# QUALITATIVE ANALYSIS OF THE MASS TRANSFER OF AN ORDERED SYSTEM OF SOLID REACTING PARTICLES AT HIGH PECLÉT NUMBERS

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Taking account of changes in the velocity and concentration fields due to the presence of other particles, a qualitative analysis is given of the mass transfer of a concentrated ordered system of reacting solid particles at high Peclét numbers.

The structure of singular streamlines starting and terminating at particle surfaces [1-4] plays an essential role in problems on convective diffusion to a system of particles at high Peclét numbers. It hence turns out that particle chains exist in a flow in which the internal mass transfer is strongly retarded by the interaction between the diffusion wakes and the boundary layers of the particle belonging to the chain. The problem of diffusion to a sufficiently sparse system of spheres arranged at the nodes of a cubic array was considered in [5].

A qualitative analysis is given below of the mass transfer of a concentrated ordered system of reacting solid particles at high Peclét numbers. Taking account of changes in the velocity and concentration fields due to the presence of other particles, the mean diffusion flux is determined on the particle surfaces. The constant of the effective volume reaction is calculated, and its dependence on the concentration and distance along the flow is found.

The qualitative investigation of the system mass transfer is executed as a function of the Reynolds number. It is shown that at low Reynolds numbers (in the absence of a developed attached vortex behind the particles) the mean diffusion flux can be considerably less than that calculated from the results in [6, 7], where the flow field near the particle surface is determined within the framework of the nuclear model [8-10], and the change in the concentration field due to the presence of other particles is neglected.

1. Let us consider stationary convective diffusion in a laminar viscous incompressible fluid flow being filtered through a system of spheres of equal radius $a$ arranged at the sites of a regular array formed by the regular duplication of the fundamental cell. For simplicity we consider the fundamental cell to have the shape of a parallelepiped and to be given by three vectors $al_i\mathbf{e}_i$, $|\mathbf{e}_i| = 1$, $i = 1, 2, 3$ and that the mean velocity of the filtration stream is parallel to the vector $\mathbf{e}_1$ and equal to U in the spacings between the spheres. We select the origin in a corner of the fundamental cell, hence the radius-vector of any particle will be determined by the relationship

$$\mathbf{r} = a\sum_{i=1}^{3} k_i l_i \mathbf{e}_i, \quad k_i = 0, 1, 2, \ldots, N_i \quad (i = 1, 2, 3).$$

Let us assume that the flow field in the array is periodic (quasiperiodic) and given by the flow in the fundamental cell.

As an illustration, let us consider the Stokes flow around particles in which their mutual influence on each other is taken into account, particularly by the following two approximate methods.

The nuclear model [8-10] considers the flow around a sphere of radius $a$ surrounded by a certain concentric shell of fluid with outer radius b. Let us assume that the fluid on the outer surface bounding the liquid sphere does not spread to the adjacent domains where others of the same spheres are found and the adhesion condition is satisfied on the particle surface. The boundary conditions on the cell surface are posed differently in different models. For instance, it is considered that there are no vortices [8] or tangential stresses

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[9, 10] on the cell boundary. In the case of a regular array (in the plane case) the models of [8-10] have been studied experimentally [11] and theoretically [12]. It has been shown that the model in [8] describes the flow field near the particle surface well.

Another method is suitable for $l_1^{-1} \ll 1$ and is based on introducing the density of the force acting from the sphere onto the fluid and given in the form of a series with constant coefficients containing the delta-function and its derivatives at the center of the spheres [5, 13]. This approach permits finding that combination of partial periodic solutions that vanishes on the sphere surfaces.

The dimensionless stream function near the sphere surfaces in these models can be represented in the form

$$\psi = 3/4\,(1+T)(r-1)^2\sin^2\theta, \tag{1}$$

where the stream function is written in a spherical coordinate system coupled to the center of the sphere, the angle $\theta$ is measured from the free stream direction, and the sphere radius $a$ and the velocity U are taken as the characteristic scales. The constant T depends on the volume concentration of the spheres in the array and can, in particular, be determined from [5, 8-10, 13].

The distribution of the concentration c in the stream is determined by the solution of the stationary convective diffusion equation

$$(\mathbf{v}\nabla)c = \mathrm{Pe}^{-1}\Delta c, \quad \mathrm{Pe} = aUD^{-1} \tag{2}$$

with constancy of the concentration far from the array and total absorption of the substance dissolved in the stream on the sphere surfaces, as boundary condition. The Peclét number Pe is henceforth considered high.

The hydrodynamic model of the flow [8-10] was used in [6, 7], where the self-similar solution of the diffusion boundary layer [14] was used to compute the diffusion influx to the sphere. Such an approach takes account of the change in the concentration field produced by other reacting particles in the stream. In particular, such an approach does not take into account the depletion of the solution because of the reaction at the particles arranged in the array in the stream, and corresponds to one active reacting sphere in a system of passive (nonreacting) spheres. In conformity with the experimental results [15, 16], such a calculation procedure for moderate Reynolds numbers ($\mathrm{Re} \lesssim 10$) can result in a severalfold exaggeration of the mean flow at the particles. The individual (at each sphere) and mean total diffusion fluxes on a sphere obtained by such a method will agree:

$$I(\mathbf{k}) = \langle I \rangle = A\mathrm{Pe}^{1/3} = I_*, \quad A = \frac{1}{2}(3\pi)^{5/3}\Gamma^{-1}\left(\frac{1}{3}\right)(1+T)^{1/3}, \tag{3}$$

$$\langle I \rangle = [N_1N_2N_3]^{-1}\sum_{k_1=1}^{N_1}\sum_{k_2=1}^{N_2}\sum_{k_3=1}^{N_3} I(\mathbf{k}), \quad \mathbf{k} = \{k_1, k_2, k_3\}.$$

In order to take account of the change in the concentration field due to the absorption of other particles, it is necessary to consider the particle diffusion wakes whose presence results in the fact that the leakage condition for the concentration of dissolved substance in the free stream for each particle will depend on its relative location in the array and is found from the solution of the problem of diffusion to particles located upstream [1-4].

Under the assumption of no closed circulation domains, the diffusion wake behind a solid sphere is investigated in [17-19] (by the method of mergeable asymptotic expansions in the high Peclét number), where it was shown that it consists, in turn, of four characteristic subdomains $W^{(i)}$ (i = 1, 2, 3, 4).

The concentration in the convective-boundary layer domain $W^{(1)}$ remains constant on the streamlines and is determined by the concentration at the exit from the diffusion boundary layer. The orders of the dimensionless length $\delta_1$ and width $\Delta_1$ of this domain are given by the inequalities $O(\mathrm{Pe}^{-1/3}) < \delta_1 < O(\mathrm{Pe}^{1/3})$. $O(\mathrm{Pe}^{-1/2}) < \Delta_1 < O(\mathrm{Pe}^{1/3})$ and the concentration $c^{(1)}$ has the order of one.

The inner domain of the diffusion wake $W^{(2)}$ is characterized by the fact that the tangential transfer of substance along the surface plays an essential part therein while the radial transfer is inessential. The fluid flow in $W^{(2)}$ is considerably depleted and the concentration is $c^{(2)} = O(\mathrm{Pe}^{-1/6})$ and grows proportionately to the square root of the distance

to the particle surface on the stream axis. The characteristic dimensions of this domain are $O(Pe^{-1/3}) < \delta_2 < O(Pe^{1/3})$, $\Delta_2 < O(Pe^{-1/2})$.

Both the tangential and radial transfer of substance to the surface must be taken into account in the domain of the rear stagnation point $W^{(3)}$. The characteristic dimensions of this domain are $\delta_3 < O(Pe^{-1/3})$, $\Delta_3 < O(Pe^{-1/3})$, and the stream is depleted maximally with $c^{(3)} = O(Pe^{-1/3})$.

In the mixing domain of the diffusion wake $W^{(4)}$ (the characteristic dimensions are $\delta_4 > O(Pe^{1/3})$, $\Delta_4 < O(Pe^{-1/3})$), the radial substance transfer is inessential and the concentration is $c^{(4)} = O(1)$.

Such a diffusion wake is destroyed when a closed circulation domain is formed behind the particles. A new diffusion wake $W_*$ with a simpler structure consisting of two subdomains, the domain of the rear critical line $W_*^{(3)}$ and the mixing domain $W_*^{(4)}$ [3], is formed on the common boundary between the attached vortex and the outer flow for finite dimensions of this domain. Both the radial and tangential substance transfers along the particle surface are essential in the domain $W_*^{(3)}$, while the radial transfer can be neglected in $W_*^{(4)}$. The concentrations in these domains are on the order of $c_*^{(3)} \sim Pe^{-1/6}$, $c_*^{(4)} \sim 1$, and the characteristic dimensions are determined by the inequalities $\delta_3 < O(Pe^{-1/3})$, $\Delta_3 < O(Pe^{-1/3})$; $O(Pe^{-1/3}) < \delta_4 < O(Pe^{-1/9})$, $\Delta_4 < O(Pe^{-1/3})$.

The diffusion wakes of particles located upstream deplete the solution of the fluid arriving next in the diffusion boundary layer, which can result in an abrupt reduction in the total diffusion flux in this fluid as compared to that which is calculated by using the self-similar solution [14].

Let us consider a compact array for which the condition $l_1 \ll Pe^{1/3}$ is satisfied.

Using the results in [1-4], we obtain the total diffusion flux per particle with number $\mathbf{k}$ analogously to the formulas obtained for the chain of spheres with period $l \ll Pe^{1/3}$ located on the stream axis:

$$I(\mathbf{k}) = I_*[k_1^{2/3} - (k_1 - 1)^{2/3}], \quad \mathbf{k} = \{k_1, k_2, k_3\}, \tag{4}$$

where $I_*$ is the total diffusion flux per sphere determined by means of the self-similar solution [14]. In particular, it is determined by (3) in the models of [5, 8-10, 13].

It is seen from (4) that the total diffusion flux per sphere diminishes with the increase in the ordinal number $k_1$, and tends to zero in inverse proportion to the one-third root of $k_1$ for large numbers. This indicates that the diffusion wakes exert considerable influence on the mass transfer of a fixed particle with the medium in the case under consideration.

2. The mean diffusion flux per sphere is determined by the formula

$$\langle I \rangle = I_* N_1^{-1/3}. \tag{5}$$

It is seen that the mean diffusion flux calculated taking account of interaction between the diffusion wakes and the particle boundary layer is always less than that calculated by means of (3), where this interaction is neglected. As the number of particles in the array increases, the mean flux per sphere tends to zero.

As Reynolds number increases, (5) remains valid until there are no closed circulation domains behind the particles. The diffusion wake starts to be reconstructed with their appearance, and this influence is exerted in distances on the order of $Pe^{-1/3}$ [3] for finite dimensions of this domain. Hence, if the array period $l_1$ is considerably greater than this quantity, the influence of the diffusion wakes ceases to be felt by the particle mass transfer and the mean value for the diffusion flux agrees (in the main approximation in the characteristic diffusion parameter) with the individual flux per particle. This can be a qualitative explanation of the fact that the results of [6, 7] agree fairly well with the experimental results for sufficiently large Reynolds numbers [15, 16].

For the case when the dimensions of the closed circulation domain are less than or commensurate with $Pe^{-1/3}$, the mean diffusion flux per particle of the array can be represented in the form [2]

$$\langle I \rangle = I_* N_1^{-\gamma(\mathrm{Re},\,\rho)}, \quad 0 \leqslant \gamma(\mathrm{Re},\,\rho) \leqslant 1/3, \tag{6}$$

where $I_*(\mathrm{Re}, \rho)$ is the total diffusion flux per individual particle of the array calculated without taking account of the change in the concentration field in the stream due to the

presence of the other spheres. The specific value of $\gamma$ hence depends on the Reynolds number Re, the volume concentration of the particles $\rho$, and the geometry of their mutual arrangement, and can be determined from experimental results.

For $Re_1$ let a closed circulation domain first occur behind the spheres and for $Re_2$ let the dimensions of the attached vortex considerably exceed $Pe^{-1/3}$. The values of $Re_1$ and $Re_2$ depend on the volume concentration of the spheres in the array. There are different experimental data (see [16, 20, 21], for example) for $Re_1$ for a very low concentration (a single sphere). As an illustration we use the result in [22], obtained by the method of merged asymptotic expansions in a small Reynolds number for the viscous flow around a single sphere. The two-term expansion in the Reynolds number for the vortex boundary agrees well with the data in [21] to Re = 60 [23]. The distance between the sphere surface and the vortex boundary along the stream axis is hence determined by the formula

$$\lambda = 0.25\ (\sqrt{1 + 3\mathrm{Re}} - 5).$$

It is seen that $Re_1 = 8$. Since Pe = ReSc, then $Pe^{-1/3}$ varies slowly with the increase in the Reynolds number, where Sc is the Schmidt number, and for ordinary fluids $Sc \sim 10^6$. Assuming $10 < Re_2 < 30$, we obtain $0.131 < Pe^{-1/3} < 0.135$. Hence by considering $\lambda = 4Pe^{-1/3}$ the selection criterion for $Re_2$, we obtain $Re_2 = 17$.

Using the linear approximation, we obtain the following approximate dependence

$$3\gamma = \begin{cases} 1, & \mathrm{Re} < \mathrm{Re}_1, \\ (\mathrm{Re}_1 - \mathrm{Re})\,(\mathrm{Re}_1 - \mathrm{Re}_2)^{-1}, & \mathrm{Re}_1 \leqslant \mathrm{Re} \leqslant \mathrm{Re}_2, \\ 0, & \mathrm{Re}_2 < \mathrm{Re} \end{cases} \tag{7}$$

for the index $\gamma = \gamma(\mathrm{Re}, \rho)$ (6) in the whole range of stationary laminar flow around a sphere.

Expressions (6) and (7) take account of both the change in the velocity field and the concentration due to the presence of other particles in the stream, and yield qualitative agreement with experimental results. They show that for moderate Reynolds numbers the mean diffusion flux per array particle can be considerably less than the individual flux calculated without taking account of the diffusion interaction, and for sufficiently high Reynolds numbers agrees with that value.

3. The results obtained permit determination of the mean values characterizing the chemical reaction rate observed in a disperse system with regular packing of the spherical particles. We still consider $Re < Re_1$, i.e., there is no attached vortex in the neighborhood of the rear stagnation point.

Let us assume that the mean concentration of the diffusing substance varies sufficiently slowly so that sufficiently many particles are contained at distances of the characteristic change therein. Let us introduce a representative volume sufficiently smaller than the scale of the change in concentration but containing a large quantity of particles.

Taking into account that the mean concentration (the concentration outside the diffusion boundary layers and wakes of the particles is also later called the concentration in the flow core) does not vary in practice at distances on the order of the distance between particles, we obtain from (5) for the mean diffusion flux per individual particle (here and below dimensional quantities are used; the quantities $a$, D, and c are used as scales for the dimensionless diffusion flux)

$$\langle I \rangle = A(n)\, aD\mathrm{Pe}^{1/3} n^{-1/9} x^{-1/3} c(x) \quad (c(0) = c_0). \tag{8}$$

Here c = c(x) is the concentration in the flow core; x, coordinate along the stream, x = 0 corresponds to the entrance to the layer, while the constant A is defined in (3); n, number of particles per unit volume. The relationship $N_1 = xn^{1/3}$ is used in deriving (8).

From (8) and the equation for the concentration (in the flow core)

$$-Udc/dx = n\langle I \rangle; \quad x = 0,\ c = c_0 \tag{9}$$

we obtain the distribution of the concentration along the stream in the flow core

$$c = c_0 \exp(-Fx^{2/3}), \quad F = 3/2A(n)\, a^{4/3} U^{-2/3} D^{2/3} n^{8/9}. \tag{10}$$

Hence we obtain for the distribution of the mean diffusion flux in a dispersed system

$$\langle I \rangle = c_0 A(n)\, a^{4/3} U^{1/3} D^{2/3} n^{-1/9} x^{-1/3} \exp(-Fx^{2/3}). \tag{11}$$

The results obtained above for the mean flux also permit calculation of the effective volume reaction rate constant in a dispersed system. The reaction is of first order, and is due to the progress of the surface reactions at the particles. From (9) and (11) we obtain an expression for the effective volume reaction rate constant

$$k = k(x) = A(n)a^{4/3}U^{1/3}D^{2/3}n^{8/9}x^{-1/3}. \quad (12)$$

Let us note that the rate constant depends substantially on the particle concentration and decreases rapidly along the flow. This distinguishes it considerably from the rate constant calculated without taking account of the diffusion interaction; in this case the rate constant is constant.

Therefore, the diffusion interaction of particles in a dispersed system with regular packing of the particles results in a change of the effective volume reaction rate constant for high Peclét numbers.

Results (8)-(12) can easily be extended to the case when there is a stationary attached vortex behind each sphere, i.e., $\mathrm{Re}_1 \leqslant \mathrm{Re} \leqslant \mathrm{Re}_2$. In particular, for the effective volume reaction rate constant

$$k = A(n)a^{4/3}U^{1/3}D^{2/3}n^{5/9+\nu}x^{-\nu}.$$

It is seen that in this case the reaction rate constant decreases more slowly with distance along the stream.

## NOTATION

$a$, sphere radius; $\mathbf{e}_i$, directional vector of the fundamental cell; $l_i$, length of the parallelepiped edge; U, mean stream velocity; v, stream velocity, $\psi$, stream function; r, $\theta$, spherical coordinate system connected to a sphere; c, concentration; D, coefficient of diffusion; $\mathrm{Pe} = aU/D$, Peclét number; Re, Reynolds number; $\mathrm{I}(\mathbf{k})$, total diffusion flux at the $\mathbf{k}$-th sphere; $\langle \mathrm{I} \rangle$, mean diffusion flux; $W^{(i)}$ $(i = 1, 2, 3, 4)$, diffusion wake domains; $\lambda$, distance from a sphere surface to the vortex boundary along the stream axis; n, number of spheres per unit volume; x, coordinate along the stream.

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# STATIONARY TRANSFER IN FIBROUS COMPOSITE MATERIALS

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The effective heat-conduction coefficients are calculated for fibrous materials of different structure.

The general methods of investigating transfer processes in heterogeneous systems developed in [1, 2] in application to dispersed media with spherical particles can be used successfully also in describing these processes in materials of a different structure. Stationary transfer in fibrous materials, which is used extensively in engineering and being of considerable applied interest, is examined below in the example of the heat-conduction process.

In the general case, materials consisting of a continuous medium and fibers with different physical properties distributed therein are not isotropic, where the nature and degree of the anisotropy are determined by the fiber packing features. Taking into account that the mean heat flux $\mathbf{q}$ and the mean temperature gradient $\tau$ are real vectors, we see that the quantity $\Lambda$ must be considered a real tensor of the second rank in the linear relationship

$$\mathbf{q} = -\Lambda\nabla\tau, \tag{1}$$

replacing the Fourier law in the case under consideration. The effective heat-conduction coefficients can comprise a nonglobal tensor in other dispersed media also, e.g., in media with spheroidal particles having a preferred direction of orientation of their axes of symmetry [3]. Equation (1) can be obtained strictly by taking the average of the local Fourier relationships, which are valid within and outside the fibers in either small physical volume (in this case the linear scale of the quantities $\mathbf{q}$ and $\tau$ should considerably exceed the internal structural scale of the material) as was done in [1], or in the ensemble of admissible fiber configurations analogous in meaning to the ensemble of configurations of systems of rigid spheres studied in [2].

Let us investigate a material with extended parallel fibers first. The cross section of each fiber is a circle of radius $a$; the centers of such circles are arranged randomly

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