

MASS TRANSFER IN THE VOID SPACE OF A
FIXED GRANULAR BED

V. A. Kirillov and Yu. Sh. Matros

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The transfer processes in the void space of a granular bed are examined. The parameters of the hydrodynamic model are experimentally determined.

In simulating processes in chemical reactors it is necessary to solve problems associated with the flow of the reaction mixture through a fixed bed of nonporous particles. In this case it is important to consider the hydrodynamic conditions in the void space of the granular bed, especially if the input parameters of the process (temperature, concentration) vary with time.

1. Choice of Model. In order to understand the processes taking place in the void space and choose an appropriate mathematical model, we made a Schlieren study [1] of the flow of a tracer gas using a Karl-Zeiss Jena-80 instrument. The plane model represented the most characteristic part of the bed between two transverse surfaces. During the experiment the gas flow velocity was varied on the interval 5-100 cm/sec, which corresponded to $Re = 20-600$. It was established that over the entire range of Re numbers the void space was nonhomogeneous: there are two regions - a flow region consisting of streams of gas in the space between grains and a nonflow region in the vicinity of particle contact points. In the nonflow zones formed as a result of the separation of the gas stream from the surface of the grain the gas is vigorously mixed owing to the presence of eddies. Accordingly, the concentration of tracer gas in the nonflow zones is approximately constant over its entire volume. Mass transfer between zones is linked with eddy fluctuations, whose intensity depends on the linear gas flow velocity.

On the basis of this experimental investigation, with allowance for the presence of molecular and turbulent diffusion in the flow zone the material balance in an element of the granular bed may be written in the following form

$$\frac{1}{Pe} \cdot \frac{\partial^2 C_1}{\partial \xi^2} - \frac{\partial C_1}{\partial \xi} - \gamma \frac{\varepsilon_2}{\varepsilon_1} (C_1 - C_2) - \frac{\partial C_1}{\partial t'} = 0, \quad (1)$$

$$\gamma (C_1 - C_2) = \frac{\partial C_2}{\partial t'}$$

where

$$\xi = \frac{l}{d}; \quad t' = \frac{U \varepsilon t}{d \varepsilon_1}; \quad \gamma = \frac{\varepsilon_1}{\varepsilon} \cdot \frac{\beta d}{U}; \quad Pe = \frac{U d K^2}{D_1 + D_2} \cdot \frac{\varepsilon}{\varepsilon_1}; \quad \beta = \alpha \frac{F}{V}.$$

The unknown parameters of this model are as follows: the fraction of nonflow zones in the bed, the coefficient β of interzonal mass transfer, the turbulent diffusion coefficient D_2 , and the tortuosity factor K .

It should be noted that a system of equations similar to (1) was previously proposed in [2]. However, the unknown parameters of that model were determined from the same experimental data, for liquids only, and on a narrow range of variation of velocity and particle geometry.

2. Turbulent and Molecular Diffusion. Mass transfer in the longitudinal direction of the bed is realized only in the flow zones as a result of viscous flow and molecular and turbulent diffusion. The flow region is a system of intersecting streams, whose dimensions are determined by the geometric structure of the bed. The available data [3] indicate that D_2 can be quite accurately determined from the expression

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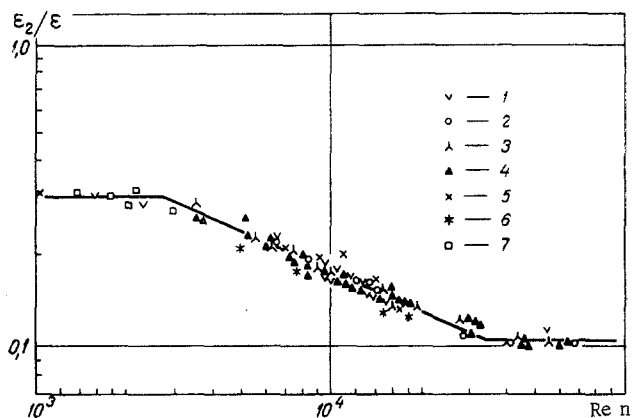


Fig. 1. Fraction of nonflow zones as a function of $Re \cdot n$ for glass beads. Air-H₂(He): 1) $d = 1.3$ mm; 2) $d = 2$ mm; 3) $d = 5.0$ mm; 4) $d = 8.0$ mm. At $d = 8.0$ mm: 5) CO₂-H₂(He); 6) Ar-H₂(He); 7) H₂-N₂(Ar).

$$D_2 = q\lambda. \quad (2)$$

The turbulent fluctuations q represent approximately 10% of the flow velocity [4], and the scale λ is determined by the dimensions of the voids between grains. For rhombohedral packing $\lambda = (0.2-0.4)d$.

The tortuosity K is exclusively determined by the structure of the bed and can be obtained as a result of relatively simple constructions for a known coordination number or found from the empirical relation [5]

$$K = \varepsilon^{-0}. \quad (3)$$

The possible errors in determining D_2 and K are unimportant owing to the low sensitivity of the solution of system (1) to these quantities.

3. Experimental Determination of the Fraction of Nonflow Zones. The fraction of nonflow zones can be determined on the basis of the geometric characteristics of the bed [6]

$$\frac{\varepsilon_2}{\varepsilon} = \frac{\pi(3 \sin^2 \varphi_1 + 2 \cos^3 \varphi_1 - 2)}{2[6(1 - \cos \theta) \sqrt{1 + 2 \cos \theta} - \pi]}. \quad (4)$$

However, the lack of data on the separation angles for such complex systems as a granular bed makes it necessary to determine them experimentally.

We have determined the fraction $\varepsilon_2/\varepsilon$ for spherical packings and gas flows with different physical properties on a certain range of linear velocities. A carrier gas is blown through the granular bed and at a certain moment of time a tracer gas is introduced at the inlet. The time interval Δt that elapses before this tracer gas is first detected in the sections l_1 and l_2 is determined. Then

$$\varepsilon_2 = \varepsilon - \left(\frac{l_2 - l_1}{U \Delta t} \right)^{-1}. \quad (5)$$

For a sufficiently steep tracer concentration front the accuracy of determination of the time of appearance of the tracer may be affected by the sensitivity of the measuring apparatus and, moreover, by molecular and turbulent diffusion. Accordingly, in experimentally determining ε_2 , on the basis of an analysis of system (1), we selected conditions such that the time of appearance of the tracer was practically independent of its concentration at a given sensitivity of the measuring apparatus. In this case Δt is directly proportional to the length of the bed and inversely proportional to the gas velocity in the flow zone.

The experimental apparatus consisted of a column of sufficiently large diameter (150-200 mm) filled with granular material. A carrier gas was blown through the column. Then at a certain moment the tracer gas was injected into the flow and its time of appearance at several points along the length of the column was registered by detectors. The experimental curves were recorded simultaneously for all the sampling points on an N-700 loop oscillograph.

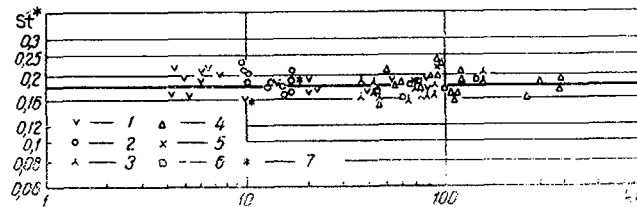


Fig. 2. The dependence $St^* = f(Re)$. Air-H₂(He): 1) $d = 1.3$ mm; 2) $d = 2.0$ mm; 3) $d = 5.0$ mm; 4) $d = 8.0$ mm. At $d = 80$ mm: 5) CO₂-H₂; 6) Ar-H₂(He); 7) H₂-N₂(Ar).

The experiments were conducted with glass beads of diameter $d = 1.3, 2.0, 5.0,$ and 8.0 mm. During the experiment the Re number was varied on the interval $10-400$. The gases investigated included air-H₂, air-He, CO₂-H₂, Ar-H₂(He), H₂-N₂, H₂-Ar (the second gas was the tracer). An analysis of the experimental data based on expression (5) showed that the fraction $\varepsilon_2/\varepsilon$ depends on the linear flow velocity in the void space and on the viscosity of the gas mixture. Varying the grain size on the interval $1.3-8.0$ mm did not have much effect on the ratio $\varepsilon_2/\varepsilon$. It was found that the results obtained are described with satisfactory accuracy by a single dependence, if the expression Ren (where $n = L/d$ gives the number of grains per meter) is plotted along the ordinate axis. It is clear from Fig. 1 that at $Ren \leq 2500$ $\varepsilon_2/\varepsilon = 0.31$; if $Ren \geq 37,000$, then $\varepsilon_2/\varepsilon = 0.1$. On the intermediate range of values

$$\varepsilon_2/\varepsilon = 8.15 (Ren)^{-0.42} \quad \text{when } 2500 \leq Ren \leq 37000. \quad (6)$$

4. Investigation of Interzonal Mass Transfer. As a result of the presence of eddies mixing in the nonflow zone is practically complete. As a result of eddy fluctuations a certain amount of gas is transferred from the nonflow to the flow zones. These fluctuations occur even at gas velocities corresponding to laminar flow [7]. Only at a flow velocity quite close to zero may interzonal transfer become purely diffusional.

Physically, the transfer coefficient β in (1) coincides with the frequency of the eddy fluctuations in the nonflow zones. In fact, on the interval $T = 1/\omega$ (T is the fluctuation period) a certain fraction of volume ηV ($\eta \leq 1$) is ejected into the flow zone, as a result of which a volume of gas equal to $F\alpha T$ enters the flow zone from the nonflow zone. Equating these two fluxes, we obtain

$$T = \eta \frac{V}{F\alpha}$$

or

$$\omega = \frac{1}{\eta} \beta. \quad (7)$$

The coefficient η denotes the fraction of gas transferred to the flow zone during a time equal to one eddy fluctuation period. On the basis of the experiments described in Sec. 1 it was found that $\eta \approx 0.2-0.3$.

The Strouhal number $St = \omega l_0/U_1$ ($l_0 \approx d/2$ is the roughness of the granular bed) is known to be constant for a given geometric structure [8]. On the basis of [8] it may be assumed that for a bed with a smooth surface $St \approx 0.3$; then, using (7),

$$St = \frac{\varepsilon_1}{\varepsilon \eta} \cdot \frac{\beta d}{U}. \quad (8)$$

As follows from Fig. 1, the flow zone fraction increases with velocity by approximately 20%. The coefficient η is also approximately constant and chiefly determined by the geometric structure of the bed. Accordingly, it follows from (8) that the mass transfer coefficient is directly proportional to the grain size. For the indicated values of the parameters in (1)

$$St^* = \frac{\beta d}{U} \approx 0.15 - 0.25. \quad (9)$$

In order to check our results we experimentally determined the mass transfer coefficients. For this purpose we used the experimental apparatus described in Sec. 3. At the entrance to the bed we introduced a signal in the form of a tracer gas pulse, which was detected at several points along the length of the

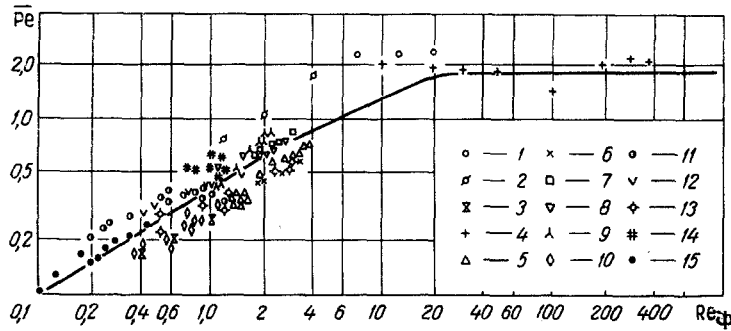


Fig. 3. The dependence $\bar{Pe} = f(Re_p)$ for various gases [9]: 1) air-mercury, $d = 1.4$ mm; 2) $d = 0.434$ mm; [10]: 3) air-He; [11]: 4) $N_2-C_2H_4$, N_2-H_2 , $d = 3$ mm; [12]: 5) N_2-He , $d = 2.0$ mm; 6) N_2-H_2 ; 7) $Ar-N_2$; 8) N_2-Ar ; 9) $N_2-C_2H_4$; $d = 0.8$: 10) N_2-He ; 11) $N_2-C_2H_4$; [13]: 12) CH_4-Kr ; 13) O_2-Kr ; 14) SO_2-Kr ; 15) H_2-Kr .

column and recorded by two analogue-digital converters of a JRA-5 computer. The computer processed the experimental data in accordance with a special program and transferred the results in digital code to a puncher. Then system of equations (1) was solved on a Minsk-2 digital computer for boundary conditions corresponding to the experimental conditions, and a coefficient β that gave the best fit between the experimental and theoretical curves was selected.

The mass transfer coefficients β were determined for the same conditions as ϵ_2/ϵ . The results were processed in the form $St^* = \beta d/U$ and are presented in Fig. 2 for various values of the Re number.

As follows from the figure, St^* is actually independent of Re and is a constant equal to 0.18, which confirms our previous conclusions.

5. Investigation of Transfer Processes in the Bed. In the nonstationary regime the longitudinal mass transfer is uniquely determined by the residence time distribution function $f(\xi, t')$ of the tracer gas, which is introduced in the section $\xi = 0$ in the form of a delta function $\delta(t')$. It can be found by solving (1) for the boundary conditions (for a bed of infinite length)

$$\begin{aligned} t' = 0 \quad C_1 = C_2 = 0, \\ \xi = 0 \quad C_1 = \delta(t'). \end{aligned} \quad (10)$$

The solution of system (1) with conditions (10) in Laplace transforms takes the form

$$\bar{f}(\xi, t') = \exp \left[\frac{Pe}{2} \left(1 - \sqrt{1 + \frac{4S}{Pe} \left(\frac{\gamma}{\gamma + S} \frac{\epsilon_2}{\epsilon} + 1 \right)} \right) \xi \right]. \quad (11)$$

On the basis of an analysis of (11) we obtained the first four initial moments and from them the principal statistical characteristics:

residence time

$$\bar{t}' = l/U, \quad (12)$$

variance

$$\sigma^2 = 2\xi \left(\frac{\epsilon_2}{\epsilon_1} \cdot \frac{1}{\gamma} + \frac{\epsilon^2}{\epsilon_1^2} \cdot \frac{1}{Pe} \right), \quad (13)$$

coefficient of skewness

$$Sk = \frac{6 \frac{\epsilon_2}{\epsilon_1 \gamma^2} + 12 \frac{\epsilon^3}{\epsilon_1^3 Pe} + 12 \frac{\epsilon_2}{\epsilon_1^2} \cdot \frac{\epsilon}{\gamma Pe}}{\sqrt{\left(2 \frac{\epsilon_2}{\epsilon_1 \gamma} + 2 \frac{\epsilon^2}{\epsilon_1^2 Pe} \right)^3} \sqrt{\xi}}, \quad (14)$$

coefficient of excess

$$\varepsilon_x = 6 \frac{\frac{\varepsilon}{\varepsilon_1 \gamma^2} + 2 \frac{\varepsilon_2 \varepsilon}{\varepsilon_1^2 \bar{P}e \gamma^2} + \frac{\varepsilon_2^2}{\varepsilon_1^2 \gamma^2 \bar{P}e} + \frac{6\varepsilon_2 \varepsilon^2}{\varepsilon_1^3 \bar{P}e^2 \gamma} + \frac{5\varepsilon^4}{\varepsilon_1^4 \bar{P}e^3}}{\left(\frac{\varepsilon_2^3}{\varepsilon_1^2 \gamma^2} + 2 \frac{\varepsilon_2 \varepsilon^2}{\varepsilon_1^3 \bar{P}e} + \frac{\varepsilon^4}{\varepsilon_1^4 \bar{P}e^2} \right) \xi} \quad (15)$$

A similar analysis was made for a diffusion model with the mathematical description

$$\frac{1}{\bar{P}e} \cdot \frac{\partial^2 C}{\partial \xi^2} - \frac{\partial C}{\partial \xi} - \frac{\varepsilon}{\varepsilon_1} \cdot \frac{\partial C}{\partial t'} = 0 \quad (16)$$

$$\text{at } t' = 0 \quad C = 0, \quad \xi = 0 \quad C = \delta(t'), \quad (17)$$

$$\bar{t} = l/U, \quad (18)$$

$$\bar{\sigma}^2 = 2\xi \frac{\varepsilon^2}{\varepsilon_1^2} \cdot \frac{1}{\bar{P}e}, \quad (19)$$

$$\bar{S}k = 3\sqrt{2}/\sqrt{\bar{P}e \xi}, \quad (20)$$

$$\varepsilon_x = 30/\bar{P}e \xi. \quad (21)$$

Comparing (12) and (18), we find that the mean residence time for these two models is the same. The variances are equal if

$$\bar{P}e = \frac{1}{\frac{1}{\text{Re Sc}'} \cdot \frac{1}{K^2} \left(1 - \frac{\varepsilon_2}{\varepsilon} \right) + \frac{1}{\text{St}^*} \cdot \frac{\varepsilon_2}{\varepsilon}} \quad (22)$$

If (22) is satisfied, the coefficients of skewness and excess for the two models may differ considerably even at comparatively large ξ . Accordingly, the distribution functions will also differ. An analysis showed that these functions approach each other only at $\xi \geq 500-800$.

There have been numerous experimental determinations of $\bar{P}e$ under various conditions [9-13]. The results of a comparison with relation (22) are presented in Fig. 3. The continuous curve corresponds to the values of $\bar{P}e$ calculated from (22).

As follows from the figure, the agreement between the calculated and experimental values of \bar{P} is quite good. It is interesting to note that the dependence obtained successfully predicts the values of $\bar{P}e$ over the entire range of variation of $\bar{R}e_f$. As follows from (22), the value $\bar{P}e = 2.0$ given in the literature corresponds to a constant volume of the nonflow zones. Thus, substituting $\varepsilon_2/\varepsilon = 0.1$ and $\text{St}^* = 0.18$ in (22), we obtain $\bar{P}e = 1.8$.

NOTATION

l	is the variable length of the bed, m;
d	is the grain diameter, m;
U	is the gas velocity in the free cross section of the bed, m/sec;
\bar{U}	is the gas velocity over the total cross section of the column, m/sec;
U_1	is the gas velocity in the flow zones, m/sec;
G	is the mass gas flow rate over the total cross section of the column, $\text{m}^3/\text{kg} \cdot \text{sec}$;
ε_1 and ε_2	are the flow and nonflow zone fractions, respectively;
β	is the interzonal transfer coefficient, sec^{-1} ;
V	is the volume of the nonflow zones, m^3 ;
F	is the interzonal transfer surface, m^2 ;
K	is the tortuosity factor;
α	is the mass transfer coefficient referred to the transfer surface, m/sec;
D_1 and D_2	are the molecular and turbulent diffusion coefficients, respectively, m^2/sec ;
t	is the time, sec;
C_1 and C_2	are the tracer gas concentrations in the flow and nonflow zones, respectively, fractions;
S	is the Laplace variable;
μ	is the dynamic viscosity, $\text{kg} \cdot \text{sec}/\text{m}^2$;
ν	is the kinematic viscosity, m^2/sec ;

θ is the angle between the sides of the rhomb formed by the lines connecting the centers of the spheres;
 φ is the flow separation angle, $\varphi_1 = 180^\circ - \varphi$;
 \bar{D} is the longitudinal diffusion coefficient, m^2/sec ;
 $\text{Re} = \text{Ud}/\nu$;
 $\text{Re}_f = \text{Gd}/\mu$;
 $\text{Sc}' = \gamma/(\text{D}_1 + \text{D}_2)$.

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