HYDRODYNAMIC MIXING IN A GRANULAR BED WITH IRREGULAR PACKING

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In investigating longitudinal [1-4] and transverse [5] mixing in a granular bed described by a cellular model it has so far been assumed that all the cells are identical, while the parameters of the distribution function of the residence time in the individual cell (henceforth called the microdistribution) are strictly fixed. In an actual granular bed with irregular packing the identical cell condition is obviously not satisfied and the microdistribution parameters may be regarded as random quantities. This paper is concerned with an investigation of the effect of the spread of microdistribution parameters on the characteristics of the process of longitudinal and transverse transport of a neutral fluid in a granular bed.

While retaining the general structure of the cellular model [5], we assume that the gas or liquid passing through the bed flows from the cells on one horizontal level into the cells on the next level downstream, its motion in the transverse direction being random. Obviously, a particle of the fluid passing through the granular bed can then occupy one cell on each horizontal level. Dropping the identical cell condition, we assume that the bed is homogeneous and that the parameters of the cells successively occupied by a fluid particle are statistically independent. This assumption means that cells with different values of the parameters have been "well mixed," so that inside the bed there are no fluctuations significantly exceeding the cell dimension in extent.

Let the microdistribution function f(t|s) and the corresponding characteristic function

$$g(p|s) = \int_{0}^{\infty} e^{-pt} f(t|s) dt \qquad (0.1)$$

depend on the random vector s. The components of this this vector are the different microdistribution parameters which vary randomly from cell to cell. In the simplest case of ideal mixing there is only one such parameter—the mean residence time in the cell s == V/v, where V is the volume of the cell and v the volume rate of flow through it. We will characterize the distribution of the random vector s by the distribution function $\varphi(s)$ which determines the probability of a fluid particle entering a cell with given value of the vector s. We will call the function $\varphi(s)$ the parametric distribution. Another possible characteristic of the distribution of the vector s is the function $\psi(s)$ which determines the probability of finding a given value of the vector s in a cell selected from the bed at random. The first characteristic is more convenient for our purposes, since it is directly related with the motion of the flow and is in some sense dynamic, as distinct from the second characteristic which is static. The two distribution functions are related by the expression

$$\varphi(\mathbf{s}) = \frac{\langle v_{\mathbf{s}} \rangle}{v_0} \, \psi(\mathbf{s}), \ \varphi(s) = \frac{s_0}{s} \, \psi(s) \text{ when } v_s \sim s^{-1}, \ (0.2)$$

where $\langle v_s \rangle$ is the mean volume flow rate in cells with a given value of the vector s, v_0 is the mean volume flow

rate in all the cells, and s_0 is the value of the mean cell residence time s averaged over all the cells. The second of equations (0.2) is obtained in the special case of ideal mixing on the assumption that the volume of the cell and the volume rate of flow through it are noncorrelated. In accordance with (0.2), the probability of a fluid particle entering cells with small mean residence times will be much greater than the fraction of such cells in the bed. The opposite will hold true for cells with large values of s.

1. Longitudinal transport. We will first consider the process of longitudinal transport of a neutral fluid. Let the subscript k indicate the number of the horizontal plane along the direction of motion of the flow. It is required to determine the probability of a fluid particle entering one of the cells in the plane k = 0, leaving any cell in the plane k = n after time t. We will call the corresponding probability density the macrodistribution and denote it by $F_n(t)$. The most important characteristics of the macrodistribution from our standpoint are its variance and the coefficient of skewness, which characterizes the deviation of the macrodistribution from the normal law. These quantities are most easily found starting from the characteristic macrodistribution function $G_n(p)$ which is related to $F_n(t)$ by an expression analogous to (0.1).

We will consider some random fluid-particle trajectory passing through cells with values $s_k(k=1, 2, ..., n)$ of the random vector s. The residence-time distribution function corresponding to this trajectory is determined in the same way as for a one-dimensional chain of cells. Since the residence times in successive cells are mutually independent, the characteristic function for the given trajectory has the form

$$G_n(p | \mathbf{s}_1, \mathbf{s}_2, \dots, \mathbf{s}_n) = \prod_{k=1}^n g(p | \mathbf{s}_k).$$
 (1.1)

In order to find the characteristic macrodistribution function, it is sufficient to perform summation over all the trajectories:

$$\langle G_n(p) \rangle = \int \dots \int \varphi (\mathbf{s}_1, \dots, \mathbf{s}_n) \prod_{k=1}^n g(p \mid \mathbf{s}_k) d\mathbf{s}_k$$
, (1.2)

where the integration is performed over the entire region of variation of the vector s. Strictly speaking, $\langle G_n(p) \rangle$, thus defined, should be regarded as the characteristic macrodistribution function averaged over the ensemble of realizations of the granular bed. However, if the number of cells in the bed is sufficiently large, this function will be almost the same for all realizations.

In accordance with our assumption concerning the statistical independence of the parameters of succes-

sive occupied cells,

$$\varphi(\mathbf{s}_1, \dots, \mathbf{s}_n) = \prod_{k=1}^n \varphi(\mathbf{s}_k),$$
$$\langle G_n(p) \rangle = \left[\int g(p \mid \mathbf{s}) \varphi(\mathbf{s}) \, d\mathbf{s} \right]^n = \langle g(p \mid \mathbf{s}) \rangle^n, \quad (1.3)$$

where the angle brackets denote averaging with respect to the parametric distribution. We will consider the following basic characteristics of the macrodistribution: the mean dwell time in the bed, variance, the third central moment, and the coefficient of skewness Sk_n, characterizing the deviation of the macrodistribution from the normal law. The first three quantities coincide with the semiinvariants of the macrodistribution \varkappa_j (j = 1, 2, 3), while the last is equal to \varkappa_3 , $\varkappa_2^{-3/2}$. Using the formula for the semiinvariants of a random quantity

$$\kappa_n = (-1)^j \left[\frac{d^j}{dp^j} \ln \langle G_n (p) \rangle \right]_{p=0} =$$
$$= (-1)^j n \left[\frac{d^j}{dp^j} \ln \langle g (p \mid s) \rangle \right]_{p=0}$$
(1.4)

and the formula for the moments of the characteristic microdistribution function

$$\alpha_j(\mathbf{s}) = (-1)^j \left[\frac{d^j g\left(p \mid \mathbf{s} \right)}{d p^j} \right]_{p=0}, \qquad (1.5)$$

we find

$$\begin{aligned} \varkappa_{1n} &= -n \left[\frac{d}{dp} \ln \langle g(p \mid s) \rangle \right]_{p=0} = n \langle \alpha_1(s) \rangle, \\ \varkappa_{2n} &= n \left[\langle \alpha_2(s) \rangle - \langle \alpha_1(s) \rangle^2 \right], \\ \varkappa_{3n} &= n \left[\langle \alpha_3(s) \rangle - 3 \langle \alpha_2(s) \rangle \langle \alpha_1(s) \rangle + 2 \langle \alpha_1(s) \rangle^3 \right], \\ \mathbf{Sk}_n &= \frac{1}{\sqrt{n}} \frac{\langle \alpha_3 \rangle - 3 \langle \alpha_2 \rangle \langle \alpha_1 \rangle + 2 \langle \alpha_1 \rangle^3}{\langle \langle \alpha_2 \rangle - \langle \alpha_1 \rangle^2 \rangle^{d_2}}. \end{aligned}$$
(1.6)

Thus, the macrodistribution characteristics are related with the mean moments of the microdistribution.

We will consider the specific case of ideal mixing. In this case there is only one random parameter—the mean residence time s. Then

$$g(p \mid s) = (1 + ps)^{-1}, \ \alpha_1 = s, \ \alpha_2 = 2s^2, \ \alpha_3 = 6s^3.$$
 (1.7)

Substituting (1.7) into (1.6), we find

$$\begin{aligned} \varkappa_{1n} &= n \langle s \rangle = n \int_{0}^{\infty} s\varphi(s) \, ds = ns_{0}, \\ \varkappa_{2n} &= n \left(2 \langle s^{2} \rangle - \langle s \rangle^{2} \right) = n \left(2v_{2} + s_{0}^{2} \right) = ns_{0}^{2} \left(1 + 2\gamma \right), \\ \varkappa_{3n} &= 2n \left(3v_{3} + 6v_{2}s_{0} + s_{0}^{3} \right), \\ \mathrm{Sk}_{n} &= \frac{2}{\sqrt{n}} \frac{s_{0}^{3} + 6s_{0}v_{2} + 3v_{3}}{(s_{0}^{2} + 2v_{2})^{3/2}} = \frac{2}{\sqrt{n}} \frac{1 + 6\gamma + 3\sigma\gamma^{3/2}}{(1 + 2\gamma)^{3/2}}, \\ \gamma &= v_{2}s_{0}^{-2}, \quad \sigma = v_{3}v_{2}^{-3/2}, \quad v_{j} = \int_{0}^{\infty} \left(s - s_{0} \right)^{j} \varphi(s) \, ds, \quad (1) \end{aligned}$$

where v_i is the j-th central moment of the parametric

.8)

distribution, while γ and σ are the variance and coefficient of skewness of that distribution, respectively.

In a system of identical cells the variance of the macrodistribution is equal to ns_0^2 . It is clear from (1.8) that the increase in the variance of the macrodistribution owing to the nonuniformity of the cells is proportional to the variance of the parametric distribution ν_2 . Upon investigating the expression for the coefficient of skewness of the macrodistribution in (1.8), we quickly note that in the two limiting cases this formula admits simplifications:

$$Sk_{n} = \frac{2}{\sqrt{n}} (1 + 3\gamma^{y_{z}} 5) \text{ when } \gamma \ll 1,$$

$$Sk_{n} = \frac{3\sigma}{\sqrt{2n}} \text{ when } \gamma \gg 1.$$
(1.9)

Let m be a number of cells along the length of the bed such that $Sk_m = 1$. Obviously, if the number of cells along the length of the bed considerably exceeds m, the macrodistribution will be close to normal. Equations (1.9) make it possible to express the number m in terms of the characteristics of the parametric distribution. It is clear from (1.9) that the number m can be large as compared with unity only if the parametric distribution is strongly asymmetrical. In this case positive skewness of the parametric distribution corresponding to the presence in the bed of a considerable number of cells with very large mean residence times (i.e., almost stagnant) leads to positive skewness of the macrodistribution corresponding to the apperance of macrodistributions with long "tails."

The effect of the characteristics of the parametric distribution on the shape of the macrodistribution can easily be followed with reference to the example of a bimodal distribution

$$\varphi (s) = a\delta (s - s_1) + (1 - a) \delta (s - s_2) =$$

= $a\delta (s - s_0 - (1 - a)\Delta s) +$
+ $(1 - a\delta) (s - s_0 - a\Delta s).$ (1.10)

Here, $\Delta s = s_2 - s_1$ is the difference between the mean residence times s_1 and s_2 for two groups of cells, *a* is the probability of entering cells of the first group, and $\delta(s - s_2)$ is the Dirac delta function.

For distribution (1.10) we have

$$v_{2} = \Delta s^{2}a (1 - a), v_{3} = \Delta s^{3}a (1 - a) (2a - 1),$$

$$\gamma = \beta^{2} (1 - a) a,$$

$$\sigma = (2a - 1)/\sqrt{a (1 - a)}, \beta = s_{0}^{-1}\Delta s. \quad (1.11)$$

Since s_1 must be positive, $\beta^{-1} > 1 - a$ always. The formulas for the variance and the coefficient of skewness of the macrodistribution now take the form

$$\varkappa_{2n} = n s_0^2 \left[1 + 2\beta^2 a \left(1 - a \right) \right],$$

$$\operatorname{Sk}_n = \frac{2}{\sqrt{n}} \frac{1 + 6\beta^2 a \left(1 - a \right) \left[1 + \beta \left(a - \frac{1}{2} \right) \right]}{\left[1 + 2\beta^2 a \left(1 - a \right) \right]^{3/2}}.$$
 (1.12)

At $\beta \ll 1$ the variance and the coefficient of skewness of the macrodistribution are almost the same as

in the analogous system of identical cells. When $\beta \gg 1$ the characteristics of the macrodistribution depend importantly on the quantity *a*. The following approximate formulas for three different regions of variation of *a* can be derived from (1.12):

$$\kappa_{2n} / ns_0^2 = 2\beta^2 (1-a) \sim \beta \gg 1 ,$$

$$\operatorname{Sk}_n = 3 / \sqrt{n (1-a)} \sim \sqrt{\beta / n}$$

when $\beta^{-2} \ll 1 - a \leqslant \beta^{-1}$, (1.13)

$$\varkappa_{2n} / n s_0^2 \sim 1, \quad \mathrm{Sk}_n = 3\beta^3 \left(1 - a\right) / \sqrt{n} \sim \beta / \sqrt{n}$$

when $\beta^{-3} \ll 1 - a \leqslant \beta^{-2}$, (1.14)

$$\kappa_{2n} / n s_0^2 = 1$$
, $Sk_n = 2 / \sqrt{n}$ when $1 - a \leq \beta^{-3}$. (1.15)

In the first region, in accordance with (1.13), the spread of the cell parameters leads to a considerable increase in the variance of the macrodistribution as compared with a system of identical cells. In this case $m \sim \beta$ and a normal distribution can be established only in a sufficiently long bed. In the second region, where formulas (1.14) apply, the variance of the macrodistribution is of the same order as the corresponding quantity for a system of identical cells, but the skewness of the macrodistribution is considerable, so that $m \sim \beta^2$, and the normal law is approached only very slowly. Finally, at an even smaller fraction of cells with large mean residence times, in accordance with (1.15), both the variance and the coefficient of skewness of the macrodistribution become the same as in a system of identical cells and the normuliformity of the bed ceases to make itself felt.

These results can be used to determine the characteristics of the macrodistribution from experimental observations of the residence time distribution function. However, in analyzing the experimental data the following question may arise: is the increase in the variance and skewness of the macrodistribution connected with the nonuniformity of the cells or the retention of neutral fluid in stagnant zones [3,4]? The following criteria may be used to solve this problem. Firstly, the effect of stagnant zones is always to lead not only to an increase in variance but also to the appearance of asymmetrical (at moderate n) macrodistributions. At the same time, the spread of the cell parameters may cause an increase in the variance of the macrodistribution without leading to an appreciable deviation from the normal law. Secondly, the action of stagnant zones is closely linked with the nature of the flow and is manifested in liquids much more strongly than in gases [4]. However, if the skewness of the macrodistribution is caused by the skewness of the parametric distribution, this effect will be purely hydrodynamic and, other conditions being equal, should be the same for liquid and gas flows.

2. Transverse transport. We will consider the problem of the transverse transport of a fluid caught in a certain cell of the bed. The macrodistribution $F_n(x, t)$ is now understood to represent the probability of finding a fluid particle entering a cell with coordinate x == 0 in the horizontal plane k = 0 at time t = 0 from the cell with coordinate x in the plane k = n at time t. Since the horizontal planes are isotropic, it is sufficient to follow the transverse motion of the particle along one coordinate axis x only. All the properties of the macrodistribution of interest can be found from an analysis of the two-dimensional characteristic function

$$G_n(\lambda, p) = \int_{-\infty}^{\infty} e^{i\lambda x} dx \int_{0}^{\infty} e^{-pt} F_n(x, t) dt. \qquad (2.1)$$

The two-dimensional characteristic function for a trajectory passing through n cells of the granular bed

will be the product of the two-dimensional characteristic functions of the following elementary events: a) r transitions from a cell of the k-th horizontal plane to a cell of the (k + 1)-th plane (k = 0, 1, ..., n) with trans verse displacement through a distance l_k at a random angle α to the x-axis, and b) retention in the n-th cell.

This is justified by the fact that all the above-mentioned elementary events are mutually independent.

The residence time in an individual cell, which is equal to the interval between two successive transitions, and the direction of transverse displacement ar also obviously independent and therefore the two-dime sional characteristic function of each of events (a) will be a product of one-dimensional functions.

Assuming that the angle formed by the direction of transverse displacement and the x-axis is uniformly distributed on the interval $(0, \pi)$, we find that the probability of displacement along the x-axis through the distance from x to x + dx in the course of a single transition with step l_k is determined by

$$dx / \pi \sqrt{l_k^2 - x^2}, \qquad -l_k \leqslant x \leqslant l_k.$$
 (2.2)

Hence we find the coordinate characteristic function of the event (a)

$$\frac{1}{\pi} \int_{-l_k}^{l_k} \frac{e^{i\lambda x} dx}{\sqrt{l_k^2 - x^2}} = J_0(\lambda l_k), \qquad (2.3)$$

where J_0 is a Bessel function of zero order.

The corresponding time characteristic function coincides with the characteristic function of the micrc distribution $g(p|s_k)$. The characteristic function of event (b) has only a time factor and is equal to (1 - g)/p.

We will consider a fixed trajectory passing through cells with values s_k of the parameter vector and under going a transverse displacement through the distance l_k at each transition. In view of what was said above, for this trajectory the two-dimensional characteristic function is given by

$$G_{n}(\lambda, p \mid \mathbf{s}_{0}, \mathbf{s}_{1}, \dots, \mathbf{s}_{n}) =$$

$$= \frac{1}{p} \left[1 - g\left(p \mid \mathbf{s}_{n} \right) \right] \prod_{k=0}^{n-1} g\left(p \mid \mathbf{s}_{k} \right) J_{0}\left(\lambda l_{k}\right). \tag{2.4}$$

In order to find the two-dimensional characteristic function of the macrodistribution averaged over the ensemble of realizations of the granular bed it is necessary to carry out summation over all possible trajectories. Using, as in \$1, the assumption concerning the homogeneity of the bed, which leads to the mutual independence of the parameters \mathbf{s}_k and l_k in successively occupied cells, we have

$$\langle G_n(\lambda, p) \rangle = p^{-1} \left[1 - \langle g(p \mid \mathbf{s}) \rangle \right] \langle g(p \mid \mathbf{s}) J_0(\lambda l) \rangle^n, \quad (2.5)$$

where, as before, the corner brackets denote averaging with respect to the parametric distribution, so that

$$\langle a(p, \lambda | \mathbf{s}, l) \rangle = \int a(p, \lambda | \mathbf{s}, l) \varphi(\mathbf{s}, l) d\mathbf{s} dl$$
. (2.6)

Here, the integration is performed over the entire region of variation of the value of the parameters s and l.

In investigating the process of transverse transport we will consider, as in [5], the probability of detecting a particle at a given time in a cell with a given transverse coordinate irrespective of its position along the longitudinal axis. In order to find the corresponding two-dimensional transverse macrodistribution function, it is necessary to sum (2.5) with respect to n:

$$\langle G(\lambda, p) \rangle = \sum_{n=0}^{\infty} \langle G_n(\lambda, p) \rangle =$$
$$= \frac{1}{p} \frac{1 - \langle g(p \mid \mathbf{s}) \rangle}{1 - \langle g(p \mid \mathbf{s}) J_0(\lambda l) \rangle}.$$
(2.7)

Differentiating (2.7), we easily find the Laplace transforms of the moments of the transverse macrodistribution

$$\mu_{j}(p) = i^{-j} \left[\frac{\partial^{j}}{\partial \lambda^{j}} \langle G(\lambda, p) \rangle \right]_{\lambda=0}.$$
 (2.8)

Because of symmetry only the even moments are nonzero. In particular, for the variance of the transverse macrodistribution we have

$$\boldsymbol{\mu}_{2}(p) = \frac{\langle l^{2}g(p \mid \mathbf{s}) \rangle}{2p(1 - \langle g(p \mid \mathbf{s}) \rangle)} \,. \tag{2.9}$$

If we assume that the parameters of the microdistribution are fixed and set $l = (2)^{1/2}$, this formula will coincide with the corresponding expression of [5]. As shown in [5], function (2.9) does not have singularities in the right half-plane or on the imaginary axis, but at the point p = 0 it has a second-order pole. Consequently, the asymptotic expression for the variance $\mu_2(t)$ has the form

$$\mu_{2}(t) = \operatorname{Res}\left[\frac{e^{pt}\langle l^{2}g\left(p\mid s\right)\rangle}{2p\left(1-\langle g\left(p\mid s\right)\rangle\right)}\right]_{p=0} = \frac{\langle l^{2}\rangle}{2}\frac{t}{\langle \alpha_{1}\rangle} + \frac{\langle l^{2}\rangle}{2}\frac{\langle \alpha_{2}\rangle}{2\langle \alpha_{1}\rangle^{2}} - \frac{\langle l^{2}\alpha_{1}\rangle}{2\langle \alpha_{1}\rangle}, \quad (2.10)$$

where α_j are the moments of the microdistribution determined in accordance with (1.5).

The asymptotic expression for the fourth moment of the transverse macrodistribution is similarly determined. In this case, as in [5], it is found that the coefficient of excess of the transverse distribution tends to zero as $t \rightarrow \infty$. Thus, at sufficiently large times there is established a normal distribution with variance $t \langle l^2 \rangle / 2s_0$ that does not depend either on the shape of the microdistribution or on the shape

of the parametric distribution and is determined only by the meansquare transverse displacement in each transition (i.e., by the mean packing structure of the bed) and by the mean value over the bed of the mean residence time in the cell s_0 . The shape of the parametric distribution may affect the time required to establish a normal law.

The macrodistribution function in the steady state $\langle F_n(x) \rangle$ can be obtained from the function $\langle F_n(x,t) \rangle$ by integrating it with respect to t from 0 to ∞ . The corresponding characteristic function $\langle G_n(\lambda) \rangle$ is obtainable directly from (2.5) by setting p = 0,

$$\langle G_n(\lambda) \rangle = s_0^{-1} \langle G_n(0, \lambda) \rangle = \langle J_0(\lambda l) \rangle^n . \tag{2.11}$$

It is clear from (2.11) that in the steady state the form of the microdistribution function and hence the spread of its parameters is unimportant. From (2.11) it is easy to find the variance, the fourth moment and the coefficient of excess of the stationary macrodistribution

$$\mu_{2n} = \frac{1}{2} n \langle l^2 \rangle, \quad \mu_{4n} = \frac{3}{8} n \langle l^4 \rangle + \frac{3}{4} n (n-1) \langle l^2 \rangle^2,$$

$$Ex_n = \frac{\mu_{4n}}{\mu_{2n}^2} - 3 = \frac{3}{2n} \left(\frac{\langle l^4 \rangle}{\langle l^2 \rangle^2} - 2 \right) =$$

$$= \frac{3}{2n} (\chi + 1) \left(\chi = \frac{\langle l^4 \rangle}{\langle l^2 \rangle^2} - 3 \right), \quad (2.12)$$

where χ is the kurtosis of the distribution of step length l. Obviously, when $\langle l \rangle \sim \langle l^2 \rangle^2$ at moderate distances from the point of admission of the fluid a normal distribution with variance proportional to n and $\langle l^2 \rangle$ is always established. Only at a considerable kurtosis of the step-length distribution (which is physically improbable) will a normal distribution be established at great distances from the point of admission.

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REFERENCES

1. H. Kramers and G. Alberda, "Frequency response analysis of continuous flow systems," Chem. Engng. Sci., vol. 2, no. 3. 1953.

2. N. R. Amundson and R. Aris, "Some remarks on longitudinal mixing or diffusion in fixed beds," Amer. Instit. Chem. Engrs. J., vol. 3, p. 280, 1957.

3. V. G. Levich, L. M. Pis'men, and S. Kuchanov, "Hydrodynamic mixing in a granular bed. Physical model of stagnant zones," DAN SSSR, 168, no. 2, 1966.

4. L. M. Pis'men, S. I. Kuchanov, and V. G. Levich, "Transverse diffusion in a granular bed," DAN SSSR, 174, no. 3, 1967.

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