FLUCTUATION METHOD OF DETERMINING THE MIXING COEFFICIENT FOR A SOLID PHASE IN A TWO-DIMENSIONAL FLUIDIZATION BED MODEL

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The effectiveness of the fluctuation method of determining the mixing coefficient D as well as the feasibility of automating the measurements and the subsequent data evaluation are confirmed experimentally on a "one-grain-thick" model.

The random mixing of solid particles in a fluidization bed is characterized by the effective mixing (diffusion) coefficient D. Earlier methods of determining this quantity [1-8] were based on measuring how the space distribution of labeled particles changes with time and comparing the test data with theoretical data obtained from the transient diffusion model of the process. Such methods are suitable for practical purposes when measurements are made at low values of D, when the changing concentration distribution of labeled particles can be measured a few times before steady-state has been reached. In addition to procedural difficulties, there are also some fundamental problems involved here in regard to the applicability and the suitability of the diffusion model itself [9]. It has been shown in the authors' earlier studies that the motion of a single particle can be broken down into a circulation, together with adjacent particles, and a pulsation; the characteristic parameters of both modes v_{circ} and D_{puls} respectively have been defined in [10]. Naturally, one may ask which of these two parameters determines the overall mixing rate of the solid phase in the apparatus as a whole.

We believe it worthwhile to analyze these problems in terms of two-dimensional "one-grain-thick" models [10-12], which represent the simplest and most convenient to analyze systems. On such models it will also be easy to develop and prove out basically new measuring procedures.

The models used here were made up of Plexiglas beakers rectangular in cross section and containing aluminum disks, the thickness of each disk slightly smaller than the width of the beaker. A bed was fluidized by means of a rising stream of water. For comparison with an air-induced fluidization bed, the governing Archimedes number was lowered by mixing the water with high-viscosity glycerine. Since such solutions are very hygroscopic and, besides, their viscosity depends strongly on the temperature, we maintained their composition constant during a test by keeping the circulation system (reactor-pump) closed with the viscosity of the circulating solution checked periodically and by holding the temperature constant. The cross-sectional area of a beaker was $150 \times 6 \cdot 10^{-6}$ m^2 and its height was 0.48 m. The diameter of the aluminum disks was 8 mm and their thickness was 3.8 mm. The total number of disks in such a two-dimensional fluidization bed was 200. The viscosity of the solutions was varied from $1 \cdot 10^{-6}$ m^2/sec (water) to $50 \cdot 10^{-6}$ m^2/sec (90% glycerine with 10% water) and the Archimedes number varied correspondingly from $1.3 \cdot 10^7$ to $4.7 \cdot 10^3$. The flow and the mixing of particles were studied with the aid of a kinematograph taking 36-48 frames/sec of the entire bed and by subsequently examining each frame magnified and projected on a screen. From the markers denoting successive positions of the center of a labeled particle on each successive frame, we plotted broken lines representing the trajectory of this particle. By varying the evaluation time interval $\Delta\tau$ (every frame, then every second frame, every third frame, ...), we constructed for each fluidization mode a curve of the average-over-the-interval velocity $\bar{v} = \Delta l/\Delta\tau$ as a function of $\Delta\tau$ in $\bar{v}$, $1/\sqrt{\Delta\tau}$ coordinates [10, 11], according to the equation

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$$\bar{v} = v_{\text{circ}} + \sqrt{D_{\text{puls}}} \cdot \frac{1}{\sqrt{\Delta\tau}} \tag{1}$$

and then determined the sought parameters. Values of the order of $9 \cdot 10^{-2}$ m/sec for v_{circ} and $0.6 \cdot 10^{-4}$ m^2/sec for D_{puls} were obtained at a fluidization number from 2 to 4 with water and with up to 70% glycerine solutions in water. With the most viscous solution of 90% glycerine in water the respective values were $v_{\text{circ}} \approx 5 \cdot 10^{-2}$ m/sec and $D_{\text{puls}} \approx (0.1\text{–}0.2) \cdot 10^{-4}$ m^2/sec. For each point on the straight line (1) we processed 100-150 frames and even this, in addition to being very laborious, was not adequate enough for the desired accuracy. We did not immediately succeed in our attempts to mechanize and automate the processing of test data.

In order to measure the mixing coefficient referred to grains throughout an entire bed, we first tried our conventional methods. In order to produce a "homogeneous source," 15 of the 200 disks were painted and, comprising the lowest row of particles, were held in this position with a special device (horizontal wire). After fluidization had begun, the wire was quickly withdrawn so that the labeled particles were now free to move upward and mix with the other ones.

For scanning the kinema frames, the bed was vertically subdivided into 10 mm high layers and the number of labeled particles n(x) was counted from level 0 to height x. A sample count is shown in Table 1.

According to the data in Table 1, the mixing of a solid phase in a two-dimensional model retained all the basic characteristics observed in a conventional three-dimensional bed. Labeled particles rose in groups or "packets" of 5 to 10. These groups dispersed and then again agglomerated. Thus, after 1/8 sec, in the bottom layer up to x = 0.01 m there remained only one particle while a group of 12 particles appeared between x = 0.02 m and x = 0.04 m. After 1 sec between x = 0.04 m and x = 0.05 m there appeared a group of 7 labeled particles, while up to x = 0.01 m there remained, as before, only one particle. On the last frame, after 6 sec, the labeled particles had distributed themselves rather uniformly over the height up to x = 0.19 m, but in the bottom layer up to x = 0.01 m there had accumulated 4 particles.

From the approximate formula

$$D \approx \frac{H^2}{\pi^2 \tau}, \tag{2}$$

and considering that almost complete mixing has taken place up to the H = 0.2 m height within τ = 4 sec, one can roughly estimate the value of the overall mixing coefficient for the entire bed: $D \approx 1 \cdot 10^{-3}$ m^2/sec, which is by one order of magnitude higher than D_{puls} characterizing the "vibratory" motion of a single particle. The measured value of D came closer to an estimate based on the group circulation velocity of particles:

TABLE 1. Distribution of Labeled Particles in Horizontal Layers of a Fluidization Bed. Fluidizing Agent: Glycerine (70%) and Water (30%). Height of Suspension Layer 0.23 m

$x \cdot 10^2$, m	t, sec														
	0	0,125	0.250	0,375	0,500	0,625	0,750	0,875	1,00	1,5	2,0	2,5	3,0	4,0	6,0
1	15	1	1	1	1	1	1	1	1	1	1	1	3	4	4
2		1	1	1	1	1	1	3	1	1	1	3	3	4	4
3		8	6	6	2	3	3	3	2	1	2	3	5	4	5
4		13	13	13	9	10	6	5	4	2	3	4	5	4	6
5		15	15	15	14	13	12	12	11	5	4	6	7	6	7
6					15	14	14	13	13	10	7	7	7	7	7
7						15	15	14	14	13	7	7	9	9	9
8								15	15	14	13	9	10	10	9
9										15	14	10	11	11	9
10											15	14	12	12	11
11												14	13	12	13
12												15	14	13	14
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17															14
18															14
19															15

$$D_{\text{circ}} \approx \frac{1}{10} v_{\text{circ}} H, \tag{3}$$

which in this case was $D \approx 2 \cdot 10^{-3}$ m^2/sec.

The relatively small number of particles (15) and even small total number of disks (200) in our model would not justify replacing the approximate relation (2) or (3) by more exact ones based on transient solutions for the continuous (rather than discrete) diffusion model. Instead, we attempted to experimentally study the feasibility of applying our new fluctuation method to the measurement of mixing coefficients [13, 14].

This method is based on measuring the average fluctuations of the number of particles within some designated volume element, and it does not require labeling the particles. We consider the case where particles in the entire volume w have already been completely mixed but where, owing to the statistical nature of the process, the number of particles entering the designated volume w during the time interval τ may be either larger or smaller than the number of particles leaving it during this time. Having measured the initial number of particles in this volume n and also their number n_τ after a time τ, and having repeated such measurements many times at fixed intervals τ = const, one can calculate the magnitude of the mean-squared fluctuations:

$$\overline{(\Delta n_\tau)^2} = \overline{(n_\tau - n)^2}. \tag{4}$$

Let $\bar{n}$ denote the average number of particles within the given volume. For long intervals τ the numbers n_τ and n will be mutually independent, i.e., there will be no correlation between them and

$$\overline{(\Delta n_\tau)^2}_{\tau\to\infty} = \overline{n_\tau^{\,2}} - 2\bar{n}_\tau \bar{n} + \bar{n}^2 = 2\,(\overline{n^2} - \bar{n}^2) = 2\,\overline{(n - \bar{n})^2} = 2\overline{(\Delta n)^2}\,, \tag{5}$$

where $\overline{(\Delta n)^2}$ is the mean-squared fluctuation of the number of particles about their average number $\bar{n}$ within the given volume. For short intervals τ the numbers n_τ and n will be correlated according to the diffusion law [11]:

$$\overline{(\Delta n_\tau)^2} = \overline{(\Delta n)^2}\,\frac{D\tau}{2\pi l^2}\,, \tag{6}$$

where l is the mean radius of the designated volume w. Relation (6) remains valid as long as $\overline{(\Delta n_\tau)^2} \ll \overline{(\Delta n)^2}$, and in this case the sought coefficient D can be calculated according to that relation on the basis of $\overline{(\Delta n_\tau)^2}$ values measured in the experiment at various time intervals τ.

For such a calculation we used the same photographic films from which v_{circ} and D_{puls} had been determined for a given fluidization mode. With a film frame projected on the screen, a circle was drawn here at a radius corresponding to the cylindrical volume element designated inside the reactor ($R = 2.5$ cm) and the number of particles n within this circle was counted. In the case of a stationary bed there were 33 disks of radius $r = 0.4$ cm inside such a circle. This corresponded to a fill factor of $\sigma = 1 - \varepsilon = 33r^2/R^2 = 0.845$, indicating a random distribution of particles and the effect of walls, since the closest possible packing would have yielded $\sigma_{\max} = \pi/2\sqrt{3} = 0.907$.

During fluidization some particles dispersed and the bed expanded, the number $\bar{n}$ becoming accordingly smaller than 33. We must note here that, in counting the number n one by one, it was difficult to determine exactly the fraction of any disk on the border of the drawn circle and only partially inside it.

On the basis of a frame-to-frame examination of the photographic film, successive values of n were tabulated after 1/36 sec, whereupon the average $\bar{n}$ and the mean-squared $\overline{(\Delta n)^2}$ or the mean absolute deviation from the average $\overline{|\Delta n|} = \overline{|n - \bar{n}|}$ were calculated. Thus, increasing the time intervals stepwise from $\tau = 1/36$ sec to $\tau = 2/36$ sec, $\tau = 3/36$ sec, etc., we calculated the fluctuations $\Delta n_\tau = n_\tau - n$ and their mean values $\overline{(\Delta n_\tau)^2}$ or $\overline{|\Delta n_\tau|}$.

Such a procedure revealed at once that the relation

$$\overline{(\Delta n)^2} = \bar{n}, \tag{7}$$

while applicable to unexpandable particles in classical statistical physics, would not be valid in our case of adjoining disks and that much closer here would be the approximate relation for expandable particles

$$\overline{(\Delta n)^2} = \bar{n}\,(\sigma_m - \sigma) \tag{8}$$

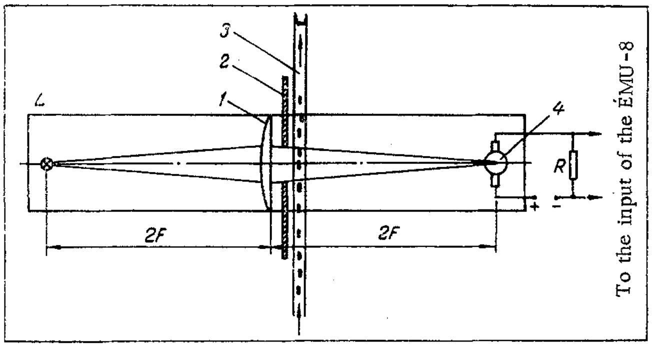

Fig. 1. Basic schematic diagram of the apparatus for determining the mixing coefficient.

according to Buevich [15]. Unfortunately, specific calculations showed that, even for the shortest time interval $\tau = 1/36$ sec (every frame) $\overline{\Delta n}_T$ had reached about 40-60% of its limit $\overline{\Delta n}_{T\to\infty}$ and that, consequently, the assumptions made in deriving formula (6) had not been satisfied.

The necessity of further reducing the interval τ and also the desirability of automating the rather unwieldy and laborious "one-by-one" computations have provided the stimulus for further improving the measurement procedure. The "one-by-one" count of particles was replaced by an automatic recording of the decrease in the intensity of light transmitted through the given reactor segment with suspended particles. A photocell then converted the optical signals to electrical ones, and the latter were fed to an analog computer for direct processing with the aid of one type BTD complementer and for an immediate readout of the desired result.

The optical–mechanical part of the apparatus is shown schematically in Fig. 1. The lamp (12 V, 90 W) was supplied from a storage battery and interference arising from fluctuations of the line voltage was thus avoided. This lamp was placed at double the focal distance from the optical center of lens 1. Light from this source was passing through the lens, a circular aperture of radius R = 2.5 cm in the opaque screen 2, and reactor 3, from where it was then focused on the photocell 4. The moving opaque particles (disks) affected the magnitude of the luminous flux impinging on the photocell, while a variable voltage U(t) proportional to the transmitted luminous flux was picked off the load resistor R and applied to the input of the EMU-8 analog computer with a switching network as shown schematically in [9]. The computer averaged an incoming signal over a prescribed time interval T_1 = 5 sec and the result of the attendant integration was put out in terms of voltmeter readings. From this we determined the average voltage

$$\bar{U} = \frac{1}{T_1}\int_0^{T_1} U(\tau)\,d\tau.$$

For the purpose of calibration, we determined the average value of a signal without particles in the bed:

$$\bar{U}_0 = \frac{1}{T_1}\int_0^{T_1} U^*(\tau)\,d\tau.$$

The signal reduction $\bar{U}_0 - \bar{U}$, as a result of partial blocking of the aperture area by particles, allowed us to calculate the proportional to it average number of particles in the bed:

$$\bar{n} = \frac{S_0}{S_1}\cdot\frac{\bar{U}_0 - \bar{U}}{\bar{U}_0}, \tag{9}$$

with S_0 denoting the full aperture area in the screen and S_1 denoting the area of one aluminum disk.

With the ÉMU-8 computer set up for solving the equation of a high-pass filter [16], it was possible to eliminate the dc component $\bar{U}$ of the total act signal $U(\tau)$ and to feed the difference $\Delta U = U(\tau) - \bar{U}$ to the integrating networks, after the positive deviations ΔU^+ had been separated from the negative deviations ΔU^- by means of semiconductor diodes and both had been integrated separately [16]. In this way, we automated the measurement of the average absolute deviation over a sufficiently long time interval T_2 = 50 sec:

TABLE 2. Mixing Coefficient for Solid Particles, at Various Values of Fluid Viscosity and Stream Velocity, Obtained in Tests with the Aid of an ÉMU-8 Computer and a BTD Unit

Fluidizing agent	Ar	$\nu \cdot 10^6$, m²/sec	$u \cdot 10^2$, m/sec	$D \cdot 10^4$, m²/sec
Water	$1{,}3 \cdot 10^7$	1	13	16
			18	20
			23	45
50% Glycerine + 50% water	$5{,}0 \cdot 10^5$	5	13	11
70% Glycerine + 30% water	$5{,}4 \cdot 10^4$	16	10	9
90% Glycerine + 10% water	$4{,}7 \cdot 10^3$	50	5	3

$$\overline{\Delta U}| = \frac{1}{T_2}\int_0^{T_2} |\Delta U|\, dt,$$

from which the mean absolute deviation could then be calculated:

$$|\overline{\Delta n}| = \frac{S_0}{S_1} \cdot \frac{1}{U_0} |\overline{\Delta U}|. \tag{10}$$

In order to determine the difference fluctuation Δn_τ, a standard time-delay block (BTD) was used in the ÉMU-8 complementer for converting the variable voltage into an identical function shifted in time by a given interval. Into the summator was then fed the original signal U(t) and the inverted and delayed signal $-U(t-\tau)$, whereupon the resulting difference $\Delta U_\tau = U(t)-U(t-\tau)$ was processed in the same manner as $\Delta U = U(t)-\bar{U}$ and the average absolute fluctuation $|\overline{\Delta n_\tau}|$ was found over various delay times τ from 0.005 to 0.150 sec. In view of the proportionality between average absolute and mean-squared fluctuations, formula (6) for calculating the effective diffusion coefficient for a solid phase was replaced by

$$D = \frac{\pi l^2}{\tau}\left\{\frac{|\overline{\Delta n_\tau}|}{|\overline{\Delta n}|}\right\}^2, \tag{11}$$

where l = R/2 in the case of a circle.

As has been noted earlier, the quantity inside the brackets in (11) begins to approach its limit (~10%) at τ = 0.030 sec and ceases to be directly proportional to τ. On the other hand, at τ = 0.005 sec the BTD introduces an excessive measurement error. For this reason, it was necessary to calculate D essentially only from the data obtained during τ = 0.01-0.02 sec. Results of these computations are shown in Table 2.

As can be seen here, the mixing coefficient D is of the order of $2 \cdot 10^{-3}$ m²/sec for fluidization with water or with up to 50% glycerine solution in water, which agrees with the earlier estimates based on relations (2) and (3). On the other hand, it appears that $D \gg D_{\text{puls}}$ and, consequently, each of these two coefficients characterizes a different aspect of the random motion of particles.

It was crucial in our tests that the dimension of the designated volume (L = 2R = 5 cm) containing about 30 particles exceeded the dimension of particle "packets" agglomerating in their motion (about 5 particles together in our case). The length of the time interval $\tau \approx 0.1$ sec, after which almost no correlation would be observed any more, indicates that during such a long period the particles within the test volume have completely mixed with surrounding particles. The estimate according to (2) will then yield only the order of magnitude of the mixing coefficient:

$$D \approx \frac{(5\text{ cm})^2}{10 \cdot 0.1\text{ sec}} \approx 25\text{ cm}^2/\text{sec} = 2.5 \cdot 10^{-3}\text{ m}^2/\text{sec}.$$

The preceding analysis has confirmed the effectiveness of the proposed fluctuation method of measuring the mixing coefficient and has confirmed the feasibility of automating the measurements as well as the data evaluation. Application of this method to the two-dimensional "one grain thick" fluidization bed model has made it possible to establish a relation between the mixing of a solid phase and the characteristics of the circulatory motion of its particles.

NOTATION

D, D_{circ}, D_{puls}	are the overall, circulatory, and pulsatory mixing coefficients;
v_{circ}, $\bar{v}$	are the circulatory and average velocity of a particle;
$\bar{u}$	is the velocity of the fluidizing stream;
Δl	is a segment of a particle trajectory;
τ, $\Delta\tau$	are time intervals;
H, W	are the height and volume of the entire bed;
w	is the volume of the designated bed segment;
n, n_τ, $\bar{n}$	are the initial number, the number after time τ sec, and the average number of particles;
$\overline{(\Delta n_\tau)^2}$	is the mean-squared fluctuation;
$\underline{l}$	is the mean radius of the designated volume;
U_0, $U(t)$	are the voltage without particles and with moving particles, respectively;
T_1, T_2	are integration periods;
r, R	are the radius of the disk and the radius of diaphragm (cylinder);
L	is the scale dimension of the designated volume;
ν	is the kinematic viscosity;
σ, σ_m	are the fill factor and maximum fill factor of the designated volume with particles;
ε	is the porosity of the bed.

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