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## INFLUENCE OF ALCOHOL ADDITION ON GAS HOLD-UP IN BUBBLE COLUMNS: DEVELOPMENT OF A SCALE UP MODEL

R. Krishna, A.J. Dreher and M.I. Urseau  
Department of Chemical Engineering, University of Amsterdam  
Nieuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands

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### ABSTRACT

Measurements were carried out in bubble columns with diameters of 0.1, 0.15 and 0.38 m with air – water system to which ethanol was added in concentrations ranging from 0.03 – 1 volume %. Alcohol addition results in a significant increase in the gas hold-up,  $\epsilon$ . This increase in  $\epsilon$  can be attributed to a delay in the point of transition from homogeneous to heterogeneous flow regime. The model of Krishna *et al.* (*Chem. Eng. Sci.*, **54**, 171 (1999)) was found applicable for the range of column diameters studied, after accounting for the influence of alcohol addition on the regime transition parameters. © 2000 Elsevier Science Ltd

### Introduction

There is considerable industrial interest in design and scale up of bubble column reactors in view of many practical applications in natural gas conversion technologies [1,2,3]. A bubble column reactor can be operated in either the homogeneous or heterogeneous flow regime; see Fig. 1. When a column filled with a liquid is sparged with gas the bed of liquid begins to expand as soon as gas is introduced. As the gas velocity is increased the bed height increases almost linearly with the superficial gas velocity,  $U$ , provided the value of  $U$  stays below a certain value  $U_{\text{trans}}$ . This regime of operation of a bubble column is called the *homogeneous bubbly flow regime*. The bubble size distribution is narrow and a roughly uniform bubble size, generally in the range 1 - 7 mm, is found. When the superficial gas velocity  $U$  reaches the value  $U_{\text{trans}}$  coalescence of the bubbles takes place to produce the first fast-rising "large" bubble. The appearance of the first large bubble changes the hydrodynamic picture dramatically. The hydrodynamic picture in a gas-liquid system for velocities exceeding  $U_{\text{trans}}$  is commonly referred to as the *heterogeneous or churn-turbulent flow regime*. In the heterogeneous regime small bubbles combine in clusters to form large bubbles in the size range 20 - 70 mm. These large bubbles travel up through the column at high velocities (in the range 1 - 2 m/s), in a more or less plug flow manner. These large bubbles have the effect of churning up the liquid phase. The large bubbles are mainly responsible for the throughput of gas

through the reactor. Small bubbles, which co-exist, with large bubbles in the churn-turbulent regime, are “entrained” in the liquid phase and as a good approximation have the same backmixing characteristics of the liquid phase. The two regimes are portrayed in Fig. 1 which shows also in a qualitative way the variation of the gas hold-up  $\epsilon$  as a function of the superficial gas velocity  $U$ . When the gas distribution is very good the regime transition region is often characterised by a maximum in the gas hold-up. The transition between homogenous and churn-turbulent regime is often difficult to characterise and there is a transition region as shown in Fig. 1.

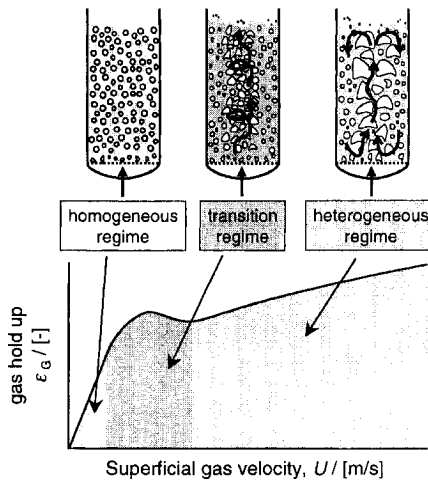


FIG. 1  
Flow regimes in bubble column reactors.

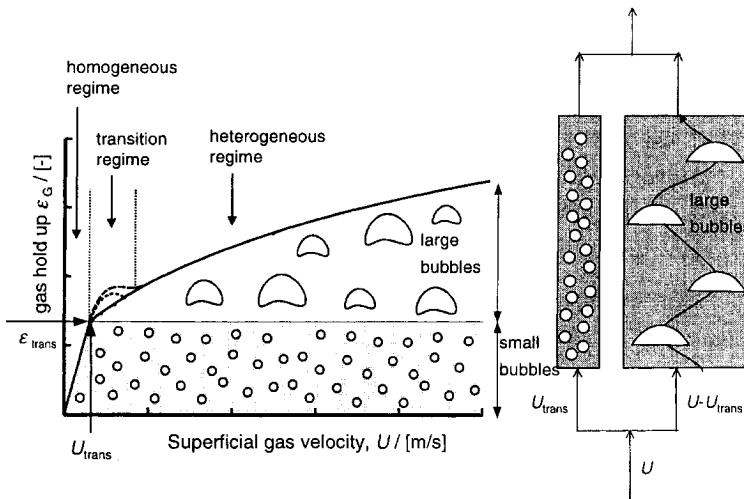


FIG. 2  
Model for gas hold-up according to Krishna and co-workers [4,5,6].

Though laboratory scale studies are often restricted to the homogeneous bubbly flow regime, prevailing at relatively low superficial gas velocities, industrial reactors are often operated at higher gas velocities in the heterogeneous flow regime. Krishna and co-workers [4,5,6] have developed a two-phase model for description of the hydrodynamics of bubble columns operating in the churn-turbulent flow regime. Their model is shown qualitatively in Fig. 2. In this model a sharp transition from homogeneous to heterogeneous flow regime is assumed and the superficial gas velocity through the small bubbles is taken to be the transition gas velocity  $U_{trans}$ . The superficial gas velocity through the "large" bubbles is remaining gas with a superficial gas velocity  $(U - U_{trans})$ .

For superficial gas velocities  $U \leq U_{trans}$ , the gas hold-up in the homogeneous flow regime is given by

$$\varepsilon = U / V_{slip}; \quad V_{slip} = v_{\infty}(1 - \varepsilon) \quad (1)$$

In the heterogeneous flow regime, i.e. for  $U \geq U_{trans}$  the total gas hold-up is given by

$$\varepsilon = \varepsilon_{b,large} + \varepsilon_{trans}(1 - \varepsilon_{b,large}); \quad \varepsilon_{b,large} = (U - U_{trans}) / V_{b,large} \quad (2)$$

where  $V_{b,large}$  is the rise velocity of the swarm of large bubbles. Krishna *et al.* [6] obtained a correlation for the large bubble velocity in the form

$$V_{b,large} = 0.71 \sqrt{g d_{b,large}} (SF)(AF) \quad (3)$$

where two correction factors are introduced into the classical Davies-Taylor [7] relation for the rise of a single spherical cap bubble in an infinite volume of liquid. Bubbles in a liquid assume a spherical cap shape when the criterion Eötvös number,  $Eö > 40$ , is met; see Clift *et al.* [8]. For the air-water system, the criterion  $Eö > 40$  is met for bubbles larger than 17 mm in diameter. The scale correction factor ( $SF$ ), accounts for the influence of the column diameter and is taken from the work of Collins [9] to be

$$\begin{aligned} SF &= 1 && \text{for } d_{b,large} / D_T < 0.125 \\ SF &= 1.13 \exp(-d_{b,large} / D_T) && \text{for } 0.125 < d_{b,large} / D_T < 0.6 \\ SF &= 0.496 \sqrt{D_T / d_{b,large}} && \text{for } d_{b,large} / D_T > 0.6 \end{aligned} \quad (4)$$

The acceleration factor  $AF$  accounts for the increase in the large bubble velocity over that of a single, isolated, bubble due to wake interactions. This factor increases as the distance between the large bubbles decreases [11]. Since the average distance between large bubbles will decrease as the superficial gas velocity through the large bubble phase increases, a linear relation is postulated for  $AF$ :

$$AF = \alpha + \beta(U - U_{trans}) \quad (5)$$

and a power-law dependence of the bubble size on  $(U - U_{trans})$ :

$$d_{b,large} = \gamma(U - U_{trans})^{\delta} \quad (6)$$

The model parameters  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $\delta$  were determined by Krishna *et al.* [6] by multiple regression of the measured data on the rise velocity of large bubbles in low viscosity liquids such as water. The fitted values are:

$$\alpha = 2.73; \quad \beta = 4.505; \quad \gamma = 0.069; \quad \delta = 0.376 \quad (7)$$

Krishna *et al.* [6] presented data to validate their model for the air-water system.

The objective of the present communication is to examine the extent to which the above model is valid to describe the hold-up in air-water systems to which small quantities of ethanol are added. A study of the published literature on the influence of alcohol addition on the gas hold-up in bubble columns [10 – 15] shows that alcohol addition has the effect of stabilising the homogeneous bubbly flow regime. Published experimental data on alcohol addition in bubble columns are largely restricted to the homogeneous flow regime and we are interested in the present communication to extending the data to the heterogeneous flow regime.

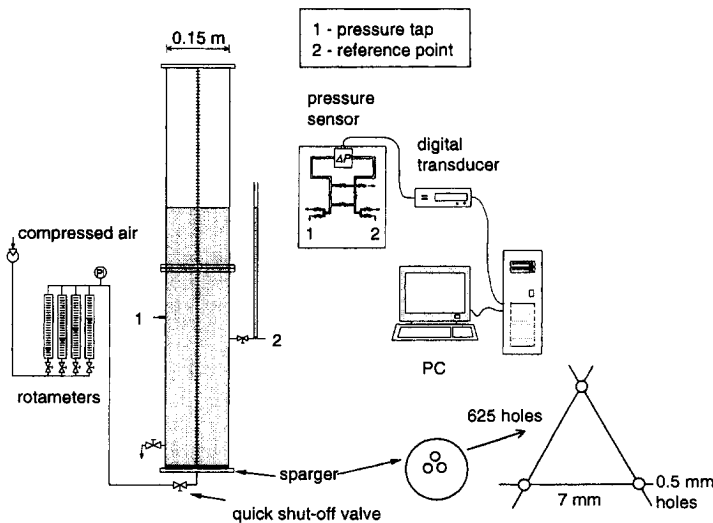


FIG. 3  
Experimental set-up for the 0.15 m diameter column.

### Experimental

Experiments were carried out in columns of 0.1, 0.15 and 0.38 m in diameter. All columns were 4 m high and air was used as the gas phase. The liquid phase used in the experiments consisted of demineralised water to which ethanol was added in various concentrations. A typical column configuration for the 0.15 m diameter column is shown in Fig. 3. All columns were fitted with similar sieve plate distributors with 0.5 mm diameter holes on a triangular pitch of 7 mm. In the 0.15 m diameter column, for example, a total of 625 holes were drilled. The total gas hold-up was determined by means of measuring the hydrostatic pressure using a Validyne pressure sensor.

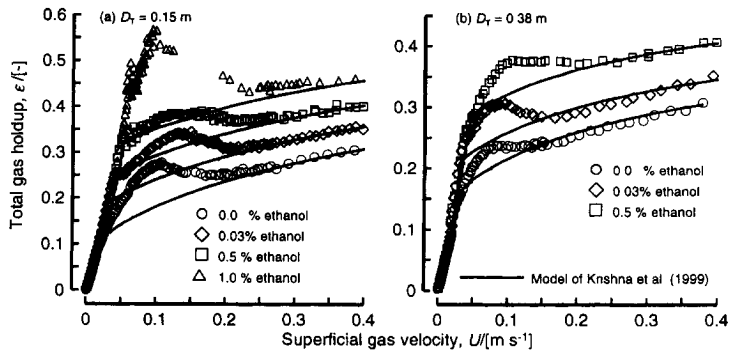


FIG. 4

Influence of ethanol addition on gas hold-up in bubble columns.

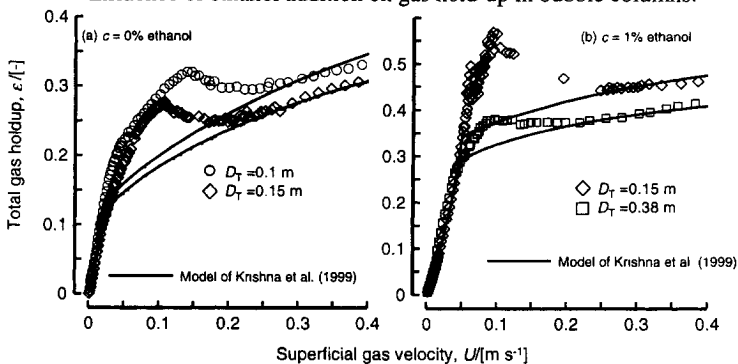


FIG. 5

Influence of column diameter on gas hold-up in bubble columns.

### Experimental Results with Alcohol Addition

Figure 4 shows the gas hold-up  $\epsilon$  as a function of the superficial gas velocity  $U$  obtained with four typical ethanol concentrations used in our experiments in the 0.15 and 0.38 m diameter columns. We see a significant increase in the total gas hold-up with increasing ethanol addition. In Fig. 5 (a) a comparison is made of the gas hold-up with demineralised water in the 0.1 and 0.15 m diameter columns. We note that there is a significant decrease in the total gas hold-up with increasing column diameter. The column diameter influence persists when considering data for systems with ethanol addition; see Fig. 5 (b) for 1% ethanol solution in the 0.15 and 0.38 m diameter columns.

In order to interpret the results, the first task is to find out the values of the regime transition parameters. The regime transition parameters  $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$  can be determined from a Wallis plot [16]. In the Wallis plot the “drift flux”,  $U(1 - \epsilon)$ , is plotted against the gas hold-up,  $\epsilon$ , as shown in Fig. 6 for the experimental data in the 0.15 m diameter column with 0% ethanol and 0.5% ethanol concentrations. The smooth curve in Figure 6 are drawn using the Richardson-Zaki formulation [17], i.e.

drift flux =  $v_{\infty} \epsilon (1 - \epsilon)^n$  where  $n$  is the Richardson-Zaki index and  $v_{\infty}$  is the rise velocity of a single gas (small) bubble. For air –water system in the homogeneous regime,  $n = 2$  and the rise velocity of a single bubble  $v_{\infty}$  can be obtained from the Wallis plot by data fitting; see Fig. 6. The Richardson-Zaki formulation is valid for the homogeneous bubbly flow regime. The point of deviation of the experimental values from the Richardson-Zaki curve is taken to indicate the regime transition point; see Fig. 6. The influence of ethanol concentration on the values of the transition parameters  $\epsilon_{trans}$  and  $U_{trans}$  is shown in Fig. 7 for the whole range of concentrations studied in our experiments with the 0.15 m diameter column. A similar trend is obtained with the other columns of 0.1 and 0.38 m diameter. It is clear that the addition of small amounts of alcohol has the effect of increasing the values of  $\epsilon_{trans}$  and  $U_{trans}$ . In other words, the addition of alcohol has the effect of stabilizing the homogeneous bubbly flow regime. This stabilisation is caused by suppression of the coalescence tendency of small bubbles.

We now determine whether the shift in the regime transition point with alcohol addition is able to

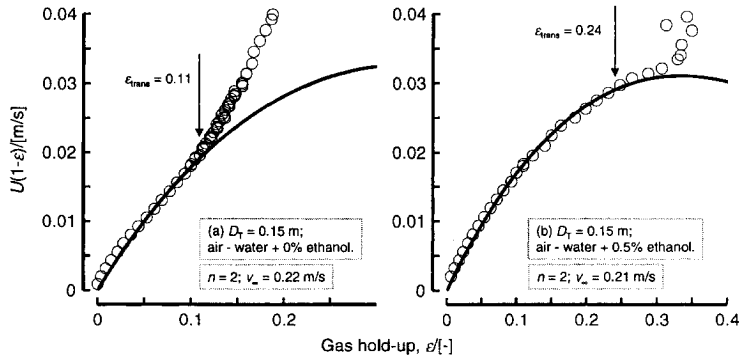


FIG. 6

Wallis plot to determine transition parameters. (a)  $c = 0$  vol % ethanol. (b)  $c = 0.5$  vol % ethanol.

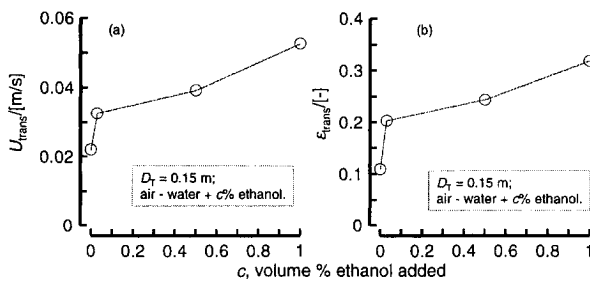


FIG. 7

Variation of regime transition parameters with ethanol concentration.

explain the increased hold-up observed experimentally and reported in Figures 4 and 5. To answer this question we apply the model of Krishna *et al.* [6] to calculate the gas hold-up. The smooth curves shown in Figs 4 and 5 have been drawn using Eqs (1) – (7), taking the fitted values of the transition parameters

( $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$ ) and the single small bubble rise velocity  $v_{\infty}$ . We note that the model works well when considering superficial gas velocities well above the transition gas velocity  $U_{\text{trans}}$ . However, the model fails to describe the transition region. The influence of column diameter on the total gas hold-up is well catered for by the model of Krishna *et al.* [6].

### Conclusions

The addition of small amounts of ethanol to water tends to result in a significant increase in the gas hold-up. This increase in the gas hold-up is entirely attributable to the delay in the regime transition point. By incorporating the appropriate values of  $\epsilon_{\text{trans}}$  and  $U_{\text{trans}}$ , the model of Krishna *et al.* [6] provides a very good description of the measured data well beyond  $U_{\text{trans}}$ . The transition regime is poorly represented by the model and there is a need for development of a gas hold-up model to describe this region. The influence of column diameter is properly taken account of in the model.

### Nomenclature

$AF$	acceleration factor, dimensionless
$c$	concentration of ethanol in water, volume %
$d_{b,\text{large}}$	large bubble diameter, m
$D_T$	column diameter, m
$E\ddot{o}$	Eötvös number, $g(\rho_L - \rho_G)d_b^2/\sigma$
$g$	gravitational acceleration, $9.81 \text{ m s}^{-2}$
$n$	Richardson-Zaki index, dimensionless
$p$	system pressure, Pa
$SF$	scale factor given by the Collins relations, eq. (4), dimensionless
$U$	superficial gas velocity, $\text{m s}^{-1}$
$U_{\text{trans}}$	superficial gas velocity at regime transition, $\text{m s}^{-1}$
$(U - U_{\text{trans}})$	superficial gas velocity through the large bubble phase, $\text{m s}^{-1}$
$v_{\infty}$	rise velocity of "small" bubbles, $\text{m s}^{-1}$
$V_{b,\text{large}}$	rise velocity of "large" bubbles in a swarm, $\text{m s}^{-1}$
$\alpha, \beta, \gamma, \delta$	parameters defined by eqs (5) – (7)
$\epsilon$	total gas hold-up, dimensionless
$\epsilon_{b,\text{large}}$	hold-up of large bubbles, dimensionless
$\epsilon_{\text{trans}}$	gas hold-up at the regime transition point, dimensionless

**Subscripts**

G	referring to gas
L	referring to liquid
b,large	referring to "large" bubbles
trans	referring to the transition point
T	tower or column

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