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A MODEL FOR GAS HOLDUP IN BUBBLE COLUMNS INCORPORATING THE INFLUENCE OF GAS DENSITY ON FLOW REGIME TRANSITIONS

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Abstract—The aim of this study was to develop a practically usable model to describe the influence of increased gas density on the gas holdup in bubble column reactors. In order to develop an insight into this effect, we performed extensive sets of experiments at pressures ranging from 0.1 MPa to 2 MPa and with several gases (nitrogen, carbon dioxide, argon, helium and sulphur hexafluoride) in de-ionized water in a 0.16 m diameter bubble column. A careful analysis of the experimental results shows that the major effect of increased gas density is to stabilize the regime of homogeneous bubble flow and, consequently, to delay the transition to the churn-turbulent flow regime. The superficial gas velocity at this regime transition point, U_{trans}, was found to be a unique function of the gas density, encompassing both effects of pressure and molar mass. To elucidate the hydrodynamics in the two regimes, dynamic gas disengagement experiments were carried out in a 0.19 m diameter bubble column with four liquids (water, turpentine, n-butanol and mono-ethylene glycol) using nitrogen at 0.1 MPa. These results showed that the churn-turbulent regime is characterized by a bi-modal bubble size distribution, consisting of fast rising large bubbles (typically 5 cm diameter or larger) and small bubbles (typically $\lesssim$ 5 mm diameter). In the churn-turbulent regime the holdup of the small bubbles was found to be virtually constant. The regime transition velocity U_{trans} was found to depend on the liquid properties. A simple model for describing the gas holdup is also proposed.

INTRODUCTION

Though industrial bubble column reactors are often operated at high pressures, it is only relatively recently that experimental data have become available on the influence of increased gas density on gas holdup in bubble columns (Clark, 1990; Hikita *et al.*, 1980; Idogawa *et al.*, 1986; Oyevaar, 1989; Öztürk *et al.*, 1987; Reilly *et al.*, 1986; Wilkinson and van Dierendonck, 1990). These data demonstrate the severe shortcomings of available literature correlations in portraying the influence of either increased reactor pressure or increased gas density. The objective of the present paper is to examine the influence of gas density on the gas holdup and to develop a model which can be used for industrial reactor scale-up purposes. The simple model proposed here for describing the gas holdup incorporates the following experimentally verified concepts:

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

(ii) At a superficial velocity greater than U_{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_{trans} and (b) the holdup of large fast rising bubbles. This holdup varies with $(U_g - U_{trans})$ and is virtually independent of the gas density or liquid properties.

(iii) The influence of the gas and liquid properties (gas density, liquid viscosity, liquid density and liquid surface tension) is reflected in their influence on the regime transition velocity. In order to develop such a model we have performed two series of experiments as described below.

EXPERIMENTAL

The first series of experiments was carried out in two metal 0.16 m diameter columns with de-ionized water at pressures ranging from 0.1 MPa to 2 MPa and with several gases (nitrogen, carbon dioxide, argon, helium and sulphur dioxide); the details of the experimental set-up and procedure has been reported elsewhere (Wilkinson and van Dierendonck, 1990). Additionally, to obtain insight into the hydrodynamics in the churn-turbulent regime of operation, dynamic gas disengagement experiments were performed in a 0.19 m diameter column of 4 m height with four liquids (turpentine, water, n-butanol and mono-ethylene glycol); the details of this second experimental set-up and procedure have been reported earlier (Vermeer and Krishna, 1981) and are therefore

Table 1. Range of conditions used for gas hold up experiments

Gas–liquid system	Pressure (MPa)	Column dimensions (m)
Water–nitrogen	0.1–2.0	$H = 1.2$ $D = 0.16$
Water–helium	0.1–0.7	$H = 1.2$ $D = 0.16$
Water–CO_2	0.1–0.3	$H = 1.2$ $D = 0.16$
Water–argon	0.1–0.3	$H = 1.2$ $D = 0.16$
Water SF_6	0.1	$H = 1.2$ $D = 0.16$
Water–nitrogen	0.1	$H = 4.0$ $D = 0.19$
Mono-ethylene glycol	0.1	$H = 4.0$ $D = 0.19$
n-Butanol	0.1	$H = 4.0$ $D = 0.19$
Turpentine	0.1	$H = 4.0$ $D = 0.19$

not repeated here. Table 1 summarizes the range of conditions studied in the two sets of experiments.

DISCUSSION ON THE INFLUENCE OF GAS DENSITY ON REGIME TRANSITIONS

The influence of pressure on gas holdup for the N_2–de-ionized water system is summarized in Fig. 1. The gas holdup is initially a linear function of the superficial gas velocity, typical of the homogeneous bubble flow regime. With increasing gas velocity a point is reached where the transition to the churn-turbulent regime occurs (heterogeneous regime). In the churn-turbulent regime the dependence of the gas holdup on the gas velocity is no longer linear. Examination of the results shown in Fig. 1 leads to the conclusion that the influence of increased pressure is to "delay" the transition to the heterogeneous or churn-turbulent regime of operation; this is the line of reasoning we follow in this paper to develop our model to describe the gas holdup. The results in Fig. 2 show that gas holdup is uniquely dependent on the gas density for any given superficial gas velocity. In other words, it does not matter whether the increased gas density is a result of operation at a higher pressure or due to the use of a gas with a higher molar mass. The experimental ε_g versus U_g data were analyzed to determine the superficial gas velocity (U_{trans}) at which the transition from homogeneous to the churn-turbulent regime occurs. We demonstrate this for N_2 in water at 1.5 MPa by plotting the swarm rise velocity (U_g/ε_g) versus V_g, as shown in Fig. 3. At U_g below 0.09 m s^{-1}, the swarm rise velocity is virtually constant at a value of 0.25 m s^{-1}, characterizing the homogeneous bubble flow regime. For a gas velocity higher than $U_g = U_{trans} = 0.09$ m s^{-1}, the swarm velocity is seen to increase beyond the value of 0.25 m s^{-1} and we enter the heterogeneous or churn-turbulent regime of operation. The rise velocity of 0.25 m s^{-1} (characteristic for water) implies that the dependence of the total gas holdup on the superficial gas velocity follows the simple linear form:

$$\varepsilon_g = 4U_g, \qquad U_g \leqslant U_{trans} \tag{1}$$

in the homogeneous bubble flow regime. Though we cannot expect eq. (1) to strictly hold for liquids other than water and for experiments with higher gas densities, we may regard eq. (1) as a good approximation for the data sets used in this study.

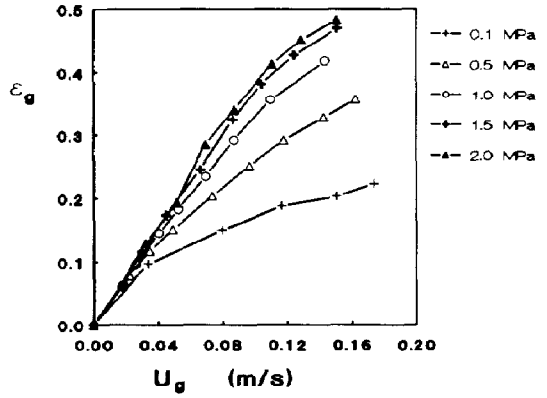

Fig. 1. Gas holdup (ε_g) versus superficial gas velocity (U_g) for the water–nitrogen system at various pressures; data obtained in 0.16 m diameter column.

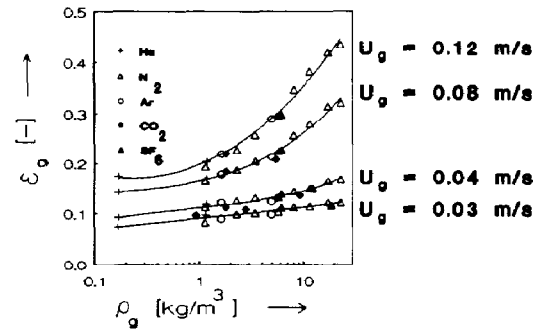

Fig. 2. Gas holdup in water versus gas density for four different superficial gas velocities. From top to bottom: $U_g = 0.12$, 0.08, 0.04 and 0.03 m s^{-1}, respectively. Experimental data obtained in 0.16 m diameter column.

For every set of measurements we determined regime transition velocity (U_{trans}) along the manner depicted in Fig. 3. The regime transition velocity (U_{trans}) was found to be a unique function of the gas density for all experiments in water as shown in Fig. 4. The stabilizing influence of increased gas density on the homogeneous bubble regime has a parallel in

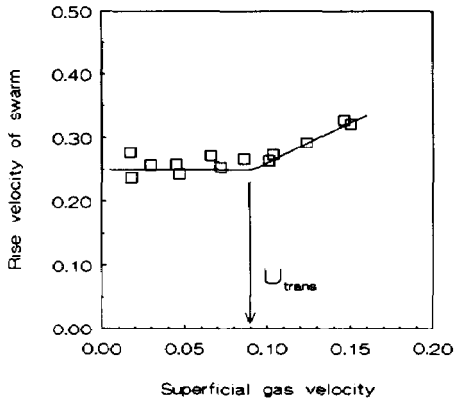

Fig. 3. Rise velocity of swarm (U_g/ε_g) versus superficial gas velocity (U_g) for nitrogen–water system at 1.5 MPa pressure.

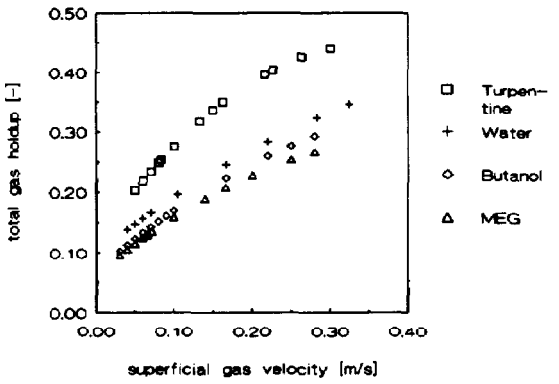

Fig. 5. Total gas holdup (ε_g) versus superficial gas velocity (U_g) for four different liquids with nitrogen at 0.1 MPa pressure. Data obtained in 0.19 m diameter column.

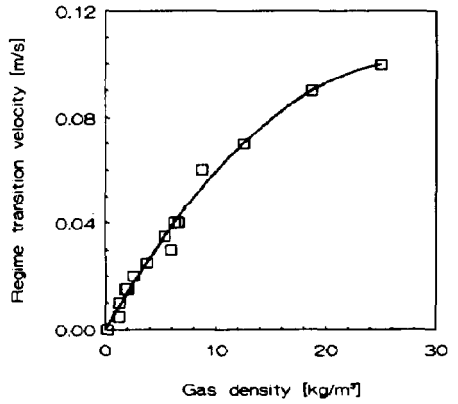

Fig. 4. Dependence of the regime transition velocity (U_{trans}) on the gas density (ρ_g) in water.

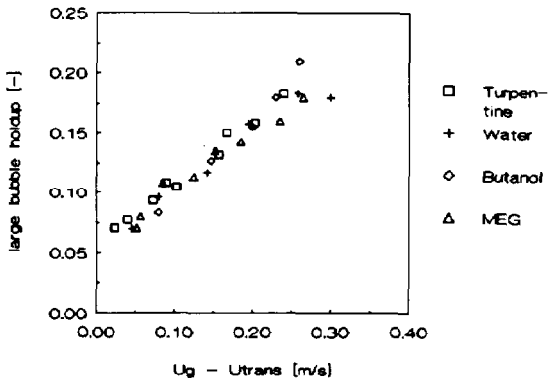

Fig. 6. Large bubble holdup (ε_{large}) versus ($U_g - U_{trans}$) for four different liquids with nitrogen at 0.1 MPa pressure. Data obtained in 0.19 m diameter column.

gas–solid fluidized beds where it is now generally accepted that increased pressure tends to delay the appearance of bubbles and increases the rate of bubble break-up. A very similar observation for the influence of gas density on bubble break-up has also been recently observed in bubble columns (Wilkinson and van Dierendonck, 1990).

DISCUSSION OF DYNAMIC GAS DISENGAGEMENT EXPERIMENTS

Before it is possible to develop a model for gas holdup it is necessary to characterize the churn-turbulent regime, i.e. the regime prevailing beyond U_{trans}. To achieve this we performed dynamic gas disengagement experiments in the 0.19 m diameter column with nitrogen at 0.1 MPa and four liquids of varying properties: turpentine, water, n-butanol and mono-ethylene glycol. The dynamic gas disengagement results confirmed our earlier findings (Vermeer and Krishna, 1981) that in the churn-turbulent regime we have a bi-modal bubble size distribution: (i) a small bubble population consisting of bubbles of about 5 mm in diameter and (ii) a fast rising bubble population consisting of bubbles of ill-defined shapes and sizes of the order of 50 mm. Visually it could be determined that the small bubbles are entrained in the liquid phase and have the backmixing characteristics of the liquid. The dynamic gas disengagement results allowed the determination of the large and small bubble holdups. From a reactor design and scale-up view point it is vital to make a distinction between the two bubble populations in view of the differing residence time distributions. The large bubbles rise fast through the column virtually in plug flow (similar to bubbles in a gas solid fluidized bed) while the small bubbles display a wide residence time distribution, approaching well mixed character in vessels of large diameter.

A most interesting aspect of the experimental results is that while the total gas holdups for the four liquids are widely different (cf. Fig. 5), as shown in Fig. 6, the large bubble holdup is practically independent

of the properties of the liquid and depends only on the parameter $(U_g - U_{trans})$, where U_{trans} was estimated as discussed before. We, therefore, assume a relation of the form

$$\varepsilon_{large} = A(U_g - U_{trans})^n, \; U_g \geq U_{trans} \qquad (2)$$

where A and n are to be determined from an empirical curve fit.

Beyond the regime transition velocity, the small bubble holdup was found to be practically constant; this is demonstrated in Fig. 7 for the experiments with water. Results similar to those with water were obtained for the other three liquids and, further, these findings are in agreement with recent literature [e.g. Wezorke, 1986]. In the light of the foregoing experimental evidence we assume that the holdup of the small bubbles equals the gas holdup at the regime transition point U_{trans} and is independent of the gas velocity beyond U_{trans}, i.e. we have

$$\varepsilon_{small} = \varepsilon_{trans}, \; U_g \geq U_{trans}. \qquad (3)$$

Since the large bubble holdups for the four liquids are virtually independent of the liquid phase properties, it can be concluded that the main effect of liquid phase properties is to alter the regime transition point U_{trans}.

The total holdup in the heterogeneous flow regime is given by

$$\varepsilon_g = \varepsilon_{trans} + A(U_g - U_{trans})^n, \; U_g \geq U_{trans}. \qquad (4)$$

Our model is pictorially represented in Fig. 8.

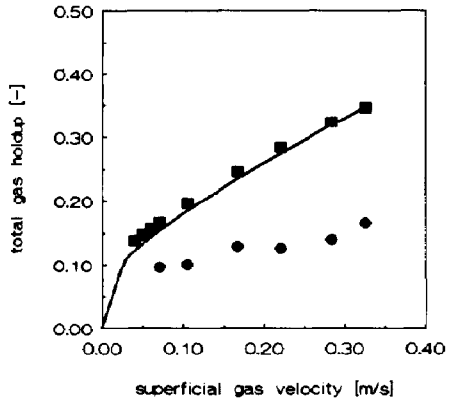

Fig. 7. Total and small bubble gas holdup versus superficial gas velocity for the nitrogen–water system at 0.1 MPa pressure. Data obtained in 0.19 m diameter column. (■ = total gas holdup ε_g; ● = small bubble holdup ε_{small}).

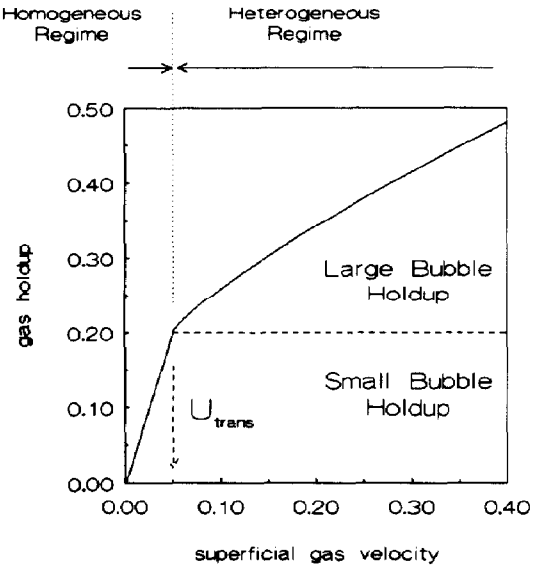

Fig. 8. Proposed model for gas holdup in bubble columns.

GAS DENSITY EFFECTS RE-ANALYZED

Since it was infeasible to perform dynamic disengagement experiments at high pressures in the metal column we accepted the hydrodynamic picture emerging from the dynamic gas disengagement experiments obtained in the 0.19 m column and took the large bubble population (ε_{large}) to be given by

$$\varepsilon_{large} = \varepsilon_g - \varepsilon_{trans} \qquad (5)$$

where, following eq. (1), we take

$$\varepsilon_{trans} = 4U_{trans}. \qquad (6)$$

Figure 9 shows that the large bubble holdup in water calculated from eq. (5) for different gases at low as well as high pressure depends only on $(U_g - U_{trans})$. Consequently it appears that neither the gas density nor the liquid properties (Fig. 6) have an influence on the relation between $U_g - U_{trans}$ and the large bubble holdup. The large bubble data could be correlated by eq. (2) with $A = 1$ and $n = 0.8$. The exponent $n = 0.8$ was chosen because of the expected analogy with the hydrodynamics of gas–solid fluidized beds; Krishna (1987) has demonstrated that use of a bubble growth model leads to a four-fifths power relation for ε_{large}. Equations (1), (3), (4) and (6) provide a complete description of the model for the gas holdup wherein the only parameter influenced by the gas density is the regime transition velocity U_{trans} (cf. Fig. 4). Figure 10 presents a parity plot of the experimental total gas holdup in water for fifteen different sets of experiments [nitrogen (0.1, 0.2, 0.3, 0.5, 0.7, 1.0, 1.5 and 2 MPa pressure), helium (0.1 and 0.7 MPa pressure), CO_2 (0.1 and 0.3 MPa pressure), argon (0.1 and 0.3 MPa pressure) and SF_6 (0.1 MPa pressure)] and the predictions of the simple model given by eqs (1), (3), (4) and (6) which has only three parameters: $A = 1$, $n = 0.8$ and U_{trans}, given by Fig. 4. The predictions can be considered extremely good and moreover the model provides a simple scale-up rule for design of industrial columns operating at high pressures. The important parameter U_{trans} has been shown to be very sensitive to liquid phase properties, and gas density, and it is expected to be influenced by the distributor design and scale of operation (e.g. column diameter).

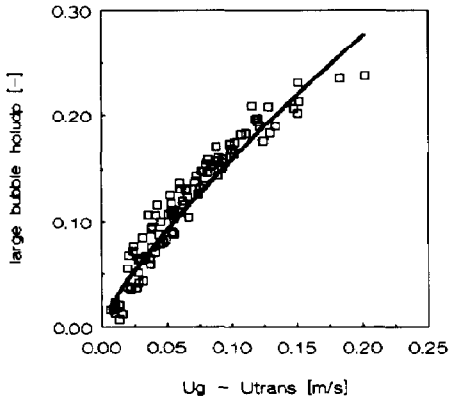

Fig. 9. Large bubble holdup ε_{large} versus $(U_g - U_{trans})$ for measurements in 0.16 m diameter column for fifteen different data sets with varying density. In all cases liquid used is water.

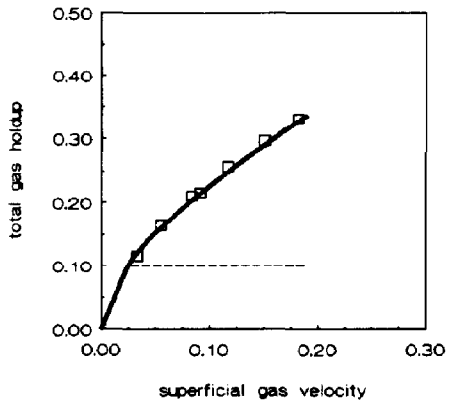

Fig. 11. Gas holdup versus superficial gas velocity for nitrogen–water system at 0.3 MPa pressure in 0.16 m diameter column.

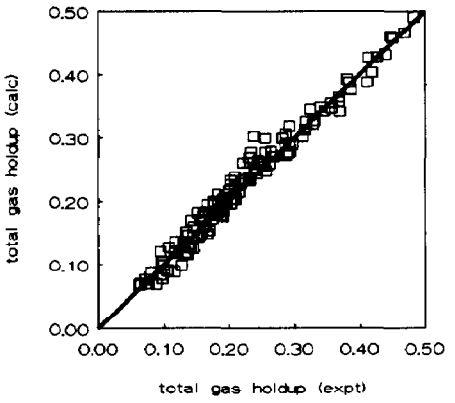

Fig. 10. Experimental total gas holdup in water versus model predictions for fifteen different data sets with varying gas density. Measurements in 0.16 m diameter column.

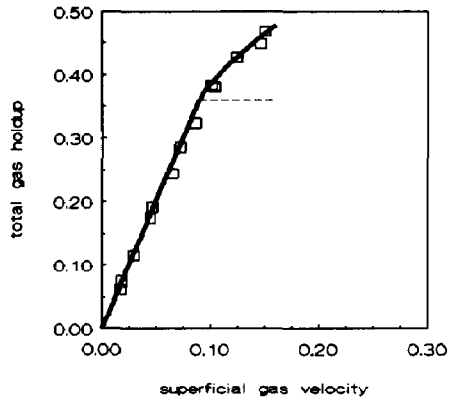

Fig. 12. Gas holdup versus superficial gas velocity for nitrogen–water system at 1.5 MPa pressure in 0.16 m diameter column.

To illustrate further the strength of our simple model we compare measured data with model predictions for three different runs: nitrogen at 0.3 MPa (Fig. 11), nitrogen at 1.5 MPa (Fig. 12) and argon at 0.3 MPa (Fig. 13). The point to note is the success with which the model is able to handle the flow regime transitions with systems of varying gas density, characterized by the change in the slope of the ε_g versus U_g curve.

CONCLUDING REMARKS

In developing a model to describe the total gas holdup in bubble columns it is essential to make a distinction between the homogeneous and heterogeneous, or churn-turbulent, regimes of operation. Equations (1), (3), (4) and (6) summarize the model developed on the basis of extensive experimentation. The gas velocity at the regime transition point U_{trans} is a key parameter in the model developed in this paper.

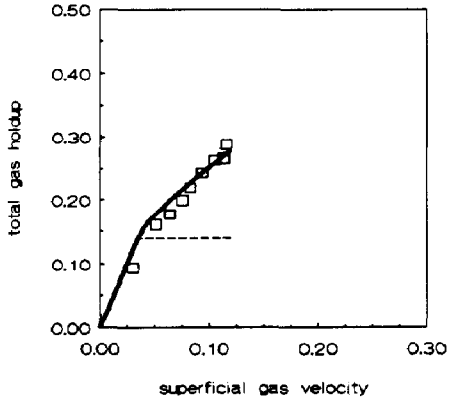

Fig. 13. Gas holdup versus superficial gas velocity for argon–water system at 0.3 MPa pressure in 0.16 m diameter column.

This parameter is found to be significantly influenced by the gas density and physical properties of the liquid. Though not demonstrated in this paper it is to be expected that U_{trans} can also depend on the gas distributor design and scale of operation (i.e. column diameter). Further experimental work is required to study the influence of scale and distributor design on U_{trans}.

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NOTATION

A	constant in eq. (4) for large bubble holdup, dimensionless
n	exponent in eq. (4) for large bubble holdup, dimensionless
U_g	superficial gas velocity, m s^{-1}
U_{trans}	regime transition velocity, m s^{-1}

Greek letters

ε_g	total gas holdup, dimensionless
ε_{large}	holdup of large fast rising bubbles, dimensionless
ε_{small}	holdup of small bubbles, dimensionless
ε_{trans}	gas holdup at $U_g = U_{trans}$, dimensionless
ρ_g	gas density, kg m^{-3}

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