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The present paper deals with a study on the lateral diffusion of a hydraulically neutral tracer injected near the center of a granular bed. The pore space of the granular bed is considered to consist of a set of cells connected by channels. The liquid or gas passing through the bed flows from the cells in one horizontal cross-section to the adjacent cells of the next lower level, and is any time mixed at random, since it is displaced laterally over a certain distance determined by the type of the layer packing. It is assumed that displacement in any lateral direction occurs with equal probability. Since the admixed substance is washed out of the cell by the directed flow of gas or liquid, the lateral velocity precisely determines the lateral transfer of the admixed substance.

Every cell is characterized by a definite differential distribution function $f(\tau)$ of the residence time in the cell; this differential function will be further called the microdistribution. The various models of granular beds yield different types of microdistributions. In the present paper we investigate the characteristics of lateral diffusion which are independent of the shape of the microdistribution $f(\tau)$; therefore, the shape of this function need not be specified.

We shall consider an infinitely extended network of identical cells. The index n indicates the number of the plane perpendicular to the flow direction, and index m the position of the cell in this plane. It is inevitable to consider random walks of the introduced tracer particle across the cell. According to the above considerations, if the number of transitions from one cell to another is fixed, the longitudinal displacement of the tracer particle is defined unambiguously, but the lateral displacement is a stochastic variable. According to the classical theory of random walk [1], the probability that a tracer particle from the cell with $m = 0$, $n = 0$ will arrive in the cell with the indices m , n equals

$$n! / 2^n \binom{n+m}{2}! \binom{n-m}{2}! \quad (1)$$

if n and m are even, whereas this probability is zero for uneven n and m . Since the process is symmetrical for n -values of opposite sign, we may further consider solely positive m values. Unlike in the classical theory of random walk, where every transition takes place at an accurately defined moment, the time interval between successive transitions in the system considered is a stochastic variable determined by the microdistribution $f(\tau)$. The probability that exactly n transitions have taken place at the moment t equals

$$\int_0^t f_n(\tau) \left[1 - \int_0^{t-\tau} f(\xi) d\xi \right] d\tau, \quad (2)$$

where $f_n(\tau)$ is the differential distribution function of the residence time in a chain of n cells. Multiplying the probabilities (1) and (2), we find the probability that the tracer particle is located in the cell with indices n , m at the moment t :

$$F_{mn}(t) = \frac{n!}{2^n \binom{n+m}{2}! \binom{n-m}{2}!} \int_0^t f_n(\tau) \left[1 - \int_0^{t-\tau} f(\xi) d\xi \right] d\tau. \quad (3)$$

For the further discussion it is convenient to carry out a Laplace transformation, changing over from $F_{mn}(t)$ to its Laplace transform. Transformation of Eq. (3) yields:

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$$\tilde{F}_{mn}(p) = \frac{n!}{2^n \left(\frac{n+m}{2}\right)! \left(\frac{n-m}{2}\right)!} \frac{1-g(p)}{p} g^n(p), \quad (4)$$

where $g(p)$ denotes the characteristic microdistribution function of a separate cell*:

$$g(p) = \int_0^{\infty} f(\tau) e^{-p\tau} d\tau. \quad (5)$$

Upon passing from (3) to (4), we utilize the circumstance that the residence times in the cells are independent, so that the characteristic function of a chain of n cells equals the product of the characteristic functions of the separate cells.

Summing (4) over all values of index n , we can determine the Laplace transform of the probability that the tracer particle located in the cell with index $m = 0$ at $t = 0$ occupies the m -th cell at the moment t , i.e., find the transform of the macrodistribution function

$$\tilde{F}_m(p) = \sum_n \tilde{F}_{mn}(p) = \frac{1-g(p)}{p} \sum_{k=0}^{\infty} \frac{(m+2k)!}{(m+k)! k!} \left[\frac{g(p)}{2} \right]^{m+2k}, \quad (6)$$

where $k = (n-m)/2$. Calculation of the sum in (6) yields

$$\tilde{F}_m(p) = \frac{g^{-1}-1}{p \sqrt{g^{-2}-1}} (g^{-1} - \sqrt{g^{-2}-1})^m. \quad (7)$$

Starting from (7), we may also find the Laplace transform of the characteristic macrodistribution function $\tilde{G}(\lambda, p)$. Taking into account that the macrodistribution functions for m values of identical absolute magnitude and opposite sign are identical, we derive

$$\tilde{G}(\lambda, p) = \tilde{F}_0(m) + 2 \sum_{m=1}^{\infty} \tilde{F}_m(p) \cos m\lambda = \frac{1}{p} \frac{g^{-1}-1}{g^{-1}-\cos\lambda}. \quad (8)$$

From (8) it is evident that the function $\tilde{G}(\lambda, p)$ satisfies the normalization condition $\tilde{G}(0, p) = 1/p$, which implies that the macrodistribution $F_m(t)$ is normalized to unity. The Laplace transforms of the moments of the macrodistribution are defined by the formula

$$\tilde{\mu}_k(p) = i^k \left[\frac{\partial^k}{\partial \lambda^k} \tilde{G}(\lambda, p) \right]_{\lambda=0}. \quad (9)$$

It is obvious that, owing to the symmetry, only the even macrodistribution moments differ from zero. In particular, the Laplace transform of the variance of the macrodistribution equals

$$\tilde{\mu}_2(p) = 1/p(g^{-1}-1). \quad (10)$$

We shall utilize the formula of reverse Laplace transformation

$$\mu_2(t) = \frac{1}{2\pi i} \int_{a-i\infty}^{a+i\infty} \frac{e^{pt}}{p(g^{-1}-1)} dp \quad (11)$$

and investigate the asymptotic behavior of the variance $\mu_2(t)$ at $t \rightarrow \infty$ without specifying the function $g(p)$. As is well known, the shape of the function $\mu_2(t)$ is determined by the peculiar points of the function under the integral sign in (11). We shall assume that all these peculiar points are poles and number them in the sequence of decreasing real parts p_0, p_1, p_2, \dots . Assuming that the conditions of the residue theory are fulfilled, we may then represent $\mu_2(t)$ by the series

$$\mu_2(t) = \sum_i \text{Res} \left[\frac{e^{pt}}{p(g^{-1}-1)} \right]_{p=p_i}. \quad (12)$$

*To simplify the further discussion, we do not use the Fourier transform as the characteristic function, but employ the Laplace transform. It is evident that all properties of the characteristic function remain unaltered.

As can be easily seen, the asymptotic behavior of the function $\mu_2(t)$ is determined by the position of point p_0 . It can be proved that p_0 always equals zero, independently of the shape of the macrodistribution. In fact, since $f(t)$ is definite, the exponent equals zero, and, therefore the function $g(p)$ in semiplane with $\text{Re } p > 0$ is determined by integral (5). Assuming that $p = x + iy$ and bearing in mind that the microdistribution function $f(t)$ is positive and normalized to unity, we find

$$\text{Re } g(p) = \int_0^{\infty} e^{-xt} \cos yt f(t) dt < \int_0^{\infty} f(t) dt = 1.$$

Consequently, the asymptotic expression for the variancy $\mu_2(t)$ derived from formula (12) equals the residue in the point $p_0 = 0$.

It follows directly from the definition of the characteristic function $g(p)$ that $g(0) = 1$. Therefore, the function $\Delta(p) = g^{-1} - 1$ is analytical in the region around the point $p = 0$ and can be expanded there into a Taylor series

$$\Delta(p) = \sum_{k=1}^{\infty} \frac{(-1)^{k+1} \alpha_k}{k!} p^k. \quad (13)$$

It can be easily proved that the first moment α_1 equals the mean residence time in a cell \bar{t} , and, since this mean time differs from zero, function $\Delta(p)$ has a first-order root in the point $p = 0$. Consequently, the point $p = 0$ is a second-order pole of the function under the integral sign in (11). Owing to this, the asymptotic formula for the variancy at $t \rightarrow \infty$ reads

$$\mu_2(t) = \text{Res} \left[\frac{e^{pt}}{p\Delta(p)} \right]_{p=0} = \frac{d}{dp} \left[\frac{pe^{pt}}{\Delta} \right]_{p=0} = \frac{t}{\alpha_1} + \frac{\alpha_2}{2\alpha_1^2}. \quad (14)$$

Asymptotic formulas for the main even moments of the macrodistribution $F_m(t)$ can be derived in an analogous way. Utilizing (8), (9), we find the Laplace transform of the fourth moment

$$\tilde{\mu}_4(p) = \frac{1}{p\Delta} \left(1 + \frac{6}{\Delta} \right). \quad (15)$$

Changing over from the transform to the original function, we get

$$\mu_4(t) = \text{Res} \left[\left(\frac{1}{p\Delta} + \frac{6}{p\Delta^2} \right) e^{pt} \right]_{p=0} = \frac{3t^2}{\alpha_1^2} + \frac{t}{\alpha_1} \left(1 + \frac{6\alpha_2}{\alpha_1^2} \right) + \frac{\alpha_2}{2\alpha_1^2} + \frac{6\alpha_2^2}{\alpha_1^4} - \frac{2\alpha_3}{\alpha_1^3} \quad (16)$$

The excess coefficient, which characterizes the deviation of the symmetrical distribution from the normal law [2], equals

$$\text{Ex}(t) = \frac{\mu_4(t)}{\mu_2^2(t)} - 3 = \frac{\frac{t}{\alpha_1} \left(1 + \frac{3\alpha_2}{\alpha_1^2} \right) + \frac{\alpha_2}{\alpha_1^2} + \frac{21}{4} \frac{\alpha_2^2}{\alpha_1^4} - \frac{2\alpha_3}{\alpha_1^3}}{(t/\alpha_1 + \alpha_2/2\alpha_1^2)^2}. \quad (17)$$

From the asymptotic formulas for the moments of the macrodistribution it follows that a normal distribution which has a variancy equal to t/\bar{t} and is independent of the shape of the microdistribution establishes at $t \rightarrow \infty$. The time needed for establishing the normal distribution generally depends on two factors. Assume that point $p = p_1$ (according to the above considerations, $\text{Re } p_1 < 0$) represents the root of function $(p) = g^{-1} - 1$ which comes closest to the imaginary axis. Then, the asymptotic formulas hold for times $t \gg t_1$, where $t_1 = -1/\text{Re } p_1$. From this it follows that the normal distribution cannot be established before the condition $t \gg t_1$ is fulfilled. However, this is a necessary, though not a sufficient condition. A sufficient condition for establishment of the normal distribution is that the excess coefficient $\text{Ex}(t)$ defined by formula (17) is close to zero and the second term in the asymptotic formula for the variancy (14) is small compared with the first term. Consequently, the time in which the normal distribution establishes depends on the shape of the microdistribution and is determined by the parameters α_k .

We shall finally consider the stationary regime of lateral transfer. Here, the following problem is interesting. A tracer is continuously admixed to the cell with indices $n = 0$, $m = 0$. The probability that the tracer is located in the cell with indices n, m , must be determined. Since tracer particles injected into the bed at an arbitrary moment $t = \tau$ may be located in any cell at the moment t , the probability sought for evidently equals

$$F_{mn} = \int_0^{\infty} F_{mn}(\tau) I(t - \tau) d\tau, \quad (18)$$

where $I(t-\tau)d\tau$ represents the probability that a particle arrives in the zero cell in the time interval between $t-\tau$ and $t-\tau+d\tau$. In the stationary regime this probability is constant and equals $d\tau/\bar{t}$. Substituting the function $F_{mn}(t)$ given by formula (4) into Eq. (18) and integrating, we find

$$F_{mn} = n! / 2^n \binom{n+m}{2}! \left(\frac{n-m}{2}\right)!. \quad (19)$$

Hence it follows that the stationary distribution of the tracer does not depend on the shape of the microdistribution $f(\tau)$. At $n \gg 1$ and $m \ll n$ the binomial distribution (19) comes close to the normal distribution. In fact, utilizing Stirling's formula, we derive from (19) that

$$F_{mn} = \frac{1}{\sqrt{2\pi n}} \exp\left\{-\frac{m^2}{2n}\right\}. \quad (20)$$

The general formulas derived in this paper are applicable to various processes of lateral transfer of mass and heat in a granular bed.

LITERATURE CITED

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