different sources.



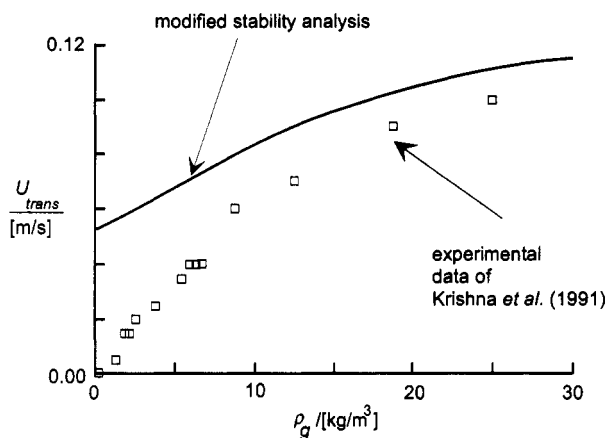
**Figure 7.** Transition gas holdup  $\epsilon_{\text{trans}}$  as a function of the gas density. Comparison of the values predicted by the modified stability analysis and those experimentally determined by Krishna et al. (1991).

$$n = 2 \exp(-0.1\rho_g) \quad (9)$$

This correlation for the Richardson-Zaki exponent  $n$  was then used in the Biesheuvel-Gorissen stability analysis to determine the values of  $\epsilon_{\text{trans}}$  as a function of the gas density. The results are shown in Figure 7, which, in sharp contrast to the results with the  $n = 1$  assumption (cf. Figure 3), predicts a significant increase in the transition gas holdup  $\epsilon_{\text{trans}}$  with increasing gas density. The  $\epsilon_{\text{trans}}$  values thus obtained represent an *upper bound* of the practically realizable transition holdups. In practice due to imperfect gas distribution there will be an additional tendency toward destabilization of homogeneous bubbly flow, leading to an "earlier" transition to the heterogeneous flow regime. This point is further emphasized if we compare the  $\epsilon_{\text{trans}}$  values obtained by the modified stability analysis with the measured transition holdup values of Krishna et al. (1991); the measured values lie consistently below those anticipated by the stability analysis, as indeed they should. Figure 8 shows the predictions of the velocity at the regime transition point  $U_{\text{trans}}$  as a function of the gas density. Predictions following the modified stability analysis lie consistently above the values reported by Krishna et al. (1991). Use of improved gas distribution would tend to yield transition values  $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$  approaching the predictions of the modified stability analysis.

### Concluding Remarks

The Richardson-Zaki exponent  $n$  has been shown to be an important parameter determining the stability of homogeneous bubbly flows in a bubble column. With



**Figure 8.** Transition gas velocity as a function of the gas density. Comparison of the values predicted by the modified stability analysis and those experimentally determined by Krishna et al. (1991).

increasing gas density the value of this exponent decreases; physically this means that increasing gas density results in reduced interaction between neighboring bubbles and, consequently, reduced chance of propagation of instabilities. We have modified the Biesheuvel–Gorissen stability analysis to incorporate the influence of the gas density on the exponent  $n$ ; the modified stability provides upper bounds for  $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$ . Our analysis provides a rationalization for the observed stabilizing influence of increased gas density on the homogeneous bubbly flow regime.

### Nomenclature

- $a$  = radius of gas bubble, m  
 $d$  = bubble diameter, m  
 $g$  = acceleration due to gravity,  $9.81 \text{ m s}^{-2}$   
 $m_0$  = voidage correction for the added mass term, dimensionless  
 $n$  = exponent in the bubble swarm rise velocity relationship (3) in laboratory fixed reference frame, dimensionless  
 $p$  = exponent in the bubble swarm rise velocity relationship (2), in the zero volume flux reference frame, dimensionless  
 $p_e$  = kinetic contribution of the effective pressure,  $\text{kg m}^{-1} \text{ s}^{-2}$   
 $U$  = superficial gas velocity,  $\text{m s}^{-1}$   
 $U_{\text{mb}}$  = minimum bubbling velocity for gas–solid fluid bed,  $\text{m s}^{-1}$   
 $U_{\text{swarm}}$  = rise velocity of bubble swarm in the laboratory fixed reference frame,  $\text{m s}^{-1}$   
 $U_{\text{trans}}$  = superficial gas velocity at regime transition,  $\text{m s}^{-1}$   
 $v_0$  = rise velocity of bubble swarm in the zero volume flux reference frame,  $\text{m s}^{-1}$   
 $v_{\infty}$  = single bubble rise velocity,  $\text{m s}^{-1}$   
 $\Delta v^2$  = mean square velocity fluctuation,  $\text{m}^2 \text{ s}^{-2}$
- Greek Letters**
- $\alpha$  = constant of proportionality in eq 4;  $\alpha = 1$ , dimensionless  
 $\delta_e$  = effective diffusivity of bubble swarm voidage,  $\text{m}^2 \text{ s}^{-1}$   
 $\epsilon$  = fractional holdup of gas bubbles, dimensionless

- $\epsilon_{\text{cp}}$  = fractional holdup of gas bubbles at closest packing,  $\epsilon_{\text{cp}} = 0.62$ , dimensionless  
 $\mu_L$  = liquid viscosity, Pa s  
 $\rho_G$  = density of gaseous phase,  $\text{kg m}^{-3}$   
 $\rho_L$  = liquid density,  $\text{kg m}^{-3}$   
 $\sigma$  = surface tension of the liquid phase,  $\text{N m}^{-1}$

### Subscripts

- cp = closest packing  
e = effective  
G = referring to gas phase  
L = referring to liquid phase  
mb = minimum bubbling  
small = referring to small bubbles in gas–liquid systems  
trans = referring to the transition point  
 $\infty$  = single bubble parameter

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