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CONVECTIVE MASS TRANSFER BETWEEN A SOLID SPHERICAL PARTICLE AND A FLUID AT LARGE PÉCLET NUMBERS

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Introduction

Mass exchange between a solid particle and a fluid is the main productive factor in many branches of chemical engineering and microbiology. This process is complicated and depends on many factors – radial diffusion, forced convection, free convection, the shape of the particle, the physical properties of the fluid and the particle, etc. Under actual conditions these factors act simultaneously, and many investigators have been confined, as a rule, to the study of mass exchange under the conditions of a uniform oncoming stream, where forced convection makes the largest contribution to mass transfer. This is explained by the relative simplicity of the investigation of processes of this kind, both theoretically and experimentally.

All the experimental methods used to investigate mass exchange in fluid–solid-body systems can be divided into two major groups, in which the following are used: the dissolving of solids in fluid streams; chemical transitions taking place at the surfaces of metallic electrodes under the action of an electric current. The advantages of the first group are the simplicity of the experimental procedure, the possibility of forming the desired hydrodynamic pattern with a high degree of accuracy, and the presence of a wide class of substances suitable for such an experiment, which enables one to vary the physical properties of the fluid and bodies within wide ranges. The main drawback is that a fixed phase interface is absent, as a consequence of which the hydrodynamics of flow over the particle constantly varies. Electrochemical methods are free from such a drawback, since the processes taking place at the electrodes proceed in opposite directions and the surface of the sensor is always clean, but there is the technical problem of fastening the sensor and reducing the influence of conducting contacts.

A number of reports are known which are devoted to the investigation of the intensity of mass exchange in uniform streams. The good agreement between results obtained by the electrochemical method and the data of other authors for Re = 400–12,500 was noted in [1]. The test data published in [2] cover a far wider range of Reynolds numbers, Re = 2–23,000. The approximation curve generalizing the data for $2 < \mathrm{Re} < 20$,

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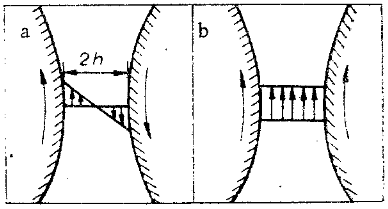

Fig. 1

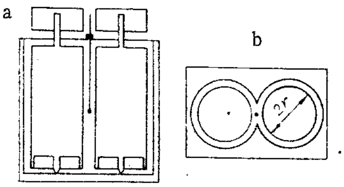

Fig. 2

$$\mathrm{Nu} = 1.032\ \mathrm{Re}^{0.385} \cdot \mathrm{Pr}^{1/3},$$

agrees well with theoretical functions obtained in [3, 4]. But unfortunately, there is very little experimental work carried out in the range of $\mathrm{Re} < 1$, since only here can a comparison be made with the existing theoretical functions. In [5] data are presented for the range of $0 < \mathrm{Re} < 2.5$, and they are well described by an approximation curve in the form

$$\mathrm{Nu} = 1.10 \mathrm{Re}^{1/3} \mathrm{Pr}^{1/3}.$$

An expression for the intensity of mass exchange between a sphere and a fluid under the conditions of a uniform oncoming fluid stream with $\mathrm{Re} \ll 1$ and $\mathrm{Pe} \gg 1$ was obtained in [3] in the boundary-layer approximation:

$$\mathrm{Nu} = 1.075 \mathrm{Pe}^{1/3}. \tag{1}$$

There this relation was also confirmed experimentally by dissolving a sphere of pressed benzoic acid in vegetable oil ($\mathrm{Re} = 0.1$–2.5). The value of the coefficient was found from the experiment to be 1.1, which is close to the theoretical 1.075.

The study of mass exchange in a fluid–solid-body system in the presence of a transverse velocity gradient is of far greater interest, however. A start for this was laid down in [6], where mass diffusion from an instantaneous point source in a horizontal stream whose velocity varies linearly along the vertical was considered. In it an expression was obtained for the concentration distribution in the vicinity of the source. The next step was to solve the problem [7] of heat and mass transfer from a sphere and a cylinder which are in a suspended state in a stream containing a velocity gradient (with shear). In the case of $\mathrm{Pe}_* \ll 1$ a solution was obtained both for a cylinder and for a sphere, while for $\mathrm{Pe}_* \gg 1$ a solution was obtained only for a cylinder. The difficulties arising in solving the problem for a sphere are explained by the three-dimensionality of the flow pattern. In [8] the results obtained earlier for a cylinder were confirmed experimentally. The suspended state was simulated by placing the cylinder on the null streamline between two walls moving in opposite directions. In [9] the problem of mass exchange between a fluid and a sphere rotating freely with an angular velocity equal to the curl of the fluid velocity was solved, and a ratio $\mathrm{Nu} = 9.0$ was found. The solution has such a form because in this case a region with closed streamlines is formed around the sphere through which mass transfer occurs by means of diffusion. Finally, the problem of diffusion to a stationary sphere in a shear stream of viscous fluid with $\mathrm{Pe}_* \gg 1$ was solved in [10]. This problem was solved in the most complete statement in [11]. The equation of convective diffusion

$$\mathbf{v} \cdot \nabla c = D \nabla^2 c$$

with the boundary conditions $c = c_0$ at the surface and $c = c_\infty$ far from the sphere where the velocity $\mathbf{v}$ varies linearly with distance was solved for two limiting cases. For $\mathrm{Pe}_* \ll 1$ an expression of the type

$$\mathrm{Nu} = \alpha\, \mathrm{Nu}_0^2 \mathrm{Pe}_*^{1/2}$$

was obtained, where Nu_0 is the Nusselt number for $\mathrm{Pe}_* = 0$ ($\mathrm{Pe}_* = \varepsilon d^2/D$ and ε is the value of the velocity gradient). For $\mathrm{Pe}_* \gg 1$ the expression for the intensity of mass exchange has the form

$$\mathrm{Nu} = \beta\, \mathrm{Pe}_*^{1/3}, \tag{2}$$

where $\beta = 1.134$ for any type of linear velocity field.

A single attempt to experimentally test the functions obtained is known so far. The process of heat exchange between a solid spherical particle and a fluid under the conditions of two-dimensional, purely deformation flow, which was modeled by the rotation of four round cylinders, was investigated in [12]. The spherical sensor also comprised the heater, the heat from which spread through the entire volume of fluid. It was mounted at the center of the working volume. The data for Pe = 0.33-24.0 agree qualitatively with Batchelor's theoretical function for $Pe_* \ll 1$, but there is some quantitative disagreement.

As seen from the review, the amount of experimental work devoted to this question is not very great. Therefore, it is hard to judge the validity of one or another theory, and a careful and repeated testing of the available theoretical functions is necessary.

Experimental Apparatus

The main difficulty in the preparation of the experiment consisted in the choice of the means of obtaining the appropriate hydrodynamic conditions, the realization of which would require little expense and the construction of which would not be complicated. As a result, we executed the idea of modeling plane Couette flow, when the walls move in opposite directions, using cylinders large enough in size compared with the test object. If large cylinders are placed side by side in the plane passing through their rotation axes and they are rotated in the same direction, then one obtains the pattern shown in Fig. 1a, i.e., flow of the type of plane Couette flow with a linear profile of velocity $\mathbf{v}$. When the cylinders are rotated in opposite directions the uniform velocity profile shown in Fig. 1b is formed between them. Naturally, this is valid only in a certain region, the size of which can be increased by increasing the diameter of the rotating cylinders. In our case the size of such a region has the order of the size of the gap between the cylinders and is far greater than the size of the test object, the sphere.

In practice, this was embodied in the experimental apparatus for which a diagram is presented in Fig. 2, where a is a side view and b is a top view. The entire experimental apparatus was made of plastic, which provides transparency and stability against active media, which the working fluid is. The cylinders have a height of $2 \cdot 10^{-1}$ m and a diameter $2r = 10^{-1}$ m and the minimum distance between them is $2h = 10^{-2}$ m. The rotation period of the cylinders was recorded with photodiodes and was varied in the range of T = 8.0-130.0 sec. The measurement error was 2-3% at the largest values of T. To avoid the influence of free convection due to a temperature gradient, the working section was fully immersed in a jacket containing a liquid, where a temperature of 20°C was maintained with an accuracy of 0.1°C with the help of a thermostat. To avoid the disturbing influence of the bottom, we did the following: Cavities were made in the lower part of the cylinders which remained full of air when the working volume was filled with fluid. Thus, we created an effect as if there is a free surface at the bottom, and only the sharp rim and not the entire bottom part of the cylinders is a source of disturbances.

An important feature is the possibility of obtaining the required hydrodynamic environment between the rotating cylinders. For this purpose we used the method of stroboscopic visualization to obtain velocity profiles of the fluid, one of which is shown in Fig. 3. About 200 points were required to obtain each velocity profile, with points being taken from a region representing a square with sides of 10^{-2} m. Since the measured values of the velocity from both parts of the profile are shown on one graph, the flow pattern between the cylinders is symmetric and the null streamline lies at the center of the gap for any plane cross section. We add that the average relative error in measuring the velocity in this way was 3% in the given experiment. For certain cylinder rotation periods, however, Taylor vortices were formed in the system and the mechanism of mass transfer was sharply altered, causing an abrupt increase in the intensity of mass exchange. Therefore, measurements were not made under such conditions.

In the case when the cylinders rotate in opposite directions the profile of mass-exchange intensity was measured along the line connecting their rotation axes. For each fixed value of T the value of Nu was constant with an accuracy of about 5%. From this it follows that uniform fluid flow occurs between the cylinders.

We certainly must note the fact that the streamlines around the cylinders are approximately circular, so that the necessary conditions are satisfied only in a limited region. But the ratio of the size of the cylinder to that of the solid spherical test particle is very large; depending on the size of the sensor sphere it varied in the range of 100-200. Then the streamlines can be taken as straight in a certain vicinity of the sphere. As the experiment showed, this assumption was fully justified.

Experimental Procedure

The intensity of mass exchange between a spherical particle and a fluid was measured by the electrochemical method [13]. Its essence consists in measuring the rate of an oxidation–reduction reaction taking place at an electrode in a diffusion mode. It is very convenient to investigate mass-exchange processes by the electrochemical method, since the form of the reaction is such that the sensor surface is always clean. In addi-

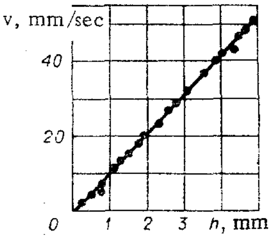

Fig. 3

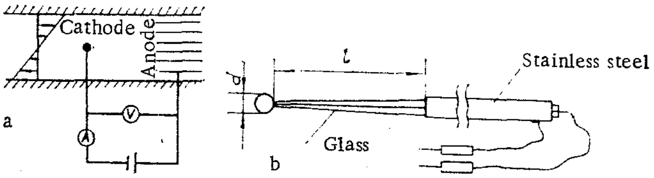

Fig. 4

tion, the detector itself is the test object, which considerably simplifies the construction of the measurement part of the apparatus.

The electrochemical cell used in the experiment is shown schematically in Fig. 4a. The spherical sensor was located at the center of the gap between cylinders, i.e., on the null streamline. The position of the center was determined from the minimum of the mass-exchange intensity by moving the sensor along the line connecting the centers of the cylindrs. The electrochemical sensor has the form shown in Fig. 4b and is made by the following technique: A platinum wire $1.5 \cdot 10^{-4}$ m in diameter was melted from the end with a gas burner until a sphere of the required size and desired shape was obtained. Then the wire, which serves as a conducting contact in this case, was covered with chemically resistant glass with a thickness on the order of several dozen microns and which gradually thickened and reached a diameter of $1.5 \cdot 10^{-3}$ m at a distance of $2 \cdot 10^{-2}$ m. Then the sensor was cemented into a stainless-steel tube $2.5 \cdot 10^{-3}$ m in diameter, with the conducting contact being carefully insulated. We note that the metal tube serves as the anode in this case, and the ratio of the anode and cathode areas varied in the range of 100-500, depending on the size of the sensor.

The mass-exchange intensity in the experiment was determined from the formula

$$\mathrm{Nu} = Id/(FSc_f D), \tag{3}$$

where I is the diffusion current; d is the diameter of the spherical sensor; F is the Faraday constant; S is the area of the sphere; c_f is the concentration of reactive ions; D is the coefficient of diffusion of reactive ions. We measured c_f by the method of potentiometric titration, and the value of c_f was determined from the formula $c_f = V_k c_k / V$, where V is the volume of solution taken for titration; V_k is the volume of the cobalt solution with a concentration c_k added to the equivalence point. This method is quite accurate; its error is no more than 0.5%. The coefficient of viscosity of the solution was measured with a standard viscosimeter. To determine the values of the coefficient of diffusion D we used the formula

$$\mu D/t = 2.36 \cdot 10^{-15} \ \mathrm{kg \cdot m/sec^2 \cdot K},$$

obtained in [14] for the same electrolyte. The error of this formula was calculated in the same article and does not exceed 4%. This relation was also used for solutions containing a weight concentration of glycerin in the electrolyte of up to 40%; the corresponding range of Prandtl numbers $\mathrm{Pr} = \nu/D$ is 1700-18,400. The diameter of the spherical sensor was measured with a measuring microscope with a scale division of 10^{-6} m, and the difference between the maximum and minimum values of the diameters of any sensor used did not exceed 10^{-5} m. In the calculations the average was taken as the true value of the diameter.

To ascertain the correctness of the results obtained for mass exchange in streams with a velocity gradient, the results obtained were analyzed by two independent means. In the first case the analysis was carried out in accordance with Eq. (3) and in the second case a calibration curve obtained in a uniform stream was used. There is no doubt of the correctness of the theoretical formula

$$\mathrm{Nu} = 0.991 \, \mathrm{Pe}^{1/3} \tag{4}$$

from [4], since it has been confirmed repeatedly in experiments, including the one in the present work. Then graphic functions of the type Nu = f(I) were constructed for a uniform stream and then the mass-exchange intensity in a stream with a transverse velocity gradient, i.e., the Nusselt number, was determined from the diffusional current.

Discussion of Results

The present work was devoted to an experimental study of the influence of a transverse velocity gradient on the mass exchange between a fixed spherical particle and a fluid. In addition, we made measurements, which had an auxiliary character, of the intensity of mass exchange between a sphere and a fluid, since they make it

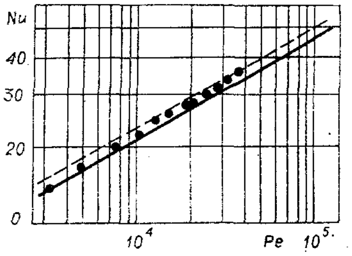

Fig. 5

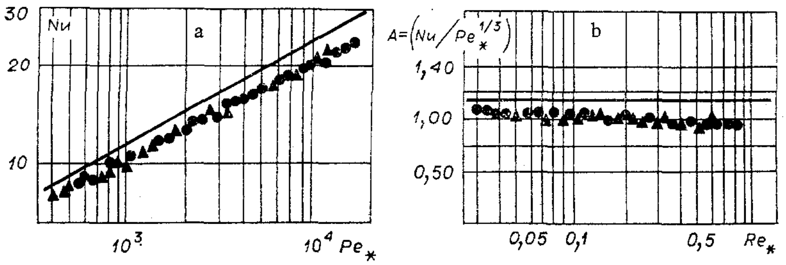

Fig. 6

possible to judge the validity of the results for fluid flows with a velocity gradient. Data for a uniform stream are presented in Fig. 5 and correspond to Re = 0.4-20; nevertheless, their good agreement with the theoretical functions (1) and (4) (dashed and solid lines, respectively) is evident in the graph. It is seen that the experimental data lie in the space between these curves. From this the conclusion follows that either formula can be used with equal success in practical calculations with an error of ±4%. Here the dimensionless complexes are chosen as follows: $Pe = RePr$ and $Re = vd/\nu$, where $v = 2\pi r/T$.

Data on measurement of the intensity of mass exchange between a sphere and a fluid in a stream with a transverse velocity gradient are presented in Fig. 6a. The simplest analysis on the basis of this graph allows us to draw the following conclusion: The theoretical function (2) obtained by Batchelor and the experimental data of the present work agree sufficiently well with each other. Consequently, the range of Péclet numbers Pe_* realized in the experiment satisfies the assumptions which were made in the theory. The slope of these curves on the logarithmic scale are different, however, which indicates the influence of Re_*.

Since the theory was obtained for the "creeping" mode of flow over particles, with a decrease in Re_* the experimental and theoretical functions almost coincide. The tendency of the theoretical and experimental curves to approach with a decrease in $Re_* = \varepsilon d^2/\nu$ (where $\varepsilon = v/h$ is the velocity gradient) is well seen in Fig. 6b. For $Re_* < 0.1$ the mean deviation of the coefficient A from the straight line corresponding to A = 1.134 is less than 6-7%. Unfortunately, it was not possible to widen the interval through a further decrease in Re_*, since a glycerin content of more than 40% in the electrolyte results in instability of the solution. On the other hand, the size of the sensor cannot be reduced further, since the conducting wires have a finite size and their disturbing influence increases with a decrease in the sphere. In the sensors used the area of contact between the conducting wires and the spherical sensor did not exceed 2% of the entire area of the sphere. For this case $Re_* = 0.03-1.0$.

It was already mentioned above that for streams with a velocity gradient the measured values were analyzed by two independent means, and therefore two types of points are presented in Fig. 6. It is seen from the graphs that both types of points lie on one curve with a high degree of accuracy.

Thus, the experimental data obtained agree well with theoretical functions for the intensity of mass transfer from a single sphere to a fluid in a uniform stream and in a flow of the simple shear type, obtained in the boundary-layer approximation.

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MIXING OF FINELY DISPERSED VAPORIZING PARTICLES WITH A DEFLECTING GAS STREAM

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In a number of problems of aerogas dynamics it becomes necessary to introduce finely dispersed particles into a gas stream. Such problems are connected, for example, with the design of prospective cryogenic and high-enthalpy wind tunnels [1, 2], visualizing devices [3, 4], accelerators of macroscopic particles for the investigation of surface erosion, the creation of active media for gasdynamic lasers using multiphase mixing [5, 6], etc. In this case certain demands may be placed on the distribution of parameters in the mixing zone, such as the macroparticles or the mass density of the gas formed as a result of their vaporization, the momentum of the particles, or any other aspect of their mass spectrum. In particular, the investigation of the conditions required to realize the advantages of multiphase mixing in gasdynamic lasers (GDL) [5, 6] led to the need to solve new gasdynamic problems which did not arise in the traditional devices for amplifying radiation [7, 8]. The statement and solution of a number of these problems are given in [9-11]. They include the problem of the maximum possible acceleration of aerosol particles by different gases, the problem of determining the depth of penetration of particles into a comoving stream and the time of their vaporization as a function of diameter, and problems of the influence of compression shocks and viscous effects, including detachment zones, the mutual influence of vaporization and vibrational relaxation, and some others. One of the central problems is the obtainment of the flow field in the mixing zone which is closest to uniform, since constancy of the gasdynamic parameters and concentrations of the components of the gas mixture is very important, for example, in the zone of action of a GDL resonator.

In [6, 10] the mixing problem was considered in an integral statement based on the use of conservation laws with allowance for the supply of a certain mass, momentum, and energy to the working stream. Such an approach makes it possible to determine the variation of pressure, temperature, and other parameters as a result of mixing, although the flow structure directly in the mixing zone remains indefinite. The detailed investigation of this structure is needed in connection with the finiteness of the vibrational relaxation time τ, and hence with the boundedness of the region of mixing of two streams in which population inversion and radiation amplification can occur. Equalization of the profiles of different parameters across a mixing zone must take place just in a region of $l \lesssim u\tau$, where u is the characteristic velocity in the mixing zone. And the present

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