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A special-correlation analysis is made of the trajectories of the motion of a particle marked with a radioactive isotope in a fluidized bed containing an insert. The movement of the particles is investigated and the parameters of the circulation and diffusion models are determined.

The mathematical modeling of the processes of transfer of matter and heat in a fluidized bed (FB), in a catalytic reactor in particular, is developed on the basis of extensive experimental material. The majority of the known methods allow one to obtain information about the local or integral properties of FB or serve to identify the parameters of the proposed mathematical models. The methods of a thermal marker [1, 2] or a magnetic [3, 4], chemical [5], etc., marker (see Fluidization, edited by I. F. Davidson and D. Harrison) are widely used in the investigation of the characteristics of the solid phase of FB. At the same time, only two experimental methods allow the direct study of particle motion without disrupting the structure of the bed with detectors. The first is based on following a particle which is a source of radioactive emission [6, 7] while the second is based on the observation of a particle which is opaque to x-ray [8] or visible [9] light.

In the present work we have developed an experimental means of investigating particle motion in fluidization, made a detailed analysis of particle behavior in an organized fluidized bed (OFB), and determined the parameters of the notable models.

Experimental Procedure

We used a method whose foundations were developed in [6]: following a particle marked by a radioactive cobalt isotope. The experimental installation was coupled with a Dnepr-2 computer-control system (CCS) [10, 11]. The random nature of radioactive decay introduces an additional and, when the activity of the particle is low, a very significant error into the determination of the coordinates. The separation of the useful random signal (the coordinates) from the random interference represents a complicated mathematical problem. But to study the character of the behavior of a particle it proved possible to determine estimates of the statistical characteristics of the random process of its motion without finding the true trajectory but having available the total signal and the corresponding estimates of the statistical characteristics of the interference signal.

The conditions under which the experiments were conducted were: activity of particle about 0.2 mCi, diameter of apparatus 0.18 m, bulk height of bed 0.2 m, through cross section of distribution grid 5.2%, fluidized material: cationite ($d = 0.8 \cdot 10^{-3}$ m, $\rho = 650$ kg/m³), silica gel ($d = 1.2 \cdot 10^{-3}$ m, $\rho = 750$ kg/m³). We varied the gas (air) velocity and the type of organizing device, the characteristics of which are presented in Table 1. The bed was calibrated under working conditions. Problems involving the statistical reliability of the results obtained, verifying the ergodicity of the process, estimating the minimum observation time, etc. are discussed in [8] for a freely boiling bed. We adhered to the methods suggested there.

Before finding estimates of the statistical characteristics of the random process of particle motion it is useful to make a qualitative analysis of the trajectories. Typical synchronously recorded sections of the trajectories of motion of the marked particle in an OFB with a quantization time of 0.3 sec are presented in Fig. 1. One can conclude that the marked particle moves through the entire volume of the bed, the zones of its ascent and descent not being fixed. The intensity of particle motion in an OFB is considerably lower than that in a free bed, and in the trajectories of a particle in an OFB one notes sections of several seconds duration when the particle undergoes oscillatory motions within the confines of a small section of the bed on the order of the dimensions of the insert. Such sections alternate with sections of directed motion and are evidently explained by a short-lived aggregation of particles. Vertical movements can reach the boundaries of

TABLE 1. Geometrical Characteristics of Inserts

Insert No.	Type	Diameter of element, mm	Height, mm	Wire diam. or wall thickness, mm
1	Pall rings	30	30	0,5
2	Double wire spirals	25	40	2,0
3	Double wire spirals	20	20	1,0
4	Double wire spirals	12	15	1,0

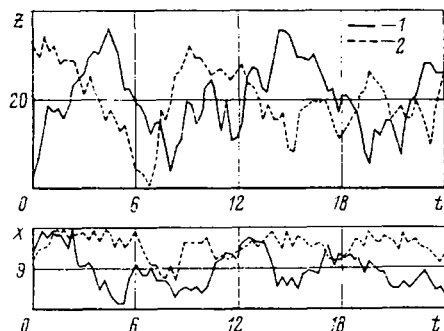


Fig. 1

Fig. 1. Trajectories of motion of a marked particle in the vertical (z , cm) and horizontal (x , cm) directions in an OFB. $U_g = 1.17$ m/sec; 1) insert No. 2; 2) No. 3.

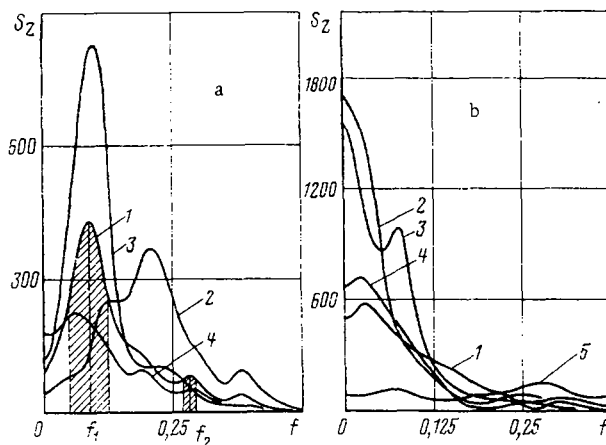


Fig. 2

Fig. 2. Spectral-density functions S_z ($\text{cm}^2 \cdot \text{sec}$) for the random process of particle motion in the vertical direction: a) $U_g = 1.05$ m/sec; b) 1.42 m/sec; 1, 2, 3, 4) inserts Nos. 1, 2, 3, and 4, respectively; 5) free bed, $U_g = 0.7$ m/sec.

the bed, whereas large horizontal displacements are very rare. The velocities on the sections of ascending motion can exceed the velocities on the sections of descending motion by two to three times. At high gas velocities (near the carry-off velocity) the particle motion slows with an increase in gas velocity. The form and dimensions of the insert affect the character of the particle motion. For example, sections of directed motion are encountered far more rarely for inserts Nos. 1 and 4 than for inserts Nos. 2 and 3.

Spectral-Correlation Analysis

A spectral-correlation analysis of the trajectories of a marked particle was carried out to investigate the mechanism of particle motion in an OFB. Such an approach was employed earlier in [12, 13], but the question of the error of the resulting statistical characteristics was not treated, as a rule. An analysis of the interference signal, conducted for its subsequent filtration, showed that it is a steady uncorrelated random process with a nearly normal distribution law. The autocorrelation function of the random process of motion of the marked particle, determined from the recorded particle coordinates with allowance for the autocorrelation function of the interference signal, approaches zero with an increase in the time shift. Hence this process is a steady ergodic one, which can be considered as an argument in favor of the applicability of the ergodic hypothesis of statistical physics to OFB.

Hemming's estimate [14] was used to calculate the estimates of the spectral density. A quantization time of 0.1 sec for the trajectories made it possible to analyze spectra with an upper limiting frequency of 5 Hz. The equivalent frequency band and the relative root-mean-square error of the estimates were 0.0625 and 0.23 Hz, respectively. Typical graphs of the spectral densities of the random process of motion of the marked particle are presented in Fig. 2.

Most of the energy of particle movement in an OFB falls in the region of low frequencies up to 0.2 Hz. A peak, which shifts into the lower-frequency region with an increase in gas velocity, down to the appearance of a maximum at the zero frequency, is clearly expressed on the spectra in this region, which may be explained

TABLE 2. Parameters of Circulation Model

Insert No.	Gas velocity, m/sec	$U, m/sec$	α	β, sec^{-1}
1	1,05	0,095	2,24	1,65
	1,05	0,072	1,66	1,75
2	1,25	0,045	1,8	0,62
	1,35	0,045	1,43	1,12
	0,53	0,12	1,15	1,57
Free bed	0,7	0,232	1,26	3,7

by the random phase distribution of the large-scale movements. A more uniform frequency distribution of the energy of particle movement was observed in the free fluidized bed in the range up to 1 Hz (in Fig. 2b, curve 5 up to 0.375 Hz).

The frequency region of 0.2-0.5 Hz, where there is a maximum, as a rule, is distinguished in all the spectra. Some part of the energy of the random process is distributed in the form of "white noise." This spectral form reflects the complicated character of the particle's motion and its participation in oscillatory processes with different amplitudes and frequencies.

The spatial scales of these processes can be estimated in the following way. The hatched areas in the vicinities of the frequencies f_1 and f_2 (Fig. 2a) are approximately equal to the dispersions of the amplitudes of the respective harmonics. The square roots of the dispersions (the standard deviations of the respective amplitudes from zero mathematical expectations) are 55 and 14 mm. The first of these quantities allows one to find the scale of the low-frequency circulation movements, which are comparable with the height of the expanded bed in order of magnitude. The second determines the scale of the collective particle interaction, approximately equal to the dimensions of the insert. It is interesting to note that the first quantity grows with an increase in the gas velocity (and the degree of expansion of the bed) while the second remains almost unchanged. Moreover, as shown by a calculation of the spectral densities of the random process of particle motion in the horizontal direction, there is also a maximum in the frequency region of 0.15-0.4 Hz.

The following conclusions about the main forms of particle motion in OFB can be drawn on the basis of the spectral densities and trajectories. The low-frequency region of the spectrum corresponds to directed large-scale (usually periodic) particle movements. They are the result of the action of forces on the part of the fluidizing stream. The rise of particles in the hydrodynamic wake of a bubble is compensated for by their descending flow, since circulating particle motion occurs, which is clearly seen in the trajectories of the motion. The horizontal low-frequency movements of a particle, in contrast to the vertical ones, are nonperiodic, while the type of insert and the gas velocity exert a weak influence on the character of the spectrum.

The oscillatory processes with low amplitudes are represented on the spectra by the frequency region of 0.2-0.5 Hz and are evidently due to the interaction of the marked particle with aggregates of particles. The reason for the occurrence of this form of motion consists in the following. In a free FB particle aggregates can move without hindrance through the entire volume of the apparatus. An organizing device prevents the growth and motion of aggregates and promotes the development of localized ordered particle motions, which prove to be stable for short time intervals under the action of hydrodynamic forces. This is indicated by the rough equality of the characteristic frequencies of variation of the vertical and horizontal coordinates of a particle in the region under consideration. The peaks in the OFB spectra at these frequencies reflect the fact that some of the energy of collective oscillations of particles along the height of the bed is scattered by the insert in a direction transverse to the stream.

The collision of particles with each other and with elements of the insert, the interaction of the boundary layers surrounding the particles, etc. give rise to their oscillating motion with very low amplitudes.

The difference between the characteristic times of the two main forms of particle motion in an OFB (circulating motion and aggregation in an ensemble) permits a sounder approach to the description of the nonsteady and thermal fields in reactors containing OFB. Thus, for "fast" processes of mass transfer it is sufficient to allow only for the particle motion occurring at the ensemble level [15] while large-scale motions can be ignored, since the temperature inside an ensemble is not able to vary noticeably during its existence. The large-scale particle motion determines the dynamics of the thermal fields in a reactor, since the characteristic time of variation of the temperature of an ensemble exceeds its mean lifetime by two to three orders of magnitude. The model for this motion, the circulation model, for example, will be the basis for the temperature-field model.

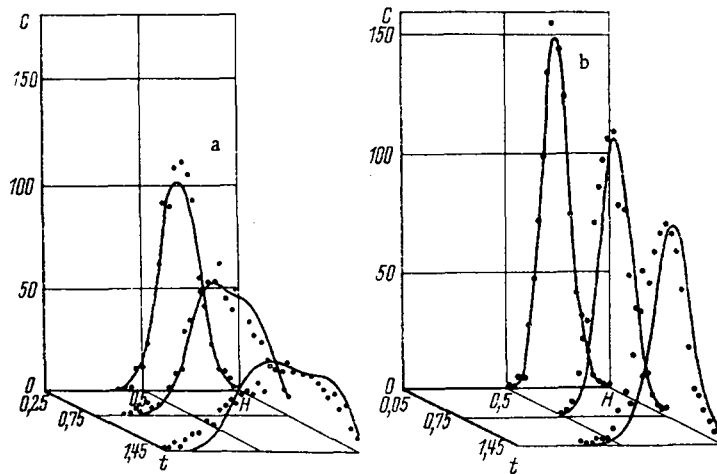


Fig. 3. Density functions C (arb. units) of distribution of marked particles over bed height $H = l/L$ (dimensionless) at different times t , sec, experimental and calculated from the circulation model: $U_g = 1.25$ m/sec; a) insert No. 2, $L = 0.38$ m; b) No. 4, $L = 0.44$ m.

Usually the particle motion in an FB is described by a diffusional model of the parabolic type (Brownian motion) or the hyperbolic type, a diffusional model with a convective flow of particles ("vectorized" Brownian motion), or by a circulation model of opposing streams. The method of observing a marked particle allows one to compare the experimental results with model concepts and to determine the parameters of the models. For this we developed a method of constructing distribution density functions of the particles with respect to the coordinates and time at a fixed gas velocity.

Particle Motion

The essence of the method consists in making a time recording of the coordinates of marked particles initially concentrated in some cross section along the height of the bed using the CCS, and then histograms of the distribution of particles over the height of the bed at different times relative to the starting point are constructed. Since the random process of particle motion is a steady ergodic process, the averaging over individual sections of trajectories having an origin in the same cross section of the bed can be treated as averaging over a set of statistically independent realizations.

The organization of the introduction of a "batch" of marked particles was performed using a system of lateral detectors screened by lead plates from the entire fluidized bed except for a narrow horizontal slit about 3 mm wide. The fact of the arrival of the marked particle at the assigned cross section of the bed was recorded sufficiently accurately by the side detectors, after which the recording by the upper and lower detectors of the vertical coordinates of the particle began, starting from the time of its arrival at the given cross section until the lapse of a time interval chosen in advance. Then the time the particle crossed the slit was again recorded and the entire procedure was repeated. Crossing the slit by the particle N times corresponds to the introduction of a batch of N marked particles. It takes about 10–15 h to conduct experiments with a "batch" of 1000 particles, which is due to the physical properties of the OFB investigated. After the accumulation of the necessary amount of information histograms of the distribution of "marked" particles with respect to the coordinates and time were constructed.

Examples of distribution-density functions of marked particles, complicated by random interference, are presented in Fig. 3. In free FB and FB organized by large inserts (Nos. 1 and 2) one can distinguish two maxima, which gradually shift from the point of insertion of the particle "batch" to the edges of the bed and disappear with time. In an OFB containing a relatively small insert (No. 4) the distribution-density functions are nearly unimodal. Bimodality of the distribution-density functions indicates a correlation of the velocities of particle motion, with the correlation being the stronger, the more clearly expressed the maxima.

Determination of Parameters of Mathematical Models

The best agreement was obtained in a comparison of the experimental particle distribution-density functions with those calculated from the circulation model. In the case of the OFB with insert No. 4 the experiments

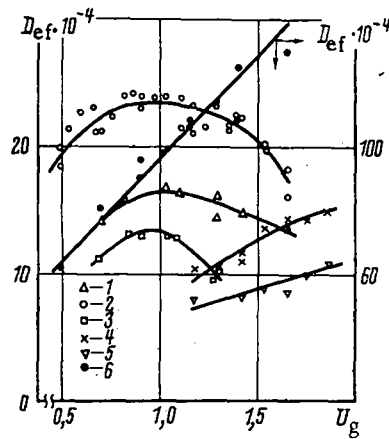


Fig. 4. Dependence of effective coefficient of diffusion D_{ef} (m^2/sec) of particles in the vertical direction on the gas velocity: 1, 2, 3, 6) cationite particles with inserts Nos. 1, 2, and 4 and a free FB, respectively; 4, 5) silica gel particles with inserts Nos. 2 and 3.

are also satisfactorily described by a diffusional model of the parabolic type. A number of the experiments were investigated with the help of a diffusional equation of the hyperbolic type [16] (inserts Nos. 1, 2, and 4).

The circulation model was used in the form

$$U \frac{\partial C_1}{\partial t} + \frac{\partial C_1}{\partial t} + \beta (C_1 - C_2) = 0, \quad (1)$$

$$U \frac{\partial C_2}{\partial t} - \alpha \frac{\partial C_2}{\partial t} + \beta (C_1 - C_2) = 0 \quad (2)$$

with the boundary conditions $l = 0, L; C_1 = C_2$. Here U is the velocity of the ascending particle motion; α is the ratio of velocities of the ascending and descending motions; β is the coefficient of particle exchange between streams; C_1 and C_2 are the concentrations of "marked" particles in the ascending and descending streams.

The particle distribution densities were calculated from (1) and (2) with allowance for the dispersion of the interference, which was determined in independent experiments. The parameters of the circulation model were determined by minimizing the sum of the squares of the deviations of the experimental points from functions with unit weights calculated for six times. The velocities of the ascending and descending motions of a particle estimated from its trajectory were taken as the initial approximations. The parameters of the model are presented in Table 2. In [2] these parameters were found using the method of an instantaneous heat source.

It is known that the diffusional approximation is valid over time intervals longer than the correlation time of the particle velocity. The sizes of these intervals depend on the type of insert in the OFB, the gas velocity, and the characteristics of the fluidized material. The diffusional model of particle motion can be used in modeling steady processes with small temperature gradients over the bed. In this case the particles provide sufficiently intense heat transfer in the FB and the detailed mechanism of their motion is not important.

We made a direct calculation of the rms displacement of a particle along its trajectory with a subsequent determination of the coefficient of diffusion of the particle from the Einstein equation. It was preliminarily established that the dispersion of the recorded signal can be represented by the sum of the dispersions of the useful signal (the coordinates of the particle) and the interference:

$$\overline{\Delta z^2} = a \cdot \overline{\Delta h^2} + \overline{\Delta \varepsilon^2}. \quad (3)$$

Here $z(t)$ is the recorded signal; $h(t)$ are the coordinates of the particle; $\varepsilon(t)$ is the interference signal; a is some coefficient. It was established that the dispersion of the interference does not depend on the time interval Δt and the error in its determination does not exceed 16%.

We constructed the dependences of $\overline{\Delta z^2}$ on Δt for each hydrodynamic mode, and then from the tangent of the slope of the initial straight section we calculated the mean value of the effective coefficient of diffusion of the particles. At large Δt the rms displacement of a particle decreases owing to the influence of the boundaries of the bed, so that the Einstein equation cannot be applied.

The dependence of the effective coefficient of particle diffusion on the velocity of the gas stream for a free bed and a bed organized by inserts Nos. 1-4 is presented in Fig. 4. This dependence is monotonic in the first case and extremal in the second. A decrease in the characteristic dimensions of the insert and an increase

in the diameter and density of the particles of fluidized material lead to a decrease in the effective coefficient of diffusion of the particles.

The extremal character of the dependence of the coefficient of diffusion of particles in the vertical direction on the gas velocity in OFB can be explained by the fact that at high linear velocities of the gas the latter is distributed more evenly than at low velocities. The decrease in inhomogeneities leads to the fact that the particles participate predominantly in oscillatory motions with low amplitudes and, as a consequence, their rms displacement decreases. In a free FB an increase in the linear velocity of the gas leads to an increase in the inhomogeneities, which promote the large-scale transport motion of particles, as a result of which their rms displacement increases. It should be noted that in experiments with an instantaneous heat source [2] they noted a monotonic character of the dependence of the effective thermal diffusivity of the bed on the gas velocity, whereas a maximum was observed when the effective longitudinal thermal conductivity was determined [4, 17].

Calculations of the effective coefficient of particle diffusion in the horizontal direction showed that it hardly depends on the gas velocity and the type of insert and equals $1.5 \cdot 10^{-4}$ – $3 \cdot 10^{-4}$ m²/sec under the test conditions.

NOTATION

z and x , vertical and horizontal coordinates of marked particle, cm; t , time, sec; S_z , spectral density of random process $z(t)$, cm²·sec; f , frequency, Hz; U_g , linear gas velocity, m/sec; d , mean diameter of fluidized particles, m; ρ , bulk density of particles, kg/m³; l , coordinate along bed, m; L , bed height, m; D_{ef} , effective coefficient of particle diffusion, m²/sec.

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