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HYDRODYNAMIC SITUATION AND TRANSFER PROCESSES IN THE STATIC GRANULAR BED OF A CATALYST

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Experimental data on the distribution of the local mass-transfer coefficients on the external surface of a spherical particle in a monodisperse static granular layer are considered. The observed rate of chemical change for a nonequiaccessible surface of the catalyst grains is determined.

It is shown in [1–2] that the free volume of a fixed layer of particles through which a gas or liquid flows is nonuniform – there is flowing region which is a jet, and a nonflowing region situated in the neighborhood of the points of contact between the particles. This hydrodynamic situation leads to nonequiaccessibility of the external surfaces of the particles, which has been experimentally studied in a number of papers [2–8]. In this paper we investigate the flow and transfer processes in a static granular layer and their effect of the rate of chemical change on the surface of the grains of catalyst. For this purpose we will use the experimental relations between the local Nusselt numbers on a grain in the static granular layer and the Reynolds number, which were electrochemically obtained [3], and we will compare them with the well-known theoretical relation

$$\mathrm{Nu} = A\,\mathrm{Re}^{n},$$

corresponding to a certain structure of the flow and transfer mechanism. Experimental curves of $\mathrm{Nu} = A\mathrm{Re}^{n}$ for certain points on the surface of the particle are shown in Fig. 1. It can be seen from Fig. 1 that for points I ($\Theta = 22°$) the exponent $n = \tan\alpha = 0.5$, which corresponds to transfer processes in the laminar boundary layer in the region of running against the flow [9]. For points II ($\Theta = 135°$) the nature of this dependence changes: as the Reynolds number increases for $10 < \mathrm{Re} < 200$, $n = 0.5$, while for $200 < \mathrm{Re} < 2500$, $n = 0.65$, which corresponds to transfer processes in the laminar boundary layer in the region of separation for turbulent basic flow [9]. For points III ($\Theta = 157°$) up to $\mathrm{Re} \approx 250$ the local Nusselt number is independent of the Reynolds number, and it is then found to increase slightly. In this case $n \approx 0.3$, which corresponds to a viscous type of flow around this point, for which [10]

$$\mathrm{Re}_{l} = \frac{U_{l} d}{\nu} \ll 1.$$

The absence of a dependence of Nu on Re in the region $10 < \mathrm{Re} < 250$ indicates a molecular transfer mechanism at this point. It follows from this that the point considered is in the contact region between solid particles. However, as the Re number increases this region contracts, and the same point will lie in the viscous flow region.

For points IV ($\Theta = 225°$) the Nusselt number is independent of the Reynolds number over the whole range of numbers $10 < \mathrm{Re} < 2500$, i.e., the point IV is in the contact region.

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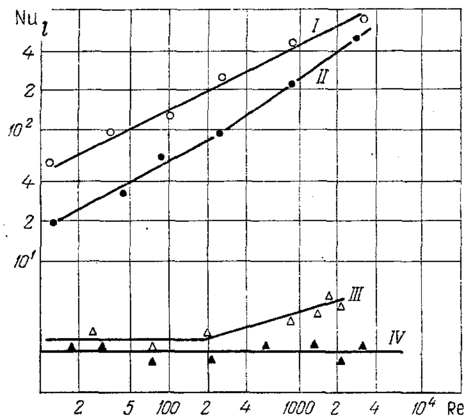

Fig. 1. Dependence of the local Nusselt numbers (Nu_l) on the Reynolds number ($Re = ud/\psi\nu$) for different values of the angle Θ with respect to the direction of flow: I) $\Theta = 22°$; II) 135°; III) 157°; and IV) 225°.

Carrying out a similar analysis for different points on the surface of the particle in the layer and taking into account the fundamental properties of detached flow, the local structure of the flow can be represented in the form of separate regions: I, region where the flow runs against the particle; II, region of displacement; III, region of vortex formation; IV, region of viscous flow; and V, region of contact.

The region of running against the particle and of vortex formation consists of the main body of the flow and the boundary layer.

On the basis of the results of an analysis of the distribution function of the exponent corresponding to a definite transfer mechanism and flow structure (Fig. 2), we can represent the difference in the flow and the mechanism of the transfer processes as the Reynolds number increases.

For small Reynolds numbers $10 < Re < 90$ the main flow is laminar and the surface of the particle in the region where the flow is against the particle is about 15% (of which 10% is on the upper part of the frontal surface and 5% close to the equator in the stern part). On the front part in the region where the flow is against the particle, a laminar boundary layer is formed which then breaks away due to the presence of a pressure gradient. The flow here has a viscous nature, i.e., the local Reynolds number $Re_l < 1$. The vortex region with a viscous nature of the flow occupies about 35% of the whole surface, of which 15% is on the frontal part, and 20% on the stern part. The remaining part (15% of the surface) is in the region of static liquid close to the point of contact, and is ≈25% on the frontal part of the surface and 25% on the rear part of the surface.

As Re increases the velocity in the main flow and in the vortex-formation region increases. The fraction of the surface around which the laminar boundary layer flows increases up to 40%. Obviously, this increase is due to the formation of the laminar boundary layer in the region of the vortex formation. In this region secondary separation of the boundary layer and the secondary vortex occurs with a viscous nature of the flow, which flows around about 35% of the surface of the particle (of which 15% is on the frontal surface and 20% on the rear surface).

The fraction of the surface which is in the static liquid is reduced to 25%.

When the Reynolds number is increased further in the region $900 < Re < 2500$, part of the vortex breaks away from the particle surface and is carried downward with respect to the flow, causing turbulence in the main flow.

The boundary layer remains laminar. The region of flow against the particles broadens to 40%, the part of the vortex-formation region with a laminar boundary layer broadens to 40%, the other part with viscous flow decreases to 15%, while the region with static liquid decreases to 5%. In this region of the Reynolds numbers in the vortex breakaway zone pulsations of the local mass-transfer coefficients are observed. When Re is increased further there is a transition to turbulent flow both in the main flow and in the region of vortex formation, and correspondingly a transition from a laminar to a turbulent transfer mechanism in the boundary layer. In accordance with the change in the flow pattern and the change in the Reynolds number, the nonequiaccessibility of the external surface of the particles in the granular layer also changes (Fig. 3).

Figure 3 shows examples of the distribution functions of the local mass-transfer coefficients on the surface of a spherical particle in an unordered layer of similar spheres constructed on the basis of experimental data. As follows from the figure, the local exchange coefficients may differ from one another by a factor of 5-10, and this difference decreases as Re increases. The data obtained agree well with the data given in [4, 8]. It follows from the experiments that in the $Re = 20\text{-}10^5$ the maximum values of the exchange coefficients may

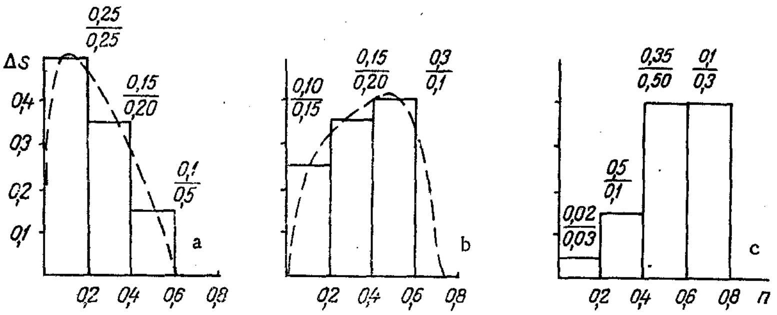

Fig. 2. Distribution function of the exponent n of the local Reynolds number Re in the relation $Nu_l = ARe^n$ (the dashed curve), and a histogram (a – Re = 9–90, b – 90–250, c – 250–2500).

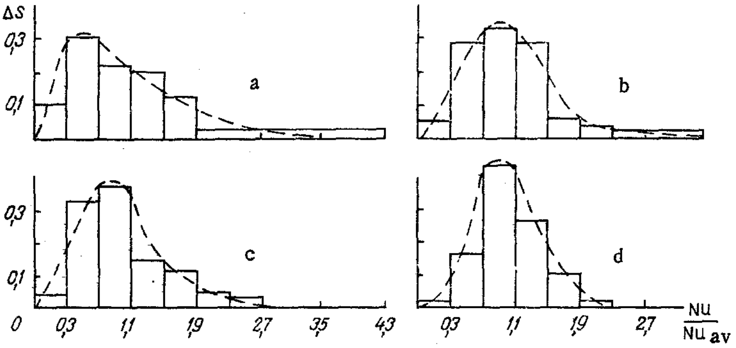

Fig. 3. Distribution function of the local mass-transfer coefficients on the external surface of a spherical particle in a granular layer for Re equal to 9 (a), 90 (b), 250 (c), and 2500 (d) (the dashed curves) and a histogram.

differ from the average by a factor of 1.5–2.5, and this difference increases as Re increases.

In this range of Re numbers the variation coefficient ϑ_{Nu} determining the degree of nonequiaccessibility of the surface of the particle varies from ~100 to 20%. It also turns out that the direction of the flow of gas or liquid acquires a certain order under exchange conditions – the exchange coefficients on the front part of a grain is greater than the exchange coefficients on the rear part by a factor of ≈1.5.

We will now consider the effect of nonequiaccessibility along the external surface of the grains of the catalyst in a static granular layer on the observed rate of chemical change. Suppose the rate of the chemical reaction is $w(c_r)$. We can then write the following expression at any point of the surface of the particle:

$$\beta(s)\,[c_0 - c_r(s)] = w\,[c_r(s)]. \tag{1}$$

If $w(c) = kc$, then

$$c_r(s) = \frac{Nu(s)\,c_0}{\delta + Nu(s)};$$

if $w(c) = kc^2$,

$$c_r(s) = -\frac{1}{2}\,\frac{Nu(s)}{\delta} + \sqrt{\frac{1}{4}\,\frac{Nu(s)^2}{\delta^2} + \frac{Nu(s)\,c_0}{\delta}};$$

if $w(c) = kc^{1/2}$,

$$c_r(s) = \frac{1}{2}\left(2c_0 + \frac{\delta^2}{Nu(s)}\right) - \sqrt{\frac{1}{4}\left(2c_0 + \frac{\delta^2}{Nu(s)^2}\right) - c_0^2};$$

if $w(c) = kc/(1 + Ac)$, then

$$c_r(s) = -\frac{1}{2}\left(\frac{\delta}{Nu(s)\,A} + \frac{1}{A} - c_0\right) + \sqrt{\frac{1}{4}\left(\frac{\delta}{Nu(s)\,A} + \frac{1}{A} - c_0\right)^2 + \frac{c_0}{A}}.$$

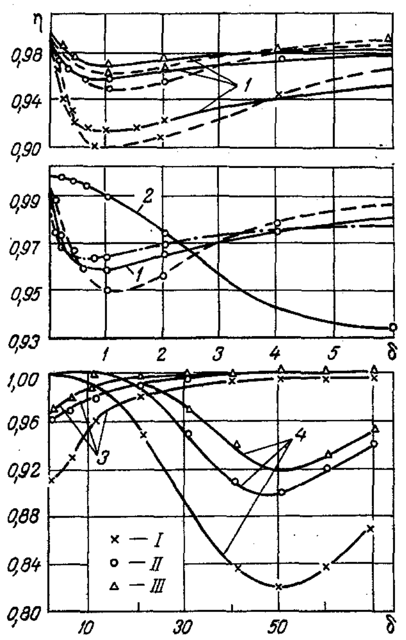

Fig. 4. Effect of nonequiaccessibility of the external surface of a grain of the catalyst on the observed rate of chemical change: 1) A = 0, 2) 10, 3) 20, 4) 50; I) Re 9, II) 90, III) 900; the continuous curves are $w - kc/(1 + Ac)$; the dashed curves are $w = kc^2$, and the dash-dot curves are $w = kc^{1/2}$.

The observed rate of the chemical change referred to unit external surface of the grain is

$$w_{\text{av}} = \frac{1}{s}\int_0^s w\,[c_{\text{r}}(s)]\,ds. \tag{2}$$

Usually when calculating catalytic processes, the following expression is used for the rate of the reaction:

$$w\,(c_{\beta_{\text{av}}}) = \beta_{\text{av}}(c_0 - c_{\beta_{\text{av}}}), \tag{3}$$

which is calculated from the average value of the mass-transfer coefficient

$$\beta_{\text{av}} = \frac{1}{s}\int_0^s \beta\,(s)\,ds. \tag{4}$$

The relation

$$\eta = w_{\text{av}}/w\,(c_{\beta\text{av}}) \tag{5}$$

can serve as a measure of the effect of the nonequiaccessibility of the external surface of a grain of the catalyst on the observed rate of chemical change. The results of calculations of this coefficient, taking into account the experimental distribution function of the mass-transfer coefficients for different kinetic behaviors of w(c), are shown in Fig. 4.

As can be seen from the figure the nonequiaccessibility of the external surface of a grain of the catalyst under isothermal conditions always reduces the degree of utilization of the catalyst compared with equiaccessible grains. However, this effect does not exceed 10% for reactions of the second, first, and half order. Only when the kinetic dependence of the process approximates to zero order, which corresponds to large values of the adsorption coefficient A, may this reduction reach 20%. Since the degree of nonequiaccessibility decreases as Re increases, it effect on the coefficient also decreases, which is well illustrated in Fig. 4. The greatest effect of nonequiaccessibility is observed when $w/c_{\text{r}}\beta_{\text{av}} \approx 1$. When the catalystic process occurs in the region of external diffusion, when the ratio $w/c_{\text{r}}\beta_{\text{av}} \gg 1$, this effect is negligible.

Hence, we can conclude that in practical situations when steady-state catalytic processes occur under conditions close to isothermal, when problems of selectivity, stability, and ignition are not important [11, 12], the effect of nonequiaccessibility on the degree of utilization of the external surface of grains of catalyst can be ignored.

NOTATION

$\mathrm{Re}=\frac{ud}{\psi\nu}$; $\mathrm{Nu}=jd/D(c_0-c_r)$; $\vartheta_{\mathrm{Nu}}=\frac{\sigma}{\mathrm{Nu_f}}100$; $\sigma=\sqrt{\frac{1}{n}\sum_{i=1}^{n}(\mathrm{Nu}_i-\mathrm{Nu_{av}})^2}$; $\delta=k/\bar{\beta}$; c, concentration; D, diffusion coefficient; d, diameter of a grain; j, specific flow of the material to the surface of the grain; k, reaction rate constant; s, external surface of the grain; $\Delta s(\mathrm{Nu})$, distribution function of the local mass-transfer coefficients on the external surface of the grain; u, rate of flow referred to the whole cross section of the layer; w, rate of chemical reaction; ψ, minimum useful cross section (0.17); β, mass-transfer coefficient; and ν, coefficient of kinematic viscosity. Indices: f and K, front and rear parts of the surface; av, average value; max, maximum value; r, surface; and o, free volume of the layer.

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EXPERIMENTAL INVESTIGATION OF THE RADIANT AND CONDUCTIVE–CONVECTIVE COMPONENTS OF EXTERNAL HEAT EXCHANGE IN A HIGH-TEMPERATURE FLUIDIZED BED

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The results of the experimental investigation of the components of complex external heat exchange in a high-temperature fluidized bed, by means of a radiometer and two α calorimeters with a different degree of surface blackness, are given.

In order to separate complex heat exchange into radiant and conductive–convective components, in this paper the combined heat-transfer coefficients to two α calorimeters, differing only in the degree of surface blackness, were measured. Each α calorimeter was a box with dimensions of 80 × 80 × 40 mm, made of Nichrome with a thickness of 5 mm, and filled inside with kaolin wadding.

The heat-transfer coefficients were determined by the heating or cooling rate of the side walls of the α calorimeter, in which a thermocouple was calked. In order to reduce thermal inflows from the end walls of the α calorimeter, which could be heated up differently than the side walls because of differences in their flow

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