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The axial distribution of holdups in an industrial-scale bubble column with evaluated pressure using γ-ray attenuation approach

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Abstract

The axial distribution of gas holdups is measured using a γ-ray densitometry in the pressured bubble column of 0.3 m diameter and 6.6 m height. The principle of γ-ray measurement and data processing is discussed. The axial and average holdups in the two-phase system are obtained in the churn-turbulent flow regime with a gas velocity up to 0.40 m s^{-1} and a system pressure up to 1.0 MPa, which are in agreement with results obtained by a conventional method (differential pressure measurement along the column height). The effects of superficial gas velocity, liquid surface tension, liquid viscosity, and system pressure on the axial gas holdup are investigated in this study. The axial gas holdups decrease with the increase of liquid viscosity and liquid surface tension, and increase with the increase of pressure and superficial gas velocity. Furthermore, it is demonstrated that γ-ray attenuation method can be used to quantify the gas distribution and fingerprint the characteristics of the flow within such a reactor. The methodology proposed here could be also used as a tool to quantify and optimize the performance of other types of complex reaction systems.

Keywords: Gas–liquid column; γ-Ray attenuation; Holdup; Axial distribution

1. Introduction

In recent years, bubble column reactors have been playing an important role in the development of liquid phase catalytic oxidation processes for the manufacture of commodity and chemicals because they offer many advantages over other multiphase reactors—simple construction, no mechanically moving parts, good mass transfer properties, high thermal stability, low energy supply, and hence low construction and operation costs [1,2]. Some of the well-known examples of liquid phase oxidation in the industry are oxidation of *p*-xylene to terephthalic acid, cyclohexane to cyclohexanol, glucose to glutonic, wet oxidation of waste water containing organic compounds, and so on [3,4]. However, the distribution of holdup is one of the most important parameters for the oxidation process, which would seriously affect the reaction rate and product distribution. Thus, it is necessary to understand and study the axial distribution characteristics of holdup in bubble columns.

Many investigations have reported such measurements using different techniques, such as differential pressure, DGD, conductivity probe, electro-resistivity probe, optical probe, ultrasonic techniques, electrical capacitance and resistance tomography, X-ray and γ-ray densitometry technology, etc. [5]. But the γ-ray densitometry technique is a non-intrusive technique that does not disturb the flow. This technique has been widely employed in a variety of tower equipment examination of petrochemical and chemical processes [6]. However, γ-ray densitometry and tomography technique have also been extended to gas–solid and gas–liquid–solid systems with certain assumptions. The procedure has been applied by Chan and Banerjee [7] in the design of gamma densitometer for two transient experiments: refilling and rewetting experiments and flow boiling experiments. Eberle et al. [8] applied a novel theoretical method for optimization of a gamma densitometer to measure the area-averaged void fraction in gas–liquid flow. Schollenberger et al. [9] have studied the holdup radial profiles in a two-phase bubble column using gamma densitometer tomography; Veera et al. [10–12] have measured the holdup radial profiles in stirred bubble column. Bukur et al. [13] measured the gas holdup and flow regime transition in bubble column using γ-ray attenuation. Xie et al. [14] investigated experimentally flow structure,

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Nomenclature

H	the axial height (mm)
I_0, I	the intensities of the incident and emerging beams, respectively (n s^{-1})
L	the thickness of the absorbing medium (cm)
P	pressure (MPa)
T	temperature (°C)
u_g	gas velocity (m s^{-1})

Greek letters

ε_g	gas holdup
η	liquid viscosity (Pa s)
μ	the linear absorption coefficient (cm^2 g^{-1})
σ	liquid surface tension (N m^{-1})
ρ	the medium density (g cm^{-3})

gas holdup and the geometric and population characteristic of gas bubbles in gas–liquid–paper pulp three-phase flow using γ-ray densitometry. Kemoun et al. [15,16] studied gas holdup in a pressurized bubble column using non-invasive γ-ray based computed tomography. It was found that the cross-sectional average gas holdup increases with pressure and superficial gas velocity. Kumar et al. [17] reviewed the progress of the gamma densitometer tomography. γ-Ray approaches have played an important role in measurement technology for gas–liquid two-phase system and gas–liquid–solid three-phase system.

In the present paper, a systematic and simple measurement principle for γ-ray densitometer and comparison with differential pressure method were presented for two-phase bubble column with air–water system and air–acetic acid system. The effect of operating conditions and liquid properties on the distribution of holdup was discussed.

2. Experimental

2.1. Experimental setup

Experimental system mainly includes a pressured bubble column, a gas-supplying compressor, a gas-cycling compressor, a γ-ray measurement system, and an on-line pressure-difference sampling system as shown in Fig. 1. The bubble column of 0.30 m inside diameter and 6.6 m height was made of stainless steel. A four-nozzle distributor was placed at the bottom of the bubble column. The inner diameter of each nozzle was 20 mm and its distance to the central axis was 0.075 m. In order to ensure a steady gas flux, two gas tanks were, respectively, installed at the entrance and exit of the gas-cycling compressor. A stainless steel pipe between two gas tanks was placed to avoid the overload of the gas-cycling compressor when a ball valve at the bottom shut down during the bed collapse. The gas was introduced into the distributor after measured by the flow meter from the cycling compressor, then entered the bubble column, and a gas–liquid separator and a packed tower to absorb acetic acid from exit gas, and finally went back the cycling compressor. Five differential pressure sensors for measuring the

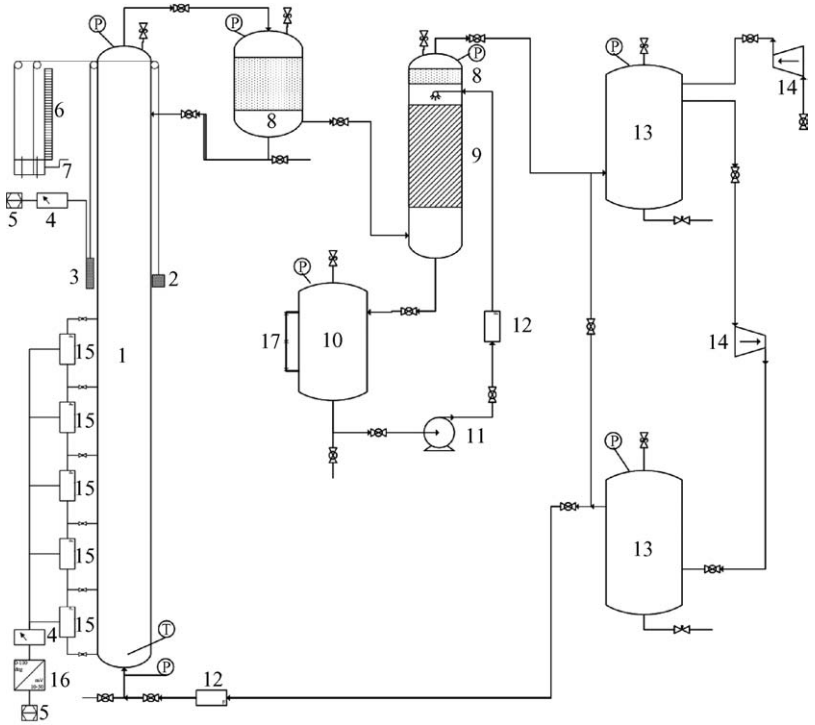

Fig. 1. Scheme of experiment apparatus: (1) bubble column, (2) Cs^{137} source, (3) γ-ray detector, (4) amplifier, (5) computer, (6) ruler, (7) manual pulley, (8) gas–liquid separator, (9) absorption tower, (10) tank, (11) pump, (12) flowmeter, (13) tank, (14) compressor, (15) pressure transducer, (16) A/D converter, and (17) level indicator.

differential pressure drop were placed along the column at 0.25, 0.75, 1.25, 1.75, and 2.25 m above the distributor. The differential pressure-sampling frequency was 115 Hz. Two neighboring pressure-sampling ports were connected to the high end and low end of each different-pressure transducer, respectively.

The γ-ray attenuation consists of a 100 mCi Cs^{137} gamma source, a sodium iodide (NaI) with activator scintillation detector, a pre-amplifier, acquisition/analysis hardware, and a computer controlled traverse. The disc source, collimated with a lead brick, is a sealed source of 5 mm in diameter. The ray source and ray detector were suspended on both sides of bubble column. Two-chain wheel and introduction slot at the top of the column provided the manual displacement synchronization for ray source and ray detector in vertical direction. The sampling distance can be measured by ruler, the sampling step is 0.1 m under normal condition and the sampling time is about 50–60 s. In order to decrease both the experimental error and two-phase flow influence, the source was set to provide as large a counting rate as possible. Each point was measured three times, and the mean intensity of the γ-ray was obtained by averaging.

2.2. Experimental systems and operating conditions

All experiments were carried out at a room temperature of approximately 25 °C. Air–water system and air–acetic acid system were, respectively, employed. Sodium oleate was used to reduce the water surface tension. The system pressure ranged from 0.5 to 1.0 MPa and the superficial gas velocity varied from 0.05 to 0.40 m s^{-1}.

2.3. Measurements and estimation of holdup profile

When γ-ray is introduced into a medium, its attenuation depends on radiation energy, and the type and thickness of absorbing material. The attenuation of ray beam passing through a thin homogeneous absorbing medium of uniform thickness is given by Lambert–Beer's law, as follows (Eq. (1)):

$$I = I_0 \, e^{-\mu\rho L} \tag{1}$$

where I_0 is the intensity of the incident beam, I the intensity of emerging beam, μ the linear absorption coefficient, ρ the medium density, and L is the thickness of the absorbing medium.

When the γ-ray passes through the gas liquid mixture and the wall of column, its attenuation varies with difference in the gas holdup and liquid holdup. The attenuation value of wall is constant, and an increased intensity of the γ-ray is used to compensate the lost energy. If the attenuation of column wall is not taken into account, the attenuation of the beam through a tube full of gas and liquid and with two-phase flow is given by the following expressions (Eqs. (2)–(4)):

$$I_l = I_0 \, e^{-\mu_l \rho_l L} \tag{2}$$

$$I_g = I_0 \, e^{-\mu_g \rho_g L} \tag{3}$$

$$I_\varepsilon = I_0 \, e^{-\mu_l \rho_l (1-\varepsilon_g) L - \mu_g \rho_g \varepsilon_g L} \tag{4}$$

Integration of Eqs. (2)–(4) yields the calculation of gas holdup, as follows (Eq. (5)):

$$\varepsilon_g = \frac{\ln I_\varepsilon - \ln I_l}{\ln I_g - \ln I_l} = \frac{\ln(I_\varepsilon / I_l)}{\ln(I_g / I_l)} \tag{5}$$

According to the above Eq. (5), the local holdup can be estimated, where I_ε, I_g, and I_l are the intensities of the γ-rays in two phase, i.e. only the gas phase and the liquid phase.

According to the relationship between local holdup and height, the bubbly height can be determined, and the overall gas holdup can be easily obtained from Eq. (6).

$$\bar{\varepsilon}_g = \frac{H - H_0}{H} \tag{6}$$

3. Results and discussion

3.1. The feasibility of γ-ray measurement methods

The average column gas holdups as a function of gas velocity figured out using the differential pressure method and the γ-ray attenuation method with acetic acid in the batch mode of operation are shown in Fig. 2. Results obtained using the two methods are generally in agreement, with the results of the differential pressure method slightly smaller in general than those of the γ-ray attenuation method. This discrepancy can be attributed to the different principles of estimated values and the different measurement methods. On the other hand, gas holdups using differential pressure method only were measured under static liquid height. Since local gas holdup increases as the axial distance increases, an error may occur in the averaged value of gas holdup along the axial height.

3.2. The axial distribution of gas holdup

Fig. 3 shows the relationship between the local gas holdup and the axial distance. The local gas holdup increases with the axial height and the gas superficial velocity. The gas holdup sharply increases when transiting from the heterogeneous regime to the whole gas regime. From increasing trend, there is an

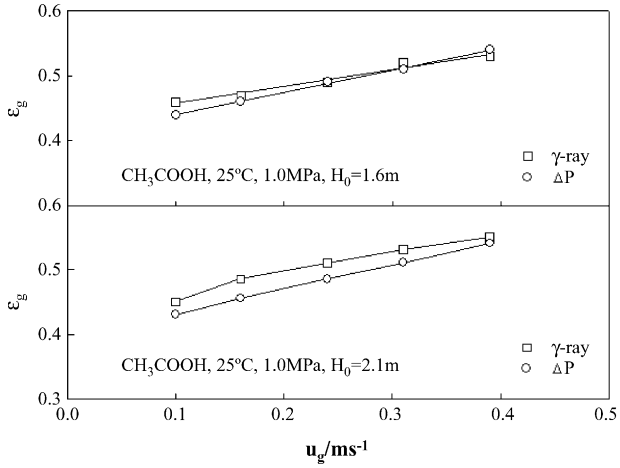

Fig. 2. Comparison of average gas holdup using two measurement methods.

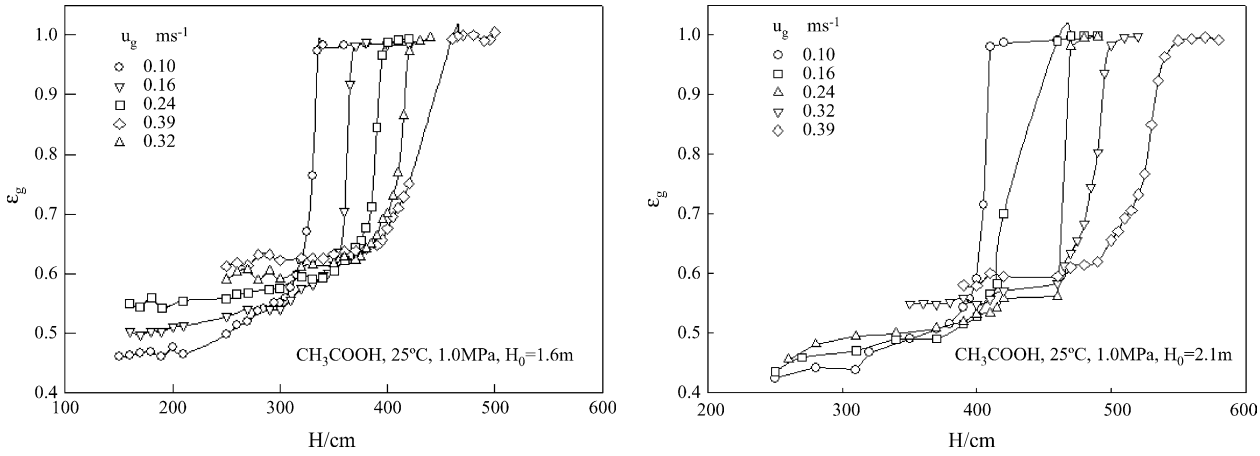

Fig. 3. The axial distribution of gas holdup.

accumulation of foam in the upper section of the column, where the gas holdup is larger than 0.8. The same trend is in agreement with the results produced by Bukur and Patel [18,19], and had been observed in our previous studies in bubble column with ambient condition. So the γ-ray method is able to measure the foam height of bubble column while the differential pressure does not react to foam height.

3.3. The effect of liquid properties on gas holdup

Experimental holdups profile for different liquid surface tension is shown in Fig. 4. The gas holdup increases almost linearly with the axial height in lower gas superficial velocity. The result further explains the accumulation of foam in the upper section of the column in lower liquid surface tension. As the gas velocity

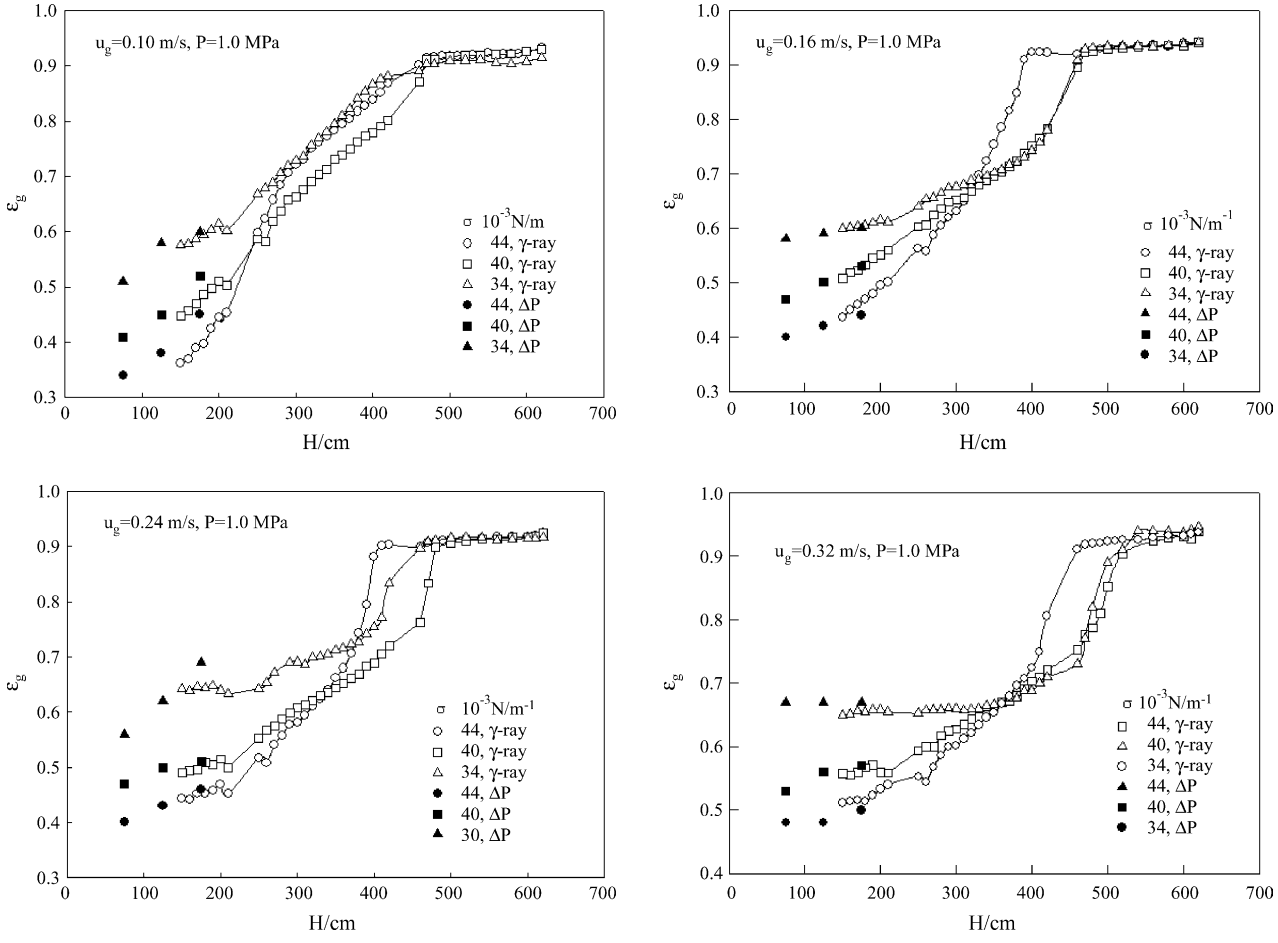

Fig. 4. Effect of different surface tension on the axial gas holdup.

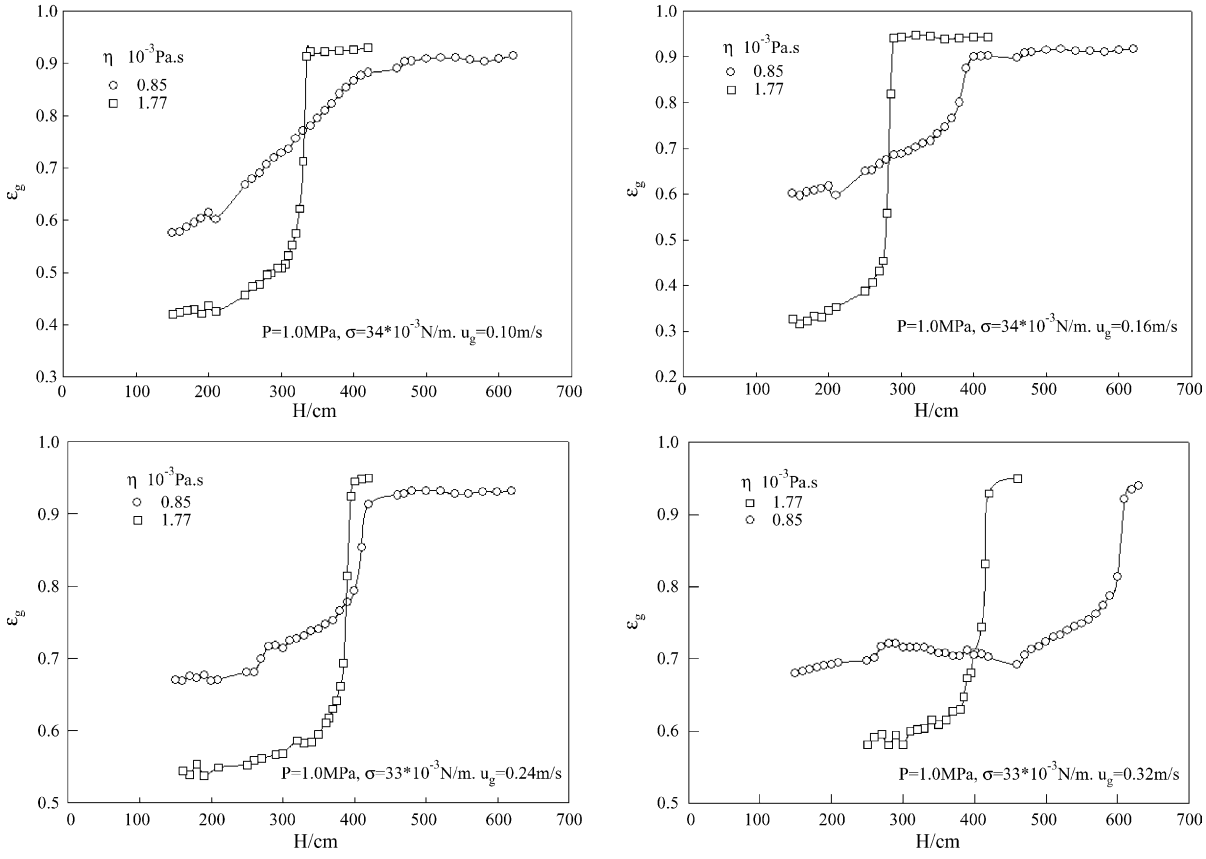

Fig. 5. Effect of viscosity on the axial gas holdup.

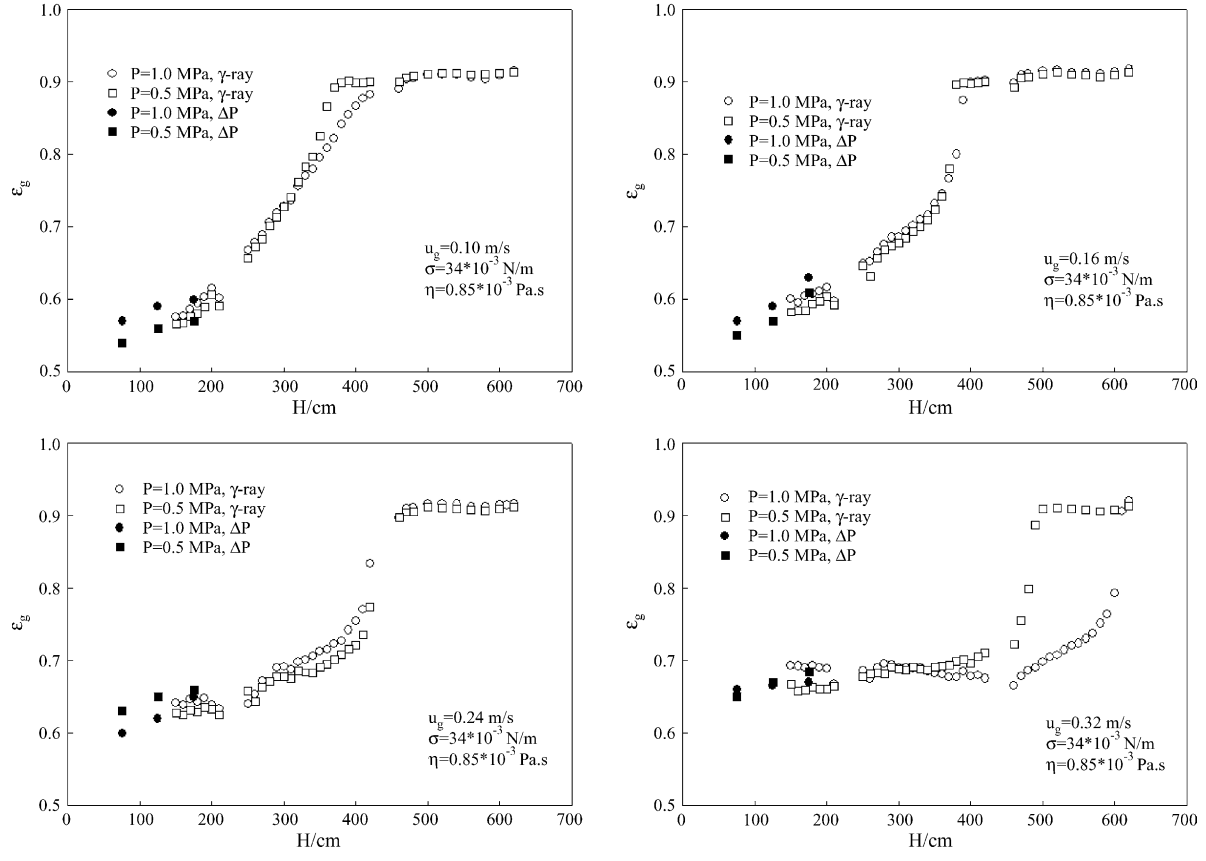

Fig. 6. Effect of pressure on the axial gas holdup.

increases, the accumulation height of foam becomes smaller and smaller because the breakup rates of bubbles further grows with an increase of gas drag force and liquid turbulent flow. The experimental results obtained from time-averaged γ-ray signals were in good agreement with measurement results using the conventional differential pressure method.

The effect of liquid viscosity on the axial distribution of local gas holdup for the same liquid surface tension with acetic acid and water used to reduce the liquid surface tension by sodium oleate can be seen in Fig. 5. Generally, the axial gas holdup and the accumulation height of foam decrease as the liquid viscosity increases at a given superficial gas velocity. Not only the viscous media has a positive effect on formation of large bubble at distributor, but also they promote bubble coalescence. In a word, the liquid viscosity also affects the distribution of local holdup.

3.4. Effect of system pressure on gas holdup

Fig. 6 illustrates results from experiments conducted at different system pressures. As the system pressure increases, the axial local holdup increases, too. When the gas superficial velocity is low, the pressure almost affects the foam height, such $u_g = 0.10$ and $0.16\ m\ s^{-1}$. With an increasing superficial velocity, the accumulation height of foam will decrease. Meanwhile the accumulation height of foam in 0.5 MPa is smaller than that in 1.0 MPa at a certain superficial gas velocity. The most plausible explanation is that the breakup probability of bubbles increases and the coalescence of bubbles decreases as the system pressure goes up, resulting in an increasing holdup of small bubbles and foam accumulation height.

4. Conclusions

Both differential pressure method and γ-ray attenuation method were used in the measurement of the axial distribution of holdups. The effects of superficial gas velocity, liquid viscosity, liquid surface tension, and system pressure were investigated in this paper. Here presented as follows are the conclusions, which are quite remarkable.

Although it is known that additional ray measurements are needed to obtain an accurate estimate of the average holdup for the entire column, the experimental results of the pressure and the ray measurements in this study were found in a very good agreement, when the axial holdup varies considerably along the height of the column.

The axial gas holdup increases with the decrease of liquid surface tension and liquid viscosity and the increase of system pressure. According to the axial distribution of gas holdup, the accumulation part of foam in the upper section of the column was obtained, and the gas holdup in foaming period is in linear growth as the axial height increases. The foaming height should be dependent on the gas and liquid properties and the operating condition.

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