tion,  $\phi = h_0 \sqrt{\frac{r_0}{r}}$ , and either  $y(r) = \tanh \phi$  or  $\sqrt{r} \tanh \phi$ will generate Equations (6) and (7) but with  $\psi = h_0 \sqrt{r_0} M_{n-1}/M_{n-1/2}.$ 

#### **ACKNOWLEDGMENTS**

This study was made possible by support from the Canadian Federal Department of Energy, Mines and Resources, Faculty of Engineering, University of Waterloo, and the Japanese Ministry

#### NOTATION

f(r) = pore size density distribution function

= rate of gasification function

y(r) = function occurring in Equation (1)  $h_0$  = Thiele modulus for a first-order reaction based on the most probable pore radius at t = 0 in a transient system

 $M_n = n$ th moment of f(r)

r,  $r_0$  = pore radius, most probable radius for f(r) at time

β = coefficient in Gaussian distribution

= pseudo effectiveness factor  $e_n$ 

= value of r satisfying Equation (1)

= (superscript) approximation

#### LITERATURE CITED

Korn, G. A., and T. M. Korn, Mathematical Handbook for Scientists and Engineers, p. 116, McGraw-Hill, New York

Hashimoto, K., and P. L. Silveston, "Gasification—Part II: Extension to Diffusion Control," AIChE J., 19, 00 (1973).

—————, "An Effectiveness Factor Approximation for

Pore Size Distributions," ibid., 17, 745 (1971).

Mingle, J. O., and J. M. Smith, "Effectiveness Factors for Porous Catalysts," ibid., 7, 243 (1961).

Manuscript received April 2, 1971; revision received August 29, 1972; note accepted October 13, 1972.

# Behavior of Gas Bubbles in Fluidized Beds

## ALOYSIUS GARCIA, JOHN R. GRACE, and ROLAND CLIFT

**Department of Chemical Engineering** McGill University, Montreal, Canada

In a recent paper, Rieke and Pigford (1971) described experiments on injected bubbles in a two-dimensional gasfluidized bed which were interpreted as contradicting commonly held theories of fluidization. In particular, they surmised that the flow of gas inside bubbles rising in fluidized beds is downwards, whereas fluid mechanical theories suggest that the gas flow inside a bubble is upwards and of order  $U_{mf}$  (the minimum fluidization velocity) with respect to a frame of reference moving with the bubble (Davidson, 1961; Davidson and Harrison, 1963; Jackson, 1963; Murray, 1965). Moreover, Rieke and Pigford interpreted their photographs as showing that there is a large wake of gas which moves with a bubble and grows with time. By contrast, theory predicts that for a bubble in steady motion, even when there is a particle wake, the size of the gas cloud enveloping the bubble and its particle wake is constant (Clift et al., 1972; Collins, 1965). Finally, Rieke and Pigford concluded that there is a substantial flow of particles through a bubble falling from the roof to the floor, whereas the theoretical studies either assume or deduce that the particle motion relative to a bubble is approximately as in an incompressible irrotational flow.

In this note, we show that it is possible to interpret the photographs of Rieke and Pigford as being consistent with all essential conclusions of fluid mechanical theories of fluidization. We also present direct experimental evidence that the velocity of gas inside a rising bubble is upwards and of order of magnitude  $U_{mf}$ , in agreement with theory.

## EXPERIMENTAL DETAILS

In observing coal particles of very wide size distribution fluidized in a two-dimensional column, one of the present authors noted that the resulting bubbles appeared grey, apparently because fines were being entrained in the rising bubble. This suggested that small particles could be used as tracers to provide a rough measure of throughflow velocities in fluidized beds. In the present work, a simple experiment was devised in which small glass beads were used as tracer particles in a bed of larger glass beads.

The experiments were performed in a transparent acrylic column of inside dimensions  $1.5~\mathrm{cm} \times 51~\mathrm{cm} \times 120~\mathrm{cm}$  deep having a sintered bronze distributor. The column was filled to a depth of 90 cm with glass beads having a size range of 420 to 600 μm and a minimum fluidization velocity (with air) of 24 cm/s. The tracer particles were glass beads which had been sieved to four narrow size ranges having surface mean diameters of 20, 23, 27, 35, and 44  $\mu$ m, respectively. These particles were dyed red to allow them to be distinguished clearly from the larger particles which formed the bulk of the

In order to find whether the tracer particles were entrained in rising bubbles, it was desirable to form a bubble with a wake rich in tracer particles. This was accomplished by admitting a pulse of air which entrained about 10 g of tracer particles as it entered the bed from a pressurized reservoir. Before each plug of air and tracer particles was injected into the column, the bed was fluidized vigorously for several minutes and then the air flow rate was reduced until the superficial velocity was only slightly in excess of the minimum fluidization velocity. Each injector together with the subsequent bubble and tracer particle motion was recorded by cinephotography on color film at 64 frames per second.

## RESULTS

When bubbles were formed as described above, a red particle wake could be clearly distinguished. The films showed that, for all but the largest tracer particles, a thin red mantle formed at the roof of the bubble as it rose. This mantle was most clearly defined for the smallest tracer particles. The labeled particles could only have reached the roof to form such a mantle by being entrained from the wake. They subsequently moved downwards around the periphery of the bubble, some of them appearing to pass into the wake of particles and some being left behind as the bubble rose.

These observations clearly indicate upwards gas flow inside a bubble relative to the bubble and ordered particle motion in the dense phase. The experiments do not provide a precise estimate of the throughflow velocity inside a rising bubble since the tracer particles were not monodisperse and, in any case, the terminal velocity of the tracer particles is bound to depend strongly on particle concentration. An order of magnitude of the gas velocity upwards relative to a bubble was obtained by putting thin horizontal layers of the four smaller sizes of tracer particles on top of the larger bed particles and gradually increasing the superficial gas velocity. Tests of this sort revealed that the tracer particles began to be entrained from the bed at a superficial velocity close to the minimum fluidization velocity for the large particles. It can therefore be concluded that the gas velocity inside a bubble rising in a two-dimensional fluidized bed is of the same order as the minimum fluidization velocity.

## DISCUSSION

The experiments described above indicate clearly that the gas motion inside a bubble in a fluidized bed is inwards at the rear and outwards at the roof. Since this evidence is directly counter to the interpretation which Rieke and Pigford (1971) placed on their photographs of bubbles containing nitrogen dioxide, it is of interest to re-examine their findings.

Rieke and Pigford used 100 µ glass beads with a minimum fluidization velocity of 1.5 cm/s. For a typical bubble radius of 1.6 cm and corresponding velocity of 28 cm/s, the model developed by Murray (1969) which has been found to agree most closely with experimentally determined cloud sizes (Rowe et al., 1964) predicts a cloud thickness of 0.15 cm or 15 particle diameters at the front of the bubble. With such a thin cloud, significant thickening of the cloud with angular position would be expected due to mixing processes occurring on the scale of a single particle (Grace, 1971) augmented by molecular diffusion and adsorption-desorption of NO2 on the particles. Thus, according to the established theories of fluidization, one would expect the photographs to show an almost imperceptible cloud at the front of the bubble, thickening at the sides as the tracer gas is transferred across the formal cloud boundary and a spreading dark region

behind the bubble formed of the gas transferred out by mixing. This is exactly what Rieke and Pigford report. It might also be expected that the volumetric rate of spreading of the dark wake region would be of the same order as the flow rate out of the upper surface of the bubble, since mixing will be strongest in this region where the concentration profile is undeveloped. It may be noted that the reported rate of growth of nitrogen dioxide-containing gas is 1.2 to 1.5 cu in/s, while the gas flow out of the bubble predicted by the model of Davidson and Harrison (1963) is 0.72 cu in/s.

The reasoning which led Rieke and Pigford to deduce that the gas flow is downward in the bubble can now be reassessed. This conclusion followed from a mass balance based on the assumption that there is no flow of gas across the boundary of the region marked by nitrogen dioxide. Since this boundary simply represents the visible mixing front, the assumption is unfounded. The conclusion that there is a substantial flow of particles through the bubble followed from a solids balance, this time based on the additional assumption that no solids move across the boundaries of the dark regions. Regardless of gas mixing processes, this assumption is obviously not valid: it amounts to assuming that the gas and solids streamlines coincide. Since the basic assumptions behind the respective material balances are in error, conclusions based on such material balances have no validity. Established theories must certainly be put to the test, but it is essential that in doing so, workers do not neglect important phenomena such as the mass transfer occurring across cloud boundaries in fluidized beds.

## CONCLUSIONS

It has been shown that the gas motion inside a bubble in a two-dimensional fluidized bed is upwards, with a velocity of the same order as the superficial gas velocity at minimum fluidization. The experiments of Rieke and Pigford are consistent with established theories of fluidization and demonstrate that in some circumstances there can be significant transfer of gas across the cloud boundary due to mixing processes occurring on the scale of a single particle augmented by molecular diffusion and adsorptiondesorption.

#### LITERATURE CITED

Clift, R., J. R. Grace, L. Cheung, and T. H. Do, "Gas and solids motion around deformed and interacting bubbles in fluidized beds," J. Fluid Mech., 51, 187 (1972).

Collins, R., "An extension of Davidson's theory of bubbles in

fluidised beds," Chem. Eng. Sci., 20, 747 (1965).

Davidson, J. F., "The flow of fluidising fluid round a spherical void in a fluidised bed," Trans. Instn. Chem. Engrs., 39, 230 (1961).

-., and D. Harrison, Fluidised Particles, Cambridge Univ. Press, England (1963).

Grace, J. R., "A note on bubble clouds in fluidized beds,"

Chem. Eng. Sci., 26, 1955 (1971).

Jackson, R., "The mechanics of fluidised beds: Part II: The motion of fully developed bubbles," Trans. Instn. Chem.

Engrs., 41, 22 (1963).

Murray, J. D., "On the mathematics of fluidization: Part 2: Steady motion of fully developed bubbles," J. Fluid Mech., **22**, 57 (1965).

Rieke, R. D., and R. L. Pigford, "Behavior of gas bubbles in fluidized beds," AIChE J., 17, 1096 (1971).

Rowe P. N., B. A. Partridge, and E. Lyall, "Cloud formation

around bubbles in gas fluidised beds," Chem. Eng. Sci., 19,973 (1964).

Manuscript received August 25, 1972; note accepted November 16, 1972.