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INFLUENCE OF MASS TRANSFER ON THE CRITICAL CONDITIONS AND THE TIME OF THE COKE PARTICLE IGNITION

The time evolution of the temperature in a coke particle during its ignition in a heated gas has been analyzed in order to analytically determine the induction period with regard for the mass transfer of oxygen to the particle surface. It is shown that the ignition process can be divided into characteristic stages. But the inflection points in the time dependence of the particle temperature do not coincide with the stage boundaries and, hence, cannot be used for their identification. When determining the end of the heating stage analytically, it is better to associate it with a temperature that is lower than the inflection temperature by one characteristic interval. When considering the further heating of the particle during the chemical reaction, the mass transfer has to be taken into account. A new method for the analytical determination of the ignition time is proposed, which makes allowance for chemical reactions and the mass transfer that simultaneously run in the transition and diffusion combustion regions.

Keywords: particle, coke, ignition time, mass transfer, critical conditions.

1. Introduction

Critical conditions required for the ignition of coal particles, as well as the ignition time (the induction period), are important characteristics of the coal particle burning. For instance, when blowing the coal dust into a heated volume of the furnace, the ignition time determines the path length of a non-burning particle in the torch, which is important to ensure the complete combustion of the particle [1, 2]. The ignition time (or the induction period) is the time of the non-stationary burning of coal particles. This process occurs, first of all, if the condition of self-acceleration of the rate of chemical heat release with respect to the rate of heat removal is satisfied. In this case, the effective heat removal coefficient α_* has to be less than the critical ignition value, $\alpha_* < \alpha_{*i}$, and the oxygen concentration Y_{O_2} larger than the corresponding ignition concentration, $Y_{\mathrm{O}_2} > Y_{\mathrm{O}_2 i}$.

When analyzing the time dependence of the temperature in a coke particle in the general case ($Y_{\mathrm{O}_2} > Y_{\mathrm{O}_2 i}$ and $\alpha_* < \alpha_{*i}$), two inflection points can be observed before the particle burning has been initiated [3–5]. Those points allow the induction period to be divided into stages: the stage of inert heating (before the first inflection point) and the stage of chemical heating (from the first to the second inflection point). In work [6], when searching for the analytical solution for the induction period, a condition of reaching the inflection point in the time dependence of the particle temperature (the Todes condition) was imposed.

However, since the temperature of the particle at this point is rather high, the induction period is approximately estimated as the time required to reach an infinitely high temperature, provided that one

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chemical reaction runs in the kinetic region. This approximation made it possible to estimate the ignition time of a particle, if its initial temperature equals the temperature of the gas medium [4, 7],

$$\tau_2 \approx \tau_{ch1} \frac{\sqrt{2}\pi}{e\sqrt{1-\dfrac{\alpha_*}{e}}} \approx \tau_{ch1} \frac{\sqrt{2}\pi}{e\sqrt{1-\dfrac{Y_{\mathrm{O_2 i}}}{Y_{\mathrm{O_2}}}}}, \qquad (1)$$

where

$$\tau_{ch1} = \frac{c\rho d R T_g^2}{6E_1 Y_{\mathrm{O_2}} \rho_g Q_1 k_1 (T_g)},$$

$$\alpha_* = \frac{\alpha R T_g^2}{E_1 Q_1 k_1 (T_g) Y_{\mathrm{O_2}} \rho_g}.$$

In practice, the induction period is understood as either the total time of the non-stationary stage, when the brightness of a burning particle practically ceases to change, or the time elapsed from the beginning of the particle heating to the appearance of external attributes of the combustion reaction [4, p. 70–73]. In the former case, the additional third stage of the induction period has to be introduced, which is characterized by a specific course of the chemical reaction in the diffusion region.

It is generally accepted that, during the burning of a carbon particle, the diffusion region of its combustion is observed. This assumption usually forms a basis when studying the particle burning rate [2]. A criterion that determines the combustion regions (the kinetic and diffusion ones) is the diffusion-kinetic ratio (the Semenov number) [8, p. 89–90]

$$\mathrm{Se} = \frac{d}{DNu}(k_1 + k_2).$$

This parameter evaluates the role of a reaction kinetics at the particle surface. In particular, the diffusion region (large particles and high temperatures) is characterized by the inequality $\mathrm{Se} \gg 1$. In this region, the total reaction rate is governed by the diffusion of oxygen. In the kinetic region (small particles and low temperatures), $\mathrm{Se} \ll 1$, and the total reaction rate is governed by the reaction kinetics. However, the particle combustion temperatures can decrease, e.g., with a decrease in the oxygen concentration in the medium. Therefore, the combustion of a particle because of a significant reduction in the Semenov number may occur not in the diffusion region, but in the transition one (the intermediate region between the kinetic and diffusion ones).

Thus, in the general case, the account for the oxygen mass transfer is required to determine the ignition time of a coke particle. It is so, because the particle temperature increases during this time interval, which leads to a gradual increase in the Semenov number from the values $\mathrm{Se} \ll 1$ to the values $\mathrm{Se} \gg 1$. The aim of this work was to analyze the time dependences of the temperature of a coke particle at its ignition in the heated gas in order to analytically determine the induction period making allowance for the mass transfer of oxygen to the particle surface.

2. Formulation of the Problem

The non-isothermal and irreversible flow of the following parallel exothermic reactions of the first order in oxygen was experimentally proved and theoretically substantiated [4, 9]:

(I) $\mathrm{C} + \mathrm{O_2} \rightarrow \mathrm{CO_2}$, $\quad \Delta H_1 = -395$ kJ/mol;

(II) $2\mathrm{C} + \mathrm{O_2} \rightarrow 2\mathrm{CO}$, $\quad \Delta H_2 = -209$ kJ/mol.

In work [9], it was found that the appearance rates of the reaction products $\mathrm{CO_2}$ and CO during the combustion of coke particles are of the same order of magnitude. The time dependence of the temperature averaged over the whole coal particle volume is determined by the solution of the differential equation [4, 10]

$$\frac{\partial T}{\partial \tau} = \frac{6}{c\rho d} q_{\mathrm{eff}} \qquad (2)$$

with the initial condition

$$T(t=0) = T_{\mathrm{b}},$$

where

$$q_{\mathrm{eff}} = q_{ch12} - q_{h\Sigma},$$

$$q_{ch1,2} = (Q_1 k_1 + Q_2 k_2)(1 - X_A)\rho_{\mathrm{gs}} Y_{\mathrm{O_2,s}},$$

$$q_{h\Sigma} = \frac{\lambda_g Nu}{d}(T - T_g) + \epsilon\sigma(T^4 - T_g^4),$$

$$Y_{\mathrm{O_2,s}} = Y_{\mathrm{O_2,\infty}} \frac{1}{1+\mathrm{Se}},$$

Q_1 and Q_2 are the thermal effects of chemical reactions I and II, respectively, per unit mass of oxygen

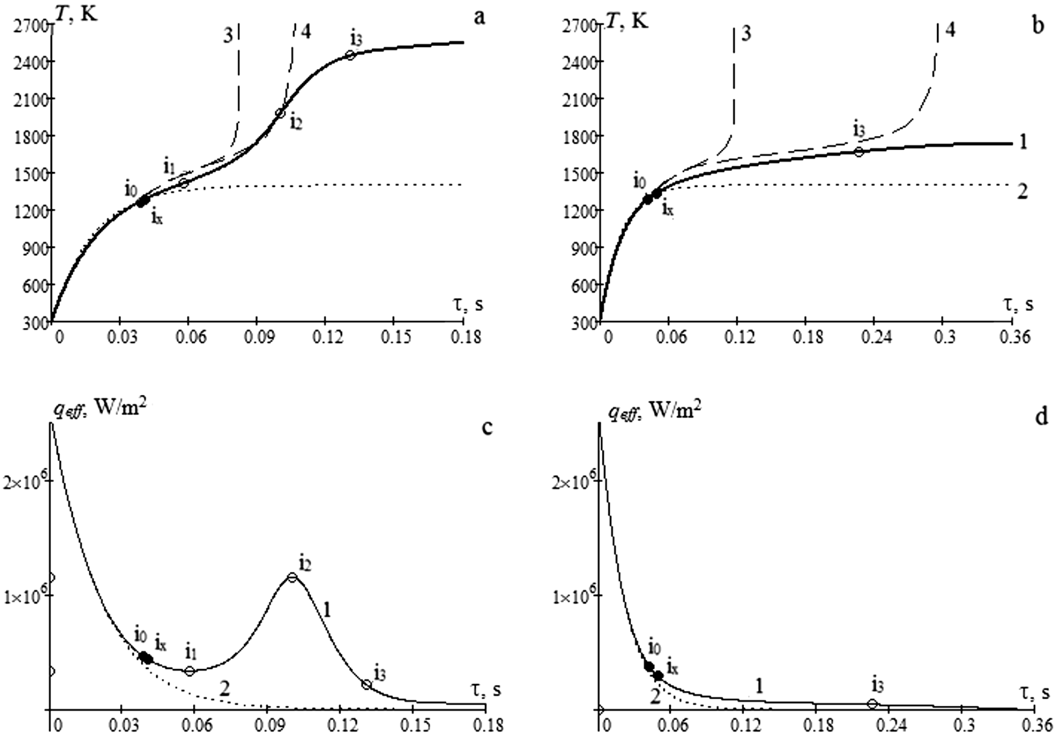

Fig. 1. Time dependences of the particle temperature (panels *a* and *b*) and the effective rate of heat release by a coal particle (panels *c* and *d*): (*1*) chemically active particle, solution of Eq. (2); (*2*) inert particle; (*3*) approximate dependence (15); and (*4*) approximate dependence (19). The calculation parameters are: the particle diameter is 100 μm, $E_1 = 140$ kJ/mol, $T_g = 1400$ K, $T_b = 300$ K, $X_A = 0$, $Y_{O_2} = 0.23$ (a, c) and 0.13 (b, d)

[J/kg O_2]; k_1 and k_2 are the rate constants of chemical reactions I and II, respectively; $Y_{O_2,s}$ is the mass fraction of oxygen in the gas near the particle surface; ρ_{gs} the density of a surrounding gas near the particle surface [kg/m^3]; c the specific heat of the particle [J/(kg K)]; T and T_g are the temperatures of the particle surface and the surrounding gas, respectively [K]; ϵ is the emittance of the particle surface; and $\sigma \approx 5.67 \times 10^{-8}$ W/(m^2 K^4) is the Stefan–Boltzmann constant.

The relevant studies [2] point to the following relationships between the activation energies E_i and the reaction-rate pre-exponential factors k_{0i}:

$$k_{\rm i} = k_{0\rm i} \exp\left[-\frac{E_{\rm i}}{RT}\right], \quad k_{0\rm i} = k_* \exp\left[\frac{E_{\rm i}}{RT_*}\right],$$
$$\frac{E_2}{E_1} = 1.1;$$

where $k_* = 100$ m/s, $T_* = 2600$ K, and $R \approx \approx 8.31$ J/(mol K) is the universal gas constant.

The change of a particle diameter during the process of particle ignition can be neglected [2, 4, 11].

3. Analysis of the Results

Let us analyze the dynamics of temperature change in a single coal particle located in the heated air (Figs. 1, *a* and *b*). As an example, the particles of ASh anthracite coke were considered. The accepted parameter values are $Q_1 = 12.3$ MJ/kg O_2, $Q_2 = = 6.84$ MJ/kg O_2, $E_1 = 140000$ J, $\gamma = 1.1$, $\rho_c = = 1440$ kg/m^3, $c_c = 2000$ J/(kg K), and $\epsilon = 0.78$.

The extremes in the time dependences of $q_{\rm eff}$ testify to changes in the heat transfer modes and the kinetics of chemical reactions, so that they determine the ignition moments. Let us briefly describe them.

When the derivative reaches a minimum value (point i_1),

$$q_{\rm eff}\big|_{i_1} > 0, \ \frac{\partial q_{\rm eff}}{\partial \tau}\bigg|_{i_1} = 0, \quad \frac{\partial^2 q_{\rm eff}}{\partial \tau^2}\bigg|_{i_1} > 0$$

Parameters of characteristic points in the time dependence of the temperature of a coal particle 100 μm in diameter at various oxygen concentrations in air. $T_g = 1400$ K

Y_{O_2}	τ_0, ms	τ_{ix}, ms	τ_{i1}	τ_{i2}	τ_{i3}	T_{i0}	T_{ix}	T_{i1}	T_{i2}	T_{i3}	Se_{i0}	Se_{i1}	Se_{i2}	Se_{i3}
0.23	39.0	40.8	57.8	100	130.8	1266	1284	1414	1975	2443	0.02	0.09	2.18	8.70
0.2	41.0	41.2	65.2	113.3	147.4	1282	1285	1447	1930	2302	0.03	0.12	1.81	6.02
0.17	43.6	41.4	79.2	137.0	176.1	1302	1285	1493	1867	2134	0.03	0.17	1.36	3.58
0.14	47.2	41.6	128.8	192.2	243	1324	1284	1587	1745	1870	0.04	0.31	0.75	1.29
0.135	47.8	41.8	175.4	180.2	252.4	1328	1285	1654	1662	1788	0.044	0.46	0.48	0.89
0.13	48.8	41.8	–	–	225.6	1333	1285	–	–	1665	0.046	–	–	0.48
0.10	55.8	42.0	–	–	150.8	1363	1284	–	–	1496	0.06	–	–	0.17

the particle ignition begins. The time required to reach this point characterizes the almost inert particle heating time. The heating mainly takes place owing to the heat exchange with a gas and the thermal radiation exchange with surrounding bodies.

After point i_1, the total chemical heat release at the particle surface becomes substantial. The temperature growth to the temperature at point i_2, which is determined by the maximum in the time dependence of $q_{\rm eff}$,

$$q_{\rm eff}\big|_{i_2} > 0, \qquad \frac{\partial q_{\rm eff}}{\partial \tau}\bigg|_{i_2} = 0, \qquad \frac{\partial^2 q_{\rm eff}}{\partial \tau^2}\bigg|_{i_2} < 0,$$

is characterized by the self-acceleration as a result of the chemical heat release and an increase of the particle temperature. Some researchers [5, 6] consider the time required to reach point i_2 as the induction period $\tau_{\rm ind0}$. In other words, after this point, the growth rate of the chemical heat release becomes lower due to the increase of the diffusion resistance (Se ≈ 1).

The time interval τ_{ind} required to reach a quasistable high-temperature mode of heat transfer (point i_3, at which the time derivative of the particle temperature becomes substantially smaller than at the previous stages and, may be, close to zero) may differ substantially from the induction period $\tau_{\rm ind0}$. Some researchers, who experimentally determined the induction period for coal particles, associated the induction time with the time required for a burning particle to achieve a constant brightness [3], in other words, when the particle temperature practically ceased to change in time.

As a condition that the particle temperature has achieved a quasistationary value (the end of the induction period), the following relation was selected:

$$\frac{c\rho d}{6q_{ch}}\frac{\partial T}{\partial \tau}\bigg|_{\tau=\tau_{\rm ind}} = 1 - \frac{q_g + q_w}{q_{ch}} = 0.05.$$

The choice of this condition as the induction period end was connected not only with visual observations of the changes in the rate of temperature growth from fast to slow, but also with the achievement by the particle temperature of a value that was close enough to the stationary value for the current particle diameter.

In Table, the parameter values for points i_1, i_2, and i_3 are quoted for various oxygen concentrations in air. The temperature corresponding to the first inflection point becomes higher than the ambient gas temperature, as the oxygen concentration decreases. This means that the particle starts to behave itself as a chemically active one (the diffusion-kinetic ratio is not small and cannot be neglected), and point i_1 is not the end point of the inert heating. The corresponding time interval has to be shorter.

One can see that, as the oxygen concentration decreases, the duration of the second stage diminishes to zero, and, below a certain Y_{O_2}-value, the time dependence of the temperature has no inflection points (Figs. 1, *a* and *b*). In addition, the combustion temperature decreases, and the particle burning mode is no more diffusion. The value of the diffusion-kinetic ratio $\mathrm{Se}|_{i_3} = 7\div10$ can be attributed to the diffusion combustion mode with reservations even at high oxygen concentrations in the mixture. At the same time, the value of $\mathrm{Se}|_{i_3}$ is close to 1 at low values of this parameter.

Thus, it is evident that if the oxygen concentration in the gas mixture becomes lower, the method based on the inflection points for determining the duration of separate stages is inapplicable. In what follows, while analytically determining the ignition time, this interval will be divided into two parts: the inert and chemical heating stages. Their calculation is convenient to be carried out in terms of dimensionless quantities.

4. Analytical Determination of Induction Period

In order to find the duration of separate stages in the induction period, it is necessary to determine critical conditions for the high-temperature heat and mass transfer, which simultaneously satisfy two conditions [2, 4, 7, 10],

$$q_{\text{eff}} = 0 \text{ and } \left.\frac{\partial q_{\text{eff}}}{\partial T}\right|_{i,e} = 0. \tag{3}$$

The simplest approximate way to determine the critical conditions for the self-ignition of a particle in a heated gas is to include the thermal radiation into the effective Newton–Richman law,

$$\alpha(T - T_g) + \epsilon\sigma(T^4 - T_g^4) \approx \alpha_{\text{eff}}(T - T_g),$$

where the effective heat transfer coefficient is introduced,

$$\alpha_{\text{eff}} = \alpha + 4\epsilon\sigma T_g^3.$$

In this case, if the chemical reaction is assumed to take place in the kinetic region, the effective heat-release density [the right-hand side of Eq. (2)] looks like

$$q_{\text{eff}} = (Q_1k_1 + Q_2k_2)\, Y_{O_2}\rho_g - \alpha_{\text{eff}}(T - T_g). \tag{4}$$

Applying the critical conditions (3), we obtain the system of equations

$$\begin{cases}(Q_1k_1 + Q_2k_2)\, Y_{\text{O}_2}\rho_g = \alpha_{\text{eff}}\,(T - T_g),\\ (Q_1k_1E_1 + Q_2k_2E_2)\, \dfrac{Y_{\text{O}_2}\rho_g}{RT^2} = \alpha_{\text{eff}}.\end{cases} \tag{5}$$

Dividing the upper equation by the lower one, we arrive at the transcendental equation for the critical temperature of a particle, which corresponds to its self-ignition,

$$T_i = T_g + \frac{RT^2}{E_1}\left(\frac{Q_1k_1 + Q_2k_2}{Q_1k_1 + Q_2k_2E_2/E_1}\right). \tag{6}$$

The critical self-ignition temperature of a particle is convenient to be represented in terms of the dimensionless quantity introduced by Frank-Kamenetskii [7]

$$\theta = \frac{T - T_g}{RT_g^2}E_1.$$

At the self-ignition, the critical temperature of a particle is insignificantly different from the effective gas temperature, $|T_i - T_g| \ll T_g$. Therefore,

$$\theta_i \approx \left.\frac{T - T_g}{RT^2}E_1\right|_i.$$

Let us use the Frank-Kamenetskii expansion for the reaction constants,

$$k_1 = k_{01}\exp\left(-\frac{E_1}{RT}\right) \approx k_{01}\exp\left(-\frac{E_1}{RT_g}\right)\exp\theta,$$

$$k_2 \approx k_{02}\exp\left(-\frac{E_2}{RT_g}\right)\exp\left(\gamma\theta\right).$$

Hence, if only one chemical reaction takes place, the critical temperature equals $\theta_i = 1$. The fraction in the parentheses in Eq. (6) weakly depends on the temperature, so that it can be calculated at $\theta = 1$ in the first approximation.

After doing some transformations and introducing the parameter $\gamma = E_2/E_1$, we obtain

$$\theta_i = 1 - \frac{Be^{\gamma}\left(\gamma - 1\right)}{1 + B\gamma e^{\gamma}} \text{ or } \theta_i = 1 - A_{\theta}, \tag{7}$$

where

$$B = \frac{Q_2k_2\left(T_g\right)}{Q_1k_1\left(T_g\right)}, \quad \gamma = \frac{E_2}{E_1}.$$

In most cases describing the self-ignition of coal particles, the second term in Eq. (7) is very small. For example, for the self-ignition of a particle of ASh anthracite coke at $T_g = 1500$ K, the parameter $B = 0.351$, so that $A_{\theta} = 0.049$. Therefore, in the case of parallel chemical reactions, we may approximately consider that $\theta_i \approx 1$.

In order to determine the dimensionless critical heat transfer coefficient α_{*i} (and, therefore, the particle diameter), let us use the second equation in system (5) and the Frank-Kamenetskii expansion for the reaction constants. As a result, we obtain [12]

$$\alpha_{*,i} = e\left(1 + Be^{\gamma - 1}\right) = \frac{\alpha_{\text{eff}}RT_g^2}{E_1Q_1k_1\left(T_g\right)Y_{\text{O}_2}\rho_g}. \tag{8}$$

To determine the induction period, let us introduce the dimensionless time $\tilde{\tau} = \tau/\tau_{ch}$, where

$$\tau_{ch} = \frac{c\rho d R T_g^2}{6 E_1 Y_{\mathrm{O}_2} \rho_g Q_1 k_1 (T_g)}.$$

Then the dimensionless variant of Eq. (2) for a change of the particle temperature reads

$$\frac{\partial\theta}{\partial\tilde{\tau}} = e^{\theta} + B e^{\gamma\theta} - \alpha_* \theta$$

with

$$\theta(\tilde{\tau} = 0) = \theta_b.$$

As the induction period, we may take a time interval, during which the particle temperature increases from its initial value to an infinitely large one [6, 7]. Therefore, in the case of two parallel reactions, the induction period is evaluated by the formula

$$\tilde{\tau}_{\mathrm{ind}} = \int\limits_{\theta_b}^{\infty} \frac{d\theta}{e^{\theta} + B e^{\gamma\theta} - \alpha_* \theta}. \qquad (9)$$

Analogously, if the mass transfer is taken into account (the Semenov number is not neglected), we have

$$\tilde{\tau}_{\mathrm{ind}} = \int\limits_{\theta_b}^{\infty} \frac{d\theta}{\left(\dfrac{e^{\theta} + B e^{\gamma\theta}}{1 + \delta_i \left(e^{\theta} + \chi e^{\gamma\theta}\right)}\right) - \alpha_* \theta}. \qquad (10)$$

The integrals in Eqs. (9) and (10) are not expressed by elementary functions. Therefore, the whole temperature interval is divided into sections, using which the induction period is determined by parts with the help of some approximations.

5. Inert Heating Stage

At this stage, the chemical heat release is assumed to be low, so that it can be neglected in Eqs. (4) and (10). The main issue here is to select the upper temperature limit in integral (10). For example, as the heating time end, we may choose a moment, at which the particle temperature reaches a value remote by the characteristic unit interval RT_g^2/E_1 from the gas temperature, i.e. $\theta = -1$ (point i_x in Fig. 1). In this case, the inert heating time is evaluated as

$$\tilde{\tau}_1 \approx \int\limits_{\theta_b}^{-1} \frac{d\theta}{-\alpha_* \theta} = \frac{1}{\alpha_*} \ln(-\theta_b).$$

However, as the inert heating end, it is better to select a temperature that deviates by the characteristic unit interval from the temperature of the corresponding inflection point. For its estimation, we can determine, where the derivative of the effective heat release equals zero, $\partial\left(e^{\theta} - \alpha_* \theta\right)/\partial\theta = 0$. As a result, we obtain an estimated value of the inflection temperature, $\theta_1 = \ln\alpha_*$, and an expression evaluating the time of the inert heating,

$$\tilde{\tau}_1 \approx \int\limits_{\theta_b}^{\ln\alpha_* - 1} \frac{d\theta}{-\alpha_* \theta} = \frac{1}{\alpha_*} \ln\left(\frac{\theta_b}{\ln\alpha_* - 1}\right). \qquad (11)$$

Figure 1 shows that Eq. (11) better describes the heating stage as the time that is required to reach the dimensionless particle temperature $\ln\alpha_* - 1$ (point i_0). First, the particle temperature at the end of the heating stage is lower than the temperature of a gas mixture (the particle is not heated above the mixture temperature). Second, the diffusion-kinetic ratio is much less than 1 at the end of the stage. Third, the duration of the stage weakly depends on the oxygen concentration, unlike the time interval that is required to reach point i_1, which testifies to the influence of the oxygen concentration on the rate of chemical heat release, which, in turn, affects the beginning of the chemical reaction self-acceleration.

6. Classical Method to Determine the Ignition Time

The duration of the next ignition stage of a heated coke particle is determined by assuming the kinetic mode of particle oxidation. Then

$$\tilde{\tau}_2 \approx \int\limits_{\ln\alpha_* - 1}^{\infty} \frac{d\theta}{e^{\theta} + B e^{\theta} - \alpha_* \theta}. \qquad (12)$$

The linear expansions of the dimensionless temperature and heat transfer coefficient near their critical values are

$$\theta = 1 + x, \quad \alpha_* = e\left(1 + B e^{\gamma - 1}\right)(1 - z),$$

where $x \ll 1$ and $z \ll 1$. With the help of the Maclaurin expansions for exponential functions, namely,

$$e^{\theta} \approx e\left(1 + x + \frac{x^2}{2}\right), \quad e^{\theta(\gamma - 1)} \approx e^{\gamma - 1}$$

(the latter is valid, since the difference $\gamma - 1$ is small), we obtain an approximate expression for the duration of the second stage of the induction period,

$$\tilde{\tau}_2 \approx \frac{2}{e\left(1 + Be^{\gamma-1}\right)} \int\limits_{\ln\alpha_* - 2}^{\infty} \frac{dx}{(x+z)^2 + 2z - z^2}.$$

The extension of the lower limit of integration to infinity (the corresponding error is negligibly small) makes it possible to obtain an approximate analytic formula for calculating the duration of the second stage of the induction period,

$$\tilde{\tau}_2 \approx \frac{\sqrt{2}\pi}{e\left(1 + Be^{\gamma-1}\right)\sqrt{z - \dfrac{1}{2}z^2}},$$

where

$$z = 1 - \frac{\alpha_*}{e\left(1 + Be^{\gamma-1}\right)}.$$

Ultimately,

$$\tilde{\tau}_2 \approx \frac{2\pi}{e\left(1 + Be^{\gamma-1}\right)\sqrt{1 - \left(\dfrac{\alpha_*}{e\left(1 + Be^{\gamma-1}\right)}\right)^2}}, \qquad (13)$$

With the help of Eq. (8), it is possible to determine the critical value of the oxygen concentration for a given particle size:

$$Y_{2,i} = \frac{\alpha_{\text{eff}} RT_{g,\text{eff}}^2}{E_1 Q_1 k_1\left(T_{g,\text{eff}}\right)\rho_g} \frac{1}{e\left(1 + Be^{\gamma-1}\right)}.$$

Then the duration of the second stage can be represented as a function of the oxygen concentration,

$$\tau_2 \approx \tau_\alpha \frac{Y_{\mathrm{O}_2 i}}{Y_{\mathrm{O}_2}} \frac{2\pi}{\sqrt{1 - \left(\dfrac{Y_{\mathrm{O}_2 i}}{Y_{\mathrm{O}_2}}\right)^2}}. \qquad (14)$$

Substituting the upper integration limit in Eq. (12) by the current temperature, we obtain an approximate parametric dependence of the particle temperature on the time during the second stage,

$$\tau_2(y) = \tau_{ch} \int\limits_{-1}^{y} \frac{d\theta}{e^\theta + Be^\theta - \alpha_*\theta}, \qquad (15)$$
$$T(y) = T_g + y\,\frac{RT_g^2}{E}.$$

The corresponding plots are depicted in Figs. 1, *a* and *c* (curves *3*). One can see that the particle temperature relatively quickly reaches high values, and the ignition time is significantly shorter than the sought value. The approximate dependence (15) agrees with the more accurate one only near the first inflection point, where the influence of the mass transfer is small.

7. Account for Mass Transfer While Determining the Induction Period

In order to account for the mass transfer, we have to approximately calculate integral (10). In work [13], while evaluating the ignition time, a method was proposed, which is based on the introduction of the dimensionless temperature, but the normalization is carried out relatively to the stationary temperature (the ignition temperature), rather than to the gas one. In this work, we choose the ignition temperature T_i as the characteristic one and introduced the following dimensionless quantities:

$$\Theta = \frac{T - T_i}{RT_i^2}E_1, \quad \alpha'_* = \frac{\alpha_{\text{eff}} RT_i^2}{E_1 Q_1 k_1\left(T_i\right) Y_{\mathrm{O}_2}\rho_g},$$

$$\tau'_{ch} = \frac{c\rho d RT_i^2}{6E_1 Y_{\mathrm{O}_2}\rho_g Q_1 k_1\left(T_i\right)}, \quad \tilde{\tau}' = \frac{\tau}{\tau'_{ch}},$$

$$\chi_i = \frac{k_2\left(T_i\right)}{k_1\left(T_i\right)}, \quad B_i = \frac{Q_2 k_2\left(T_i\right)}{Q_1 k_1\left(T_i\right)}, \quad \delta'_i = d\frac{k_1\left(T_i\right)}{DNu}.$$

In this case, Eq. (2) for the particle temperature evolution is rewritten in the form

$$\frac{\partial\Theta}{\partial\tilde{\tau}'} = \frac{e^\Theta + B_i e^{\gamma\Theta}}{1 + \delta'_i\left(e^\Theta + \chi_i e^{\gamma\Theta}\right)} - \alpha'_*\left(\Theta + 1\right)$$

with

$$\Theta(\tilde{\tau}' = 0) = \Theta_b.$$

As was done in the classical case, we linearly expand the dimensionless temperature and the heat transfer coefficient near their stationary values ($x' \ll \ll 1$ and $z' \ll 1$),

$$\Theta = x', \quad \alpha'_* = \alpha'_{*i}\left(1 - z'\right),$$

where

$$\alpha'_{*i} = \alpha'_*\big|_{\Theta=0} = \frac{1 + B_i}{1 + \delta'_i\left(1 + \chi_i\right)}.$$

As a result, we obtain

$$\frac{\partial\Theta}{\partial\tilde{\tau}'} = f(\Theta) - (1 - z')(\Theta + 1), \qquad \Theta(\tilde{\tau}' = 0) = \Theta_b, \tag{16}$$

where

$$f(\Theta) = \frac{e^{\Theta} + B_i e^{\gamma\Theta}}{1 + \delta_i'(e^{\Theta} + \chi_i e^{\gamma\Theta})}\,\frac{1 + \delta_i'(1 + \chi_i)}{1 + B_i},$$

$$\tilde{\tau}_*' = \frac{\tau}{\tau_{ch}'}\,\frac{1 + B_i}{1 + \delta_i'(1 + \chi_i)}.$$

The term $f(\Theta)$ on the right-hand side of Eq. (16) can be expanded in a Maclaurin series in a vicinity of $\Theta_i = 0$,

$$f(\Theta) \approx 1 + \frac{\Theta}{1 + \delta_i'(1 + \chi_i)} + \frac{1}{2}\frac{1 - \delta_i'(1 + \chi_i)}{1 + \delta_i'(1 + \chi_i)}\left(\frac{\Theta}{1 + \delta_i'(1 + \chi_i)}\right)^2.$$

This expansion makes it possible to approximately determine the ignition time,

$$\tilde{\tau}_{\text{ind}}' = \int\limits_{\ln\alpha_* - 2}^{\infty} \frac{d\Theta}{f(\Theta) - (1 - z')(\Theta + 1)} = \int\limits_{\ln\alpha_* - 2}^{\infty} \frac{2d\Theta}{\dfrac{\Theta^2}{b} + (a + z' - 1)\Theta + 2z'}, \tag{17}$$

where

$$a = \frac{1}{1 + \delta_i'(1 + \chi_i)}, \qquad b = \frac{(1 + \delta_i'(1 + \chi_i))^3}{1 - \delta_i'(1 + \chi_i)}.$$

Like the classical case, the extension of the lower integration limit to infinity (the corresponding error increases the calculated time by 15%) makes it possible to obtain an approximate analytic formula for the induction period,

$$\tilde{\tau}_{\text{ind}}' \approx \frac{2\pi\sqrt{b}}{\sqrt{2\left(1 - \dfrac{\alpha_*'}{\alpha_{*i}'}\right) - b\left(a - \dfrac{\alpha_*'}{\alpha_{*i}'}\right)^2}}. \tag{18}$$

If the rate of mass transfer is lower than the rate of chemical reactions (the Semenov number is very small), the values of both parameters a and b tend to 1. Then expression (18) transforms into formula (13).

Again, by substituting the upper integration limit in Eq. (17) by the current temperature, it is possible to obtain an approximate parametric dependence of the particle temperature on the time during the second stage, which takes the mass transfer of oxygen to the particle surface into account,

$$\tau_2(y) = \tau_{ch}'\,\frac{1 + \delta_i'(1 + \chi_i)}{1 + B_i} \times \int\limits_{\ln\alpha_* - 2}^{y} \frac{d\Theta}{f(\Theta) - (1 - z')(\Theta + 1)},$$

$$T(y) = T_i + y\,\frac{RT_i^2}{E}. \tag{19}$$

Figures 1, *a* and *c* demonstrate the corresponding dependences (curves *4*), which are in much better agreement with the solution of problem (2).

The final third stage (or the ignition stage) with chemical reactions running in the diffusion region can be included into the calculation scheme using formula (18).

8. Influence of Mode Parameters on the Induction Period

The total ignition time of a coke particle can be estimated by summing up the durations of both stages. In Fig. 2, a comparison is made for the calculation results of the ignition time obtained by approximate formulas (11) and (13) or (18), and the times of reaching points i_2 and i_3 numerically calculated according to Eq. (2). One can see that the application of classical formula (15) reflects, in general, the dynamics of the induction period dependence on the mode parameters (the oxygen concentration and the gas mixture temperature). However, the induction period calculated by Eq. (13) is shorter than the time required to reach the second inflection point, i_2, being almost half as large as the given induction period (the time interval to point i_3) at high oxygen concentrations and gas mixture temperatures.The account for the mass transfer allows the induction period to be evaluated more accurately. If the correction terms are taken into account, the appearance of expressions (13) and (18) for the induction period are similar to each other.

As the concentration of oxygen in a gas mixture decreases (Fig. 2, *a*), the induction period does not grow infinitely (when the critical ignition concentration is reached), which was predicted by Eq. (13) and Eq. (18). At this concentration, the inflection

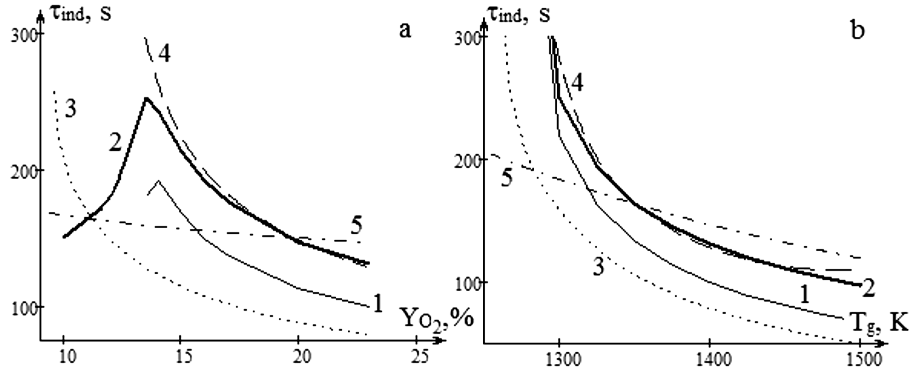

Fig. 2. Dependences of the induction period of a particle 100 μm in diameter (*a*) on the oxygen concentration ($T_g = 1400$ K and $T_w = 1500$ K) and (*b*) on the gas medium temperature ($Y_{\rm O_2} = 0.23$). Parallel reactions are taken into account. Curves: the time of reaching the second inflection point (point i_2) (*1*), induction period (point i_3) (*2*), dependence (14) (*3*), dependence (18) (*4*), dependence (20) (*5*)

points i_1 and i_2 in the time dependence of the particle temperature degenerate. The combustion mode changes from quasidiffusion (the diffusion-kinetic ratio ${\rm Se}_m > 1$ at the maximum combustion temperature) to quasikinetic (${\rm Se}_m < 1$). Accordingly, the combustion temperature gradually decreases to values close to the gas mixture temperature (Fig. 1, *b*).

As the temperature of a gas mixture decreases to the critical ignition temperature (Fig. 2, *b*), the ignition time of a coke particle increases infinitely.

Experimental studies of the ignition time of coal particles ranging from 150 to 800 μm in size were carried out in work [3]. The volume content of oxygen in a gas composition was varied from 5% to 21%, and the gas temperature was varied from 1200 to 1600 K, with the temperature of the walls in the reaction chamber being different by no more than 10–30 K from the gas temperature. The ignition time of dry coke remnants of coal particles was described by the dependence

$$\tau_{\rm ind} = 1.105 \times 10^{10}\, k_{ik} \frac{\rho d^{1.2}}{T_g^3} \left(\frac{0.23}{Y_{\rm O_2}}\right)^{0.15}. \qquad (20)$$

Here, the initial diameter of a coke particle d is expressed in meter units, the gas temperature T_g in Kelvins, the particle density ρ_c in kg/m^3 units, the oxygen concentration $Y_{\rm O_2}$ in relative units, the time in seconds, and k_{ik} is an experimentally determined coefficient, which is different for different coal blends (for anthracite, it equals 1.6). Dependence (20) was obtained experimentally. It includes the sum of the time required for a coal particle to be heated up to the ignition of volatile components, the time of the burning of those components, and the time required for the coke particle to be heated up. The formula demonstrates a weak dependence of the ignition time on the oxygen concentration in the gas and a rather strong dependence on the ambient temperature. It can be used for large particles provided constant external conditions. But it is applicable for particles smaller than 150 μm (Fig. 2), although expression (20) does not involve such parameters as the critical oxygen concentration and the temperature of gas mixture.

9. Conclusions

By analyzing the characteristic points in the time dependence of the coal particle temperature, it is shown that the inflection points cannot serve as indicators of the boundaries between separate stages during the induction period. When analytically determining the duration of the heating stage, it is better to associate its end with a temperature that is shifted from the inflection point by one characteristic interval. When calculating the duration of the further particle heating owing to chemical reactions, the mass transfer has be taken into account. In this work, we have proposed a new method which is based on the introduction of the dimensionless temperature as the ratio between the particle and ignition temperatures. This method allows us to find an analytic expression for the induction period, in which the parallel chemical reactions and mass transfer are taken into account.

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ВРАХУВАННЯ МАСООБМІНУ ПРИ ВИЗНАЧЕННІ КРИТИЧНИХ УМОВ І ЧАСУ ЗАЙМАННЯ ЧАСТИНКИ КОКСУ ВУГІЛЛЯ

Резюме

Проводиться аналіз часових залежностей температури частинки коксу при її займанні в нагрітому газі з метою аналітичного визначення періоду індукції з урахуванням масопереносу кисню до поверхні частинки. Весь час займання ділиться на характерні стадії. Показано, що точки перегину не можуть бути межами окремих стадій періоду індукції. Закінчення тривалості стадії прогрівання при її аналітичному визначенні краще співвіднести з температурою, яка менша за температуру перегину на один характерний інтервал. Для тривалості подальшого прогрівання частинки при хімічному реагуванні слід враховувати масоперенос. Запропоновано новий метод аналітичного визначення тривалості періоду індукції, який заснований на введенні безрозмірної температури відносно температури займання. Він дозволяє врахувати протікання паралельних хімічних реакцій і масоперенос (в перехідній і дифузійній областях горіння).