$$- [0.001(T - 1100)]\} + \left(\frac{0.001\,B(-\Delta H)}{3.5(1.7 \times 10^5\,C)} \right)$$

$$[3.5p]\ [1.7 \times 10^5\,k]$$

$$[1.7 \times 10^5\,k] = f\,[0.001(T - 1100)]$$

$$[0.001(T_0 - 1100)] = 0.200$$

Settings of variable diode function generator (Arrhenius approximation):

$[0.001(T - 1100)]$	$[1.7 \times 10^5\,k]$
0.0	0.0006
0.1	0.0024
0.2	0.010
0.3	0.035
0.4	0.103
0.5	0.258
0.6	0.595
0.7	1.250
0.8	↑
0.9	overload (7)
1.0	↓

Program details (see Figure 12).

Manuscript received September 29, 1969; revision received October 25, 1970; paper accepted November 13, 1970.

Behavior of Gas Bubbles in Fluidized Beds

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Apparently it is generally accepted that, of the total volumetric flow of gas upward through a fluidized bed of solid particles, only a small fraction stays in the particulate or emulsion phase where most of the solid particles are. The superficial gas velocity in this part of the bed is nearly the same as the superficial velocity at minimum fluidization u_{MF}, which is usually much smaller than the superficial velocity u corresponding to the total fluid flow rate. The excess of gas, corresponding to $u - u_{MF}$, flows through the bed as gas pockets or "bubbles."

Each of these cavities contains only a small mass of the solid phase and, for a fluidized catalytic reactor, the extent of contact between the solid catalyst and most of the fluid stream depends on the circulation of fluid in and out of the gas bubbles. Gas which does not escape the bubbles on their way through the catalyst mass will have by-passed the catalyst and will not have reacted. Thus, for the rational design of a fluidized reactor it is essential to know how rapidly gas flows between bubbles and emulsion and by what mechanism such exchange occurs.

PREVIOUS WORK

The most promising calculations of fluid flow patterns around a rising gas bubble in a fluidized bed are those of Davidson and Harrison (*6*) who conclude that the velocities of the solid particles around a bubble can be described by a velocity potential. Indeed, rising velocities of bubbles calculated on this basis were found to agree within about 10% with observed velocities. The data covered a variety of solid-particle sizes and densities and a wide range of bubble volumes. Their formula is

$$U_B = 0.71\,(g_L{}^3\,V_B)^{1/6} \qquad (1)$$

which closely resembles an earlier formula for the rising velocity of a large gas bubble in a liquid owing to Davies and Taylor (*8*). For two-dimensional gas bubbles in liquids contained between large flat plates the measurements of Collins (*13*) lead to

$$U_B = 0.66\,(g_L{}^2\,V_B/h)^{1/4} \qquad (2)$$

where h is the distance between plates.

With such surprising success in their application of potential flow methods to the bubble motion in fluidized beds, Davidson and Harrison went on to compute the relative movement of gas through the emulsion phase around a gas-filled cavity. They used the Darcy Law for this purpose, concluding that the pressure profile in the emulsion phase was represented by the Laplace equation with a uniform pressure on the surface of the cavity. In this way both gas and solid particle streamlines could be found around a cavity of assumed shape.

Figure 1 shows a few of their calculated gas streamlines around an assumed spherical cavity for the practically

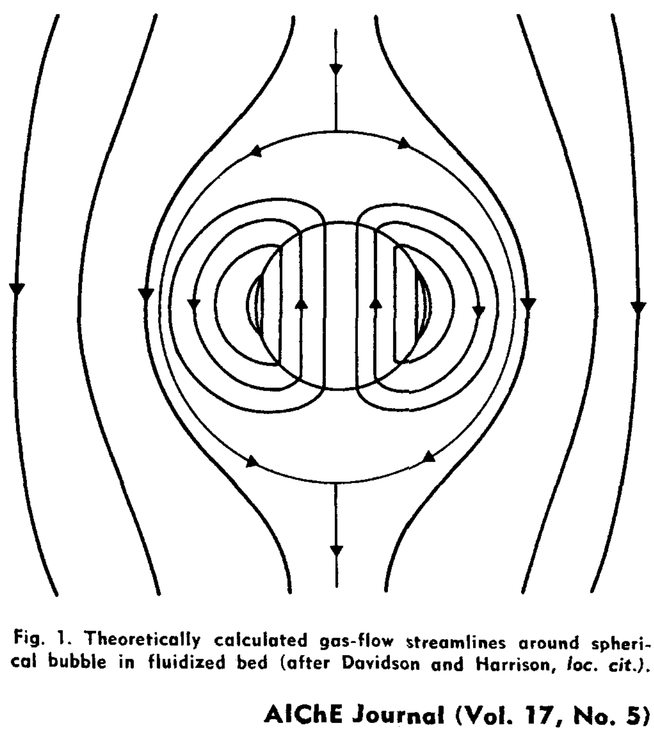

Fig. 1. Theoretically calculated gas-flow streamlines around spherical bubble in fluidized bed (after Davidson and Harrison, *loc. cit.*).

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important case in which U_B is greater than the interstitial fluid velocity $U_G = u_{MF}/\epsilon$. Under these conditions the solid particles which descend around the bubble are able to carry gas with them, the gas filling the interstices between particles. This is in spite of the pressure gradient in the distant emulsion phase, which tends to make gas flow upward at the velocity U_G. Near the lower stagnation point, however, the pressure gradient can force gas upward into the base of the cavity; similarly, across the roof of the bubble the pressure gradient forces gas to flow outward into the particulate phase. The result is a circulating "cloud" of gas which flows in a toroidal pattern into and out of the particulate phase, following the bubble as it rises.

The calculations show that there is a circular dividing streamline which contains all the toroidal flow pattern. Gas outside this streamline never enters the bubble; gas originally inside the bubble cannot cross the circular streamline and thus cannot escape into the distant particulate phase except by slow molecular diffusion. Rowe, Partridge, and Lyall (2) have plotted the ratio of the calculated radius of the dividing streamline to the radius of the spherical bubble against the ratio of the bubble's rising velocity to the velocity of the interstitial gas, as in Figure 2. For the conditions of the experiments to be reported below, in which the particles were light and their minimum fluidizing velocity was small, the radius of the spherical shell of particulate phase which is invaded by gas from the rising bubble is only a little greater than the radius of the bubble itself.

Davidson and Harrison (6) have computed the volumetric rate at which gas leaves the cylindrical cavity of a two-dimensional bubble through its upper semicircular surface. Their formula is

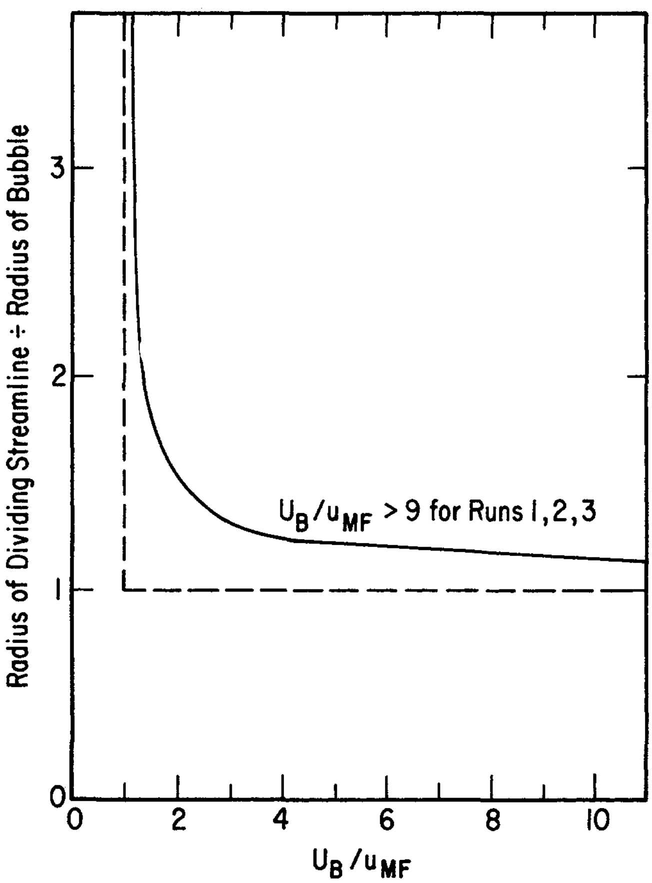

Fig. 2. Change of thickness of cloud boundary layer as bubble rising velocity changes (after Davidson and Harrison, *loc. cit.*).

$$q = 2\, u_{MF}\, d_B\, h \tag{3}$$

where d_B is the bubble diameter and h is the spacing between the flat sides of the apparatus. With this estimate of gas exchange rate Davidson and Harrison were able to compute the conversion of a reactant introduced into the gas feed stream, knowing the specific reaction rate per unit volume of the solid-filled particulate phase. There was at least qualitative agreement with some data from several experiments (6, 10); this lent additional support to the idea that the all-important bubble motion phenomena which are required in any theory of fluid bed performance had been successfully resolved. Indeed, in more recent work Kunii and Levenspiel (4) further exploited the potential flow formulas and somewhat extended the same ideas.

Nevertheless, in spite of the empirical evidence favoring the potential flow calculations, doubts about the flow phenomena themselves have been expressed occasionally. For example, evidence has been cited that bubbles sometimes split into smaller fragments because of fluid mechanical or other instability. Perhaps the splitting phenomenon may be important if it occurs frequently enough. There is also evidence (11) that bubbles grow larger as they rise at rates far exceeding the growth rates expected from the reduction of static fluid pressure. Such growth must represent influx of gas from the particulate phase; if rapid enough, the influx could have a major effect upon the exchange rates of gas to and from bubbles.

Finally, observations have sometimes shown that the bubbles are not completely void of solid particles. In fact, it seems intuitively likely that particles at the bubble roof might disentangle themselves from their neighbor particles and fall through the bubble cavity to the floor. After all, the upward fluid velocity inside the bubble corresponding to the upward volumetric flow q is usually not enough to support freely falling particles. If the upward velocity did exceed the particle terminal velocity, particles could easily be lifted from the floor, eventually resulting in disruption of the bubble itself (6).

Thus, because of the importance attached to bubble leakage phenomena and because not very many experimental observations of the phenomena have been reported, it was concluded that new experimental data might prove valuable. The experiments to be described confirm that some of the details of the theory are wrong, although the estimated rates of exchange of gas to and from rising bubbles may be about right.

EXPERIMENTAL WORK

A two-dimensional gas-fluidized bed was constructed that would make it possible to photograph single bubbles containing colored nitrogen dioxide. Such bubbles could be injected singly near the bottom of the bed and could be followed photographically as they rose and as nitrogen dioxide leaked from them into the white particulate phase. The bed, consisting of 100-μ glass beads, was contained between two vertical sheets of plate glass ½ in. apart. Each side of the rectangular channel formed between the glass plates was 10 in. wide. The bed was supported on a ⅛-in.-thick sintered bronze plate having a mean pore diameter of 30μ.

Gas bubbles were formed at the end of a stainless steel tube 1.5 mm. in diameter which was directed upward at a point about 4 in. above the distributor plate and in the center of the bed's cross section. This small tube was connected to a stainless steel chamber of 49-cc. volume in which a known quantity of a nitrogen dioxide-air mixture could be stored. The quick opening of a valve between this chamber and the tube produced a gas bubble in the bed and set off a timing mechanism which later turned on two photographic flash lamps, catching the bubble in flight through the bed. The measurement of pressure and temperature in the 49-cc. chamber before and after release of the bubble permitted the calculation of the initial volume of gas in the bubble.

Photographs were taken on Kodak Contrast Process Orthochromatic film, which is sensitive to red light reflected from the nitrogen dioxide gas in and around the bubbles. Lighting was by means of two Honeywell-600 Strobonar electronic flash guns. The delay mechanism which activated the flash guns was calibrated with an electronic timing device.

The bed was fluidized vigorously before each measurement and the flow rate of the fluidizing air stream was reduced to the minimum fluidizing point just before a bubble was injected. By adjusting the time delay setting, successive bubbles of the same initial volume could be photographed at various distances above the injection point. Three series of experiments were made, two with beds of about 10 in. and 16 in. in depth and two with initial bubble volumes of 0.19 and 0.32 cu. in. Measurements were made on the photographs of the areas covered by the bubble cavities and their nitrogen dioxide-stained wakes, using a planimeter. Figure 3 shows a typical picture of a bubble, one of a series of similar photographs and bubbles, all of the same initial size, at increasing heights above the injection point. Table 1 summarizes the experimental conditions and gives some of the derived results.

RESULTS OBTAINED FROM PHOTOGRAPHS

Figure 4 is a plot of the observed positions of bubbles, initially of 0.19 cu. in. volume, rising through 16 in. of bed. The straight line fitted to the data by least squares gave a rising velocity of 10.8 in./sec. with a standard error of 1.0 in./sec. This value and two others from the other series are shown on Figure 5 in comparison with Equation (2). The comparison supports the conclusions of Davidson and Harrison (*6*), Harrison and Pyle (*3*), and Harrison and Leung (*5*), that potential flow of the solid particles seems to occur near the forward stagnation points above the bubbles.

The most striking feature of the photographs is the rapid increase in the volume of the stained wake underneath each bubble. Figure 6 is a plot of wake volumes computed by multiplying the areas measured from photographs by the thickness of the bed. As indicated in Table 1, the wake was increasing in this series at the rate 2.82 cu. in./sec. with a standard error of 0.21 cu. in./sec. Thus,

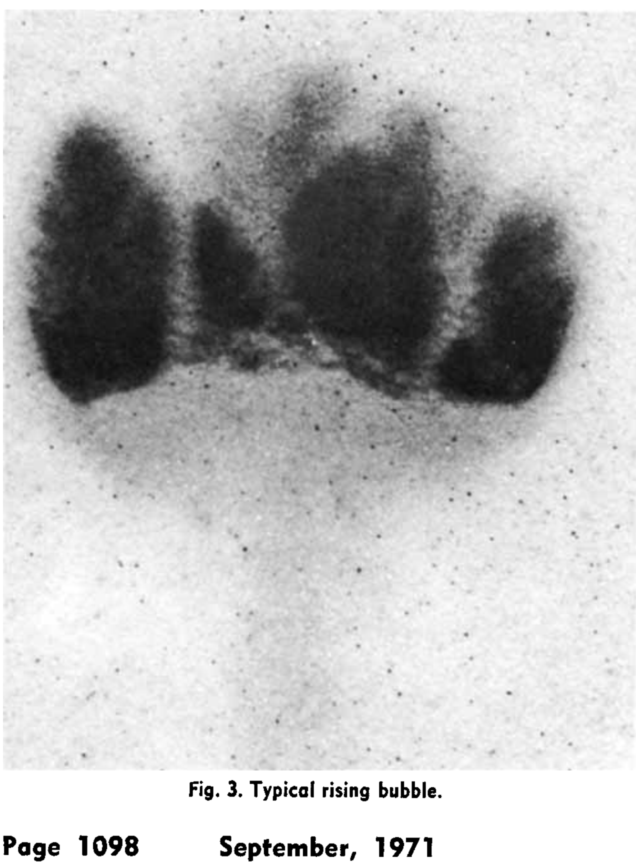

Fig. 3. Typical rising bubble.

Table 1. Summary of Experiments

	Run 1	Run 2	Run 3
Bed depth above bubble release point	5.7 in.	7.7 in.	14.0 in.
Initial bubble vol., $V_B(0)$	0.32 cu. in.	0.19 cu. in.	0.19 cu. in.
Bubble rising velocity, in./sec.	10.9 ± 0.7	11.4 ± 0.7	10.8 ± 1.0
Bubble growth rate, cu.in./sec.	0.46 ± 0.56	0.84 ± 0.17	0.08 ± 0.22
Wake growth rate, $\dot{V}_W$, cu.in/sec.;	4.66 ± 0.44	2.82 ± 0.17	2.75 ± 0.21
$\dot{V}_W/V_B(0)$, sec.$^{-1}$	14.6	14.8	14.5

* Solid particles: 100μ glass beads, ρ_s = 2.5 g./cc.
Bed dimensions: thickness = 0.5 in., height = 10 to 16 in., width = 10 in.
At minimum fluidization: u_{MF} = 0.58 in./sec., U_G = 1.3 in./sec.
ϵ = volume fraction gas in particulate phase = 0.434

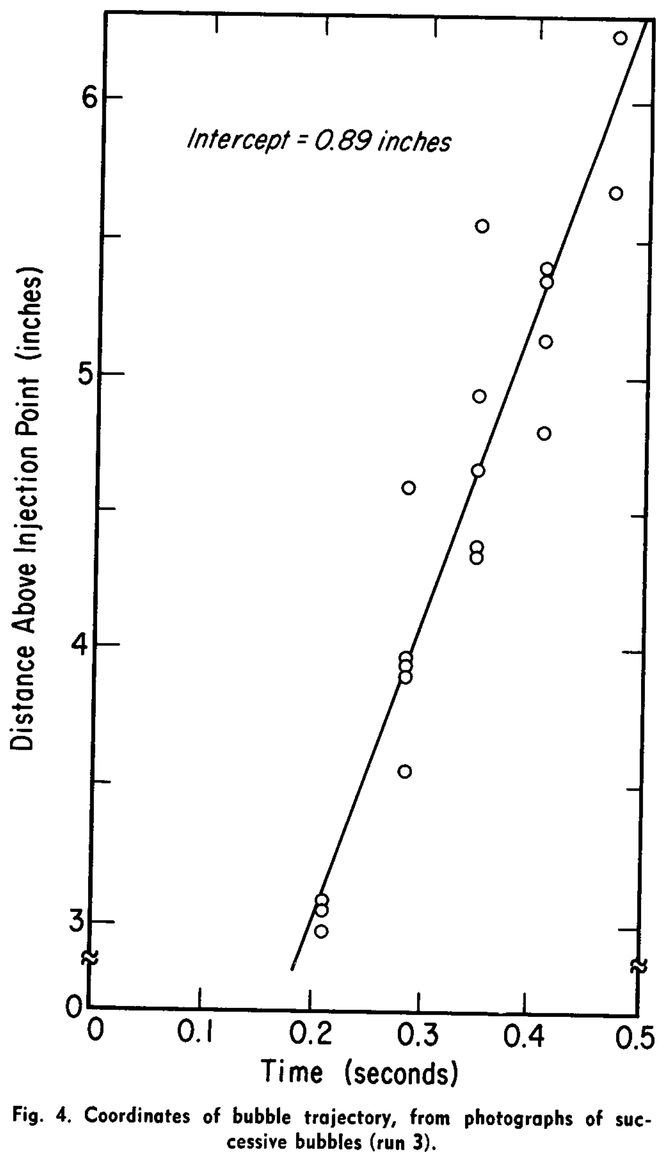

Fig. 4. Coordinates of bubble trajectory, from photographs of successive bubbles (run 3).

at the end of the 0.5-sec. life of each bubble, the tracer gas originally contained in only about 0.2 cu. in. had spread through more than ten times the original volume.

Figure 7 shows the measured volumes of the bubble cavity in the same series of experiments. The data show that there must have been an initially very rapid increase in the volume of the cavity, probably owing to draining of gas from the particulate phase. As soon as photographs could be taken and while significant data were being collected, the volume was hardly increasing. The least-squares straight line passing through the data has a slope of 0.80 cu.in./sec. with a standard error of 0.22 cu.in./sec.

In every series of experiments the same behavior was observed. The tracer gas filled a wake of rapidly increasing size behind each bubble and there was a probable increase in the volume of the bubble itself, although this increase was small. The fractional rates of increase were not influenced significantly by either the depth of the bed above the positions at which photographs were taken or by the initial volumes of the gas injected.

INTERPRETATION OF OBSERVATIONS

The large growth rates of the wake regions without a corresponding reduction in the size of the bubbles themselves imply not only that there were not equal currents of tracer gas leaving and entering the bubbles but also that there must have been a considerable flow of gas into each bubble and its wake from the adjacent particulate phase. Both of these observations are at variance with the conclusions of the potential-flow, Darcy–Law theory previously outlined. Such theory has assumed either that no wake is present or, if one exists, that the bubble behaves just as the top part of a cavity behaves, despite the wake. Obviously, something is wrong with the theory or with the

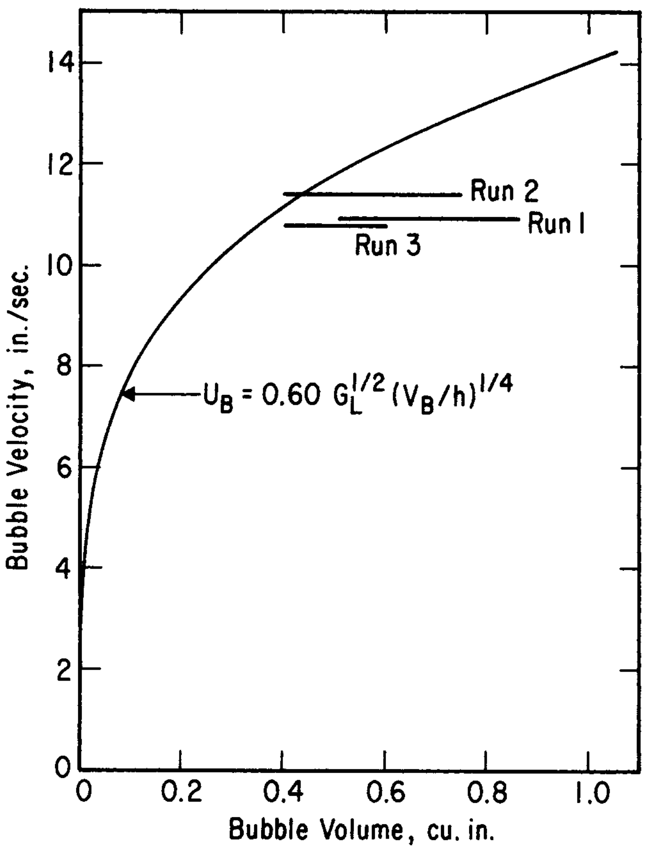

Fig. 5. Comparison of observed bubbles velocities with estimates from potential flow theory.

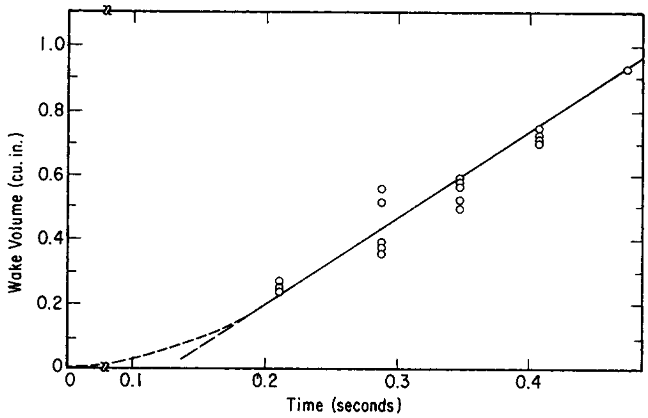

Fig. 6. Measured volumes of tracer-stained wakes behind rising bubbles (photographs from Run 3).

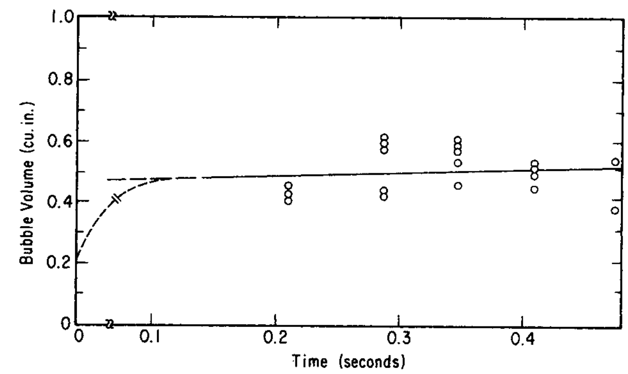

Fig. 7. Volumes of gas cavity during rise of bubbles through fluidized beds (Run 3).

assumptions which were made to derive it, even though predicted bubble rising velocities are about right.

Figure 8 shows the results of a material balance made on the wake and the bubble cavity regions for the same series of experiments to which the earlier figures refer. The gas volumetric flows q_{f2} and q_{f3} were computed from measurements of widths of the nitrogen dioxide-stained areas representing, respectively, the long tail of gas which streams out behind each bubble (0.116 ± 0.013 in.) and the rapid flow down each side of the bubble (0.116 ± 0.017 in.).

In these calculations it was assumed that the fraction of gas volume in the particulate phase near the bubbles is the same as in the whole bed at incipient fluidization. The fluid velocity affecting q_{f3} was computed from the potential flow theory; that in the long tail was computed as the difference $U_b - U_G$, where U_G is the interstitial fluid velocity at the minimum fluidization. Using the growth rate of the gas volume in the wake from the photographs, q_{f4} is found by difference. Note that it corresponds to a *downward* volumetric flow, rather than an upward flow as expected from the theory. Similarly, a material balance can be made on the whole space occupied by both bubble and wake. Numerically, it depends only on the estimated quantity q_{f2} and the measured values of $\dot{V}_W$ and $\dot{V}_B$. The large inward gas flow q_{f1} which results would be zero in the potential theory but is almost certainly present.

The important assumption in such material balance calculations is not that the nitrogen dioxide tracer concentration is constant inside the stained region of the photographs—acually, the concentration decreases gradually. The assumption is that there is no flow of gas across any of the clearly visible streamlines, including the outer

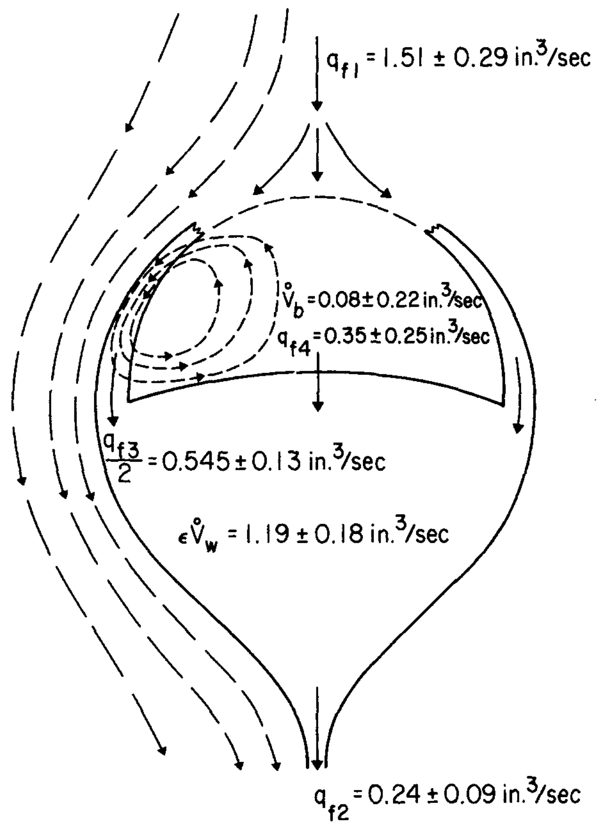

Fig. 8. Numerical results of a material balance for tracer-gas initially in gas cavity.

boundary of the wake region and its long tail as well as the boundary at the sides of the bubble. Molecular diffusion undoubtedly occurred across these boundaries but must have been too slow to affect the results.

As the photographs show, the boundaries were sharp. Thus, around the sides and bottom of the bubbles the stained area was a closed system for gas, although not for particles. On the roofs of the bubbles, however, no clear streamlines were visible in the pictures. There, flow of gas, stained or unstained, and of particles is assumed to be possible, but the numerical results show that nitrogen dioxide can hardly have escaped and that unmarked gas must have flowed in.

A similar set of material balances can be made for fluxes of solid particles (Figure 9) on the additional assumption that no solid particles move across the tracer-marked streamlines. The conclusions are similar: a strong inward flux of solid occurs across the dividing gas streamline which is expected in the theory above the bubble. Some of the solid may fall into the bubble cavity itself, and there is probably a flux of solid through the cavity and onto the floor of the bubble. Obviously these computations are not independent, for it has been assumed in making them that volumetric fluxes of solid and gas in the particulate phase are in the ratio $(1 - \epsilon)/\epsilon$.

Nevertheless, the conclusions are slightly different. It is of course expected that solid particles will cross the gas streamlines at the top of the cavity but it is not expected, if one follows the theory rigidly, that solid will fall onto the floor of the bubble. From the data, the chances that such a flow occurred are certainly better than even. The corresponding gas flows could have occurred simply because solid particles have fallen away from the roof of the bubble, leaving interstitial gas below the advancing bubble roof and trapping gas in the wake as the bubble's floor rises.

The photographs show quite clearly that there is solid inside the bubbles. White streamers of solid can be seen falling from the roof of the cavities and becoming thinner as the floor is approached, just as if the particles were falling freely through the gas and were experiencing the local gravitational acceleration. The floor of each bubble, except for those that have been freshly formed, has a mound of particles in the center, resulting possibly from the flow of particles downward from the most unstable part of the roof. A few photographs were obtained of bubbles which are in the act of splitting. These showed especially strong currents of solid particles falling from the roof, suggesting that some kind of flow instability originating there is the cause of splitting.

In every one of the photographs the cloud boundary layer marked by the nitrogen dioxide tracer can be located clearly at the sides of the bubble, but the boundary streamline becomes very hazy and confused as one tries to follow it toward the crest of the bubble's roof. There no steady flow streamlines apparently exist, as if the flow were highly chaotic. Such would be the case if the roof is continually or intermittently shedding solid particles into the gas-filled cavity underneath.

This is not the first work in which such behavior has been seen. Using essentially the same experimental technique, Rowe et al. (2) obtained similar photographs. Indeed, one of them, obtained with very heavy solid particles, for which minimum fluidizing velocity is only slightly less than the bubble rising velocity, showed a clear boundary between tracer-colored gas in the cloud boundary layer and the unmarked gas in the surrounding particulate phase, including the region just at the top of the bubble.

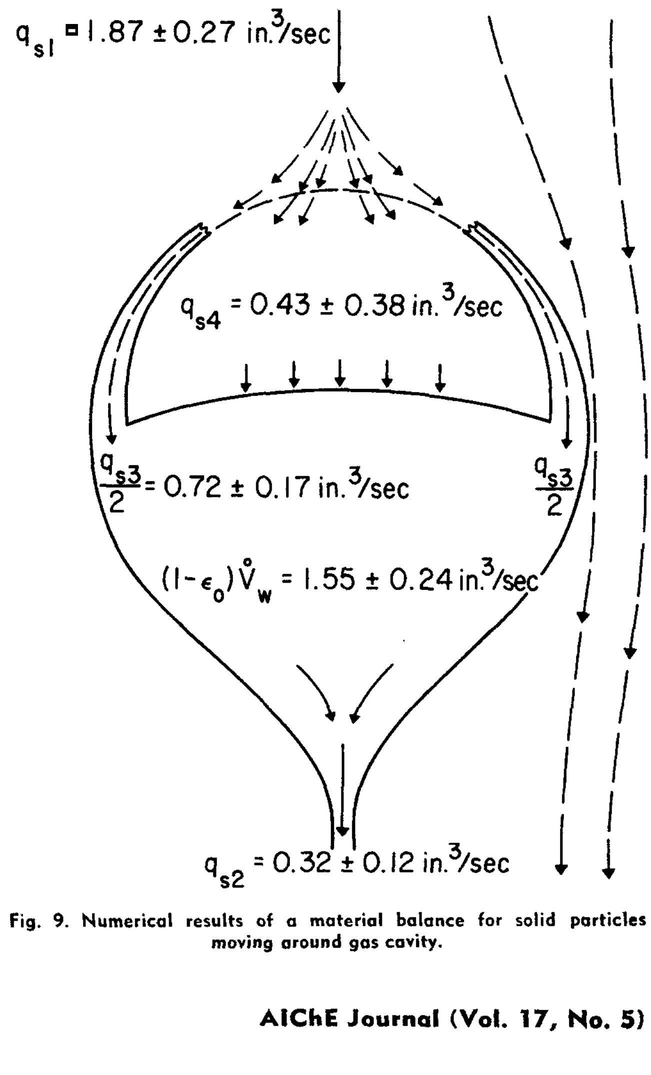

Fig. 9. Numerical results of a material balance for solid particles moving around gas cavity.

In other experiments, however, in which the ratio of velocities was greater—typical of the light particles which are used in many practical applications—Rowe and coworkers saw solid particles falling through the bubbles, just as we have. They attributed these observations to an effect of the walls of their flat-sided channel, suggesting that the solid particles that are partially immobilized by contact with the walls are free to fall under gravity and are not supported by the upward flow of gas in the cavity.

It is not possible from the evidence available to eliminate the possibility of a wall effect such as Rowe et al. assumed. On the other hand, there appears to be no clear reason why one must occur to such an extent that bubbles contained between parallel walls would behave entirely differently than they do in large beds. After all, the rising velocities of the bubbles are not affected significantly by the walls, as our measurements have shown. This indicates that the main pattern of movement of the solid particles around the sides is not altered appreciably. Furthermore, the hydrodynamic instability of an interface which is denser above and which is subject to gravity is well known (7, 9). Instability of the upper interface is expected to occur. We believe that it does.

Finally, it is interesting to compare the observed values of the total rate of gas leakage from a rising bubble with the value expected from the potential flow theory of Davidson and Harrison (6). If Equation (2) is applied to the experimental conditions of Figure 8, corresponding to Run 3, we compute $q = (2)\ (0.576)\ (1.24)\ (0.5) =$ 0.72 cu.in./sec., the bubble diameter at its base being about 1.24 in. and the minimum fluidizing velocity, 0.576 in./sec. Note that the sum of the two flow rates of nitrogen dioxide down the two sides of the bubble on Figure 8 is 1.09 ± 0.26 cu.in./sec., which probably does not differ significantly from 0.72. (If the downward flow of gas through the base of the bubble is included in the total leakage rate from the bubble, we get 1.44 ± 0.35 cu.in./sec., but the theory cannot be expected to yield anything but the flow out the roof of a bubble because the flow pattern in the wake is far different from potential flow.)

Thus, the pressure gradients in the particulate phase around a bubble evidently force gas to slip through the current of solid particles at about the rate expected, but the volume of particulate phase occupied by tracer is not constant, as the theory predicts, but increases steadily.

One final point remains. If there is a strong current of solid particles falling from the roof of a bubble onto its floor, cannot this alone be the cause of a bubble's apparent rise through the bed? After all, each differential deposit of solids on the floor causes a differential upward displacement of the floor and an equal displacement of the roof. Is it necessary to allow for a free movement of solid particles around the sides of the bubble to account for its observed rate of rise, as has been assumed in the potential flow theory?

The data given on Figures 8 and 9 provide a numerical answer to the question. The total volumetric rate of addition of gas and solid to the floor of the bubble can be found by adding q_{f4} from Figure 8 and q_{s4} from Figure 9, yielding 0.78 ± 0.45 cu.in./sec. When this is divided by the area of the bubble's floor, the velocity of the floor becomes only 1.3 ± 0.7 in./sec., which is only about 11% of the observed bubble-rising velocity and is obviously insufficient to account for the observed rise rate. The effect of the raining of solid particles on the floor should therefore be to cause the apparent velocity of bubbles to be only slightly greater than expected from the fluid mechanical analysis of Davidson and Harrison.

CONCLUSIONS

The potential flow theory agrees approximately with the observations in two respects. First, the bubble's rising velocity is within 20% of the value expected and second, the upward leakage rate through the roof is about what is expected. This rate is needed for estimating the reaction efficiency of a bed, as shown by Davidson and Harrison who were successful in their application of the potential flow theory to bubbles.

Two observations are not in agreement with the theory. First, there is a large flux of tracer gas from the particulate phase above the bubble across the expected circular dividing streamline and into the cloud boundary layer or into the bubble cavity itself. Second, there is a wake of gas behind each bubble which rises with the bubble, growing rapidly as it rises. Gas in the wake probably does not return to the bubble, and the gas velocity inside a bubble is downward rather than upward.

NOTATION

U_B = rising velocity of bubbles
g_L = local acceleration owing to gravity
V_B = volume of gas bubble
V_W = volume of wake behind bubble
u_{MF} = superficial gas velocity at minimum fluidization
u = superficial velocity of gas through bed
ϵ = fraction void space between particles in fluidized bed
U_G = interstitial gas velocity in particulate of fluidized bed
q = volumetric rate of gas leakage through upper surface of a bubble according to potential flow calculations
d_B = bubble diameter
q_{f1} (q_{s1}) = volumetric rate of flow of gas (solid) into roof of bubble as in Figure 8
q_{f2} (q_{s2}) = volumetric rate of flow of gas (solid) from wake of bubble as in Figure 8
q_{f3} (q_{s3}) = volumetric rate of flow of stained gas (solid) down sides and bubble as in Figure 8
q_{f4} (q_{s4}) = volumetric rate of flow of gas (solid) across rising floor of bubble

LITERATURE CITED

1. Davidson, J. F., R. C. Paul, M. J. S. Smith, and H. A. Druxbury, *Trans. Inst. Chem. Engrs.*, **37**, 323 (1950).
2. Rowe, P. N., B. A. Partridge, and E. Lyall, *Chem. Eng. Sci.*, **19**, 973-85 (1964).
3. Harrison, D., and D. L. Pyle, *ibid.*, **22**, 531-35 (1967).
4. Kunii, D., and O. Levenspiel, *Ind. Eng. Chem. Fundamentals*, **7**, 446-52 (1968).
5. Harrison, D., and L. S. Leung, *Trans. Inst. Chem. Engrs.*, **40**, 146 (1962).
6. Davidson, J. F., and D. Harrison, "Fluidized Particles," Cambridge Univ. Press, Cambridge, England (1963).
7. Rice, W. J., and R. H. Wilhelm, *AIChE J.*, **4**, 423-9 (1958).
8. Davies, R. M., and G. I. Taylor, *Proc. Roy. Soc. A*, **200**, 375 (1950).
9. Taylor, G. I., *Proc. Roy. Soc. (London), Ser. A*, **201**, 192 (1950).
10. Orcutt, J., J. F. Davidson, and R. L. Pigford, *Chem. Eng. Progr., Symp. Ser.*, **58**, 38, I (1962).
11. Baumgartner, P. K., and R. L. Pigford, *AIChE J.*, **6**, 115 (1960).
12. Collins, R. J., *Fluid Mech.*, **22**, 763 (1965).

Manuscript received January 16, 1970; revision received September 9, 1970; paper accepted September 11, 1970. Paper presented at AIChE New Orleans meeting.