DESCRIPTION OF SEGREGATION IN A HORIZONTAL DRUM MIXER BY USE OF THE DIFFUSION EQUATION*

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An attempt has been made to describe the axial segregation of solid particles of two dimensions in a horizontal drum mixer. For this purpose the Kolmogorov's forward diffusion equation with variable diffusion coefficient and zero drift velocity was used. For the case of "pure" segregation this approach has given good results.

At mixing of the solid phase in a rotating horizontal drum mixer individual particles are in mutual motion both in axial and radial directions. If properties of these particles do not differ significantly their motion in space and time can be described satisfactorily by use of the Fick's law

$$\frac{\partial c}{\partial t} = D \nabla c \tag{1}$$

with the constant diffusion coefficient D, while c denotes concentration of particles, indicated e.g. by a paint[1,2]. In agreement with this equation after sufficiently long time of mixing (i.e. practically in the stationary state) uniform particle distribution in the bulk of the mixer is reached.

On the contrary in systems in which the particles differ in their size or density and shape segregation of individual types takes place. This phenomena has been studied experimentally in detail[3−9], where first of all Donald and Roseman[4,5] have qualitatively explained in a satisfactory way why it takes place. Attempts have also been made to describe segregation by use of model relations[8,10−12].

Fan and Shin[8] have used for description of the segregation phenomena in the axial direction the Kolmogorov's forward diffusion equation (see e.g.[13])

$$\frac{\partial x}{\partial t} + \frac{\partial}{\partial z}\left[V(z)\,x\right] - \frac{\partial^2}{\partial z^2}\left[D(z)\,x\right] = 0 \tag{2}$$

for spherical particles of two dimensions, while by symbol x is denoted concentration (mass fraction) of smaller particles and by symbol V drift velocity of these particles. The authors have made an attempt to verify experimentally this relation so that at the beginning they estimated the shape of functions $V(z)$ and $D(z)$ as two first infinitesimal moments of transitive probabilities describing motion of particles in the axial direction. Numerical solution of Eq. (2) then gave results which were in satisfactory agreement with the made experiments.

The authors have also pointed to the fact that the classical type of Fick's law with variable diffusion coefficient (see e.g.[14])

$$\frac{\partial x}{\partial t} = \frac{\partial}{\partial z}\left[D(z)\frac{\partial x}{\partial z}\right] \tag{3}$$

does not enable to describe the segregation phenomena as solution of this equation in the stationary state leads to the uniform spacial particle distribution.

In this study the identical problem is studied, which differs first of all − as will be demonstrated in discussion − from the above quoted paper by the method of determination of coefficients of the diffusion equation.

THEORETICAL

Motion of individual solid particle in a rotating drum mixer can be generally considered to be random and continuous both in time and space. If it is assumed that it is determined only by initial conditions and is independent of intermediate states, motion of such a particle can be considered to be the diffusional Markov process[13]. If we consider the description of its kinematics as sufficiently accurate, motion of an individual particle in the three-dimensional space can be generally characterised by the Kolmogorov's forward differential equation

$$\frac{\partial f_3}{\partial t} + \nabla \cdot \left[\mathbf{V}(\mathbf{x}, t) f_3 \right] - (\nabla \circ \nabla) : \left[\mathbf{D}(\mathbf{x}, t) f_3 \right] = 0 , \tag{4}$$

in which $\mathbf{V}$ denotes the vector of drift velocity, $\mathbf{D}$ tensor of second order, describing the relative motion of the considered particle in the mixture.
The function

$$f_3 = f_3(\mathbf{x}; t | \mathbf{x}_0; t_0) \quad [t > t_0] \tag{5}$$

is the three-dimensional transitive probability density, that − roughly speaking − the particle will be located in the moment t in close vicinity of the point with the radius-vector $\mathbf{x}$ at the condition, that in some preceding moment t_0 it was in the point $\mathbf{x}_0$.

Let us state concrete conditions at which Eq. (4) will be applied and also assumptions which will lead to its simplification. Discussion of these assumptions will be also given in the discussion of results of this study.

Let us consider that mixing of solid particles takes place in a closed rotating drum mixer of cylindrical shape with radius R and length $2L$. The axis of its cylindrical symmetry is oriented strictly horizontally, so that gravitation does not affect motion of solid particles in the axial direction. The speed of rotation of the mixer is constant. The walls of the mixer could not be penetrated by solid particles and adhesivity of particles to the wall does not exist. The mixer is filled by solid particles of constant density and of two characteristic dimensions; their dimensions and number do not change significantly during the process.

From this summary first of all results the form of the boundary condition of Eq. (4): flow of particles in each point of the mixer surface is equal to zero — as concerns the terminology of random processes the surface is a reflexing boundary, i.e.

$$\left[\mathbf{V}(\mathbf{x}_S, t) f_3 - \nabla \cdot \mathbf{D}(\mathbf{x}_S, t) f_3 \right] \cdot \mathrm{d}\mathbf{S} = 0 , \quad \left[\mathbf{x}_S \in \mathbf{S} \right] \tag{6}$$

where $\mathrm{d}\mathbf{S}$ denotes an oriented element of the mixer surface.

It would be suitable to write Eqs (4) and (6) in cylindrical coordinates. But in the next part the process will be studied only in the axial direction, i.e. in the direction of horizontal coordinate z. The terms in these equations will be averaged with respect to the angular and radial coordinates φ and r. Probability density in the first term will take the form

$$f = f(z; t|z_0; t_0) = \int_0^R \int_0^{2\pi} \int_0^R \int_0^{2\pi} f_3(r, \varphi, z; t|r_0, \varphi_0, z_0; t_0) f_{30} \, \mathrm{d}\varphi_0 r_0 \, \mathrm{d}r_0 \, \mathrm{d}\varphi r \, \mathrm{d}r .$$

$$\tag{7}$$

At averaging of other terms it is possible to interchange the volume integrals for surface ones on the cylindrical part of the mixer surface. The terms denoting reflexivity on this part of the surface are with regard to Eq. (6) equal to zero; integration of other terms makes possible to define coefficients of a unidimensional equation:

$$V(z, t) = \int_H V_z(\mathbf{x}, t) f_3(\mathbf{x}; t|\mathbf{x}_0, t_0) f_{30} \, \mathrm{d}H / f(z; t|z_0; t_0) , \tag{8}$$

the integral in this relation over the region H is a simplifed expression of the quadruple integral in the form (7). $V_z(\mathbf{x}, t)$ denotes the axial component of drift velocity, so that the term on the left hand side of the equation is a scalar quantity. Similarly it would be possible to define the diffusion coefficient of unidimensional diffusion

$D(z, t)$ in the axial direction and Eq. (4) then takes the form

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial z}\left[V(z, t)f\right] - \frac{\partial^2}{\partial z^2}\left[D(z, t)f\right] = 0 \,. \tag{9}$$

This equation enables — in the case that the form of coefficients V and D is apriori known — to find the complete probability characteristics of random motion of the solid particle in axial direction.

Further on we will assume that the number of particles of larger dimensions is sufficiently large and that the probability density can be considered to be proportional to the concentration of these particles expressed in mass fractions. Then the relation holds:

$$\frac{\partial x_i}{\partial t} + \frac{\partial}{\partial z}\left[V(z, t)\,x_i\right] - \frac{\partial^2}{\partial z^2}\left[D(z, t)\,x_i\right] = 0 \,, \tag{10}$$

where

$$x_i = x_i(z, t) = \int_{-L}^{+L} f(z; t|z_0; t_0)\,x_i(z_0, t_0)\,dz_0 \quad \left[t > t_0\right] \tag{11}$$

denotes the mass fraction of particles of larger dimensions (concentration of larger component) averaged in the given point of the axial coordinate with respect to radial and angular coordinate. To Eq. (6) correspond in the single-dimensional case the boundary conditions on both ends of the mixer

$$\lim_{z \to \pm L}\left[V(z, t)\,x_i - \frac{\partial}{\partial z}\left(D(z, t)\,x_i\right)\right] = 0 \,. \tag{12}$$

Finally let us made assumptions on coefficients in Eq. (10). First of all it will be assumed that in a closed mixer the effect of drift velocity of the solid phase is negligible in comparison with the effect of diffusion coefficient. According to this assumption, it holds

$$V(z, t) \equiv 0 \,. \tag{13}$$

For the diffusion coefficient the following cases will be considered:

a) For "pure" segregation, i.e. for the case of initial uniform distribution of components in the bulk of the mixer the diffusion coefficient depends only on the length coordinate, in the way that it is the even function of this coordinate with respect to the centre of the mixer. At these assumptions, it results from Eqs (10) to (13)

$$\frac{\partial x_i}{\partial t} = \frac{\partial^2\left[D(z)\,x_i\right]}{\partial z^2} \quad \left[D(z) = D(-z)\right] \tag{14}$$

with the initial and boundary conditions

$$\lim_{z \to \pm L} \frac{\partial(Dx_i)}{\partial z} = 0 \; ; \quad \lim_{t \to 0} x_i(z, t) = x_{i0} = \text{const.} \tag{15}$$

At these conditions Eq. (14) has the solution which is also the even function with respect to the centre of the mixer and which has for the stationary case the form

$$\lim_{t \to \infty} x_i(z, t) = x_{is}(z) = \alpha/D(z) , \tag{16}$$

where α is the integration constant.

b) In the case of "pure" mixing the diffusion coefficient is constant $D_m \neq D_m(z)$. This type of relocation of the solid phase can exist only for the case of homogeneous particles differing only by their colour. Instead of relations (14) and (15) for this case the relations are obtained

$$\frac{\partial x}{\partial t} = D_m \frac{\partial^2 x}{\partial z^2} \; ; \quad \lim_{z \to \pm L} \frac{\partial x}{\partial z} = 0 \; ; \quad \lim_{t \to 0} = x_0(z) \tag{17}$$

for which even the analytical solution[2] can be found (see also e.g.[15]) while for the stationary state holds $x(z, \infty) = \text{const.}$

c) In the case of real process in the horizontal mixer, superposition of these two phenomena takes place. An attempt has been made to approximate this situation by a model, where the asymmetric concentration profile in the charge is affected by mixing. Thus first of all the term asymmetry indicator is introduced which is the difference of average concentrations in the left and right parts of the mixer, given by

$$I(t) = \frac{1}{L} \left| \int_{+\varepsilon}^{+L} x_i(z, t) \, dz - \int_{-L}^{-\varepsilon} x_i(z, t) \, dz \right| , \tag{18}$$

where ε is an arbitrary small positive number.

It is obvious that the asymmetry indicator can reach values within the range $0 \leq I(t) \leq 1$. Next it is assumed that in a real mixer the diffusion coefficient characterising the mixing is directly proportional to the asymmetry indicator; the effect of diffusion coefficient characterising segregation is directly proportional to the difference of this indicator to one. Action of both these coefficients is aditive, i.e.

$$D_m = AI(t) \; ; \quad D(z) = B(1 - I(t))/x_{is}(z) \tag{19}$$

second of these relation results with regard to Eq. (16). Both A and B are constants.

The resulting diffusion equation then becomes

$$\frac{\partial x_i}{\partial t} = \frac{\partial^2}{\partial z^2} \{[AI(t) + B(1 - I(t))/x_{is}(z)] x_i\} \tag{20}$$

with the initial and boundary conditions

$$\lim_{z \to \pm L} \frac{\partial}{\partial z} \{[AI(t) + B(1 - I(t))/x_{is}(z)] x_i\} = 0 ; \quad \lim_{t \to 0} x_i = x_{i0}(z) \tag{21}$$

The results obtained after numerical integration of these relations were compared with the experimental data.

RESULTS

The experiments were performed in a horizontal drum mixer 0·12 m long, with internal diameter 0·074 m. Speed of rotation of the mixer was 70 rpm. Two fractions of the mixing material — sea sand of diameter $0·2 - 0·25 \cdot 10^{-3}$ m and 0·385 to $0·43 \cdot 10^{-3}$ m — were used. Description of the experimental unit, sampling method and evaluation of results were already published[9]. Two experiments were performed: at the beginning of the first one the concentration of both fractions was uniformly distributed along the mixer length. At the beginning of the second one the fractions were completely segregated. At the end of the experiments, i.e. after reaching the stationary state the final particle distribution along the mixer was in both cases roughly equal, as follows from the results published in the above quoted study.

These results were compared with the proposed model with dimensionless variables introduced in Eqs (20) and (21) by relations

$$y = z/L; \quad \tau = tB/L^2 ; \quad \gamma = A/B . \tag{22}$$

The stationary distribution of concentration $x_{is}(y)$ was determined by averaging the experimental data in the stationary state, where the conditions of symmetry were taken into account

$$x_{is}(y) = x_{is}(-y) = \sum_{p=1}^{2} [x_{is}^p(|y|) + x_{is}^p(|-y|)]/4 , \quad [-1 \leqq y \leqq 1], \tag{23}$$

where p denotes measurements based on the mixed ($p = 1$) and segregated ($p = 2$) states. Comparison of this interpolation with the experimental results could be seen from Fig. 1.

As it was not possible to take samples closely at the wall of the mixer and we have not considered as possible to extrapolate with sufficient accuracy the concentration of particles closely to the wall in the stationary state, the boundary condition (21) was considered to be valid in the last sampling points. Thus the value of L was considered as equal to 0·055 m. The differential equation (20) with the boundary condition (21) thus has, after transformation the form

$$\frac{\partial x_i}{\partial \tau} = \frac{\partial^2}{\partial y^2} \left\{ \left[\gamma I(\tau) - (1 - I(\tau))/x_{is}(y) \right] x_i \right\} \tag{24a}$$

$$\lim_{y \to \pm 1} \frac{\partial}{\partial y} \left\{ \left[\gamma I(\tau) - (1 - I(\tau))/x_{is}(y) \right] x_i \right\} = 0 . \tag{24b}$$

Initial conditions were determined from the averaged initial experimental data, i.e. in the case of initial uniform mixing from

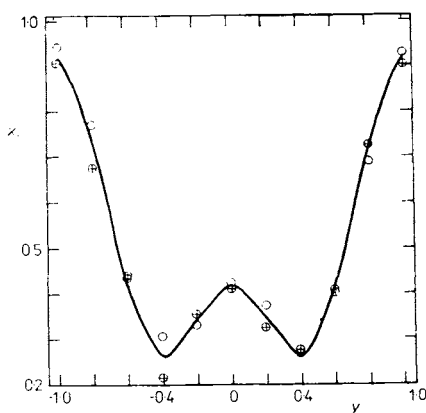$$\bar{x}_{10}^p = \frac{1}{2} \int_{-1}^{+1} x_{10}^p(y) \, dy = 0.522 \quad [p = 1] \tag{25}$$

FIG. 1

Stationary concentration of the larger component in dependence on axial coordinate; –– averaged according to Eq. (23), ○ experimental data reached from the initial uniform mixing ($p = 1$), ⊕ experimental data reached from the initial segregation ($p = 2$)
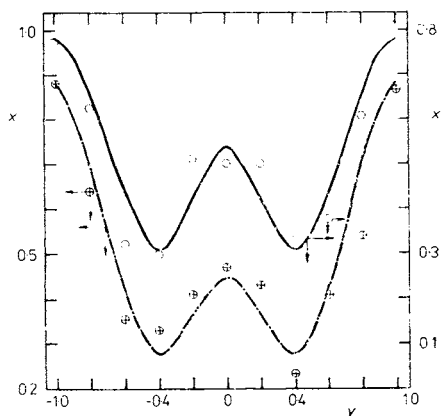
FIG. 2

Concentration of larger component in dependence on axial coordinate for the initial uniform mixing; $t = 4$ min: — result of numerical solution, ○ experimental data, $t = 10$ min: –·–·– result of numerical solution, ⊙ experimental data

and in the case of initial segregation, from

$$
\bar{x}_{io}^{p}(y) \begin{cases} = \int_{+\varepsilon}^{+1} x_{io}^{p}(y)\,\mathrm{d}y = 0{\cdot}064 & [y > 0] \\[2mm] = \quad x_{io}^{p}(0) \quad = 0{\cdot}522 & [y = 0] \\[2mm] = \int_{-1}^{-\varepsilon} x_{io}^{p}(y)\,\mathrm{d}y = 0{\cdot}98 & [y < 0] \end{cases} \Bigg\} \quad [p = 2] \qquad (26)
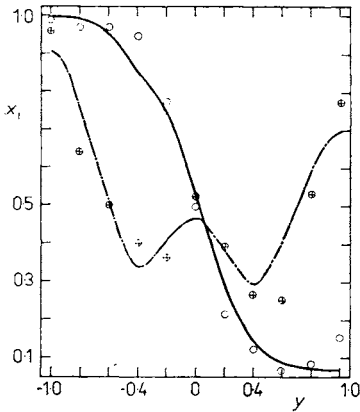$$

Fig. 3

Concentration of the larger component in dependence on axial coordinate for the case of initial segregation; $t = 4$ min: — result of numerical solution, ○ experimental data, $t = 120$ min: —·—·— result of numerical solution, ⊕ experimental data
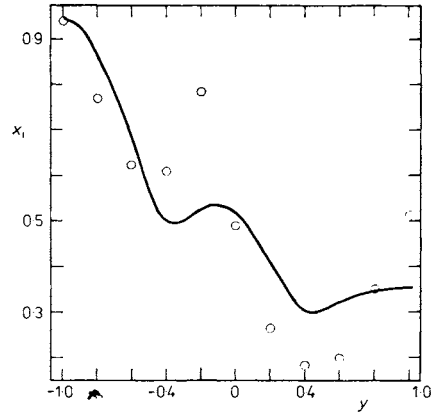

Fig. 4

Concentration of larger component in dependence on axial coordinate for the case of initial segregation; $t = 40$ min: — result of numerical solution, ○ experimental data
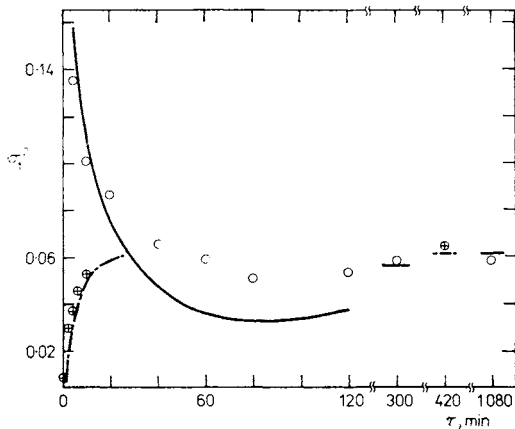

Fig. 5

Time dependence of concentration variance of the larger component with respect to the mixer length (relations (27)); ——— from numerical solution of the diffusion equation $(p = 2)$, ○ from experimental data $(p = 2)$, —·—·— from numerical solution of the diffusion equation $(p = 1)$, ⊕ from experimental data $(p = 1)$

Due to experimental inaccuracy the averaged concentrations at the beginning of the exeriment are not exactly equal to the averaged values at stationary state. These inaccuracies were thus corrected during the calculation.

In case of the first initial condition (25) the initial value of the asymmetry indicator, defined by relation (18), equals zero and the solution of differential equation (24) is then symmetrical with respect to the centre of the mixer in every next moment, which means that also the asymmetry indicator then becomes equal to zero. In the second case its value is decreasing with time.

The proposed differential equation was solved by the numerically simple explicite method, where the coefficients B and γ were determined by the method of nonlinear regression (see e.g.[16]) from the experiments measured earlier[9]. (For the initial uniform concentration distribution, the concentrations of the larger component were available, measured in time intervals 2, 4, 6, and 10 min from the beginning of the experiment and in time intervals 4, 10, 20, 40, 60, 90, 120, and 300 min after initial segregation). It was found that the coefficient B for the case of initial mixing ($p = 1$) was equal to $2\cdot47 \cdot 10^{-7}\,\mathrm{m^2\,s^{-1}}$, and for the initial segregation ($p = 2$) the values were obtained: $B = 5\cdot48 \cdot 10^{-8}\,\mathrm{m^2\,s^{-1}}$ and $\gamma = 4\cdot39$. The results of calculations and comparison with the experimental data are given in Figs 2 to 4.

The variance of concentrations with respect to the mixer length was also calculated according to relations

$$\sigma_t^2 = \int_{-1}^{+1} \left[x_i(y, tB/L^2) - \bar{x}_i \right]^2 \mathrm{d}y/2 \,; \quad \bar{x}_i = \int_{-1}^{+1} x_i(y, tB/L^2)\,\mathrm{d}y/2 \qquad (27)$$

and was compared with the variance calculated from experimental data[9] (see Fig. 5).

DISCUSSION

The assumption on the Markov property of the considered process can be considered as plausible in case that agglomerates of the solid phase do not form. Formation of such structures on a larger scale and first of all their relative stability with time would be obviously the cause of "memory" of the system. With sieved and dried sea sand used in the performed experiments such agglomerates did not form to an observable extent.

Simplification of the three-dimensional process to a unidimensional one is for the given diffusion model correct. But the transitive probability density f_3 is not symmetric with respect to the radial coordinate, due to gravitation and segregation phenomena. The corresponding experiment would thus require measurement of concentration of the larger component in a large number of measuring points at the given axial coordinate. In our experiments this requirement was approximated by the analysis of samples in three randomly selected points for 10 equidistant values

of axial coordinates and in 10 points for the centre of axial symmetry $(y = 0)$. The considered procedure can be the cause for a certain (even though random) deviation of experimental data from the values averaged with respect to the radial and angular coordinate.

At description of mixing of the solid phase it is usual to express concentration of components in the form of mass fractions for the relative simplicity with which this quantity could be determined. This method was also used in this study. But it is necessary to mention that the quantity directly proportional to the probability density for the position of the studied component is e.g. the partial density ϱ_i of this component in the given point and considered moment, i.e.

$$\varrho_c f_3(\mathbf{x}, t) = \varrho_i(\mathbf{x}, t) \approx \frac{\varrho_c n_i(t)}{\Delta U} , \tag{28}$$

where ϱ_c is density of the solid phase. Left-hand side term can be considered to be the definition, the approximate relation holds for the volume ΔU of finite dimensions with the centre in the point $\mathbf{x}$ in which in the time moment t are just located n_i particles of the component.

Function f_3 denotes three-dimensional probability density, "unidimensional" partial density of the component can be approximated by the relation

$$\varrho_i(z, t) \approx \varrho_c \frac{n_i(t)}{\pi R^2 \Delta z} . \tag{29}$$

From these considerations it is obvious that the mass fraction is approximately directly proportional to the corresponding probability density only in the case of lower concentrations of the component.

Next discussion concerning the assumptions made on coefficients of the diffusion equation (10) is related to considerations of analogous assumptions made in the study by Fan and Shin[8].

The drift velocity is in the mathematical theory of diffusion processes (see e.g.[13]) defined as the time derivative of the expected value of a small random change of position in our case in the axial direction, i.e.

$$V(z, t) = \lim_{\Delta t \to 0} \frac{1}{\Delta t} E[Z(t + \Delta t) - z(t)] , \tag{30}$$

where $Z - z$ denotes random shift of particle, which in the moment t was located in the point z.

In chemical engineering (see e.g.[14]), this expected value is approximated by use of the sum of momentum of all particles of the mixture, which in the moment t are in close vicinity of the point z, i.e.

$$V(z, t) \approx \sum_k [m_k V_k(t)] / \sum_k m_k , \qquad (31)$$

(see Eq. (29)). Such definition enables to write the total material balance of the solid phase in the form

$$\frac{\partial \varrho}{\partial t} + \frac{\partial}{\partial z} [V(z, t) \varrho] = 0 , \qquad (32)$$

where $\varrho = \varrho_i + \varrho_j = \varrho(z, t)$ is the mass concentration of the solid phase in the infinitesimal volume as equal to the sum of partial densities of the larger and smaller components which in our case form the mixture. It is obvious that also in the case of particles of two different dimensions which are otherwise equal, this concentration will in general depend on the mutual local ratio of particles in the mixture and thus it will be a function of the axial coordinate and time. The term for the drift velocity then results from integration and arrangements of Eq. (32) which gives

$$V(z, t) = \int_{-L}^{z} \frac{\partial \varrho(z', t)}{\partial t} \, dz' / \varrho(z, t) \qquad (33)$$

on the basis of which it would be possible to estimate experimentally these functions by use of the bulk density of solid phase in dependence on concentration of components. But the axial motion of the smaller and larger components at mixing in the horizontal mixer takes place in the opposite direction (see e.g.[4]) so that it is possible to assume that decrease of mass of one component in the volume $\pi R^2 \Delta z$ is approximately compensated by the increase in mass of the second component and thus ϱ is not changing significantly. Derivatives in Eq. (33) are then small and the drift velocity $V(z, t)$ does not become significant.

As long as we accept that description of concentration changes of components by use of mass fractions is sufficiently accurate, the assumption on the zero velocity drift is satisfied automatically as the sum of concentrations of both components is equal to one.

A noticable attempt for the experimental estimate of the drift velocity on basis of the probability definiton[17]

$$V(z, t) = \lim_{\Delta t \to 0} \frac{1}{\Delta t} \int_{\delta < |z' - z|} (z' - z) f(z', t + \Delta t | z, t) \, dz' ; \quad \lim_{\Delta t \to 0} \frac{1}{\Delta t} \int_{\delta \geqq |z' - z|} f \, dz' = 0 \qquad (34)$$

proposed in the study by Fan and Shin[8] cannot be considered as correct, especially at the wall of the mixer as results from the following example which enables analytical solution. It is known (see e.g.[17]) that the probability solution of Eq. (1) with the reflexing boundary in the point $z = 0$ is the relation

$$f(z'; \Delta t|z) = \frac{1}{\sqrt{(2\,\pi\Delta t)}\,\sigma} \left[\exp\left(-\frac{(z'-z)^2}{2\sigma^2\,\Delta t} \right) + \exp\left(-\frac{(z'+z)^2}{2\sigma^2\,\Delta t} \right) \right], \quad [z, z' \geqq 0],$$

$$(35)$$

where for D the term is substituted by $D = \sigma^2/2$. To satisfy the next condition, i.e. the second of Eqs (34) the relation $\sigma \ll \delta$ must hold. After substitution from relation (35) into the first one of Eqs (34) it is possible to demonstrate that the drift velocity on the reflexing boundary is given by the relation

$$V(0) = \lim_{\Delta t \to 0} \frac{1}{\Delta t} \int_0^\delta z' f(z'; \Delta t|0)\, dz' = \lim_{\Delta t \to 0} \sigma \sqrt{\left(\frac{2}{\pi \Delta t}\right)} \to \infty \qquad (36)$$

which is obviously in contradiction with Eq. (1), where this value is identically equal to zero. Solution of the same equation without the reflexing boundary is the even function with respect to z; by substitution into the first of Eqs (34) and by integration of the first moment the drift velocity is equal to zero everywhere.

However, the analogous experimental estimate of diffusion coefficient[8] (i.e. of the second moment) is from this point of view correct because the corresponding integration gives identical values of $\sigma^2/2$ as the solution without the reflexing boundary. Situation is schematically depicted in Fig. 6.
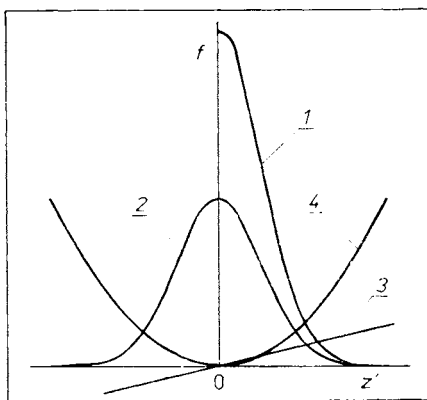

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Dependence for calculation of first two moments of the function in Eq. (35). 1 Solution with the reflexing boundary in point $z = 0$, 2 solution without the reflexing boundary, 3 $f = z'$ (for the first moment), 4 $f = z'^2$ (for the second moment)

The statement made concerning the symmetry of the diffusion coefficient with respect to the axial coordinate is based on the assumption that at constant speed of rotation of the mixer with the axis of rotation adjusted exactly horizontally is the condition for axial shift of components only their geometry and geometry of the mixer. These effects determine the field of radial velocities of the solid phase which is not homogeneous in the axial direction in the neighbourhood of horizontal walls of the mixer (see e.g.[4,5]). As is obvious from Fig. 2 the experimental data confirm validity of this assumption as they describe correctly the form of "pure" segregation in time.

It is worth mentioning that application of Eqs (14) and (16) has been proposed theoretically by Fan and Shin[8], but the initial distribution of concentrations which was used in their study would lead to the physically incorrect solution as values of mass fractions could in some moments, at the beginning of the process, be larger than one.

The assumption about mutual interaction of diffusion coefficients describing first of all mixing and segregation of weighed asymmetric mass distributions of the larger component in both halves of the mixer is an attempt for solution of the mentioned problem. It leads to an implicit dependence of the diffusion coefficient on time. In the explicite form this dependence was proposed earlier by one of the authors of this study[12].

From Fig. 3 it is obvious that in short time intervals from the beginning significantly appears first of all the mixing process, in long time intervals of the process first of all segregation. These conclusions are in a reasonably good agreement with the experiments. But the proposed model does not describe in a satisfactory manner the actual case in the medium time intervals where the effect of both coefficients is significant (see Fig. 4). This conclusion is obvious also from Fig. 5 where the time dependences of concentration variances obtained on basis of the experiments and according to the proposed model are compared. These were calculated in both cases according to relations (27). Better agreement could be obtained perhaps by application of a more complicated function instead of the linear combination of both diffusion coefficients.

It is possible to state that the part of experiments performed by Fan and Shin[8] confirms our conclusion on the prevailing effect of the symmetric segregation coefficient at the end of the experiment (see Fig. 5E of the quoted study) while is obvious the qualitative indirect proportionality between the diffusion coefficient and stationary concentration distribution.

Moreover, an attempt has been made to compare the diffusion coefficients given in literature[2,8] with the values obtained in this study.

First of all it is necessary to express the dependence of this quantity on the axial coordinate with respect to the fact that the mixer described in this study is considerably shorter than the mixer quoted in the above-mentioned study[8]. (Dimensions of the mixer described in the study by Hogg et al.[2] are from this point of view in-

significant, diffusion coefficient describing mixing of particles of equal dimensions is a constant). The dimensionless mixer length, defined as the ratio of the half mixer length related to its diameter $L/R = 2\cdot64$ for the mixer in the study by Fan and Shin[8] in comparison to $L/R = 1\cdot62$ of this study. In an attempt to eliminate the effect of this difference of dimensionless lengths in this study the results of Donald and Roseman[4,5] were used, according to which considerable changes in concentration with length take place in close vicinity of side walls of the mixer due to changes of the gradient of radial velocity. In the central part of relatively long mixers this gradient is equal to zero and does not affect mixing of the solid phase. In graphical comparison of diffusion coefficients the dimensionless length of the here used mixer was increased for the difference of dimensionless lengths $2\cdot64 - 1\cdot62 = 1\cdot02$. This difference was situated into the central part of the mixer and it was assumed that the diffusion coefficient will not change here. The situation is demonstrated in Fig. 7, on the upper coordinate axis. The line segments at the left and right hand parts of this axis taken together (without the middle line segment) correspond to the horizontal coordinate axes in Figs 1 − 4 of this study. The lower coordinate axis in Fig. 7 is identical with the horizontal coordinate axis in Fig. 3 of the study by Fan and Shin[8]. Next the diffusion coefficients calculated according to relations (19) and (20) were recalculated for concentration of the smaller component as they are given in the quoted study. For this calculation the following equation was used

$$D(z, t) = AI(t) + B[1 - I(t)]/[1 - x_{is}(z)] . \qquad (37)$$

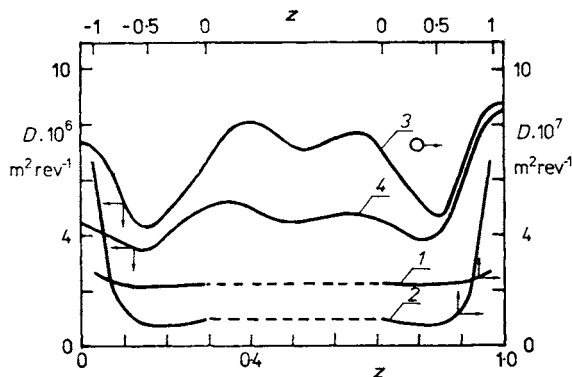These results were related to one revolution of the mixer.

FIG. 7

Diffusion coefficient in dependence on axial coordinate 1 Values at the beginning of the experiment ($p = 2$; $\omega = 0\cdot552$; $L/R = 1\cdot62$), 2 values at the end of the experiment ($p = 2$; $\omega = 0\cdot552$; $L/R = 1\cdot62$), 3 data by Fan and Shin[8] ($\omega = 0\cdot604$; $L/R = 2\cdot64$), 4 data by Fan and Shin[8] ($\omega = 0\cdot667$; $L/R = 2\cdot64$), ○ data by Hogg et al.[2] ($\omega = 1\cdot0$)

Results of these calculations are demonstrated in Fig. 7, where the dependence of the diffusion coefficient on axial coordinate is given at the beginning (curve *1*) and at the end (curve *2*) of mixing. In the central — extended — part of the axis the curves are dashed. In agreement with the considerations made in the preceding paragraph they are only an approximation and do not represent real values. The curves *3* and *4* are taken from Fig. 3 of the study[8]. The parameter is in this case the ratio of mean dimensions of smaller and larger particles, denoted by the symbol ω.

It is obvious that the dependence of the diffusion coefficient is in all cases the same: toward the mixer centre starting from the walls it at first decreases and then passes through a minimum which is for our data not very profound. Coordinates at which the dependence reaches the minimum is in all cases approximately the same $(\sim 0.4\ L/R)$ which confirms conclusions by Donald and Roseman[4] concerning the effect of walls on the velocity gradient. Diffusion coefficients for sea sand are roughly for one order of magnitude smaller than for spherical lucite particles[8]. This is because the resistance to axial motion ("viscosity") of individual particles is in the case of spheres considerably lower than with particles of nonuniform shapes. The diffusion coefficient for "pure" mixing of very small spherical particles[2] is situated approximately in between these data.

More detailed study of this problem would require experimental investigation of different systems of the charge and geometric arrangement of the mixer.

CONCLUSIONS

On basis of considerations made in this study and evaluation of experimental data obtained by earlier investigations the following most important conclusions can be made:

1) The set of assumptions was specified at which it is possible to consider the mixing of solid particles in the horizontal cylindrical mixer to be the Markov diffusion process with the adequate Kolmogorov's equation and when the diffusion coefficients in this equation could be determined.

2) The term of "pure" segregation was defined, which represents relocation of two components of the solid phase of different sizes, when concentration of these components is in each moment symmetric with respect to the vertical plane which divides the mixer into two identical halves. On basis of good agreement with the experiments it was proved that this operation can be described by use of the diffusion coefficient which is the even function of axial coordinate.

3) A model was proposed which describes mixing of two components of the solid phase of different sizes which are at the beginning nonuniformly distributed as the linear superposition of the classic diffusion and "pure" segregation and which leads

to the conclusion that the resulting diffusion coefficient is the implicite function of time. The experiments have confirmed validity of this model only in the initial and final states of mixing when only one of the mechanisms is dominant.

LIST OF SYMBOLS

c	concentration (general)
D	diffusion coefficient, $m^2 s^{-1}$
f	probability density, m^{-1}
f_3	three-dimensional probability density, m^{-3}
I	asymmetry indicator
L	half of mixer length, m
m	mass of particle, kg
n	number of particles
R	radius of mixer, m
r	radial coordinate, m
S	surface area of the mixer, m^2
t	time, s
U	volume, m^3
V	drift velocity, $m s^{-1}$
x	mass fraction
$\boldsymbol{x}$	radius-vector, m
y	dimensionless axial coordinate
Z	position of particle (random variable), m
z	axial coordinate, m
δ	integration limit (see Eq. (*34*))
ϱ	density, $kg\ m^{-3}$
φ	angular coordinate, rad
σ^2	variance
τ	dimensionless time
ω	ratio of mean dimensions of smaller and larger particles

Subscripts

c	related to solid phase
i	related to larger component
j	related to smaller component
S	related to the mixer surface
s	related to symmetric conditions
0	related to initial conditions

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Translated by M. Rylek.