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Gas Holdup in a Three-Phase Fluidized-Bed Bioreactor

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ABSTRACT

The effects of system parameters on the gas holdup in a three-phase fluidized-bed bioreactor were examined. A valve technique was used to measure the average gas holdup in the reactor. Gas holdup was found to be strongly affected by fluid superficial velocities, bead size and density, electrolyte concentration, type of gas sparger, and temperature. Quantitative and qualitative observations were made regarding the differences between conventional glass-particle systems and low-density gel-particle systems. Most notably, the degree of bubble coalescence and mode of fluidization were highly dependent on each system.

Index Entries: Bioreactor; fluidized bed; gas holdup; low-density particle; three-phase.

INTRODUCTION

Fluidized-bed bioreactors are increasingly being used in the areas of wastewater treatment and fermentations (*1*). Frequently, low-density particles composed of polysaccharide gels, such as alginate or carrageenan, are used as the solid phase. These particles provide high porosity and ease of cell entrapment. However, literature on the effects of low-density particles on fluidized-bed behavior is lacking. As with conventional fluidized-bed studies utilizing glass particles as the solid phase, characterization of the fluidized-bed bioreactor is necessary for the development of predictive mathematical models to describe the system.

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Phase holdup behavior is dependent on the following factors:

-Particle properties, such as size, density, wettability, and surface roughness.
-Fluid properties, such as gas and liquid flowrates, surface tension, viscosities, the presence of surfactants, and electrolyte concentration.
-Reactor properties, such as column geometry and sparger design.

The purpose of this investigation was to examine the effects of system parameters on the gas holdup in a three-phase fluidized-bed bioreactor. In this study, the effects of fluid superficial velocities, bead size and density, electrolyte concentration, type of gas sparger, and temperature were investigated. Results from this study will be applicable to the design and analysis of fluidized-bed bioreactors that use light, biocatalyst support beads as the solid phase.

MATERIALS AND METHODS

The experimental system is shown in Fig. 1. The fluidized-bed reactor consisted of a 5-cm ID, 155-cm long, jacketed glass column with 10 axial ports at 15-cm intervals. These ports were used to monitor the column pressure profile with water manometers. The gas sparger consisted of either a flexible rubber sparger or a glass frit. The temperature-controlled liquid phase was pumped into the reactor through a calming section of 3-mm diameter glass beads at the base of the reactor. Upon exiting the reactor, the liquid was returned to the recirculating tank. A screen was placed at the top of the column to prevent entrainment of the low-density particles. The gas phase consisted of filtered process air that was humidified at 23°C before being sparged into the reactor. A liquid seal was used at the top of the column to contain the effluent gas. The outlet gas flow rate was monitored with either a wet test meter (American Meter Company model # AL 17-1) or a graduated cylinder air/water displacement system. The gas was assumed to be saturated with water at an average temperature of the column inlet and outlet gas streams. The tank and pertinent fluid lines were insulated.

Two solid phases were used in this study. Three sizes of glass beads (specific gravity 2.5) were utilized; the average diameters of 50 beads of each size were 5.99 ± 0.07 mm, 3.03 ± 0.06 mm, and 1.30 ± 0.15 mm. Also, 4% ferric oxide enriched gel beads (2:1 ratio, 1% Gelrite to 2% alginic acid [weight ratios]) of three sizes (average diameters of 50 beads each: 3.84 ± 0.10 mm, 2.59 ± 0.14 mm, and 1.15 ± 0.15 mm) were used. The gel beads had a specific gravity of 1.015 ± 0.004. Gel beads were formed either dropwise or by the method described by Haas (2). Materials for the gel bead were chosen to give a nearly spherical, thermophilically applicable (up to 60°C), cellular support medium. Supplies were obtained from the follow-

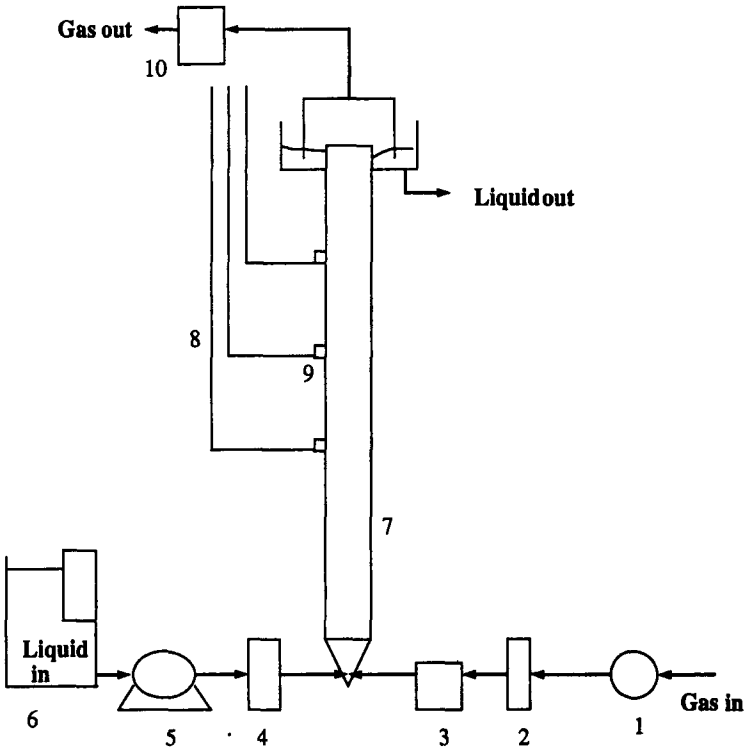

Fig. 1. Experimental system: (1) air filter, (2) flowmeter, (3) humidifier, (4) flowmeter, (5) pump, (6) liquid tank, (7) column, (8) water manometers, (9) sampling port, (10) gas measurement system.

ing manufacturers: glass geads from Jaygo Inc. (NJ) and Thomas Scientific (NJ), Gelrite from Kelco (CA), alginic acid # A-2158 from Sigma (MO), and ferric oxide # F1035 from Spectrum Chemical Mfg. Co. (CA).

An important system parameter was the liquid-phase electrolyte concentration. In order to bracket the gas holdup values expected in actual reactor operation, the method suggested by van't Riet for the scale-up and prediction of gas–liquid mass transfer coefficients was adapted to the measurement of gas holdup (3). This method entails using two ideal liquid extremes: a purely bubble-coalescing liquid, and a purely noncoalescing liquid. The gas holdup properties of a specific fermentation medium are expected to lie between these two extremes. Distilled water was used as the coalescing medium. For the noncoalescing liquid phase, a 0.2*M* $CaCl_2$ salt solution was used. The electrolyte concentration was chosen from the experimental results of Lessard and Zieminski (4). An ACS grade salt (J.T. Baker (NJ) # 1332-01) was used for all studies except for the temperature-dependent studies (E.M. Science [NJ] # CX0175-1 [anhydrous calcium chloride]).

The effects of gas and liquid superficial velocities were also studied. Liquid flow rates were varied from above the minimum fluidization veloc-

ity (determined visually) to the point of maximum solids expansion. The gas throughput was varied from about 50 mL/min of gas (at 25°C and 1 atm) to gas slugging conditions. The specific volume of the gas was calculated from an average temperature of the column inlet (approximately 23°C) and column outlet, with the moisture content of the gaseous stream assumed constant at a value equal to water-saturated air at the column inlet temperature.

Column temperature was varied between 30 and 60°C to compare gas holdup values under mesophilic and thermophilic fermentation conditions. A tolerance of 2°C was placed on this quantity (an average difference of the tank liquid temperature and the column liquid outlet temperature.)

Finally, the effects of the gas distribution system on column operation were examined. Two sparger configurations were used. The flexible gas sparger consisted of flexible amber latex tubing (1/8 in. id, 1/32 in. wall thickness, 20 cm in length) with hypodermic needle punctures (25 gge) at 0.5-cm intervals, arranged in a circular configuration. The 25-mm diamter fritted sparger (Ace Glass Inc. # 7200-06) had a maximum pore-size diameter of 25–50 μm.

A valve technique was used to measure the average gas holdup. By simultaneously stopping the gas and liquid flows into the reactor and allowing all gas bubbles to exit, the displaced volume of gas in the reactor (V_g) was measured. The average gas holdup (ϵ_g) was then calculated using the relation below

$$\epsilon_g = V_g/V_c \tag{1}$$

where V_c is the column volume. The solids loading (volume of solids added) was kept constant at 900 cm^3. In all experiments, the bed was expanded to at least 70% of the total column height to minimize the effect of the solids-free zone above the bed. The height of the fluidized bed could not accurately be measured in the low-density-particle systems because of a nondistinct two-phase/three-phase boundary. Axial pressure profiles, commonly used to study fluidized beds of dense particles, were not accurate enough to obtain an estimated fluidized height, because of the small density difference between the liquid and solid phases. In this case, the fluidized-bed height was taken as the entire column length.

RESULTS AND DISCUSSION

Glass Particle System

Several types of flow behavior were observed in these studies. In general, the gas holdup increased rapidly with gas superficial velocity at low velocities and then leveled off at higher velocities. In terms of the gas flow regimes, bubbly flow was noted at low gas velocities. The transition to churn-turbulence and slugging corresponded to the leveling off of the

gas holdup vs gas velocity curve. The larger number of bubbles produced at higher gas flow rates resulted in more frequent collisions and a greater degree of coalescence. The larger, coalesced bubbles rose faster and hence had lower residence times than smaller bubbles.

Comparison of experimental trends with literature values was excellent for the glass-particle systems (*5,6*). Bubble coalescence was generally observed for the beds of 1.30- and 3.03-mm particles. Large, coalesced bubbles were observed exiting the column, and three-phase gas holdup values were generally less than those from two-phase (gas–liquid) flow. A lack of bubble coalescence relative to the two-phase system was noticed for beds of 6-mm particles. Also, the effect of liquid velocity on column behavior was dependent on the bead size. Gas holdup was independent of liquid flow for the 1.30-mm glass beads. For beds of 3.03- and 5.99-mm particles, increasing the liquid superficial velocity decreased the gas holdup, but increased the gas velocity at which slugging first occurred.

Low Density System

Solid Bead Size

The solid bead size influenced both the gas holdup and axial movement of the solid phase. The relation between gas velocity and gas holdup for all three gel bead sizes at the same liquid superficial velocity is given in Fig. 2. The interpolating curves were obtained from a second-order polynomial fit with an α level for T statistics of 0.05. These experiments were carried out at 30°C using 0.2*M* $CaCl_2$ and the rubber sparger. Over the range of gas velocities studied, three-phase holdup values exceeded two-phase values (two-phase values are not shown). In addition, holdup values were slightly larger for the 3.84-mm gel beads than for the smaller bead sizes. Although not quantitatively measured, a large variance in solids movement was visually observed. The 1.30-mm gel beads were influenced most by the addition of gas to the liquid–solid bed. Particle mixing appeared to decrease with increasing bead diameter. The degree of solids mixing may affect the column performance by increasing the liquid axial dispersion and by subjecting the biocatalyst support particles to large changes in substrate concentration.

Gas Superficial Velocity

The effects of gas superficial velocity on gas holdup are also illustrated in Fig. 2. At low gas velocities (below 0.3 cm/s), bubbly flow was observed. Here, a sharp increase in the gas holdup was obtained with increasing gas flow. With an increase in the gas throughput, transitions to churn-turbulent and slugging flow regimes occurred. These transitions are shown by a smaller increase in gas holdup with increasing gas velocity relative to the bubbly flow regime. Trends observed for the low-density-particle systems were similar to those observed for the glass-particle systems. However, the range of gas velocities was smaller for the low-density-particle systems.

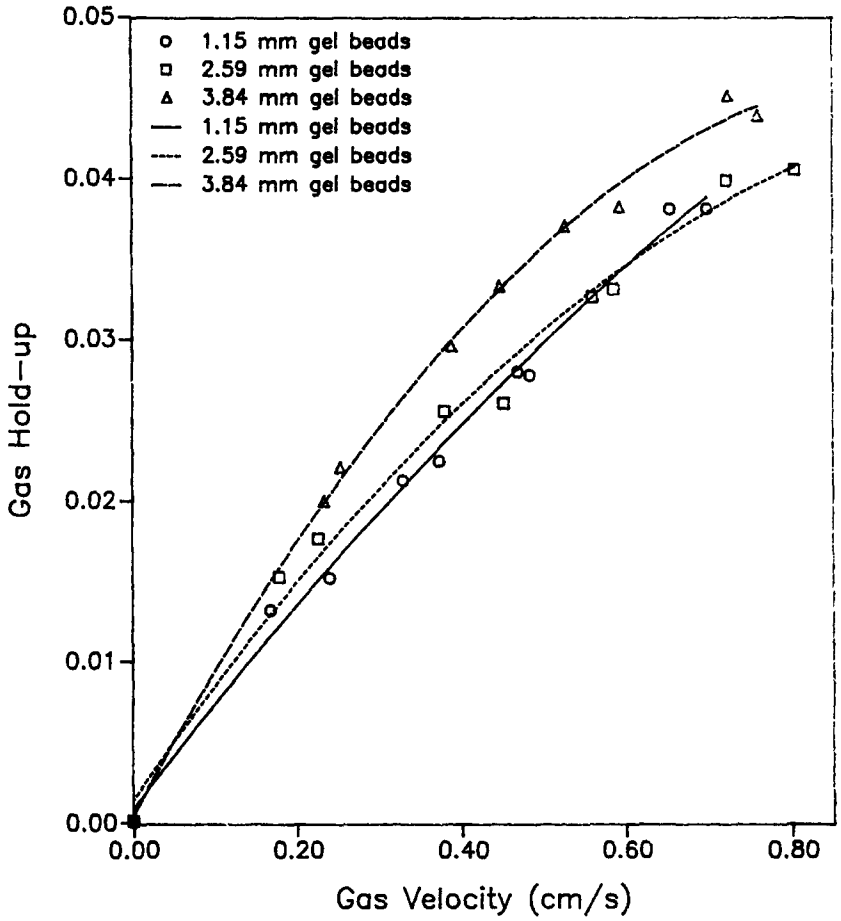

Fig. 2. Effects of gas velocity and gel bead size on gas holdup: $U_l = 0.525$ cm/s, 0.2*M* $CaCl_2$, rubber sparger, 30°C.

Liquid Superficial Velocity

Liquid velocity had a substantial effect on the reactor flow conditions, as shown in Fig. 3 for the 1.15-mm diameter gel beads in the noncoalescing medium. In general, increasing the liquid superficial velocity led to an increase in the bed expansion and an increase in the gas velocity at which gas slugging first occurred. However, increasing the liquid flow rate may decrease gas holdup by increasing the bubble-rise velocity. This effect was most noticeable for two-phase (gas–liquid) flow conditions. (Bubble-rise velocity was not measured.)

Liquid Electrolyte Concentration

Liquid characteristics strongly affected phase holdups and transitions between gas flow regimes. The effects of liquid type (coalescing or noncoalescing) on gas holdup are illustrated in Fig. 4 for the 3.84-mm gel beads. Gas holdup values differed by as much as a factor of two for a given

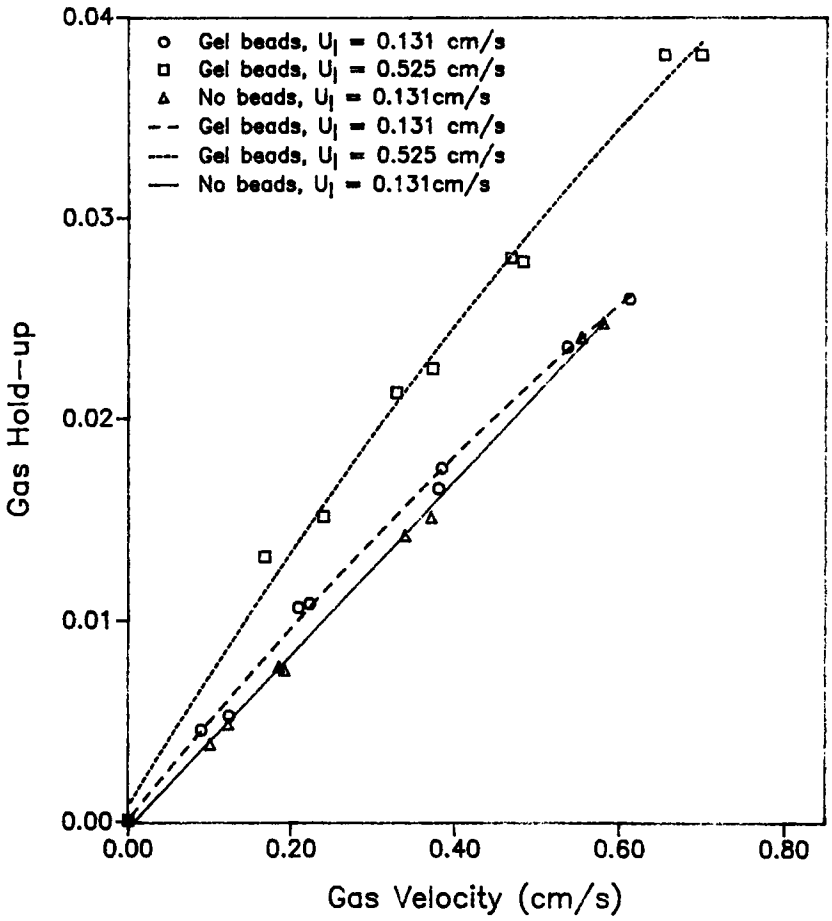

Fig. 3. Effect of liquid superficial velocity on gas holdup for 1.15-mm gel beads: 0.2*M* $CaCl_2$, rubber sparger, 30°C.

gas velocity. Smaller initial bubble sizes as well as a decrease in bubble coalescence were observed for the salt solution, resulting in increased gas holdup values. In addition, a larger gas velocity could be used before slugging occurred for the noncoalescing medium than for the coalescing medium. These results were expected for the electrolyte solution because of the noncoalescing nature of the liquid.

Sparger Comparison

Comparison of the glass frit sparger and the flexible rubber sparger is shown in Fig. 5. The glass frit sparger produced a smaller bubble size (observed visually) than the rubber sparger at a given gas velocity. In addition, identical gas velocities were achieved at a lower pressure drop for the glass frit. Using the glass frit sparger, holdup values were up to twice as large as those for the rubber sparger under three-phase flow conditions. Also, higher gas velocities were used before gas slugging was

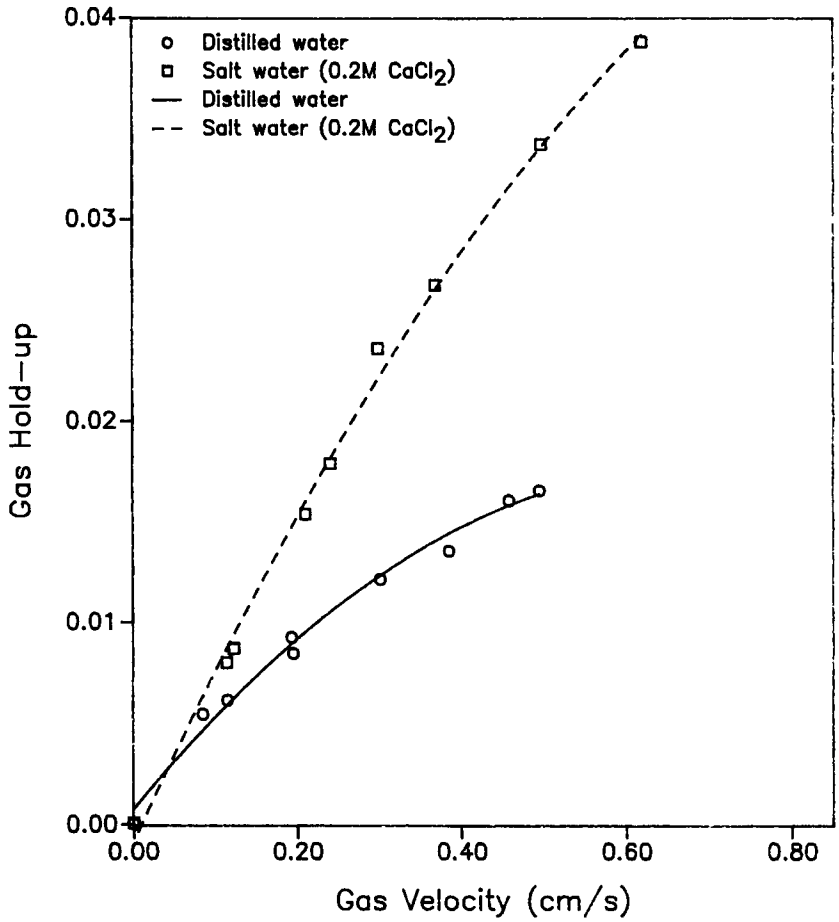

Fig. 4. Effect of electrolyte concentration on gas holdup for 3.84-mm gel beads: $U_l = 0.592$ cm/s, glass frit sparger, 30°C.

observed. These results conflict with those of Rice et al. (*7*), who studied dispersion and holdup in bubble columns with both rigid and flexible spargers. Perforated rubber sheets were found to produce more uniform emulsions, smaller bubbles, and larger gas voidages than perforated rigid plates. In addition, the onset of gas slugging was repressed for the perforated-rubber-sheet sparger system.

Temperature

The effects of temperature on gas holdup in the three-phase reactor are shown in Fig. 6. An increase in temperature led to an increase in gas holdup for both three-phase and two-phase conditions. Also, column temperature had no noticeable effect on the transition from churn-turbulent to slug flow as indicated from the gas velocity/gas holdup relation. As the surface tension and viscosity decrease with increasing temperature, an increase in bubble coalescence and thus a decrease in gas holdup was expected. However, experimental results agree with the experimental

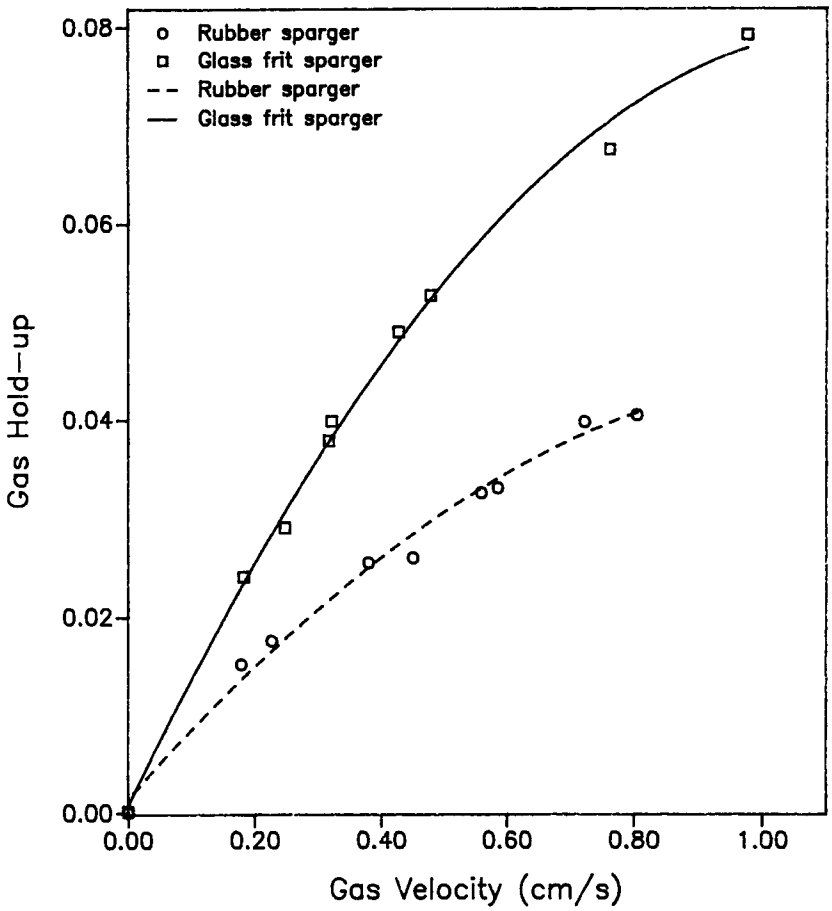

Fig. 5. Effect of sparger type on gas holdup: 2.59-mm gel beads, 0.2*M* $CaCl_2$, $U_l = 0.525$ cm/s, 30°C.

work of Grover et al. (*8*) and Smith et al. (*9*) at low gas velocities (less than 2.5 cm/s) in gas–liquid bubble columns, where an increase in gas holdup with increasing temperature was observed.

Experimental trends for the low-density biocatalyst-support-particle systems were significantly different from those observed for the conventional glass-ballotini system. Most notably, bubble coalescence was observed under most operating conditions for the gel particles. The degree of coalescence was dependent on system properties such as bead size and liquid type. For example, even though the 0.2*M* $CaCl_2$ solution was considered to be a noncoalescing liquid, bubble coalescence was sometimes apparent in the form of large bubbles as well as swarms of smaller bubbles. Another difference between the glass-particle and gel-bead systems was the mode of fluidization. The glass beads were mainly fluidized by the upflow of liquid. Gas flow did not aid in the solid-phase fluidization. The low-density particles, on the other hand, were partially fluidized by the gas phase. This effect was apparent when the gas phase was added to the

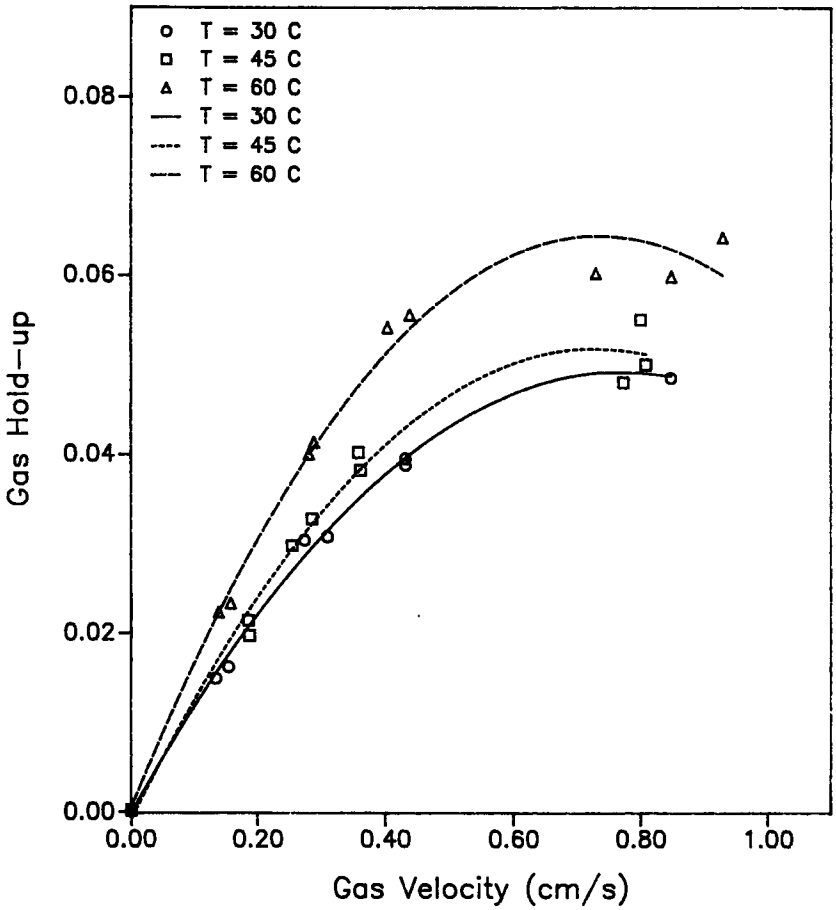

Fig. 6. Effect of temperature on gas holdup: 1.15-mm gel beads, 0.2*M* $CaCl_2$, $U_1 = 0.372$ cm/s, glass frit sparger.

two-phase (liquid–solid) system. In general, for all bead sizes, nondistinct bed heights were observed for the three-phase case. Even though a distinct bed height was obtained for the two-phase (liquid–solid) condition, addition of the gas phase produced a solids gradient throughout the column. The gradient appeared to be most pronounced for the 1.15-mm beads and least pronounced for the 3.84-mm gel beads. However, it could not be quantified manometrically because of the small density difference between the solid and liquid phases and the wall–particle frictional effects. In addition, considerable axial movement of the particles was visually observed. Movement was most notable for the small beads and decreased with increasing particle size.

The mode of fluidization, nondistinct bed height, and axial movement of solids were a result of the effects of bubble wakes; namely, small, low-density particles were influenced more by rising bubble wakes relative to larger, heavier solids.

In this study, although an increase in bubble coalescence was observed relative to gas–liquid flow conditions, three-phase gas holdup values were sometimes larger than corresponding two-phase values. This effect is illustrated in Fig. 3 for the 1.15-mm gel beads in the noncoalescing liquid. The difference between two- and three-phase gas holdup values was highly dependent upon the liquid type as well as the gas and liquid superficial velocities. This positive effect of low-density particles on gas holdup is consistent with the results of Smith et al. (*9*) for wettable solids. Differences in experimental results with the work of Verlaan et al. (*10*), and Verlaan and Tramper (*11*) may result from the different initial bubble sizes produced.

Bubble size is important in determining the reactor hydrodynamic behavior. For large bubbles, Verlaan et al. (*10*) concluded that the effect of the solid phase was to increase the bubble collision frequency by decreasing the area for flow of the gas–liquid mixture. However, when the solid particle is larger than the bubble, the effect of the neutral-density solid phase may be to impede the upward motion of the bubble. In this study, the initial bubble size was less than the equilibrium bubble size of air in water (as was used by Verlaan et al. (*10*). As a result, the primary effect of the solid phase in the sparger area may have been to impede the upward motion of the bubbles. Thus, gas holdup increased relative to two-phase flow conditions. This concept also explains the larger difference between two- and three-phase values for noncoalescing media than for coalescing media. Namely, smaller initial bubble sizes were obtained for the noncoalescing liquid. Consequently, the solid–bubble interactions were more significant. At higher gas velocities, an increase in collision frequency and bubble size led to an increase in coalescence. This effect, as was suggested by Verlaan et al. (*10*), was caused by a decrease in flow area by the presence of the solids. As a result, gas holdup values decreased relative to two-phase flow conditions.

CONCLUSION

Gas holdup was found to be strongly affected by fluid superficial velocities, bead size and density, electrolyte concentration, type of gas sparger, and temperature. Holdup values differed by up to a factor of two, depending on the gas sparger and the liquid type used. In addition, it was shown that low-density-particle systems behaved much differently than conventional glass-particle systems. Most notably, the degree of coalescence and mode of fluidization were highly dependent on each system. Research is needed to characterize important phenomena, such as axial dispersion and mass transfer, for low-density particle systems. These results will be necessary for the predictive modeling and scale-up of fluidized-bed bioreactors.

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REFERENCES

1. Andrews, G. (1988), *Biotechnol. Genet. Eng. Rev.* **6,** 151.
2. Haas, P. (1975), *AIChE J.* **21,** 383.
3. van't Riet, K. (1983), *Trends Biotechnol.* **1,** 113.
4. Lessard, R. and Zieminski, S. (1971), *Ind. Eng. Chem. Fund.* **10,** 260.
5. Michelsen, M. L. and Ostergaard, K. (1970), *Chem. Eng. J.* **1,** 37.
6. Dhanuka, V. R. and Stepanek, J. B. (1978), *Proc. Second Eng. Foundation Conf.*, Davidson, J. F., and Keairns, D. L., eds., Cambridge University Press, Cambridge, UK, 190.
7. Rice, R., Tupperainen, J., and Hedge, R. (1981), *Can. J. of Chem. Eng.* **59,** 677.
8. Grover, G., Rode, C., and Chaudhari, R. (1986), *Can. J. of Chem. Eng.* **64,** 501.
9. Smith, E. L., Jamialahmadi, M., Olajuyigbe, J. T., and Shayegan Salek, J. (1986), *Proc. of BHRA Int. Conf. on Bioreactor Fluid Dynamics,* Cambridge, England, p. 677.
10. Verlaan, P., Tramper, J., van't Riet, K., and Luyben, K. (1986), *Proc. of BHRA Int. Conf. on Bioreactor Fluid Dynamics,* Cambridge, England, p. 93.
11. Verlaan, P., and Tramper, J. (1987), *Int. Conf. on Bioreactors and Biotransformations,* Gleneagles, Scotland, p. 363.