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# INFLUENCE OF RADIATION ON THE LIMITS OF HETEROGENEOUS COMBUSTION OF A PARTICLE IN TWO PARALLEL REACTIONS ON ITS SURFACE

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*The influence of radiation on critical parameters of heterogeneous ignition and extinction of a carbon particle in air is analyzed with allowance for two heterogeneous reactions*

Heterogeneous combustion of coal particles has been considered in numerous theoretical works, in which the mechanism of heat transfer or chemical kinetics is ignored for no proper reason in interpreting experimental results [1-8]. For example, heat transfer by conduction and convection is not covered in [4]. In this case it is assumed that only one chemical reaction proceeds on the particle surface. In the known works [5, 6] concerned with thermal regimes of heterogeneous reactions heat transfer by radiation is ignored. The problem of the limits of heterogeneous combustion due to different mechanisms of heat transfer and chemical kinetics of coal particles is not, in practice, touched upon in modern textbooks [7, 8]. In this work an analysis is made of and analytical expressions are obtained for steady stable and critical conditions of heat and mass transfer with account taken of heat losses by radiation from the surface of a particle on which two chemical reactions proceed in parallel.

We determined the critical parameters from the solution of equations of unsteady heat and mass transfer (HMT) and chemical kinetics of a coal particle in heated air and from the solution of equations that govern the stability of steady HMT regimes. For the case of two parallel reactions ($C + O_2 = CO_2$ and $2C + O_2 = 2CO$) for $\mathrm{BiO} \ll 1$, the equations for the change in the coal particle temperature and diameter have the form

$$\frac{1}{6} c_1 \rho_1 d \frac{dT_1}{dt} = Q_{ch} - Q_h , \quad T_1 (t = 0) = T_{in} , \tag{1}$$

$$\frac{d(d)}{dt} = - \frac{2\rho_2}{\rho_1} n_{ox} \sum_{i=1}^{2} \Omega_i k_i \left( \frac{\sum_{i=1}^{2} k_i}{\beta} + 1 \right)^{-1} , \quad d (t = 0) = d_{in} , \tag{2}$$

$$Q_{ch} = \left( \sum_{i=1}^{2} q_i k_i \right) \left( \frac{\sum_{i=1}^{2} k_i}{\beta} + 1 \right)^{-1} n_{ox} \rho_2 , \tag{3}$$

$$k_i = k_{0i} \exp \left( - \frac{E_i}{RT_1} \right) , \quad \beta = D\mathrm{Nu}/d , \quad D = \lambda_2/(c_2 \rho_2) ,$$

$$\rho_2 = \rho_{20} (T_0/T_*) , \quad \lambda_2 = \lambda_{20} (T_*/T_0)^n ,$$

$$c_2 = c_{20} + 0.125 (T_* - T_2) ,$$

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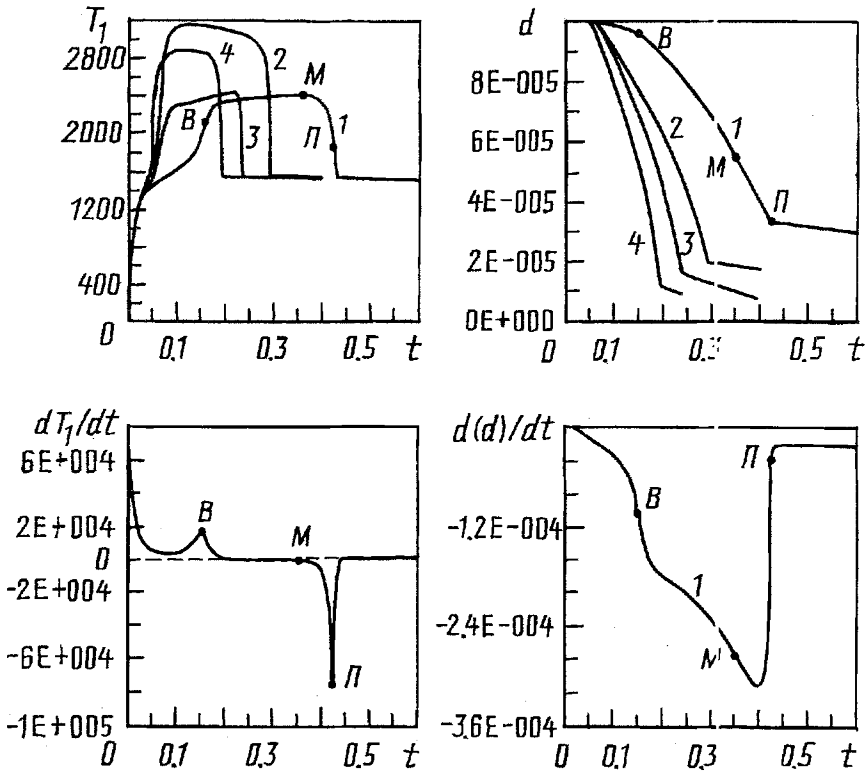



Fig. 1. Influence of radiation and heterogeneous reaction scheme on the time dependences of temperature, particle diameter, and their derivatives at $T_2 = 1400$, $T_w = T_2$, $n_{ox} = 0.23$: 1) with account taken of radiation ($\varepsilon = 0.78$), one reaction; 2) with no account taken of radiation ($\varepsilon = 0$), one reaction; 3) $\varepsilon = 0.78$, two reactions; 4) $\varepsilon = 0$, two reactions. $T$, K; $d$, m; $t$, sec.

$$Q_h = Q_{m.c} + Q_r, \quad Q_r = \varepsilon\sigma(T_1^4 - T_w^4), \quad Q_{m.c} = \alpha(T_1 - T_2), \quad \alpha = \lambda_2 \mathrm{Nu}/d. \tag{4}$$

For air $n = 0.75$, the characteristic temperature at which we take the properties of the air is equal to $T_* = 0.5(T_1 + T_2)$.

The critical parameters by the unsteady HMT model are bifurcation values that satisfy simultaneously two conditions

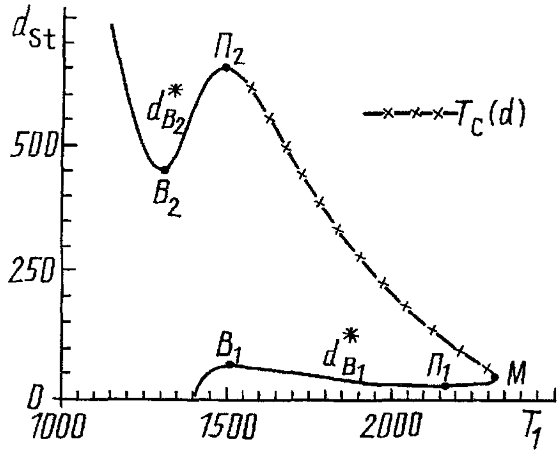
$$dT_1/dt = 0 \quad \text{and} \quad d^2T_1/dt^2 = 0.$$

From the dependences $T_1(t)$, $d(t)$, $dT_1/dt(t)$, and $d(d)/dt(t)$ given in Fig. 1 it is clear that particle ignition (point $B$) characterizes the completion of self-acceleration for the chemical reaction and attaining a high-temperature quasisteady HMT-combustion regime. The combustion temperature changes slightly with time, attains a maximum value (point $M$), and then decreases, the diameter decreasing at a high rate. Once the critical diameter $d_{ext}$( point $П$) is attained spontaneous extinction of the particle occurs. The spontaneous extinction diameter $d_{ext}$ for the case of two reactions is two times smaller than for one.

Heat losses by radiation lead to a decrease in combustion temperature and to an increase in diameter for the spontaneously extincted particle (Fig. 1).

In the same manner as in [1, 2], to analytically derive expressions for steady and critical diameters, we make an analysis of the steady diameter as a function of the particle temperature for the case of two paralel reactions. From the condition $dT_1/dt = 0$ and formulas (1)-(4) we have

$$d_{st} = \frac{\lambda_2 \mathrm{Nu}\,(T_1 - T_2)}{\left(A + \sqrt{A^2 - B}\right)\left(\sum_{i=1}^{2} k_i q_i\right) n_{ox}\rho_2}, \tag{5}$$



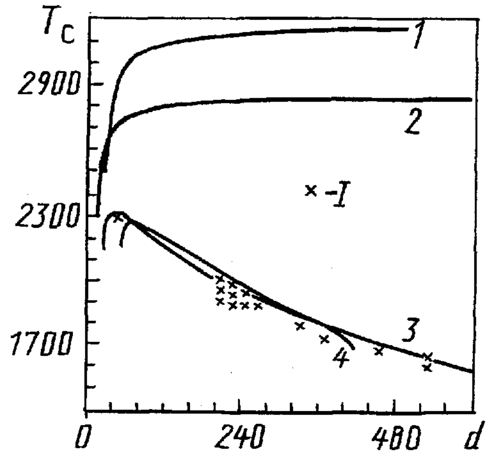




Fig. 2. $d_{st}$ vs temperature calculated with account taken of heat transfer by radiation for two heterogeneous reactions at $T_2 = 1400$, $T_w = 500$. $d_{st}$, $\mu$m; $T$, K.

Fig. 3. Influence of radiation and reaction scheme on the dependence of the combustion temperature of a coal particle on its diameter: $T_2 = 1400$, $n_{ox} = 0.23$; 1) $1 - \varepsilon = 0$, one reaction; 2) $\varepsilon = 0$, two reactions; 3) $\varepsilon = 0.78$, two reactions; 4) $\varepsilon = 0.78$, one reaction; $I$, V. I. Babii's experimental data [4]. $T_c$, K.

where

$$A = \frac{1}{2}(1 - A_1 - A_2), \quad B = A_1 A_2;$$

$$A_1 = \frac{c_2 (T_1 - T_2) \sum_{i=1}^{2} k_i}{\left(\sum_{i=1}^{2} q_i k_i\right) n_{ox}}; \quad A_2 = \frac{\varepsilon\sigma (T_1^4 - T_w^4)}{\left(\sum_{i=1}^{2} k_i q_i\right) n_{ox} \rho_2}. \tag{6}$$

we denote

$$\frac{\lambda_2 \, \mathrm{Nu} \, (T_1 - T_2)}{d \left(\sum_{i=1}^{2} k_i q_i\right) n_{ox} \rho_2} = A_3 .$$

The extrema on the curve of $d_{st}(T_1)$ are governed by the critical values of diameters and temperatures at which the particle ignition and extinction occurs (Fig. 2). The maximum at point $B_1$ and the minimum at point $B_2$ are connected respectively with the critical diameters $d_{ign\,1}$ and $d_{ign\,2}$ that limit the size interval over which particle self-ignition occurs. The critical diameter $d_{ign\,1}$ due to the heat losses by heat conduction and $d_{ign\,2}$ is due to those by radiation. The high-temperature process of HMT-combustion is governed by a branch that links points $\Pi_1$ and $\Pi_2$. Once the particle hits the combustion branch with $d_{ign1} < d_{in} < d_{ign\,2}$ it decreases its size as a result of combustion, the combustion temperature $T_c$ increasing. This behavior of the combustion temperature is explained by the fact that at small heat losses by heat conduction there is an increase in heat release on account of the increasing mass-transfer coefficient ($\beta = D\mathrm{Nu}/d$) with a decrease in particle diameter.

Once the maximum value is passed (point $M$) the combustion temperature decreases. This is connected with the increase in the heat-transfer coefficient as the particle size decreases. When the particle diameter attains the critical value of $d_{ext\,1}$ its spontaneous extinction (point $\Pi_1$, Fig. 2) occurs. Point $\Pi_2$ governs the critical diameter $d_{ext\,2}$ that characterizes forced extinction at this temperature of the medium.

At low wall temperatures in the region of large particle diameters we observe behavior of the dependence of the ignition diameter on the initial temperature of the particle (portion $B_2\Pi_2$, Fig. 2) that is different from that in the region of its small sizes (portion $B_1\Pi_1$). The increase in the particle initial temperature leads to an increase in the critical diameter of ignition $d^*_{\text{ign 2}}$. Analyzing the overall constant of the chemical reaction rate $k = k_1+k_2$, the mass-transfer coefficient $\beta$, the molecular convective heat flux $Q_{\text{m.c}}$, and the heat flux by radiation $Q_r$ on the portion $B_2\Pi_2$ of the curve $d_{\text{st}}(T_1)$ showed that $k \ll \beta$ in the vicinity of point $B_2$ (the region of the reaction is kinetic); consequently, it is the particle temperature rather than the diameter that governs mainly the change in the heat release $Q_{\text{chem}}$. The molecular convective heat flux $Q_{\text{m.c}}$ is not very small yet as compared to $Q_r$ ($Q_r/Q_{\text{m.c}} \approx 3$) and heats the particle ($T_1 < T_2$, Fig. 2). Consequently, the increase in the particle initial temperature and hence in $Q_{\text{chem}}$ is connected here with the decrease in the heat flux $Q_{\text{m.c}}$ to the particle as a result of its diameter increase.

As $d$ increases further (in the vicinity of point $\Pi_2$) the heat flux $Q_{\text{m.c}}$ that cools the particle ($T_1 > T_2$) becomes negligibly small as compared with the heat losses to the reaction chamber walls ($Q_r/Q_{\text{m.c}} \geq 10$). The heat release through the chemical reaction also decreases as a result of the decrease in the mass-transfer coefficient $\beta$. The reaction region shifts to a transition one $k \approx \beta$. Consequently, if the ignition of a particle of a larger diameter is to occur, increasing $Q_x$ is needed on account of the increase in the particle initial temperature.

A decrease in the critical diameter of ignition occurs in the small particle region $d_{\text{ext}} < d_{\text{in}}$ as the initial temperature increases. The mechanism of this phenomenon is explained by the predominant increase in the heat-transfer coefficient in the kinetic region of heterogeneous reactions with a decrease in the particle diameter, the heat losses by radiation being small. For ignition we need to increase the particle initial temperature.

The dependence of the critical diameter of ignition for $d_{\text{ext 1}} < d_{\text{in}} < d_{\text{ign 1}}$ is represented in analytical form

$$d^*_{\text{ign 1}} = \begin{cases} d_{\text{ign 1}} & \text{for} \quad T_{\text{in}} < T_{\text{ign 1}}, \\ \dfrac{\lambda_2 \,\text{Nu}\, (T_{\text{in}} - T_2)}{\left(A_{\text{in}} + \sqrt{A_{\text{in}}^2 - B}\right)\left(\sum\limits_{i=1}^{2} k_i q_i\right)\rho_2 n_{\text{ox}}} & \text{for} \quad T_{\text{ign 1}} < T_{\text{in}} < T_{\text{ext 1}}, \end{cases} \tag{7}$$

where

$$k_i = k_{0i} \exp\left(-\frac{E_i}{RT_{\text{in}}}\right).$$

In the large particle region the critical value of the initial temperature is in the $T_{\text{ign 2}} < T_{\text{in}} < T_{\text{ext 2}}$ interval; the critical diameter of ignition is $d_{\text{ign 2}} < d^*_{\text{ign 2}} < d_{\text{ext 2}}$.

To explicitly determine the critical diameter of ignition $d_{\text{ign 2}}$ as a function of $T_{\text{in}}$ we need to divide the initial temperature region into two parts: 1) $T_{\text{in}} < T_2$; 2) $T_{\text{in}} > T_2$. For the region where $T_{\text{ign 2}} < T_{\text{in}} < T_2$:

$$d^*_{\text{ign 2}} = \frac{D\text{Nu}\left(\sum\limits_{i=1}^{2} k_i q_i\right) n_{\text{ox}}\rho_2}{\left(\sum\limits_{i=1}^{2} k_i\right) \varepsilon\sigma (T_{\text{in}}^4 - T_{\text{w}}^4)} \left(\sqrt{\left((A')^2 + B'\right)} - A'\right),$$

$$A' = 0.5\,(A_2 - 1 - A_1'), \quad B' = A_1' A_2,$$

$$A_1' = \frac{c_2 (T_2 - T_{\text{in}}) \sum\limits_{i=1}^{2} k_i}{\left(\sum\limits_{i=1}^{2} k_i q_i\right) n_{\text{ox}}}, \quad A_2 = \frac{\varepsilon\sigma (T_{\text{in}}^4 - T_{\text{w}}^4)}{n_{\text{ox}}\rho_2 \left(\sum\limits_{i=1}^{2} k_i q_i\right)}.$$

TABLE 1. Parameters of the High-Temperature Process of Heat and Mass Transfer at the Maximum Temperature of Particle Combustion

| $T_2$, K | $d$, $\mu$m | $T_{c.m}$, K | $\beta/\sum_{i=1}^{2} k_i$ | $A_1$ | $A_2$ | $A_3$ |
|---|---|---|---|---|---|---|
| 1400 | 41.5 | 2320 | 0.29 | 0.61 | 0.05 | 0.13 |
| 1500 | 31 | 2430 | 0.3 | 0.63 | 0.04 | 0.14 |

In the $T_2 < T_{in} < T_{ext\,2}$ region

$$d^*_{ign\,2} = \frac{DNu}{\left(\sum_{i=1}^{2} k_i\right)} \frac{\left(\sum_{i=1}^{2} k_i q_i\right) n_{ox}\rho_2}{\varepsilon\sigma (T_{in}^4 - T_w^4)} \left(A - \sqrt{A^2 - B}\right),$$

where $A$ and $B$ are expressed by formulas (6) when $T_{in}$ is substituted for $T_1$.

The influence of the particle diameter on the combustion temperature that governs the high-temperature quasisteady TMO regime is determined, in the presence of heat losses by radiation, by formula (5), in which the value of combustion temperature should be substituted for $T_1$: $T_1 \equiv T_c$. The combustion temperature and corresponding particle diameter are limited respectively by the critical values of $T_{ext\,1}$ and $T_{ext\,2}$, $d_{ext\,1}$ and $d_{ext\,2}$. The combustion temperature $T_{c.m}$ and the corresponding diameter $d_m$ are obtained from the condition $dT_c/d(d) = 0$ or $A^2 = B$ in formula (5) as

$$T_{c.m} = T_2 + \frac{n_{ox}\left(\sum_{i=1}^{2} k_i q_i\right)}{c_2\left(\sum_{i=1}^{2} k_i\right)} \left[\sqrt{\left(\frac{\varepsilon\sigma (T_{c.m}^4 - T_w^4)}{\left(\sum_{i=1}^{2} k_i q_i\right) n_{ox}\rho_2}\right)} - 1\right]^2,$$

$$d_m = \frac{DNu}{\sum_{i=1}^{2} k_i} \left[\sqrt{\left(\frac{\left(\sum_{i=1}^{2} q_i k_i\right) n_{ox}}{\left(\sum_{i=1}^{2} k_i\right) c_2 (T_{c.m} - T_2)}\right)} - 1\right]^{-1},$$

$$k_i = k_{0i} \exp\left(-\frac{E_i}{RT_{c.m}}\right).$$

Calculation of $(\beta/\sum_{i=1}^{2} k_i)$, $A_1$, $A_2$, and $A_3$ (Table 1) for the two reactions with account taken of the dependence of the transfer coefficients, density, and heat capacity of the gas on temperature $[T_* = 0.5(T_1+T_2)]$ shows that, despite the relatively small value of heat losses by radiation ($A_2$), we cannot ignore the influence of chemical transformation kinetics on the combustion temperature and the corresponding diameter. Account taken of the heat losses by radiation and two parallel reactions yields a closer agreement with experimental results [4] (Fig. 3).

The limiting values of the combustion temperature and the corresponding particle diameters, the critical parameters of HMT that govern heterogeneous ignition and extinction of the particle are found from solving the conditions of steadiness $Q_x = Q_h$ and instability of the steady regime as $\partial Q_x/\partial T_1 = \partial Q_h/\partial T_1$. With no account

TABLE 2. Influence of $T_*$ on the Critical Diameters of Ignition $d_{ign\,1}$ and Extinction $d_{ext\,1}$; $T_2 = 1400$ K, $T_w = T_2$, $n_{ox} = 0.23$

| $T_*$ | $d_{ign\,1}$, μm | $d_{ext\,1}$, μm |
|---|---|---|
| $T_1$ | 49.8 | 31.9 |
| $0.5(T_1+T_2)$ | 44.2 | 20.7 |
| $T_*$ | 33.1 | 9.37 |

taken of the temperature dependence of the transfer coefficients and gas density ($T_* = T_2$) we write an equation to determine the dependences of the particle critical temperatures on the temperature of gas and the oxidant content in it

$$n_{ox}\rho_2\beta\frac{\left(\sum_{i=1}^{2} q_ik_iE_i\right)\left(\beta+\sum_{i=1}^{2} k_i\right)-\left(\sum_{i=1}^{2} q_ik_i\right)\left(\sum_{i=1}^{2} k_iE_i\right)}{\left(\beta+\sum_{i=1}^{2} k_i\right)^2 RT_1^2}=\beta c_2\rho_2+4\varepsilon\sigma T_1^3, \tag{8}$$

where

$$\beta=\frac{\mathrm{Nu}D}{d}=\frac{\left(A\pm\sqrt{A^2-B}\right)\left(\sum_{i=1}^{2} q_ik_i\right)n_{ox}}{c_2(T_1-T_2)}. \tag{9}$$

Taking into account the temperature dependence of the properties of the gas ($T_* = 0.5(T_1 + T_2)$) the equation for the particle critical temperatures has a more complex form

$$\left(\sum_{i=1}^{2} q_ik_i\right)n_{ox}\rho_2\left(1+\frac{\sum_{i=1}^{2} k_i}{\beta}\right)^{-1}\left\{\left(1+\frac{\sum_{i=1}^{2} k_i}{\beta}\right)^{-1}\left[\frac{\left(\sum_{i=1}^{2} k_i\right)(n+1)}{\beta(T_1+T_2)}-\frac{\left(\sum_{i=1}^{2} k_iE_i\right)}{\beta RT_1^2}\right]-\frac{1}{(T_1+T_2)}+\frac{\sum_{i=1}^{2} q_ik_iE_i}{RT_1^2\left(\sum_{i=1}^{2} q_ik_i\right)}\right\}=$$

$$=\beta c_2\rho_2\left[1+\left(\frac{T_1-T_2}{T_1+T_2}\right)n\right]+4\varepsilon\sigma T_1^3. \tag{10}$$

Calculations performed at different $T_*$ show that the influence of the characteristic temperature has the most substantial effect on the critical parameters of extinction in the region of small particle sizes (Table 2). The decrease in the diameter $d_{ext\,1}$ in the transition from $T_* = T_1$ to $T_* = 0.5(T_1 + T_2)$ and $T_* = T_2$ is explained by increased heat removal from the particle by heat conduction.

Figure 4 gives the dependences of the critical temperatures and diameters of the particle on the gas temperature calculated by formulas (10) and (9) with and without account taken of heat losses by radiation. The heat losses by radiation cause the presence of the limiting temperatures $T_{2\,lim}$ and $T'_{2\,lim}$ (points $C$ and $C'$). The

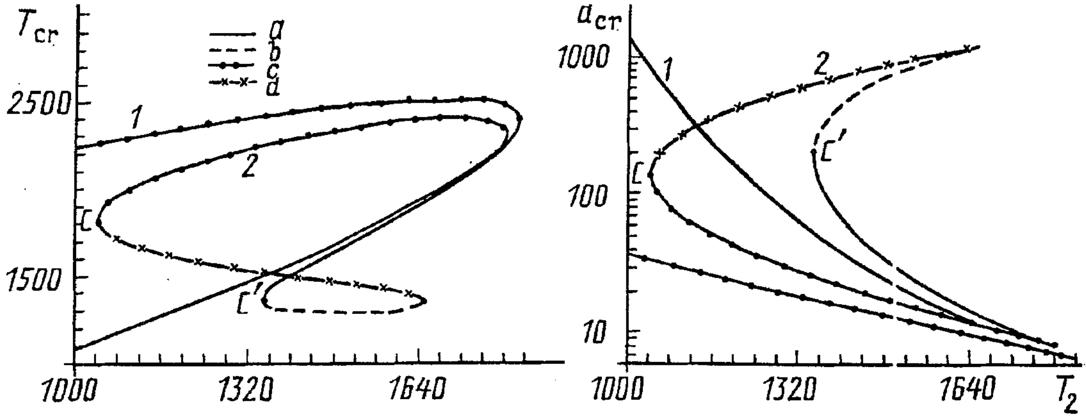



Fig. 4. Influence of heat transfer by radiation on the critical temperatures and diameters of particles as functions of the gas temperature: $n_{ox} = 0.23$; 1) $\varepsilon = 0$; 2) $\varepsilon = 0.78$; $T_w = 500$; a) $d_{ign\,1}(T_2)$, $T_{ign\,1}(T_2)$; b) $d_{ign\,2}(T_2)$, $T_{ign\,2}(T_2)$; c) $d_{ext\,1}(T_2)$, $T_{ext\,1}(T_2)$; d) $d_{ext\,2}(T_2)$, $T_{ext\,2}(T_2)$. $d_{cr}$, m; $T_{1r}$, K

limiting temperature $T'_{2\,lim}$ governs the critical temperature of gas below which the particle ignition is impossible. In the region limited by the curves $d_{ign}(T_2)$ and $d_{ext}(T_2)$ the change-over to a combustion regime is possible as a result of the increased initial temperature of the particle. Below the limiting temperature $T_{2\,lim}$(point $C$) it is impossible to get the particle to ignite only by changing its diameter or initial temperature.

The limiting temperatures of the gas are found from the condition $\partial T_{2\,cr}/\partial(d_{cr}) = 0$ and, for the case of two heterogeneous reactions, are determined by the number:

$$\frac{\varepsilon\sigma\left(T_{1\,cr}^4 - T_w^4\right)}{\left(\sum_{i=1}^{2} q_i k_i\right)\rho_2 n_{ox}} = \left[\frac{\sum_{i=1}^{2} k_i}{\beta} + 1\right]^{-2},$$

which is equivalent to the condition $\sum_{i=1}^{2} k_i\beta = Q_{m.c}/Q_r$.

For the kinetic region ($\sum_{i=1}^{2} k_i \ll \beta$) of chemical reaction, the heat losses by radiation are equal to the chemical heat release and the critical temperature of the particle is equal to the gas temperature.The limiting temperatures of the gas are found by solving the transcendental equation

$$\varepsilon\sigma\left(T_{2\,lim}^4 - T_w^4\right) = \left(\sum_{i=1}^{2} q_i k_i\right) n_{ox}\rho_2,$$

$$k_i = k_{0i}\exp\left(-\frac{E_i}{RT_{2\,lim}}\right).$$

For the case of two parallel reactions, the limiting temperatures $T_{2\,lim}$ and $T'_{2\,lim}$ are smaller than for the case of one reaction.

Therefore it is shown that account taken of the influence of radiation and the scheme of two parallel reactions leads to new qualitative and quantitative results for the critical HMO parameters that govern the heterogeneous combustion limits and for the dependence of the combustion temperature on the particle diameter.

## NOTATION

$Q_{chem}$, surface power of heat release through chemical reactions, W/m$^2$; $Q_h$, overall density of heat flux by molecular convection $Q_{m.c}$ and radiation $Q_i$, W/m$^2$; $d$, particle diameter, m; $t$, time, sec; $T_1$, $T_2$, $T_2$, $T_w$, particle, gas, and reaction chamber wall temperature respectively, K; $\rho_1$, $\rho_2$, particle and gas density, kg/m$^3$; $c_1$, $c_2$, specific heat of particle and gas, J/(kg·K); $n_{ox}$, relative mass concentration of oxidant in the gaseous medium; $q_i$, thermal effect of the first ($i$ = 1, $C + O_2 = CO_2$) and the second ($i$ = 2, $2C + O_2 = 2CO$) chemical reactions, J/kg; $\Omega_i$, stoichiometric coefficient; $E$, activation energy, J/mole; $k_{0i}$, preexponential factor, m/sec; $R$, universal gas constant, J/(mole·K); Nu, Nusselt number; $\lambda_2$, thermal conductivity coefficient of gas, W/(m·K); $D_2$, diffusion coefficient of gas, m$^2$/sec; $\rho_{20}$, $\lambda_{20}$, $D_0$, density, thermal conductivity, and diffusion coefficients of gas at $T_0$; $\varepsilon$, emissivity coefficient; $\sigma$, Stefan-Boltzmann constant, W/(m$^2$·K$^4$); $\alpha$, $\beta$, heat- and mass-transfer coefficients, W/(m$^2$·K), m/sec. Indexes: 1, particle; 2, gas; ign, ignition; ext, extinction; w, wall; st, steady; cr, critical; in, initial; c, combustion; m, maximum; lim, limiting.

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