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HEAT AND MASS TRANSFER IN POROUS AND DISPERSIVE MEDIA

PARAMETRIC INVESTIGATION OF A RECUPERATIVE REACTOR WITH A FIXED POROUS BED FOR OXIDIZATION OF ORGANIC IMPURITIES CONTAINED IN AIR

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A recuperative reactor with a fixed porous bed for oxidization of organic impurities contained in air has been simulated. This reactor represents a system consisting of two coaxial tubes filled with a porous carcass mix. The influence of the dimensions of the reactor, the heat losses through its side wall, and the diameter of the filling-material balls on the maximum temperature realized in it, the efficiency of recuperation, and the position of the combustion front has been investigated. It has been established that the maximum temperature of the indicated reactor changes nonmonotonically with increase in the heat exchanges between its side wall and the environment. The parametric-investigation results obtained can be used for optimization of the design of recuperative reactors.

Introduction. A pressing problem of many mechanical, chemical, biochemical, and other technological processes [1, 2] is the neutralization and removal of contaminating gases. Phenol, acetone, formaldehyde, benzene, and many other volatile organic compounds (VOC) have a combustion heat sufficient for their effective combustion in an inert porous medium (filtrational combustion) [3–6]. Filtrational combustion provides effective heat recirculation in a system. A stationary combustion of methane in a porous medium of a recuperative reactor with an equivalent ratio between the gas and air $\Phi = 0.026$ (which is almost 20 times lower than the threshold of combustion of this mixture in an open flame) was described in [7]. A filtrational-combustion wave can be stabilized in a porous carcass mix or as a result of a change in the direction of filtration of gases. In [3], a regenerative reactor, in which a stable combustion was obtained at $\Phi = 0.15$, was simulated.

A peculiarity of the filtrational combustion of a gas in a porous medium is that in it an internal recirculation of heat is provided in the combustion wave due to the heat exchange between the gas and the porous medium in the region of preheating of the wave. In practice, in reactors for burning of superpoor fuel mixtures, an "external" heat recirculation is provided by the heat exchange between the inflowing and outflowing gases (recuperative scheme) or it arises as a result of a change in the direction of gas filtration (regenerative scheme). Such schemes were investigated under laboratory conditions [3, 4, 7–9] and are used in industrial reactors for oxidization of volatile organic compounds of the Thermatrix [10], ReEco-Stroem [11], and other companies. The physical aspects of filtrational combustion of gases in an inert porous medium were discussed in [4, 12, 13]. Despite the fact that the indicated reactors are used in practice, their parameters were not investigated in detail. Because of this, we simulated reactors of the recuperative, regenerative, recuperative-regenerative types with the aim to compare their characteristics, such as the maximum temperature realized in the reactor, the emission of nitrogen oxides in it, and the operating range of the reactor with respect to the flow rate of a gas mixture and the concentration of deleterious impurities in it [14]. In [15], a parametric investigation of the main operating conditions of a recuperative reactor containing an electric element for additional heating element and the influence of the flow rate of a gas in the reactor, the disposition of the electric heating element, the power of this element, and the heat losses through the face of the reactor on the maximum temperatures of the porous carcass and the gas and on the concentration of a incompletely oxidized organic substance at the output of the reactor has been carried out.

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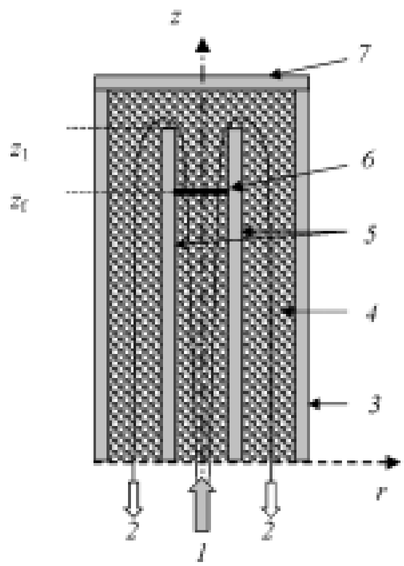

Fig. 1. Scheme of the reactor being investigated: 1) inflowing gas mixture; 2) outflowing gases; 3) side wall of the reactor; 4) porous carcass; 5) inner tube; 6) combustion front; 7) cover of the reactor.

In the present work, we investigated the dependence of the thermophysical operating parameters of a recuperative reactor for oxidization of volatile organic compounds on the design parameters of this reactor: the diameter of its inner tube, the size of the particles of a porous carcass, and the heat-transfer coefficient. Such parameters are the maximum temperature of the porous carcass, the position of the combustion front on the longitudinal axis of the reactor, and the efficiency of recuperation realized in it.

Formulation of the Problem. A recuperative reactor consisting of two coaxial tubes filled with a porous carcass mix — ceramic balls — is considered (Fig. 1). It is assumed that the wall of the inner tube and the side wall of the reactor have zero thickness. At the initial instant of time, the porous carcass located in the upper part of the reactor is heated to 1150 K. An inflowing gas mixture consisting of a low-concentration combustible gas (methane, VOC) and air passes through the inner tube and, in doing so, exchanges heat with the porous carcass and is heated due to the heat exchanged and the heat released as a result of the VOC oxidization. The partially or completely reacted gases reach the end of the inner tube, and then they flow to the output of the reactor, as shown in Fig. 1. The heat released by these gases is absorbed by the porous carcass that transfers it through the wall of the inner tube to the inflowing gas. In time, stationary distributions of temperature fields, concentrations, and flow rates are established. It is assumed that the position of the filtrational-combustion front on the z axis is the point at which the heat released by the gas as a result of the chemical reactions is maximum.

Mathematical model. The problem being considered was formulated using the method of mean-volume approximation of mutually penetrating continuums. A system described by the continuity equations for a gas and the gas filtration, the equations of mass conservation for the chemical gas components, the equations of heat conduction in the porous filling material and in the gas, and the equation of ideal-gas state [14–17] was simulated:

$$\frac{\partial \rho_g}{\partial t} + \nabla (\rho_g \mathbf{u}) = 0 , \tag{1}$$

$$-\nabla p = \frac{\mu}{k_0} \mathbf{u} + \frac{\rho_g}{k_1} |\mathbf{u}| \mathbf{u} , \tag{2}$$

$$\rho_g \frac{\partial Y_i}{\partial t} + \rho_g \mathbf{u} \nabla Y_i - \nabla \rho_g \mathbf{D} \nabla Y_i = \dot{\rho}_i , \tag{3}$$

$$\rho_g c_p \frac{\partial T_g}{\partial t} + c_p \rho_g \mathbf{u} \nabla T_g - \nabla \Lambda \nabla T_g = \frac{\alpha_{vol}}{m} (T_s - T_g) - \sum_i h_i \dot{\rho}_i , \tag{4}$$

$$(1-m)\,\rho_s c_s \frac{\partial T_s}{\partial t} - \nabla\,(\lambda \nabla T_s) = \alpha_{vol}\,(T_g - T_s)\,, \tag{5}$$

$$\rho_g = \frac{pM}{RT_g}\,. \tag{6}$$

The heat-transfer equation for the gas includes terms accounting for the dispersive diffusion $\mathbf{D} = D_g\mathbf{I} + \mathbf{D}_d$ and the heat conductivity $\Lambda = \lambda_g\mathbf{I} + c_p\rho_g\mathbf{D}_d$, where $\mathbf{D}_d = \begin{bmatrix} D_p\tau_z^2 + D_t\tau_r^2 & (D_p - D_t)\,\tau_z\tau_r \\ (D_p - D_t)\,\tau_z\tau_r & D_p\tau_z^2 + D_t\tau_z^2 \end{bmatrix}$ is the dispersive-diffusion tensor and the unit vector is determined as $\boldsymbol{\tau} = \frac{\mathbf{u}}{|\mathbf{u}|}$. The components of the tensor are related to the rate of gas filtration by the following relation:

$$D_p = 0.5d_0\,|\mathbf{u}|\,,\quad D_t = 0.1d_0\,|\mathbf{u}|\,. \tag{7}$$

The heat-conduction equation for the carcass accounts for the radial heat conduction

$$\lambda = \lambda_s + \frac{16}{3}\left(\frac{0.666m}{1-m}\right)\varepsilon\sigma d_0 T_s^3\,. \tag{8}$$

The coefficient of three-dimensional heat transfer between the porous carcass and the gas is determined as

$$\alpha_{vol} = \frac{\lambda_g 6\,(1-m)}{d_0^2}\left[2 + 1.1\left(\frac{\mu c_p}{\lambda_g}\right)^{1/3}\left(\frac{\rho_g u_g d_0}{\mu}\right)^{0.6}\right]. \tag{9}$$

Equations (2) and (3) were solved on the condition that the walls of the inner tube and the reactor are impermeable. For the equations of heat conduction in the gas (4) and in the porous carcass (5), the adiabatic boundary conditions were set at the side walls of the reactor. The heat losses through the side wall of the reactor were determined under the following adiabatic conditions for the temperature of the porous carcass:

$$-\lambda \left.\frac{\partial T_s}{\partial r}\right|_{r=0.5d_2} = \alpha\left(\left.T_s\right|_{r=0.5d_2} - T_0\right). \tag{10}$$

The problem was solved for the following standard values of the parameters: z_1 = 0.37 m, z_2 = 0.4 m, d_0 = $4.8\cdot10^{-3}$ m, d_1 = 0.028 m, d_2 = 0.04 m, T_0 = 300 K, p_0 = $1.01325\cdot10^5$ Pa, ε = 0.45, m = 0.4, ρ_s = 3987 kg/m^3, c_s = 1300 J/(kg·K), λ_s = 1.87 W/(m·K), Φ = 0.1, G = 2.21671 m^3/h, U_g = 1 m/sec.

As the VOC-containing mixture, we used a methane-air mixture with an equivalent fuel-oxidizer ratio Φ = 0.1 (the mole fraction of methane $X[CH_4]$ is ~ 0.0099). The combustion of methane was simulated by the second-order gross reaction $CH_4 + 2O_2 \xrightarrow{k} CO_2 + 2H_2O$ with a rate constant $k = 2.17\cdot10^8 \exp(-15{,}640/T_g)$ m^3/(mole·sec) [18]. The ratio between the air components $N_2{:}O_2$ = 4:1.

At the above-described standard parameters, the thickness of the front region, in which 95% of the methane is oxidized, is equal to 4 cm. The coefficient of three-dimensional heat transfer between the gas and the porous carcass, determined from the solution of the problem, is equal to $\alpha_{vol} \approx 10^5$ W/(m^3·K).

Problem (1)–(6) was solved using a 2DBurner program package for simulation of nonstationary two-dimensional processes [15, 17]. The desired dependences were constructed using the parameters corresponding to the steady-state operating conditions of the reactor.

Results of Simulation. The most important parameters of a VOC-oxidization reactor that determine its design and operation are the maximum temperatures of the gas and the porous carcass, the concentration of the incompletely oxidized organic substance at the output of the reactor, and the efficiency of heat recuperation realized in it:

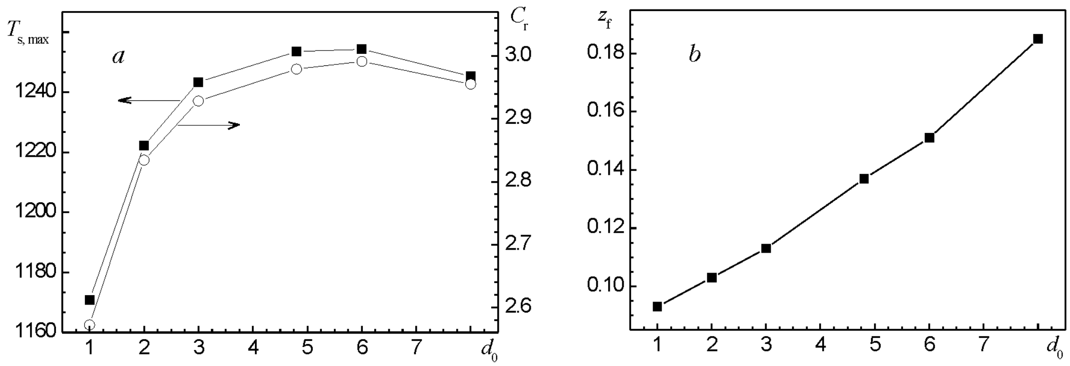

Fig. 2. Dependence of the maximum temperature of the porous carcass $T_{s,max}$ (a), the recuperation efficiency C_r (a), and the combustion-front position z_f (b) on the diameter of the filling-material balls d_0. d_0, mm; $T_{s,max}$, K; z_f, m.

$$C_r = \frac{Q_r}{\Delta h \rho_g U_g S}\,, \tag{11}$$

where $Q_r = \int\limits_{\Omega_1} \lambda \frac{\partial T}{\partial r} dS$ is the conductive heat flow generated by the outflowing burnt gas and is transferred to the inflowing cold gas through the porous carcass and the inner-tube wall Ω_1 and $\Delta h \rho_g U_g S$ is the heat flow generated by the burnt gases.

We also investigated the position of the combustion front at different parameters of the reactor being considered. By this position one can judge which region of the inner tube participates in the recuperation of the heat released by the outflowing hot gases because, in the reactor region $z \geq z_f$, the temperatures of the inflowing and outflowing gases are practically equal and all heat transfer between the gas flows is realized in the region of length $z \leq z_f$.

At the above-indicated parameters, the temperature of the gas differs insignificantly from the temperature of the porous carcass; therefore, only the maximum temperatures of the porous carcass are indicated on the graphs. We also did not indicate the concentration of the incompletely oxidized methane at the output of the reactor because, in the stable regime of operation of the reactor, it was negligibly small ($\sim 10^{-6}$ mole/mole) or there arose conditions under which the combustion decayed and practically all methane emerged from the reactor. Such cases are specially pointed out in the present work.

Influence of the diameter of the filling-material balls d_0 on the maximum temperature, the position of the combustion front, and the heat flowing through the wall of the inner tube. It follows from the data obtained (Fig. 2) that the dependences of the recuperation efficiency C_r (11) and the maximum temperature of the carcass $T_{s,max}$ on d_0 are identical. Both quantities reach maximum values when filling-material balls of diameter ~6 mm are used. The position of the front z_f depends practically linearly on d_0 in the range of its values being considered. The shift of the front downstream (Fig. 2b) is probably due to the increase in the heat conductivity of the carcass (8) and the decrease in the three-dimensional heat transfer (9). An increase in z_f leads to an increase in the degree of recuperation and, consequently, in the maximum temperature of the carcass.

Influence of the ratio between the diameters of the tubes d_1/d_2 on the maximum temperature, the position of the combustion front, and the heat flowing through the wall of the inner tube. It is seen from Fig. 3 that a decrease in the diameter of the inner tube leads to an increase in the maximum temperature of the porous carcass $T_{s,max}$ and an increase in the recuperation efficiency C_r (11). This can be explained by the shift of the combustion front downstream, which leads to an increase in the region of the inner tube participating in the heat recuperation. This shift is mainly due to the increase in the velocity of the gas flowing through the inner tube. However, when d_1 further decreases, at d_1/d_2 = 0.4, which can take place when an inner-tube diameter of 16 mm is used, the combustion front is blown out and combustion decays. It follows from the comparison of curves a and b in Fig. 3 that the curve of C_r (d_1/d_2) is entirely identical in form to the curve of z_f (d_1/d_2).

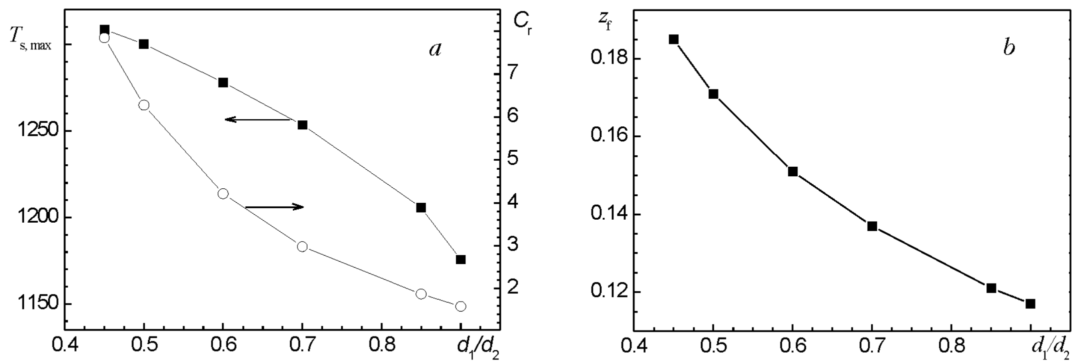

Fig. 3. Dependence of the maximum temperature of the porous carcass $T_{s,max}$ (a), the recuperation efficiency C_r (a), and the combustion-front position z_f (b) on the ratio between the diameters of the tubes d_1/d_2. $T_{s,max}$, K; z_f, m.

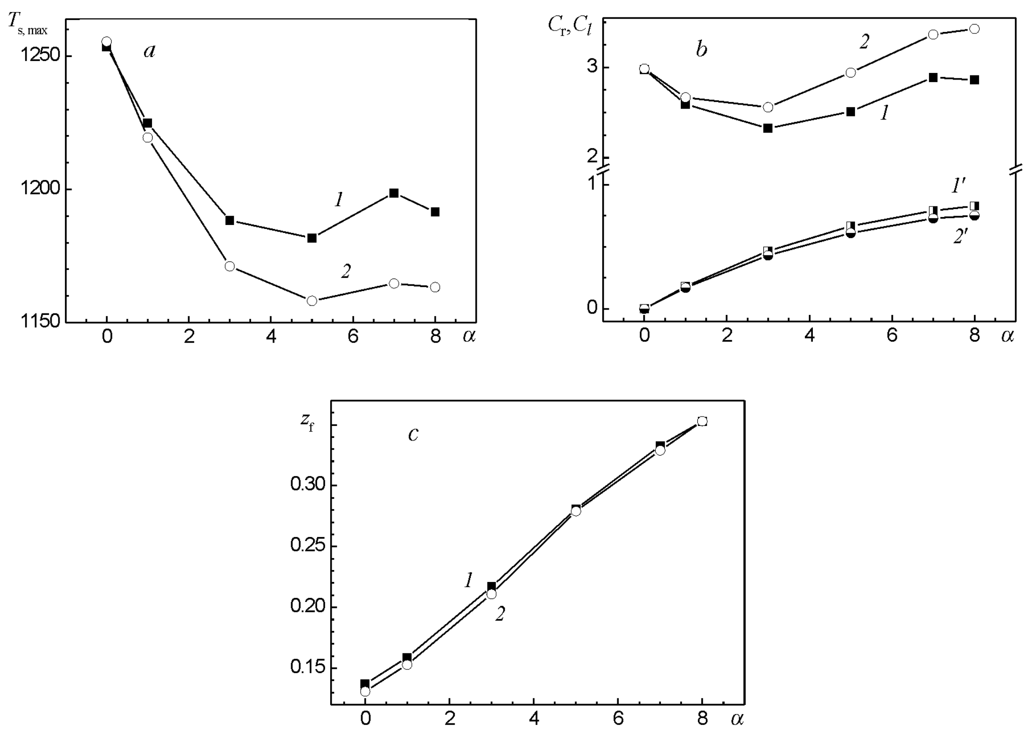

Fig. 4. Dependence of the maximum temperature of the porous carcass $T_{s,max}$ (a), the recuperation efficiency C_r (b, curves 1 and 2), the dimensionless heat losses through the side wall of the reactor C_l (b, curves 1′ and 2′), and the combustion-front position z_f (c) on the heat-transfer coefficient α in the case where a gas is supplied through the central tube of the reactor as is shown in Fig. 1 (1, 1′); and when the gas is supplied through the gap between the inner and outer tubes (2, 2′). α, W/(m^2·K); $T_{s,max}$, K; z_f, m.

The influence of the heat losses on the maximum temperature, the position of the combustion front, and the heat flowing through the wall of the inner tube. We investigated two methods of gas supply: 1) a gas is supplied through the central tube of the reactor, as shown in Fig. 1; 2) the gas is supplied through the gap between the inner tube and the body of the reactor in the direction opposite to the direction of the gas flow in case 1. The heat losses through the side wall of the reactor are presented in the dimensionless form as the ratio between the heat Q_l =

$\int_{\Omega_2} \alpha (T_s - T_0)dS$ lost through the side surface of the reactor Ω_2 as a result of the heat exchange with the environment and the heat released by the burnt gas $C_l = Q_l/\Delta h \rho_g U_g S$.

It follows from the data obtained that there is a range of values of the heat-transfer coefficient, α = 5–7 W/(m^2·K), in which the maximum temperature of the carcass $T_{s,max}$ and the recuperation efficiency C_r increase with increase in α (Fig. 4a and b). This can be explained by the fact that the length of the recuperation region increases with increase in z_f (Fig. 4c). A stabilization of the combustion front downstream also leads to a decrease in the maximum-temperature region of the reactor side wall, which leads to a decrease in the rate of increase in the heat losses through this wall and, consequently, to an increase in the temperature of the system.

Our investigations have shown that the method of gas supply has practically no influence on the position of the combustion front and the amount of heat flowing through the walls of the tube to the environment (Fig. 4b and c) but significantly influences the maximum temperature of the porous carcass $T_{s,max}$ (Fig. 4a, curves 1 and 2) and the recuperation efficiency C_r (Fig. 4b, curves 1 and 2). It should be noted that, in the case where the first method of gas supply is used, $T_{s,max}$ is higher despite the fact that the value of C_r is lower than the value of this quantity realized with the use of the second method of gas supply. The relatively high recuperation efficiency in the case where the gas is supplied through the gap between the inner tube and the body of the reactor is explained by the fact that the temperature gradient of the porous carcass in the direction from the reacted gas to the inflowing gas is larger, which is explained by the heat losses through the side wall of the reactor. It is seen from Fig. 4 that, in the case where the gas is supplied into the outer tube, the Newton heat losses C_l are somewhat smaller that those in the case where the gas is supplied into the inner tube. At α = 10 W/(m^2·K), the combustion decays.

CONCLUSIONS

1. A porous carcass with particles of diameter 6 mm is optimum for the system considered. In this case, at this size of the porous-carcass particles, all things being equal, a higher maximum temperature is attained as compared to the maximum temperature attained at other sizes of these particles.

2. When the diameter of the inner tube of the reactor decreases relative to the diameter of the reactor, the combustion front shifts for a larger distance from the input of the system, which leads to an increase in the maximum temperature of the reactor; however, the maximum allowable rate of the gas flow decreases in this case.

3. In the case where heat is lost through the side wall of the reactor, it makes sense to supply a volatile organic compound through the central tube of the reactor. In this case, a higher maximum temperature is attained, as compared to the case where the gas is supplied through the gap between the inner tube and the body of the reactor.

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NOTATION

C_l, dimensionless heat losses; C_r, coefficient of recuperation efficiency; c_p, heat capacity of a gas at a constant pressure, J/(kg·K); c_s, heat capacity of a porous carcass mix, J/(kg·K); **D**, gas-diffusion tensor including the gas-diffusion coefficient and the dispersive-diffusion tensor, m^{-1}; D_p, D_t, components of the dispersive-diffusion tensor, m^{-1}; d_0, diameter of the filling-material particles, m; d_1, diameter of the inner tube, m; d_2, diameter of the outer tube, m; G, gas flow rate, nm^3/h; h_i, mass enthalpy of the ith gas component, J/kg; Δh, specific heat of combustion of a gas mixture, J/kg; k, rate constant of a reaction, m^3/(mole·sec); k_0, k_1, Ergun permeability constants; m, porosity; M, average molar mass of the gas, kg/mole; p_0, pressure at the output of the reactor, Pa; Q_r, recuperative heat flow, W; Q_l, heat losses through the side wall of the reactor, W; r, radial coordinate, m; R, universal gas constant; Re, Reynolds number; S, area of the cross section of the inner tube, m^2; T_0, initial temperature of the system (ambient temperature), K; T, temperature, K; T_{ad}, adiabatic temperature of combustion, K; $\Delta T_{ad} = T_{ad} - T_0$, K; $T_{s,max}$, maximum temperature of the porous carcass, K; **u**, rate of gas filtration, m/sec; $U_g = G/S$, specific flow rate of the gas, m/sec; Y_i, mass fraction of the ith gas component, kg/kg; z, longitudinal coordinate, m; z_1, length of the inner tube, m; z_2, length of the reactor, m; z_f, position of the combustion front, m; α, coefficient of heat exchange between the reactor side wall and the ambient air, W/(m^2·K); α_{vol}, coefficient of three-dimensional heat transfer, W/(m^3·K); ε, degree of blackness of the porous carcass; Λ, heat-conductivity tensor of the gas; λ, effective heat conductivity of the porous carcass, W/(m·K); μ, viscosity of the gas, Pa·sec; ρ, density, kg/m^3; $\dot{\rho}_i$, rate of formation of the *i*th gas component as a result of chemical reactions, kg/sec; **τ**, unit velocity vector; Φ, fuel/oxidizer ratio; Ω_1, side surface of the inner tube, m^2; Ω_2, side surface of the reactor, m^2. Subscripts: 1) inner tube; 2) body of the reactor; ad, adiabatic; d, dispersive; f, combustion front; g, gas; *i*, number of a gas-mixture component; max, maximum; p, parallel; r, recuperative; s, porous carcass; t, transverse; vol, volumetric.

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