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Combustion and spontaneous extinction of pulverized coal particles

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Abstract
Combustion temperatures and corresponding response rate of carbon versus the diameter of porous carbon particle have been determined analytically and studied with consideration for the mass Stefan flow, radiation heat transfer, simultaneous formation of reaction products and response within the pores. This study shows the role of above processes for determining the maximum temperature of combustion and the corresponding diameter at various temperatures of gas and mass fraction of oxygen contained in it.
Key words: COMBUSTION TEMPERATURE, CARBON PARTICLE, REACTION RATE, RADIATION HEAT TRANSFER, STEFAN FLOW, POROSITY

Formulation of scientific problem and its meaning

Studying of stable and critical modes of heat and mass transfer and combustion determined by the size of coal particles in conditions of simultaneous generation of carbon monoxide and carbon dioxide still remains a crucial task [1, 2], conditioned by a wide area of use. Currently, it is generally accepted that the combustion of coal particles takes place in the diffusion mode in which the combustion temperature does not depend on the size of particles, which is contrary to numerous experimental data. However, Stephan flow arising as a result of formation of gas at simultaneous reactions contributes in an additional resistance to combustion thus providing that reactions shift to the transition zone. Shift to the transition area, in which the size of particles has a substantial influence, is also the result of neglect of radiation heat loss, which rises with the increase of particle size. It is still important to determine both the diameter of ignition and extinction which restrict the hysteretic temperature and combustion rate dependence on size of coal particles, mechanisms of impact of carbon particle size on temperature and combustion rate, rate of appearance of the reaction products and gas composition at different concentrations of an oxidizer, temperatures and pressure of the surrounding gas.

Theoretical determination of diameters of ignition and extinction, combustion temperature and rate dependence on the size of carbon particles is complicated not only by the simultaneous reactions $C + O_2 = CO_2$ (I) and $2C + O_2 = 2CO$ (II), but the non-linear dependence on the temperature of such characteristics as radiation heat loss and Stefan flow. Therefore, this problem is often solved by experimental studies or numerical simulations [1, 2].

The aim of this work was an analytical study, with an account for the radiation heat transfer and Stefan flow, of the combustion temperature, the rates of formation of carbon monoxide and carbon dioxide, depending on size of the particle, and determination of ignition and extinction diameters.

Physical and mathematical model

The time derivative of the particle temperature is proportional to the specific speed of the reaction heat radiation q_{ch} minus the total heat loss per one unit of the particle's surface q_h:

$$\frac{\partial T}{\partial t} \sim q_{eff}, \quad q_{eff} = q_{ch} - q_h.$$

$$q_{ch} = Q_1 k_1 \left(1 + Q_2 k_2 / Q_1 k_1\right)\left(\frac{k_v}{k_1 + k_2} + 1\right)\rho_{gs}\frac{Z_{O_2}}{1 + Se}, \quad Se = \frac{\left(k_1 + k_2 + k_v + U_{sk}\right)\rho_{gs}}{\beta \rho_g},$$

$$k_1 = k_{01}\exp\left(-\frac{E_1}{RT}\right), \quad k_2 = k_{02}\exp\left(-\frac{E_2}{RT}\right), \quad Q_1 > Q_2, \; E_1 < E_2,$$

$$q_h = \beta c_g \rho_g Le^{-1+s}\left(T - T_g\right) + U_{sk} c_g \rho_{gs} T_* \frac{1}{1 + Se} + \varepsilon\sigma\left(T^4 - T_w^4\right),$$

$$\beta = \frac{D_g}{d_*}, \quad d_* = \frac{d}{Sh}, \quad U_{sk} = \frac{\mu_C}{\mu_{O_2}}\left(k_1 + 2k_2\right)\left(\frac{k_v}{k_1 + k_2} + 1\right)Z_{O_2}, \quad T_* = \frac{T + T_g}{2}$$

Where q_{ch} - surface density of the chemical heat radiation, Vt/m^2; q_h - the surface density of the total heat loss Vt/m^2; Q_1, Q_2 - the heat effect of the reaction of oxidation of carbon to carbon dioxide and carbon monoxide, calculated per 1 kg of oxidant J/kgO$_2$; E_1, E_2 - the energy of activation, J/mol; k_1, k_2 - constants of chemical reactions; β - coefficient of mass transfer of the oxidizer; number Se - diffusion-kinetic relation; R - universal gas constant, J/(mol·K); ρ_g - density of gas, kg/m^3; c_g - gas specific heat, J/(kg·K); D_g - the coefficient of diffusion of the oxidant in the mixture, m^2/s; $\delta_* = \delta / Sh$ - given diameter, m, ε – coefficient of the particle surface emissivity, σ - Stefan-Boltzmann constant, Vt/(m^{2}K^4); Le - Lewis number; Sh - Sherwood number, U_{sk} - the rate of Stefan flow in the kinetic zone of the chemical reaction, m/s; T, T_g, T_w - the temperatures of surrounding gas and surrounding bodies, s – the exponent, depending on relation of Sherwood criteria to Nusselt criteria from criterion of Lewis.

Temperature properties of gas were defined according to dependences (p – gas pressure):

$$D_g = D_{g0}\left(\frac{T_*}{T_0}\right)^{n+1}\frac{p_0}{p},\ c_g = c_{g0} + \Omega\left(T_* - T_0\right),\ \rho_g = \rho_{g0}\frac{T_0}{T_*}\frac{p_0}{p},\ \rho_{gs} = \rho_{g0}\frac{T_0}{T}\frac{p_0}{p}.$$

Study of critical and steady conditions of a heat-mass exchange and coal particles combustion is based on the analysis of stability of particle temperature,

$$\frac{\partial q_{eff}}{\partial T}\partial T + \frac{\partial q_{eff}}{\partial \delta_*}\partial \delta_* = 0$$

As for the ignition and extinction

$$\frac{\partial q_{eff}}{\partial T} = 0\ ,\ \text{then}\ \left.\frac{\partial \delta_*}{\partial T}\right|_{i,e} = 0.$$

It means that extreme values on dependences of diameter of a particle on stationary temperature determine diameters of ignition and extinction and critical temperatures of a particle corresponding to them. It is simple to show that the curve connecting points of ignition and extinction characterizes influence of initial temperature on the critical diameter of ignition. In fact, critical value of initial temperature and corresponding diameter of ignition is defined as

$$q_{eff} = 0,\quad \frac{\partial q_{eff}}{\partial T} > 0\cdot$$

Then from the equation (1) it follows that the ignition diameter decreases with the growth of particle initial temperature

$$\frac{\partial \delta_*}{\partial T} = -\frac{\dfrac{\partial q_{eff}}{\partial T}}{\dfrac{\partial q_{eff}}{\partial \delta_*}} < 0\ \text{,i.e.}\ \frac{\partial q_{eff}}{\partial \delta_*} > 0.$$

And the maximum combustion temperature and diameter corresponding to it are defined from the conditions:

stationarity, $q_{eff} = 0$ [3], which in case of change of particle diameter can be presented in the form:

$$\frac{\partial \delta_*}{\partial T} = -\frac{\dfrac{\partial q_{eff}}{\partial T}}{\dfrac{\partial q_{eff}}{\partial \delta_*}} \qquad (1)$$

$$q_{eff} = 0\ \frac{\partial q_{eff}}{\partial T} < 0, \frac{\partial q_{eff}}{\partial \delta_*} = 0.$$

Then from the equation (1) it follows that:

$$\left.\frac{\partial T}{\partial \delta_*}\right|_M = 0.$$

In the general case it is impossible to receive in an analytical form the dependence of diameter of a porous particle on stationary temperature as the constant of internal reaction of k_v depends on diameter of a particle. However, in the combustion mode the constant k_v practically doesn't depend on diameter:

$$k_v \approx \sqrt{F_v D_v\left(k_1 + k_2\right)},\quad D_v = D_g m^2,$$

where F_v – specific surface of the pores, m^2/m^3; D_v, D_g – coefficient of oxygen diffusion inside the pores and in the gas, m^2/sec; m – porosity of a particle.

In this case from the stationarity of temperature of a particle $q_{eff} = 0$ it is possible to receive the dependence of carbon particle diameter on its stationary temperature describing the critical and steady modes of a heat-mass exchange in the following way [3]:

$$\delta = \frac{D_g c_g \rho_g Le^{-1+s} Sh}{Q_1 k_{01}\left(1+\frac{Q_2 k_2}{Q_1 k_1}\right)\left(1+\frac{k_v}{k_1+k_2}\right)\rho_{gs} Z_{O_2}} \cdot \frac{\left(T-T_g\right)}{\left(B+\sqrt{B^2-A_g A_w}\right)} \exp\left(\frac{E_1}{RT}\right) \tag{2}$$

and

$$\delta = \frac{D_g c_g \rho_g Le^{-1+s} Sh}{c_g \rho_{gs}} \cdot \frac{q_{ch,m}}{\left(k_1+k_2+k_v+U_{sk}\right)} \cdot \frac{\left(B+\sqrt{B^2-A_g A_w}\right)}{\varepsilon\sigma\left(T^4-T_w^4\right)}, \tag{3}$$

$$A_g = \frac{c_g \rho_{gs}\left(k_1+k_2+k_v+U_{sk}\right)\left(T-T_g\right)}{q_{ch,m}},\ A_w = \frac{\varepsilon\sigma\left(T^4-T_w^4\right)}{q_{ch,m}},\ A_{sf} = \frac{U_{sk}\rho_{gs} c_g T_*}{q_{ch,m}},$$

$$B = \frac{1}{2}\left(1-A_g-A_w-A_{sf}\right),\ q_{ch,m} = \left(Q_1 k_1 + Q_2 k_2\right)\left(1+\frac{k_v}{k_1+k_2}\right)\rho_{gs} Z_{O_2},$$

Having dependences (2) and (3), it is simple to receive in a parametrical form, stationary dependences of chemical reactions speeds for various diameters of particles. In the case under consideration, mass rates of chemical reactions (in kg/(m^{2}sec) are determined by carbon, carbon oxide and carbon dioxide as:

$$W_C = \frac{\mu_C}{\mu_{O_2}}\left(k_1+2k_2\right)\left[1+\frac{k_v}{k_1+k_2}\right]\rho_{gs} Z_{O_2,s}, \tag{4}$$

$$W_{CO_2} = \frac{\mu_{CO_2}}{\mu_{O_2}} k_1\left[1+\frac{k_v}{k_1+k_2}\right]\rho_{gs} Z_{O_2,s},\ W_{CO} = \frac{2\mu_{CO}}{\mu_{O_2}} k_2\left[1+\frac{k_v}{k_1+k_2}\right]\rho_{gs} Z_{O_2,s},$$

$$Z_{O_2 s} = \frac{Z_{O_2}}{1+Se}.$$

Where μ_C, μ_{CO}, μ_{CO2} – molar masses of carbon, carbon oxide and carbon dioxide.

Analysis of the results

Dependences of stationary temperature of a porous particle on its diameter in heated air of various temperature are presented in Figure 1. During the analysis the following parameters were used: E_1 = 140 кJ/mole, E_2 = 154 кJ/мole, k_{01} = 6.5· 10^4 m/sec, k_{02} = 13·10^4 m/sec, Q_1 = 12.3 MJ/kgO$_2$, Q_2 = 6.84 MJ/kgO$_2$, R = 8.31 J/(mole·K), z_{O_2} = 0.23, ε= 0.8, m = 0.1, Sh = 2.

For particles of the small size $\delta < \delta_M$ (Fig. 1) heat-losses from radiation are insignificant ($A_w << A_g$) and stationary dependence of the diameter on temperature is defined (2). For the carbon particles $\delta > \delta_M$ in size the main reason of heat loss is the thermal radiation from the particle. Stationary dependence of the diameter on temperature is defined (3).

As critical conditions of self-ignition and extinction of a carbon particle are defined by a condition that $\partial q_{eff}/\partial T|_{i,e} = 0$, then according to (1) it is easy to show that extreme values of a particle diameter dependence on stationary temperature determine the critical diameters of a particle, which characterize self-ignition of a particle (i-point) and limit high-temperature conditions of a heat-mass exchange (diameter of extinction, e- point).

When entering the quasi-stationary mode temperature and rate of the carbon particle combustion increase while it is burning out. The particle passes through the quasi-stationary steady states (Figure 1). Really, with diameter reduction the oxygen mass transfer to the surface unit increases. At the same time the increase in the Newtonian heat loss is broken because of radiation heat loss. Therefore, the particle combustion temperature T_{bur} and combustion rate depend non-monotonicaly on the particle size. This is confirmed experimentally [4, 5], though there is a difference between the calculated ($(T_{bur} - T_g) \sim d^{-0.48}$) and the experimental ($(T_{bur} - T_g) \sim d^{-0.16}$) data.

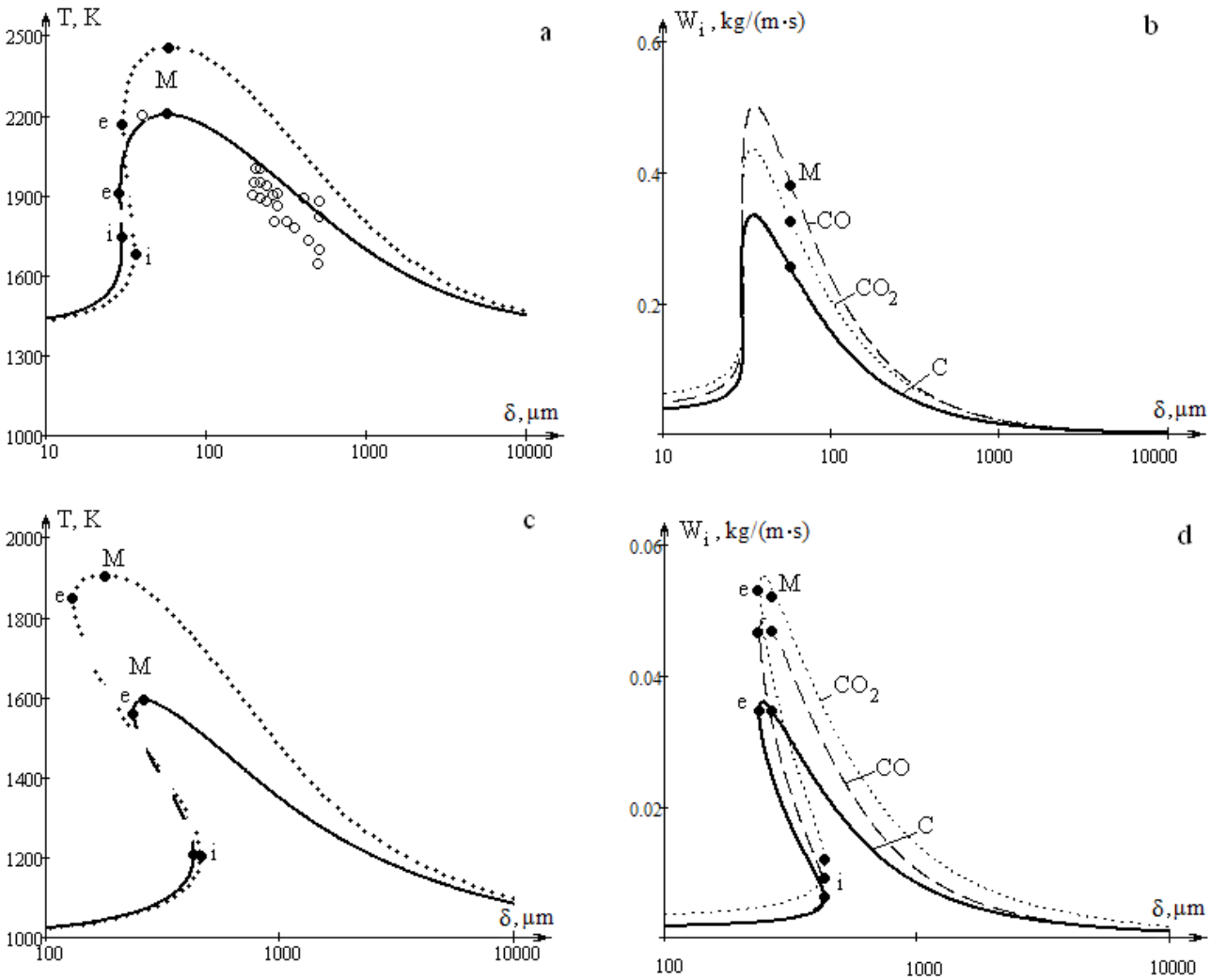

Figure 1. Stationary temperature of solid carbon particle and chemical reaction rate versus particle diameter at a, b) $T_g = T_w = 1400K$ and c,d) $T_g = T_w = 1000K$; $Z_{O_2} = 0.23$. o – test data [4]. a) —— – two reactions (I) – (II). - - - – one reaction (I).

The maximum combustion temperature T_M can be determined from the equality of rates of overall heat loss and heat generation variations at change of the diameter (($\partial q_{eff}/\partial\delta = 0$). This is identical to equality to zero of the radicand in (2) and (3). Thus, we receive equation for the maximum combustion temperature as a transcendental equation, obviously not depending on diameter of the particle:

$$T_M = T_g + \frac{(Q_1k_1 + Q_2k_2)\left(1 + \frac{k_v}{k_1 + k_2}\right)Z_{O_2}}{(k_1 + k_2 + k_v + U_{sk})c_g}\left[\sqrt{1 - A_{sf}} - \sqrt{\frac{\varepsilon\sigma(T_M^4 - T_w^4)}{q_{ch,m}}}\right]^2.$$

The following diameter of the particle corresponds to this temperature according to (2):

$$\delta_M = \frac{\lambda_g Nu}{Q_1k_{01}\left(1 + \frac{Q_2k_2}{Q_1k_1}\right)\left(1 + \frac{k_v}{k_1 + k_2}\right)\rho_{gs}Z_{O_2}} \cdot \frac{(T_M - T_g)}{B}\exp\left(\frac{E_1}{RT}\right).$$

Without radiation heat transfer ($\varepsilon = 0$), internal response (kv = 0), Stefan flow ($U_{sk} = 0$) we have classical equation for the combustion temperature in the diffusion zone of chemical reaction:

$$T_M = T_g + \frac{(Q_1k_1 + Q_2k_2)}{(k_1 + k_2)}\frac{Z_{O_2}}{c_g}$$

If to consider that constants of chemical reactions in combustion are close to each other in value, we

have a reduction of the maximum temperature with account of parallel reactions (I)-(II) in comparison with the reaction of carbon oxidation to carbon dioxide (I) (tables 1, 2).

The maximum reaction rate does not correspond to the maximum combustion temperature. The combustion temperature drop for particle diameters $\delta_e < \delta < \delta_M$ is connected with a higher increase of the heat rejection rate on a unit of surface as the diameter is reduced than the chemical heat generation rate at the expense of increase in a mass transfer of oxygen to the surface. Semenov number Se is reduced to the value of the order of unity. This increase in the surface concentration of oxygen determines the increase of the rate of steady chemical reaction.

Table 1. Action of the gas temperature on the maximum combustion temperature and critical diameters of the particle (z_{O_2} = 0.23). «(I)-(II)» – general case, «(I)» – the reaction only to CO_2, «k_v = 0» – without reaction in pores, «U_{sk}=0» – without Stefan flow.

	$T_g = T_w$ = 1400 K				$T_g = T_w$ = 1000 K			
	(I)-(II)	(I)	$k_v = 0$	U_{sk}=0	(I)-(II)	(I)	$k_v = 0$	U_{sk}=0
δ_M, micron	57	54	64	38	263	154	–	93
T_M, K	2203	2451	2172	2502	1592	1900	–	2023
$\left(W_{CO} / W_{CO_2}\right)_M$	1.19	0	1.17	1.30	0.89	0	–	1.11

Table 2. Action of mass fraction of the oxygen on the maximum combustion temperature, the relevant particle diameter and mass based chemical yield ($T_g = T_w$=1400 K). «(I)-(II)» – general case, «(I)» – the reaction only to CO_2, «k_v = 0» – without reaction in pores, «U_{sk}=0» – without Stefan flow.

	z_{O_2} = 0.15				z_{O_2} = 0.30			
	(I)-(II)	(I)	$k_v = 0$	U_{sk}=0	(I)-(II)	(I)	$k_v = 0$	U_{sk}=0
δ_M, micron	122	130	160	91	36	33	39	23
T_M, K	1848	1943	1786	1996	2472	2837	2453	2904
$\left(W_{CO} / W_{CO_2}\right)_M$	1.03	0	0.99	1.10	1.29	0	1.28	1.43

Tables 1 and 2 illustrate the action of the gas temperature and the oxygen content by weight on the maximum combustion temperature, the relevant diameter and the mass yield of CO and CO_2. With the rise of the combustion temperature as a result of gas temperature or oxygen concentration rising the share of the carbon monoxide originating on a surface of the particle increases.

Lowering of the gas temperature, of course, increases the critical diameter of self-ignition of the carbon particle. At that a hysteresis area (area of firing: ignition at increase of the initial temperature of the particle $\delta_e < \delta < \delta_i$) is expanded and shifted to the area of larger size particles. Internal response in the pores is crucial for the occurrence of forced ignition of the particle.

At high temperatures, the role of the reaction in the pores wanes, and the increase of the maximum combustion temperature becomes insignificant. However, the role of Stefan flow (mass flow directed from the particle surface) rises. As a result of the additional heat removal from the unit surface and resistance to the oxygen mass transfer to the surface of the particle the combustion temperature significantly goes down. Therefore, it must be included.

Conclusions and prospects for further researches

Thus, the used analytical dependences allow estimating, considering radiation heat transfer and Stefan flow, the combustion temperature of porous carbon particles, as well as areas of their spontaneous and induced ignition. Using an approach based on a comparison of steady and unsteady dependencies [3] in the future we expect to show the impact of the carbon gasification reaction, as well as an internal reaction in the pores in a general way.

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