Lateral Solids Mixing in Fluidized-Packed Beds

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A study was made of the mixing of particulate solids fluidized in the voids of a packed bed of larger nonfluidized bodies. In such a system heat transfer rates are dependent on the movement of fluidized particles. Recently application has been made of inert fluidized particles in chemical reactor systems to improve the removal of the heat generated by the exothermic reaction of the larger particles. A measure of the particle movement was achieved in this study by the determination of diffusion coefficients for the rates of solids mixing in the lateral direction when metal shot is fluidized in the voids of spherical and cylindrical packing. Experimental data showed that the particle mixing was related to the void structure of the packed bed by a random walk model. A general correlation was made for fluidized-solids diffusivity in spherically packed beds.

Fluidization of particulate solids with gas provides a technique for carrying on chemical reactions in a medium with high degrees of turbulence and mixing. High rates of solids mixing provide high heat transfer coefficients at the reactor wall and favor temperature uniformity of the fluidized reacting systems. Recently Argonne National Laboratory has shown interest in chemical reactors employing inert solids fluidized in the interstices of a static packed bed of larger bodies, which undergo reaction with the gas. This interest arose in connection with the development of a process for recovery of fissionable values from spent uranium dioxide reactor fuel (*4, 10*). Specifically, fluorine gas is reacted with massive uranium dioxide pellets in a packed bed, and inert fused alumina particles are fluidized in the voids of the packed bed to provide rapid heat removal. Such combination systems are referred to as *fluidized-packed beds*. The present paper describes a study of the effect of process variables on the solids-mixing behavior of such beds. It is clear from current theories (*7, 17, 18*) that the turbulence of the fluidized solids accounts for the heat transfer properties of fluidized-packed beds. This turbulence may be most directly measured in terms of the rate of solids mixing.

The baffling effect of the packing accounts for the major differences between fluidization in open tubes and fluidization in packed beds. Thus the extremely high solids circulation rates for the unbafflled fluidized beds are not found in the fluidized-packed bed. In the latter case, the void structure of the packing determines the mixing behavior. This has been shown for gas mixing in fluidized-packed beds (*5*) .

There are few literature references to such combination fluidized-packed beds. Sutherland, et al. (*14*) were interested in fluidized-packed beds because the baffling effect of the fixed packing permits smooth fluidization (without slugging) for large height-to-diameter ratios. Recently Ziegler (*19*) published data and discussed the mechanism of radial heat transfer in fluidized-packed beds; he worked at Argonne National Laboratory in cooperation with Northwestern University, Evanston, Illinois.

No works on solids mixing in fluidized-packed beds were found. However, solids mixing has been investigated for fluidized beds (without packing). Bart (*2*), Massimilla and Bracale (*8*), Hayakawa, et al. (*6*), and Talmor and Benenati (*15*) obtained quantitative measurements for longitudinal solids mixing in fluidized beds. Brötz (*3*) studied solids mixing in fluidized beds in the lateral direction. Although these references are interesting as to analysis and technique, they give no indication of the effect that fixed packing might have on the mixing rates.

In general, fluidized-packed beds would be expected to show features that are combinations of those of packed beds and of fluidized beds. The primary motion of the fluidized material is determined by particle size and gas velocity, whereas the dimensions of the packed material determine the void structure within which fluidization takes place. The present study concerns the measurement of mixing in the lateral direction of solids fluidized in the voids of a fixed bed and the effect of system variables on the rate of this mixing.

EXPERIMENTAL DESIGN

In the present study, the experimental method of measuring the rate of solids mixing was patterned after that of Brötz. The general method is based on the use of two solids similar in the physical properties that determine the mixing behavior, but different in some property usable for analysis of composition. The analytical property used by Brötz was color, but for the present study the differences in magnetic properties of nickel and copper were used. Because nickel and copper have the same density (8.9 g./cc.), the same particle size and shape should exhibit identical fluidizing characteristics.

Determination of Solids Composition by Magnetic Separation

If pure nickel and copper are placed in a rectangular cell and separated by a centrally located partition, then, when the partition is removed, mixing will occur on each side of the center line. After a certain mixing time, the fluidizing gas is cut off and mixing is arrested. Bed samples are taken at points of known distances along the line of mixing. Determination of the composition of the samples allows points on the concentration profile to be plotted. A typical set of experimental points is plotted in Figure 1. The location of the center line is shown at the distance $x = 0$.

Diffusion Equation for Solids Mixing

A diffusion coefficient for the bed of fluidized solids was used to characterize the rate of solids mixing. The usual differential equation for diffusion in one dimension is

$$\frac{\delta C}{\delta \theta} = D \frac{\delta^2 C}{\delta x^2} \tag{1}$$

The solution of this equation is given by Sherwood and Reed (*13*):

$$C = 1/2 + \frac{2}{\pi}\left[\exp\left(-\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{R^2}\right)\sin\frac{\pi x}{2R} + 1/3\exp\left(-9\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{R^2}\right)\sin\frac{3\pi x}{2R} + 1/5\exp\left(-25\left(\frac{\pi}{2}\right)^2 \frac{D\theta}{R^2}\right)\sin\frac{5\pi x}{2R} + \text{------}\right] \tag{2}$$

In this equation, the values of the concentration, C, are related to the values of the diffusion distance, x, in terms of the diffusivity D; one half the column width, R; and the time of mixing θ. Thus the diffusion coefficient, D, was found from the value of the parameter which fit the experimentally measured profile of C as a function of x. Good fits were obtained, a typical one appearing in Figure 1.

Apparatus and Procedure

A schematic diagram of the apparatus is shown in Figure 2. Nitrogen, the fluidizing gas, was distributed by a porous metal plate. A rotameter measured gas flow rates. Sampling was accomplished by a horizonal row of 1½-in. long, fixed hypodermic needles, ¾ in. above the bottom and spaced 1 in. apart, and by a 9-in.-long needle injected into the bed to the desired position. The concentration profile was invariant with respect to height because of high vertical mixing rates.

The fluidizing gas was humidified by a 1-ft packed column partially filled with water to prevent static effects.

The fluidizing material was copper and nickel shot of −40 +50, −100 +120, −120 +200, and −140 +170 mesh sizes. Fixed packing consisted of 1/2-, 3/8-, 1/4-, and 3/16-in. diam. steel spheres and 1/2 × 1/2, 3/8 × 3/8, and 1/4 × 1/4 in. brass cylinders. A set of runs was also made without fixed packing; that is, normal open-tube fluidization.

A rubber partition was inserted in the center of the column and one half of the column was charged with fixed packing and nickel shot; the other half was charged with fixed packing and copper shot. A needle valve was preset to give the desired flow rate. Then the partition was removed, and fluidization over a measured time interval was effected by opening and closing a quick-opening toggle valve in the gas line. Samples of the mixed particulate solids were withdrawn by means of the hypodermic needles. The sample mixtures were spread out on a flat surface, and the nickel particles were removed by passing a small magnet over the surface. The separated fractions were weighed directly to establish sample compositions.

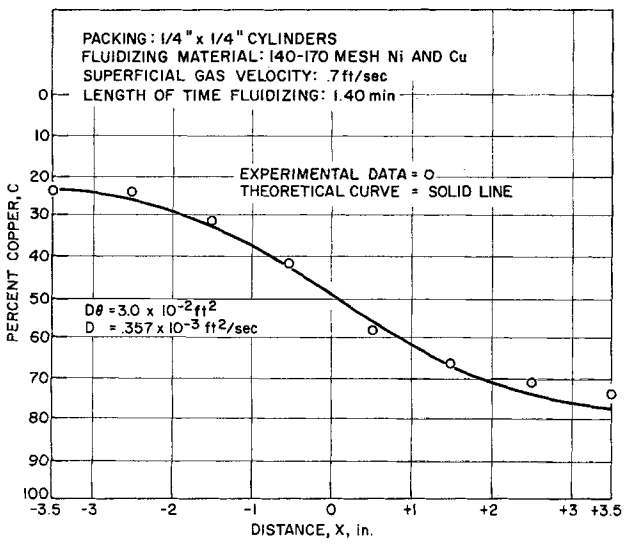

Fig. 1. Sample concentration profile.

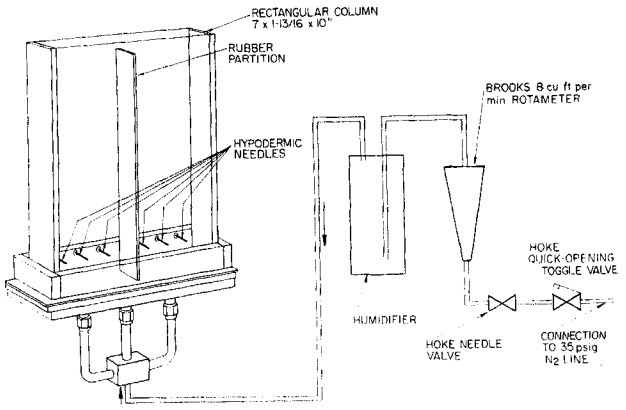

Fig. 2. Schematic diagram of apparatus.

The run time varied from about 0.5 to 20 min. depending on the gas velocity and the fluidized particle size. Smooth fluidization was attained in the order of about a second after introduction of the fluidizing gas. It was, therefore, expected that initial start-up disturbances would have a negligible effect on the data.

THEORETICAL MODELS FOR SOLIDS MIXING

Certain theoretical models were examined with respect to their ability to describe the mechanism of particulate-solids mixing in quantitative terms consistent with the experimental results. The three different diffusion models were a molecular-kinetic model, a model developed for fluidization by Trawinski, and a random-walk model.

Molecular-Kinetic Model

Because particles in a fluidized bed appeared to move randomly upon visual observation, it was considered, following Reboux (*11*), that models developed for molecular motion might be appropriate.

If the particles do move in an independent random motion, the rate of solids mixing may be predicted by analogy with the kinetic theory of gases. The number of collisions per unit time, n, is given by the equation

$$n = \underbrace{\frac{u\,\Sigma}{4\,(V-b)}}_{\text{wall collisions}} + \underbrace{\frac{\sqrt{2}\,\pi\,\nu\,d^2 u}{V-b}}_{\text{interparticle collisions}} \tag{3}$$

If the collision rate and the mean speed of the particles are known the diffusivity, D, may be predicted:

$$D = 1/3\,\frac{u^2}{n} = 1/3\,u\,\frac{(V-b)}{\Sigma/4 + \sqrt{2}\,\pi\,\nu\,d^2} \tag{4}$$

The volume, V, is determined by the void fraction of the packing, which was taken as 0.43 for the spherical packing used. The number and volume of the fluidized particles are determined from the void fraction of the particles at rest (0.36 was assumed) and the bed expansion ratio, which is a function of gas velocity.

For the case of ½-in. spherical packing and −140 +170 mesh (average diam. .0038 in.) fluidizing material,

the following fluid-bed expansion ratios were obtained experimentally as a function of superficial gas velocity:

Superficial gas velocity (ft./sec.)	Expansion ratio
0	1.00
0.3	1.16
0.5	1.39
0.7	1.54
1.0	1.85
1.4	2.62

With the above bed expansion and void fractions, solids diffusion on the basis of a molecular-kinetics model may be predicted for various mean particle speeds. In Figure 3 the solids diffusion coefficient, D, is shown as a function of gas velocity for this model. D is calculated for the case in which the mean particle speed is equal to one-half Vo/ϵ, the mean velocity of gas in the packed-bed voids.

Trawinski Model

Trawinski (*16*) presented an equation for solids diffusivity based on a model in which individual particles pass through holes or openings in the particle suspension of the fluidized bed. Trawinski's equation is

$$D = \frac{Kd\,u}{6\,(1-\epsilon_0)}\,\frac{\Delta L}{L_0} \qquad (5)$$

The Trawinski equation was then applied to the fluidized-packed bed case of ½-in. diam. spheres and 0.0038-in. diam. fluidized particles to compare it with the other models.

Random-Walk Model

Photographic work by Massimilla and Westwater (*9*) revealed that the particles do not move individually, but in agglomerates. Rowe (*12*) has shown that overall particle motion results from small portions of the particles being carried up in the wake of individual bubbles. It can then be envisaged that small aggregates of particles fluidized in the voids of a packed bed would be carried up in the wake of the bubble and deflected in a random fashion at each encounter with an element of fixed packing. Baron (*1*) predicted eddy diffusivities for gas in packed beds on the basis of lateral deflections of the stream caused by the presence of the packing. The theory of lateral deflections or random-walk displacements caused by the packing is here applied to lateral diffusion of solids in a fluidized-packed bed in a rectangular column.

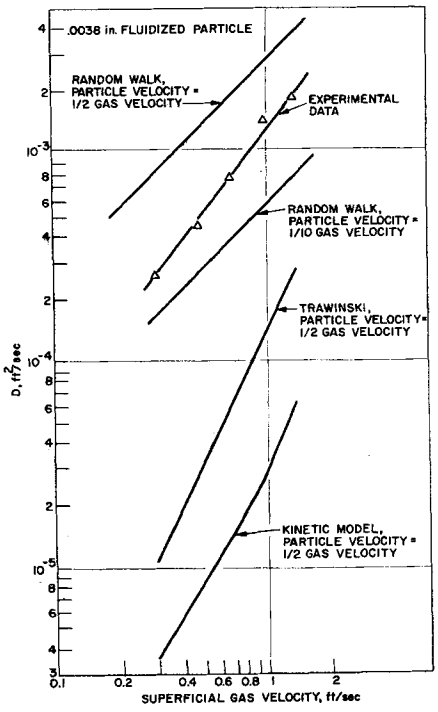

Fig. 3. Comparison of theory and data for diffusion of fluidized particles in voids of ½-in. spheres.

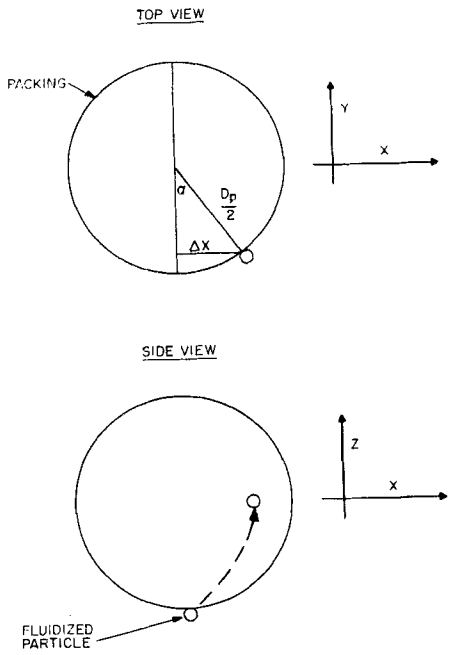

Fig. 4. Fluidized particle deflection by spherical packing.

The mean distance, $\overline{\Delta X}$, that a particle is deflected in the x direction (the direction of diffusion) upon contact with a sphere is found by integration to obtain the average distance between the circumference and the chord perpendicular to the x direction that bisects the maximum horizontal circular cross section of the packing. In Figure 4, a top and side view is shown conceptually for a fluidized particle deflected by a spherical packing element:

$$\overline{\Delta X} = \frac{\int_0^{\pi} \frac{D_p}{2} \sin\alpha\, d\alpha}{\int_0^{\pi} d\alpha} = \frac{D_p}{\pi} \qquad (6)$$

Actual solids mixing occurs as a batch system. In the model here adopted, both rising and falling particles are taken to have the same average lateral deflections.

The time, θ, between contacts for fixed packing arrayed rhombohedrally is found by dividing the vertical distance travelled between contacts by the mean particle velocity:

$$\theta = 1/2\,\frac{(1+\sqrt{2/3})\,D_p}{u} = \frac{0.909\,D_p}{u} \qquad (7)$$

The average distance of deflection per contact and the time between contacts is related to a diffusion coefficient by the random-walk (mean free path) deflection in Einstein's diffusion equations originally developed for Brownian motion:

$$\overline{\Delta X}^2 = 2\,D\,\theta \qquad (8)$$

In the fluidized-packed bed the solids diffusivity would be

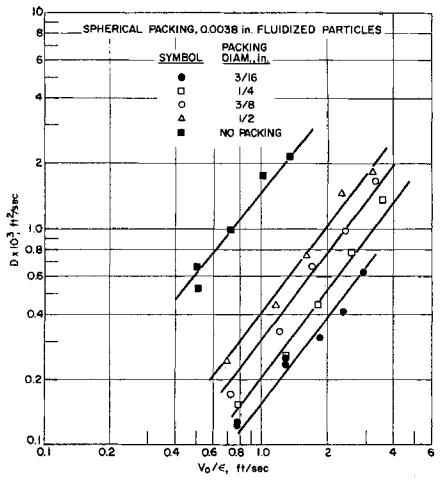

Fig. 5. Effect of fixed packing on solids mixing.

$$D = 1/2 \frac{u}{0.909\, D_p} \left(\frac{D_p}{\pi}\right)^2 \qquad (9)$$

$$= 0.0558\, D_p\, u$$

In Figure 3 the mixing diffusion coefficients are shown as a function of gas velocity, as calculated for the cases in which the average particle velocities are 1/2 and 1/10 the average gas velocity in the packing voids.

It would be expected that the random-walk model would apply over the entire range of gas-fluidization velocities for lateral solids mixing. At low flow rates, when the bubbles are uniformly distributed with little interaction, the particles carried in the bubbles would be deflected by the packing. At high flow rates, even though there may be interaction and coalesence of bubbles, the solids movement would still be in streams of particle aggregates which undergo lateral deflections by the packing. The random-walk model only applies to lateral solids mixing and not to axial mixing.

Comparison of Theoretical Models

The comparable cases calculated for each of the three models considered are shown in Figure 3, together with a plot of the experimentally measured diffusion coefficients. The models based on Trawinski's theory and on the kinetic theory differ from the experimental data by factors of approximately 10 and 10^2, respectively. The random-walk model appears to give a much closer approximation, especially since the experimental value falls within the two assumed cases of mean particle velocity (namely, 1/2 and 1/10 the average gas velocity). That this range of particle velocity is realistic is shown by further work described below in which actual particle velocities were observed.

Experimental Results

Values of the diffusion coefficient, D, as a function of gas velocities for various fluidized-packed bed systems, were obtained from the mixing experiments performed. Variations of D were investigated for different sizes of spherical and of cylindrical packing as well as different sizes and heights of fluidized material. The results are presented in the form of a general correlation closely related to the random-walk theoretical model. Reference cases for fluidization without packing are also presented.

Effect of Size of Spherical Packing

A series of runs was made with packing consisting of 1/2-, 3/8-, 1/4-, and 3/16-in. diam. spheres and 0.0038 in. diam. copper and nickel shot. The data for the several packings are plotted as D vs. average gas velocity in Figure 5. The diffusion coefficients increased with packing size over the range tested. Fluidization was also attempted in a bed of 1/8-in. spheres but the particle motion was greatly inhibited and channelling occurred. For open-tube fluidization (no packing) bed diffusion coefficients are about three and a half times higher than for 1/2-in. packing for the same velocity. This fluidized-bed data was for an expanded bed height of approximately 1 in. The open-tube fluidized-bed mixing rate increased strongly with bed height; this effect is discussed further in a later section.

The diffusion coefficient, D, was directly proportional to the packing diameter, D_p, which is consistent with the random-walk theory. For these tests the fluidizing gas velocities were varied in a range of about 5 to 25 times the minimum fluidization velocity of the 0.0038 in. particles. The gas velocity was limited by the beginning of dilute phase fluidization.

Effect of Cylindrical Packing

The solids diffusivities of 0.0038-in. fluidized particles in the voids of ½ × ½, ⅜ × ⅜, and ¼ × ¼ in. cylindrical packing were also measured. The solids diffusivities were approximately the same for cylindrical packing as for the spherical packing. However, the diffusivities changed less with increasing gas velocity. Aside from the difference in shape, the larger cylinders exhibited a greater tendency for bridging when randomly packed. This bridging resulted in nonuniform void fractions throughout the bed and caused slightly more erratic results than for the spherical packing.

In order to determine if length of time of fluidization affected the solids diffusivity, a run was made with the ¼ × ¼-in. cylindrical packing in which samples were taken after 2.15, 3.84, and 4.84 min. of fluidization. The diffusivity was found to be time invariant to within about 1.5%.

Effect of Fluidized Particle Size

Runs were made with −40 +50, −100 +120, and −140 +170 mesh copper and nickel in the voids of ⅜-in. spherical packing to determine the effect of particle size on mixing rates. Diffusivities vs. superficial gas velocities for the various particle sizes are given in Figure 6. The smaller fluidized particles showed the higher diffusivity.

Generalized Correlation

From the above data on the dependencies of solids diffusivity on system variables, a generalized correlation was

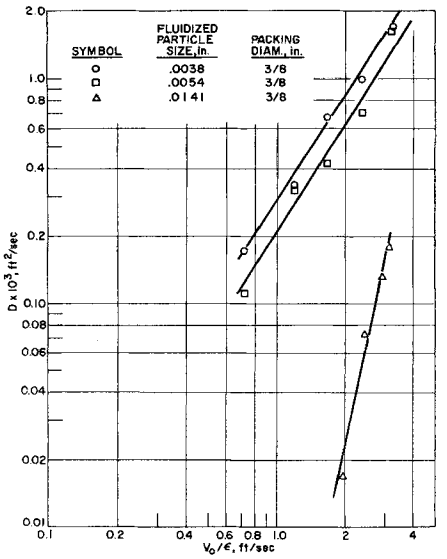

Fig. 6. Effect of fluidized particle size on solids mixing.

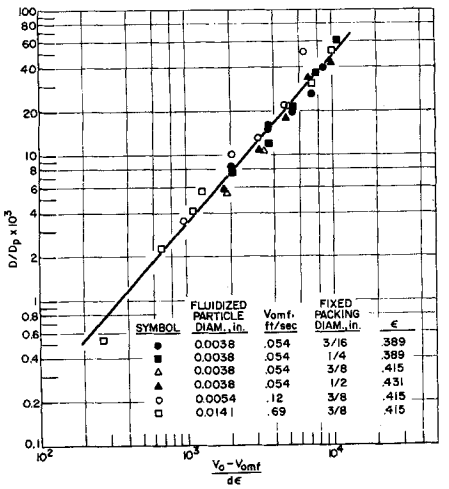

Fig. 7. General correlation for solids mixing in a fluidized-packed bed.

made relating D/D_p to the fluidized particle size, d, and the average gas velocity in the voids of the packed bed, V_o/ϵ, as corrected for the minimum fluidization average gas velocity, V_{omf}/ϵ. A plot is given in Figure 7. In numerical terms the correlation is

$$D = D_p \left[\frac{V_o - V_{omf}}{d\epsilon}\right]^{1.15} (1.22 \times 10^{-6}) \text{ sq. ft./sec.} \tag{10}$$

This is a dimensional equation, and all units of dimension are in feet and seconds. The effects of gas viscosity and fluidized particle density were not considered here.

The above correlation was made with narrow cuts in screen sizes. Several runs were made with −120 +200 mesh particles which have approximately the same average diameter as −140 +170 mesh particles (0.0038 in.) but have a greater spread in size distribution. In Figure 8 a comparison is made of the effect of particle distribution on the solids mixing for fixed packings of ½- and ¼-in. spheres. It is seen that mixing is greater with −120 +200 mesh particles, that is with the wider size spread. Also, it appears that the diffusion coefficient is no longer a simple logarithmic function of gas velocity. It was observed with the −120 +200 mesh particles that at increased velocities the fluidized bed segregated into two phases. There was a dilute, highly agitated phase in the upper portion of the packed bed and a denser phase in the lower portion of the column.

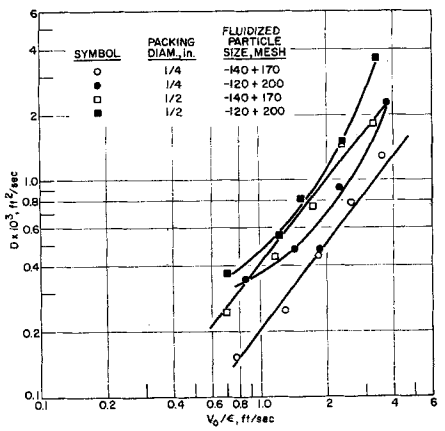

Fig. 8. Effect of fluidized particle size distribution on solids mixing.

Effect of Bed Height for Fluidized and for Fluidized-Packed Bed

A brief examination was made of fluidized bed mixing; that is, without the presence of a fixed packing. In Figure 5 a comparison is made of the mixing rates of a fluidized bed (static height 0.7 in.) and of the fluidized-packed beds. The mixing rate is greater for the fluidized bed because of the absence of fixed packing baffling effects.

It was also observed with the fluidized bed that slugging increased with depth of bed. A more vigorous motion and an increased effect of particles being thrown into the air with bubble bursts at the surface accompanied increased slugging. Runs were, therefore, made with the 0.0038-in. particles at a fluidizing gas velocity of 0.5 ft./sec. to determine the effect of bed height on solids mixing for both the fluidized bed and the fluidized-packed bed. At initial bed heights of 0.7-, 1.4-, and 2.1-in. the corresponding diffusivities were 5.2, 14.8, and 27.8 sq. ft./sec. The expansion ratio for the fluidized bed at this gas velocity was 1.4.

The mixing diffusion coefficients for three different initial fluidized particle bed depths (1⅛-, 2¼-, and 3⅜-in.) for a fluidized-packed bed with ⅜ in. spherical packing were measured. Within the experimental accuracy there was no effect of height of fluidized bed. The packing effectively baffles the slugs in the fluidized bed, and the result is a smooth, uniform fluidization. From the above data it is expected in practical cases that mixing in open-tube fluidization would be considerably greater than in fluidized-packed beds.

MEASUREMENT OF AVERAGE PARTICLE VELOCITY

The diffusion coefficient for solids mixing, D, is related to the average fluidized particle velocity by the random-walk theory according to the equation

$$u = \frac{D}{0.0558\, D_p} \tag{11}$$

From the above equation and the diffusivity data it is possible to calculate an average fluidized particle velocity. The particle velocity for the various size particles is shown in Figure 9.

As a means of independent measurement of the par-

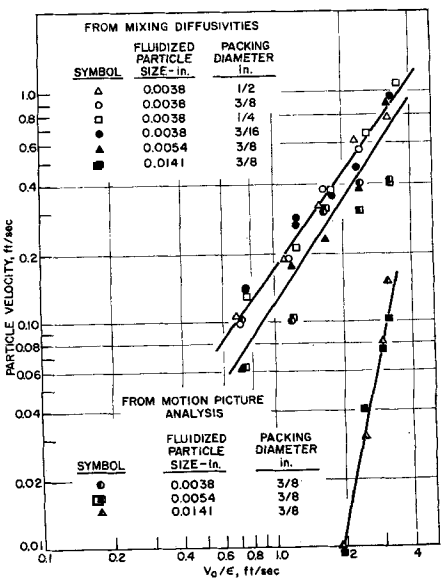

Fig. 9. Average fluidized particle velocity.

ticle velocity, high-speed motion pictures were taken of the fluidized particles. Approximately 1 sq. in. area of the particle bed at the wall was photographed at 4,000 frames/sec. One roll of film was examined for each run condition. At a given instant it was observed that large areas of the bed were relatively stagnant except where there were local disturbances caused by the bubbles. No area was permanently inactive; all areas would eventually be disturbed by passing bubbles. It was estimated that at a given instant for the beds of smaller particles at the higher velocities that about 50% of the bed was not in motion, and at low velocities with the 0.0141-in. particles that about 90% of the bed was inactive. When the particle velocities were determined, the speed of the particles in upward and downward directions and the fraction of the particles not in motion were averaged in the following manner:

$$u_{ave} = \left(\frac{\text{ave. } u_{up} + \text{ave. } u_{down}}{2}\right) \cdot (\text{active fraction}) \quad (12)$$

The movement of the particles appeared not as random movement of individual particles, but as streams of particles moving in the upward direction or falling in the downward direction because of displacement by bubbles rising through the packed bed. Only the vertical velocity component was measured.

The average particle velocity based on the photographic data is only approximate because only a small sample of particle motion in terms of time and area was examined, the proportion of particles in motion at a given instant was only estimated, and the particle motion at the wall may not have been entirely representative of the internal motion. However, as shown in Figure 9, the particle velocity indicated by the diffusion theory and that estimated by photographic means are in close agreement.

CONCLUSIONS

1. The lateral mixing behavior of particles fluidized in the voids of a packed bed is not analogous to the random motion of gas molecules but rather to eddy diffusion in a flowing gas stream.

2. A random-walk model may be used to relate these solids diffusivities to the void structure of the packed bed. Mean particle velocities calculated from this theoretical model appear consistent with observations of particle velocity made photographically. Depending on the size of the particle, the particle velocity is a factor of four to two hundred lower than the average linear gas velocity in the packed bed.

3. A dimensional correlation for solids diffusivity in a spherically packed bed was empirically deduced and has the following form:

$$D = D_p \left[\frac{V_o - V_{omf}}{d\epsilon}\right]^{1.15} (1.22 \times 10^{-6}) \text{ sq. ft./sec.}$$

Velocity dimensions are in feet per second, and particle sizes are in feet. Further experimental work involving variation of gas properties and fluidized particle densities is necessary before a completely generalized correlation can be made.

4. The rate of change of solids diffusivity with gas velocity is less for cylindrically packed beds than for spherically packed beds.

5. The rate of solids mixing increases with bed height for open-tube fluidization (no packing), but was virtually independent of height for fluidized-packed beds within the limits of the experimental conditions.

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NOTATION

b = correction for available volume, cu. ft.
C = concentration, fractional weight
d = fluidized particle diameter, ft.
D = diffusion coefficient, sq. ft./sec.
D_p = fixed packing diameter, ft.
K = correction for space occupied by particles, cu. ft.
L_o = bed height at static conditions, ft.
ΔL = bed expansion, ft.
n = number of collisions
R = 1/2 width of column, ft.
u = mean speed of particle, ft./sec.
V_o = superficial gas velocity, ft./sec.
V_{omf} = minimum superficial gas fluidization velocity, ft./sec.
V = volume, cu. ft.
x = distance from center of column, ft.
$\overline{\Delta X}$ = mean free path of particle, ft.

Greek Letters

α = angle, radians
ϵ = packing void space
ϵ_o = fluidized particles void space of static conditions
θ = time, sec.
ν = number of particles
Σ = surface area of walls, sq. ft.

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