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METHOD OF CALCULATING THE THERMAL CONDUCTIVITY OF POROUS GRANULAR MATERIALS WITH METAL FILLER IN DIFFERENT MEDIA

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The article suggests a method of calculating the thermal conductivity of porous granular materials with metallic filler. The results of the calculation are compared with the experimental data. The error of the calculation is commensurable with the error of specifying the initial data.

To intensify technological processes at high temperatures, porous granulated aluminum oxide with metallic filler is used as catalyst. The aluminum oxide is made in the form of granules with 0.8-1.25-mm diameter. In dependence on the temperature, the weight concentration of the metallic particles n_{met}, and the composition of the gaseous medium, the thermophysical properties of the material change, and this affects the conditions of its operation.

Investigation of heat transfer in charges of porous aluminum oxide with metallic filler enable us to evaluate the range of change of thermophysical properties in dependences on the above parameters.

We will carry out the analysis on a model in the form of a charge of grains (spheres) with the same diameter; the grains themselves have a porous structure, and in the walls of the pores and in its surface metal particles are embedded (Fig. 1).

We will consider the processes of heat transfer through such a structure stage by stage. At the first stage we evaluate the thermal conductivity of a porous grain of aluminum oxide without metal particles, assuming that the pores contain gas. We use the well-known model of cracked material whose components form an interpenetrating grid [1]. The thermal conductivity of cracked material is determined by the formula

$$\lambda' = \lambda_{Al_2O_3}[c^2M + \nu(1-c)^2 + 2\nu c\ (1-c)/(\nu c + 1 - c)], \quad \nu = \lambda_{por}/\lambda_{Al_2O_3}, \tag{1}$$

where the parameter c is correlated with bulk porosity by the dependence c = 0.5 − arc cos $(1 - 2m_2)$ for $m_2 < 0.5$; M is a parameter characterizing the cracked state of the material.

The thermal conductivity of the pores is determined by the radiant and molecular heat transfer, and it is equal to

$$\lambda_{por} = \lambda_r + \lambda_m. \tag{2}$$

The radiant component of thermal conductivity is evaluated by the formula for radiant heat exchange between two parallel plates whose degree of blackness is ε, and the distance between them is δ (pore size) [1]:

$$\lambda_r = \sigma_0 (T/100)^3 \varepsilon_{cd} \delta, \quad \varepsilon_{cd} = \varepsilon/(2-\varepsilon). \tag{3}$$

The molecular component of thermal conductivity depends on many parameters, and it is determined by the formula [1]

$$\lambda_m = \frac{\lambda_g}{1 + B/(H\delta)}, \quad B = \frac{4(c_p/c_v)}{1+(c_p/c_v)} \frac{2-a}{a} \Lambda H_0 \mathrm{Pr}^{-1}, \quad H_0 = 1 \cdot 10^5 \text{ Pa}. \tag{4}$$

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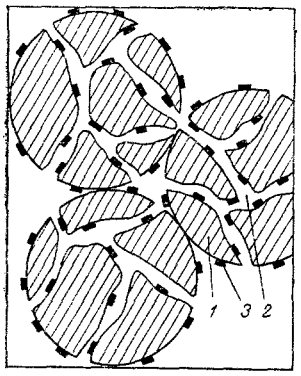

Fig. 1. Structure and model of an aluminum oxide charge containing a metallic filler: 1) aluminum oxide particles; 2) pores; 3) embedded metal.

At the second stage we evaluate the thermal conductivity of the material of the granule λ'', i.e., of cracked porous material with embedded metal particles λ_{met}. We will regard such granules as bicomponent material one component of which is aluminum oxide with thermal conductivity λ', and the other is metal. We describe the structure of the material by a model with insulated inclusions, and calculate its thermal conductivity by the formula [2]

$$\lambda''=\lambda'[1-m_{met}/((1-\nu')^{-1}-(1-m_{met})/3)], \tag{5}$$

where $\nu' = \lambda_{met}/\lambda'$, m_{met} is the bulk concentration of the metal correlated with the weight concentration by the dependence [1]

$$m_{me}=n_{met}\,[\rho_{met}\,(n_{met}/\rho_{met}+(1-n_{met})/\rho_{Al_2O_3})]^{-1}, \tag{6}$$

where ρ_{met}, $\rho_{Al_2O_3}$ is the bulk density of the metal particles and of the porous aluminum oxide, respectively.

At the concluding, the third stage we calculate the thermal conductivity λ of a granular system whose grains have thermal conductivity λ'', and between which there is gas as filler. The stacking of the grains (their coordinate number N) is determined by the porosity m'_2 for intergranular space. It is correlated with the total porosity m and the porosity of the material of the grains m_2 by an expression obtained on the basis of the following transformations:

$$V=V_1+V_2+V_3,\quad m_2'=\frac{V_1}{V},\quad m_1=\frac{V_3}{V},\quad m=\frac{V_1+V_2}{V},\quad m_1+m=1,$$

$$m_2=\frac{V_2}{V_2+V_3},\quad m_2'=\frac{Vm-V_2}{V}=m-\frac{V_2}{V-V_1}\,\frac{V-V_1}{V}=m-m_2(1-m_2'),\ m_2'=\frac{m-m_2}{1-m_2}. \tag{7}$$

The thermal conductivity of a granular system is determined by a method suggested in [1]:

$$\lambda=\lambda''((y_1^2/(0{,}5h_g+(1-0{,}5h_g)\Phi)+(D/y_3^2+(A/(1-0{,}5h_g-B+$$
$$+0{,}5h_g/\nu_{tg})+2\nu_g(D-F+w\ln((w-D)/(w-F)))/(1-\nu_g))^{-1})^{-1})+\nu_{2tp}E))/y_4^2,$$

where

$$A=y_2^2-y_1^2;\quad F=\sqrt{1-y_2^2};\quad D=\sqrt{1-y_3^2};\quad E=y_4^2-y_3^2;$$
$$w=[1-\nu_{tg}\sqrt{1-y_3^2}+B/(Hd)]/(1-\nu_{tg});\quad d=2r;$$
$$\nu_{tg}=\lambda_{tg}/\lambda_1;\quad \nu_{2tp}=\lambda_{2tp}/\lambda_1;\quad h_g=0;\quad \nu_g=\lambda_g/\lambda_1.$$

The radiant and molecular components of thermal conductivity are calculated by formulas that are analogous to (3), (4), with other dimensions of the gap taken into account. The calculation of the geometric parameters of the system is presented in [1]:

$$y_1=1\cdot10^{-2}y_2,\quad y_2=3{,}3\cdot10^{-3}\sqrt{(1-m_2')^{2/3}[p_{sp}+10\rho_1(1-m_2')h_{1r}]},$$
$$y_3=2\sqrt{N-1}/N,\quad y_4=y_3/\sqrt[3]{1-m_2'},$$
$$N=(m_2'+3+\sqrt{(m_2')^2-10m_2'+9})/(2m_2').$$

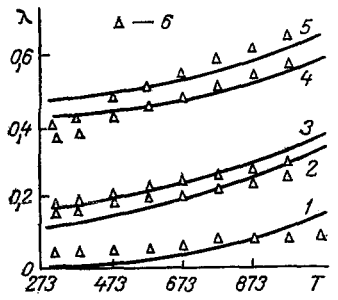

Fig. 2

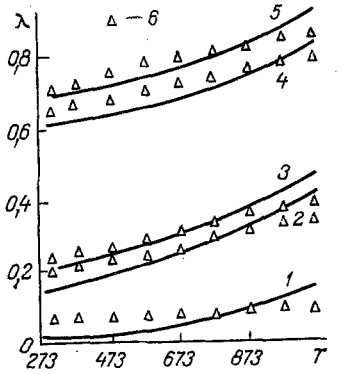

Fig. 3

Fig. 2. Thermal conductivity of porous granulated aluminum oxide without metallic fillers in different gaseous media and in vacuum: 1-5) calculated values (1) in vacuum; 2) in argon; 3) in nitrogen; 4) in helium; 5) in hydrogen); 6) experimental values. λ, W/(m·°K); T, °K.

Fig. 3. Thermal conductivity of porous, granulated aluminum oxide containing 30% embedded nickel particles in different gaseous media and in vacuum: 1-5) calculated values (1) in vacuum; 2) in argon; 3) in nitrogen; 4) in helium; 5) in hydrogen); 6) experimental values.

We used the method suggested above to calculate the thermal conductivity of granulated aluminum oxide, both in the pure state and containing nickel with a weight concentration of 30% in a medium of a gaseous filler (argon, nitrogen, helium, hydrogen) at normal pressure and in vacuum at different temperatures.

To compare the calculated data with the experimental ones, we measured the thermal conductivity of aluminum oxide in pure form and containing nickel with a weight concentration of 30% in the temperature range 293-1040°K in argon, nitrogen, helium, hydrogen at atmospheric pressure and in vacuum ($P = 8 \cdot 10^{-3}$ mm Hg).

For the investigations we took granulated aluminum oxide which is widely used as carrier in high-temperature catalytic processes (specific surface ~123 m^2/g; total pore volume ~0.35 cm^3/g; bulk density ~1 g/cm^3). The granules of aluminum oxide were of cylindrical shape and had the following dimensions: height 1.0-1.5 mm, diameter 0.80-1.25 mm.

Specimens containing nickel were made by the method of impregnating aluminum oxide with a solution of nickel nitrate with subsequent heat treatment in air and in hydrogen at 673°K.

The effective thermal conductivity of the specimens was measured by the method of the cylindrical bicalorimeter of regular thermal regime [3]. The experimental installation consisted basically of a cylindrical bicalorimeter thermostating the system, of electrical measuring instruments, a vacuum system, and a filling system.

The bicalorimeter consists of two coaxially arranged copper cylinders, an inner one (with 16.0-mm diameter) and an outer one (with the diameters of the outer and inner part 90.0 and 28.3 mm, respectively). The gap between the cylinders is filled with the investigated specimen.

The inner cylinder (the core of the bicalorimeter) consists of a measuring cylinder and two compensation cylinders which make it possible to prevent heat transfer through the upper and lower ends of the measuring cylinder. The thickness of the investigated layer is 6.2 mm. The temperature gradient on the boundaries of the investigated layer changed from 1.8 to 0.9°K. The experimental temperatures were measured with Chromel–Alumel differential thermocouples.

An analysis of the possible errors of measurement of thermal conductivity shows that the relative confidence error of measurements does not exceed ±2.5%.

In Figs. 2 and 3 the experimental data are compared with the results of calculation. It may be concluded from the comparison that the thermal conductivity of the above-described structures can be calculated by the suggested method with an error that is commensurable with the error of specifying the initial data.

Since the error of the method of calculating the thermal conductivity of complex systems, as well as the error of the method of calculating the thermal conductivity of granular systems [1] is approximately 15-20%, whereas the error of specifying the initial data varies between 5 and 100% (the error of reference data: λ_g, ε, Λ, S, c_p/c_v, , Pr, λ_{met}, ρ_{met} 5% for gases and 10% for solid materials; thermal conductivity of aluminum oxide according to literature data within the limits 50-100%; grain size of the charge 100%), the agreement between calculation and experiment may be regarded as perfectly satisfactory, both qualitatively and quantitatively.

NOTATION

n_{met}, m_{met}, weight and bulk concentration, respectively, of the metal; m, m_2, m'_2, total porosity, porosity of the material of the grains and of the charge of grains, respectively; $\lambda_{Al_2O_3}$, λ_g, λ_{met}, thermal conductivity of porous aluminum oxide, of the gaseous filler, and of the metal particles, respectively; λ_r, λ_m, λ_{por}, radiant, molecular, and total thermal conductivity, respectively, of the pores; λ', λ'', λ, thermal conductivity of cracked grains, with embedded metal, and effective thermal conductivity of the charge, respectively; λ_{tg}, λ_{2tp}, thermal conductivity of a through gap and of a through pore, respectively, W/(m·°K); V_1, V_2, V_3, volumes of pores situated between grains, volume of cracks, and volume of the grains themselves, respectively, m^3; ρ_{met}, $\rho_{Al_2O_3}$, ρ_1, bulk densities of the metallic particles, of the porous aluminum oxide, and of the grains themselves, respectively, kg/m^3; y_1, y_2, y_3, y_4, relative radii of sections of an averaged element; c, parameter depending on bulk concentration; M, parameter characterizing the cracked state of the material; ε, degree of blackness of the pore surface; δ, pore size, m; σ_0, Stefan–Boltzmann constant, $W/(m^2 \cdot °K^4)$; T, temperature of the charge, °K; Λ, free path length of the molecule of the gaseous filler, m; H, pressure of the gaseous filler, Pa; c_p/c_v, ratio of the isobaric and isochoric heat capacities of the gaseous filler; a, coefficient of accomodation of the gas to the surrounding substance; Pr, Prandtl number; N, coordiantion number; h_q, h_{1r} height of microroughnesses and of the layer of charge, respectively, m; Φ, function taking the change of thermal flux into account; p_{sp}, specific external load on the charge, N/m^2.

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