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Study of high-temperature heat-mass transfer of two-fraction gas suspensions and individual carbon particles in heated air

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In the work, the regularities of high-temperature heat-mass exchange in two-fraction gas suspensions of carbon particles in heated air are studied. These studies are relevant for predicting high-temperature processes in combustion chambers and determining the main characteristics of these processes: induction period, time and temperature of combustion, critical parameters of ignition and extinction. Physico-mathematical modeling of the problem is carried out on the basis of the equations of heat and mass transfer and chemical kinetics for the components of the gas suspension, taking into account the molecular-convective and radiation mechanisms of heat transfer. The characteristics of ignition, burning, and extinction of a two-fraction gas suspension of carbon particles with equal mass concentrations of small (60 μm) and large (120 μm) fractions in the temperature range of 1100 \div 1500 K were studied; a comparison was made with the characteristics for single particles of the corresponding diameters. It has been proven that when the gas temperature decreases, the induction period of the fine fraction can exceed the induction period of the large fraction, while the combustion temperature of small particles becomes lower than the combustion temperature of large particles. At high gas temperatures (1400 K, 1500 K), the situation changes to the opposite. The critical ignition and extinction parameters of two-fraction gas suspensions (gas temperatures, particle diameters) were found.

Keywords: heat and mass transfer, ignition, combustion, extinction, gas suspension, carbon particles.

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Introduction

Coal will continue to be one of the main energy resources for the next few decades due to its low price and abundant reserves [1, 2]. The products of coal combustion and gasification (oxides of carbon, nitrogen, sulfur and solid residues) lead to environmental pollution and are a source of anthropogenic emissions of greenhouse gases [1, 3]. Therefore, there is a need to develop new clean technologies for the use and processing of coal raw materials [4]. Currently, two ways of reducing CO₂ emissions are being considered: burning pulverized coal in different gas media, such as O₂/H₂O, O₂/C₂O, O₂/N₂, with different oxygen concentrations, and using biomass particles. Many experimental and theoretical works combine these two directions [5-10]. The physical-mathematical model of high-temperature heat-mass transfer and kinetics of chemical transformations of a two-fraction gas suspension of particles is based on the

differential equations of heat and mass balances for the particles of each of the fractions and the corresponding equations for the gas mixture containing the oxidant [1,2].

Combustion of coal fuel is a complex process that includes successive, more often sequential-parallel stages: the release of volatile substances, ignition and burning of volatile substances [8, 11, 12], ignition and burning of coke residue (carbon), the appearance of combustion products (gaseous and solid), which are harmful to the environment. The stage of heterogeneous oxidation of carbon is the main process that occurs during high-temperature processing of coal and occurs simultaneously or after the release of volatile substances, depending on the reaction conditions [13]. The time required for the combustion of a coke particle can be several orders of magnitude longer than the time for the release of volatiles and is often the rate-determining step in the total combustion rate of the pulverized fuel. The physical structure of coal, including pore structure, surface area,

particle size, and inorganic content are important parameters for understanding and modeling high-temperature coal oxidation processes. The chemical properties of coal affect the reactivity of carbon fuel, critical parameters of ignition, and are important factors for predicting and preventing self-ignition of fuel in the places of its extraction and storage [14-17]. Based on research [16], a method for determining the kinetic parameters of coal oxidation was proposed, which can be used for numerical modeling. In addition, the stages at which various factors, namely temperature or oxygen concentration, are dominant in the process of high-temperature oxidation have been determined.

The development of effective modes of using dispersed natural fuels in energy systems requires knowledge about the characteristics of high-temperature heat-mass exchange of fuel-gas systems depending on the parameters of the technological process. One of these most important parameters is temperature, which affects both the intensity of heat and mass transfer and the kinetics of the reaction of the solid phase with the active gas. Temperature is a key element for determining [18]: combustion reaction rate; calculation of the heat flow to or from the particle due to convection and radiation; concentrations of gaseous components - reaction products. The temperature of a burning particle correlates with such studied parameters as the change in particle diameter, loss of their mass, combustion time, etc.

In addition, other studied parameters can be [18]:

- particle burning time as a function of particle size under different temperature conditions of the process;
- particle ignition time (induction period), determined by the time from the beginning of particle heating to the moment of self-acceleration of the oxidation reaction on its surface [19]; in experiments, this moment is indicated by the appearance of a flame on the surface of a particle or an excess of its temperature above the gas temperature [18];
- time of separate stages of combustion of particles – time of combustion of volatile substances, time of combustion of carbon residue (coke);
- change in size and mass loss of particles during combustion.

The loss of mass of particles during combustion is due to both a decrease in their outer diameter and a decrease in density due to the burning of carbon in the pores. The coal mass is permeated with pores, which can have different sizes [20] depending on the type of coal and the number of volatile substances. Basically, these are micropores that have a sufficiently good fractal structure [21] and chemical reactions during oxidation and combustion of particles can occur in its pores. Coal with a developed inner surface of pores has a stronger tendency to ignite and burn [20-22].

Solid fuel is burned in a pulverized state in pulverized coal boilers or in a pulverized state in vortex furnaces and fluidized bed furnaces. [1, 4, 23]. During the last decades, the main one is the pulverized method of combustion, when pulverized coal fuel is fed into the furnace with the air used for combustion.

Complex processes occurring in energy systems can also be studied with the help of software modeling of physico-chemical, gas-dynamic processes and

technological features of various ways of using pulverized coal systems [24]. Development of physical-mathematical models, conducting calculation studies is the optimal method for understanding the processes that occur in real conditions. This makes it possible to also predict the behavior of fuel systems under conditions when conducting an experiment is impossible. Currently, physical and mathematical models of combustion of mixtures of coal dust particles with air and propane-air mixtures in different reaction volumes at different mass concentrations of dust and oxidant concentrations are being actively developed [14, 15, 25-27]. The analysis of experimental and theoretical works showed that there is no study of the phenomenon of physical underfire, which consists in the spontaneous extinction of burning particles when they reach critical dimensions. The phenomenon of spontaneous extinction is associated with a change in the heat balance of particles as a result of the competition between the processes of chemical heat release and heat flow from particles when their diameter decreases due to combustion [19]. Reducing the underburning of particles is one of the important tasks of increasing the efficiency and purity of coal fuel combustion. Research [22] showed that the use of carbon fuel with a developed pore structure leads to an increase in the completeness of combustion.

Pulverized coal fuel, which is blown into the reaction volume of power plants, burns in the form of gas suspensions. It is polydisperse, that is, it contains particles of different sizes. A special case of polydisperse fuel is a two-fraction gas suspension, which mainly contains particles of two sizes (small and large fractions) in different mass ratios. In the general reaction mixture, particles of different sizes react with different intensity at different temperatures of the surface and the concentration of the oxidant near it [11, 23]. The peculiarities of high-temperature processes in two-fraction carbon systems at different temperatures have not been sufficiently studied. The purpose of the work is to study the patterns of high-temperature heat-mass transfer (ignition, burning, and extinction) of two-fraction gas suspensions of carbon particles in the air at different temperatures of the heated gas and to compare them with the course of these processes for a single particle placed in an air medium heated to the same temperature. Studying the behavior of a single particle in a heated gas is necessary to assess the influence of the collective effect. In gas suspensions, oxygen is consumed by a large number of particles, and because of this, the combustion mechanism may be different [18]. To determine the characteristics of ignition, burning and extinguishing of gas suspensions and individual carbon particles, a physical and mathematical modeling of the problem and a numerical experiment were carried out under different temperature conditions and mass concentrations of fuel.

I. Theoretical part

The physical-mathematical model of high-temperature heat-mass transfer and kinetics of chemical transformations of a two-fraction gas suspension of particles is based on the differential equations of heat and mass balances for the particles of each of the fractions and

the corresponding equations for the gas mixture containing the oxidant [19, 28].

When carbon fuel enters the heated oxidizer, chemical reactions begin to occur on the surface and in the pores of the particles, as a result of which the particles ignite and burn. For gas temperatures in the range of 1100 ÷ 1500 K, you can use the scheme of two parallel chemical reactions between carbon and oxygen [23]: $C + O_2 = CO_2$ (I), $2C + O_2 = 2CO$ (II). The expression for the total density of chemical heat release for a particle of the i -th fraction as a result of surface and internal reaction in the pores is written in the form [28]:

$$q_{ch} = (k_{1i}q_1 + k_{2i}q_2)\rho_g n_{O_{2,si}}(1 + K_i), K_i = \frac{k_{vi}}{k_{1i} + k_{2i}}, \quad (1)$$

where q_{ch} – total density of chemical heat release on the surface and in the pores of the particle, k_{1i}, k_{2i} – rate constants of chemical reactions (I) i (II), q_1, q_2 – thermal effects of chemical reactions (I) i (II), ρ_g – gas density, $n_{O_{2,si}}$ – relative mass concentration of oxygen on the surface of a carbon particle; k_{vi} – effective internal response constant [19, 22].

In the combustion chamber, molecular - convective (q_{ai}) and radiative (q_{wi}) heat exchange of particles with the heated gas and with the walls of the reaction volume takes place. The densities of these heat flows can be found from the equations, respectively:

$$q_{ai} = q_{\lambda i} + q_{sfi}, \quad q_{\lambda i} = \alpha_i(T_i - T_g), \quad \alpha_i = \frac{Nu\lambda_g}{d_i}, \quad (2)$$

$$q_{wi} = \varepsilon\sigma(T_i^4 - T_w^4), \quad (3)$$

where $q_{\lambda i}$ is the component of the heat flux density due to molecular convective heat exchange, q_{sfi} is the component of the heat flux density due to the Stefan current [22], α_i is the heat exchange coefficient of the particle with the gas, T_i, T_g, T_w respectively: the temperature of the particle, the gas and the walls of the reaction chamber, λ_g is the coefficient of thermal conductivity of the gas, d_i is the diameter of the particle.

Let's write the heat and mass balance equation for a particle of the i th fraction:

$$\frac{c\rho_i d_i}{6} \frac{\partial T_i}{\partial t} = q_{chi} - q_{ai} - q_{wi}, \quad T_i(t = 0) = T_{ib}, \quad (4)$$

$$-\frac{1}{2}\rho_i \frac{\partial(d_i)}{\partial t} = W_{si}, \quad W_{si} = \frac{M_C}{M_{O_2}}(k_{1i} + 2k_{2i})n_{O_{2,si}}\rho_g, \\ d_i(t = 0) = d_{bi} \quad (5)$$

$$-\frac{1}{6}d_i \frac{\partial(\rho_i)}{\partial t} = W_{si}K_i, \quad \rho_i(t = 0) = \rho_{bi}. \quad (6)$$

Here c is the specific heat capacity of the particle, ρ_i is its density, t is time, M_C, M_{O_2} are the molar masses of carbon and oxygen, respectively.

During the combustion of pulverized coal gas suspension in the reaction volume, the gas temperature and oxygen concentration change over time. The heat and mass balance equations for gas, taking into account the external heat and mass exchange, have the following form:

$$c_g \rho_g \frac{\partial T_g}{\partial t} = \sum_{i=1}^n S_i C_{N_i} q_{ai} + \alpha_g F_g (T_g - T_{g\infty}), \quad T_g(t = 0) = T_{g\infty}, \quad (7)$$

$$-\frac{\partial n_{O_{2,g}}}{\partial t} = \sum_{i=1}^n C_{N_i} S_i n_{O_{2,si}} (k_{1i} + k_{2i} + k_{vi}) - F_g \beta_g (n_{O_{2,\infty}} - n_{O_{2,g}}), \quad n_{O_{2,g}}(t = 0) = n_{O_{2,\infty}}, \quad (8)$$

where c_g is the specific heat capacity of the gas, S_i is the surface area of the particle, F_g is the specific surface area of the gas suspension, $n_{O_{2,g}}$ is the relative mass concentration of oxygen in the gas, α_g, β_g are the coefficients of heat exchange and mass exchange of the gas suspension with the environment, C_{N_i} is the numerical concentration of particles of the i th fraction, which is related to the mass concentration of carbon fuel by the ratio:

$$C_{m_i} = \frac{1}{6}\pi \cdot d_i^3 \cdot \rho_i \cdot C_{N_i}, \quad C_m = \sum_{i=1}^n C_{m_i}.$$

The calculations of the presented equations make it possible to determine the main characteristics of the combustion of gas suspensions: the ignition delay time or the induction period, the time and temperature of combustion, the critical parameters of ignition and extinction (gas temperatures, particle diameters, fuel and oxidizer concentrations). A two-fraction gas suspension ($i = 2$) with particle diameters of the fine fraction of 60 μm and the large fraction – 120 μm with equal mass concentrations of the fractions was taken for calculations.

The studies were conducted for gas temperatures in the reaction chamber from 1100 to 1500 K.

II. Results of physical and mathematical modeling and their analysis

We will conduct a study of the influence of the heated gas temperature on the ignition characteristics of two-fraction gas suspensions and single particles of the same initial diameters. Figure 1a, b shows the dependence of the induction period t_{ind} of a two-fraction gas suspension and a single particle on the initial gas temperature. Fig. 1a shows that for gas temperatures $T_{g\infty} > 1400$ K, the induction period of the small fraction is shorter than that of the large fraction. Then, as the gas temperature decreases, the induction periods of the small and large fractions converge, and at air temperatures $T_{g\infty} < 1400$ K, the particles of the large fraction engage earlier than the small ones. The increase in the induction period of the small fraction compared to the coarse fraction for temperatures $T_{g\infty} < 1400$ K is explained by an increase in the heat transfer coefficient to the gas with a decrease in

the particle diameter (formula 2). It is known that the ignition of a particle consists of two stages [19]: the stage of inert (thermal) heating to a temperature close to the temperature of the gas, and the stage of chemical heating, during which self-acceleration of chemical reactions on the surface of the particle occurs. Therefore, the induction period is the sum of the times of inert and chemical heating: $t_{ind} = t_h + t_{ch}$.

Table 1 shows that if the gas temperature is reduced for the fine fraction, the time of the chemical stage of the induction period increases much more than for the large fraction. When the gas temperature decreased from $T_{g\infty}=1500$ K to $T_{g\infty}=1300$ K, the chemical heating time of small particles increased almost 27 times. If we compare the time of inert heating (t_h) and the time of chemical heating (t_{ch}) of small particles, it has increased by more than 7 times. The situation is different for a large fraction of gas suspension with particle diameters of $120 \mu\text{m}$. For these particles, regardless of the gas temperature, the duration of the chemical stage of the induction period is approximately 4 times shorter than the duration of the inert heating stage. Thus, in the region of high temperatures, the period of induction of a two-fraction gas suspension of particles is determined by the period of induction of small particles, and in the region of low temperatures - by the period of induction of particles of a large fraction.

Table 1.

Characteristic heating times of particles at the ignition stage of a two-fraction gas suspension at different initial gas temperatures.

$T_{g\infty}, \text{K}$	1500		1300	
$d_b, \mu\text{m}$	60	120	60	120
t_h, mc	15.6	34.1	42.8	127.4
t_{ch}, mc	12.2	23.2	335.9	99.7

At a certain critical gas temperature T_{cr} (Fig. 1 a, b), the particles do not engage (the induction period runs to infinity). This gas temperature is called the ignition temperature. From fig. 1a and 1b, it follows that the critical ignition temperature of a two-fraction gas suspension is significantly lower than the ignition temperature of single particles of the same diameter. Moreover, the critical ignition temperatures for individual large (T_{cr2}) and small (T_{cr1}) particles differ significantly (Fig. 1, b), and in the conditions of a two-fraction gas suspension, they practically coincide (Fig. 1 a).

Figure 1 c, d shows the burning time of a two-fraction gas suspension and individual particles depending on the initial temperatures of the heated gas. It can be seen that the burning times of small and large fractions in the gas suspension are weakly dependent on the gas temperature. The burning time of the large fraction is almost 4.5 times

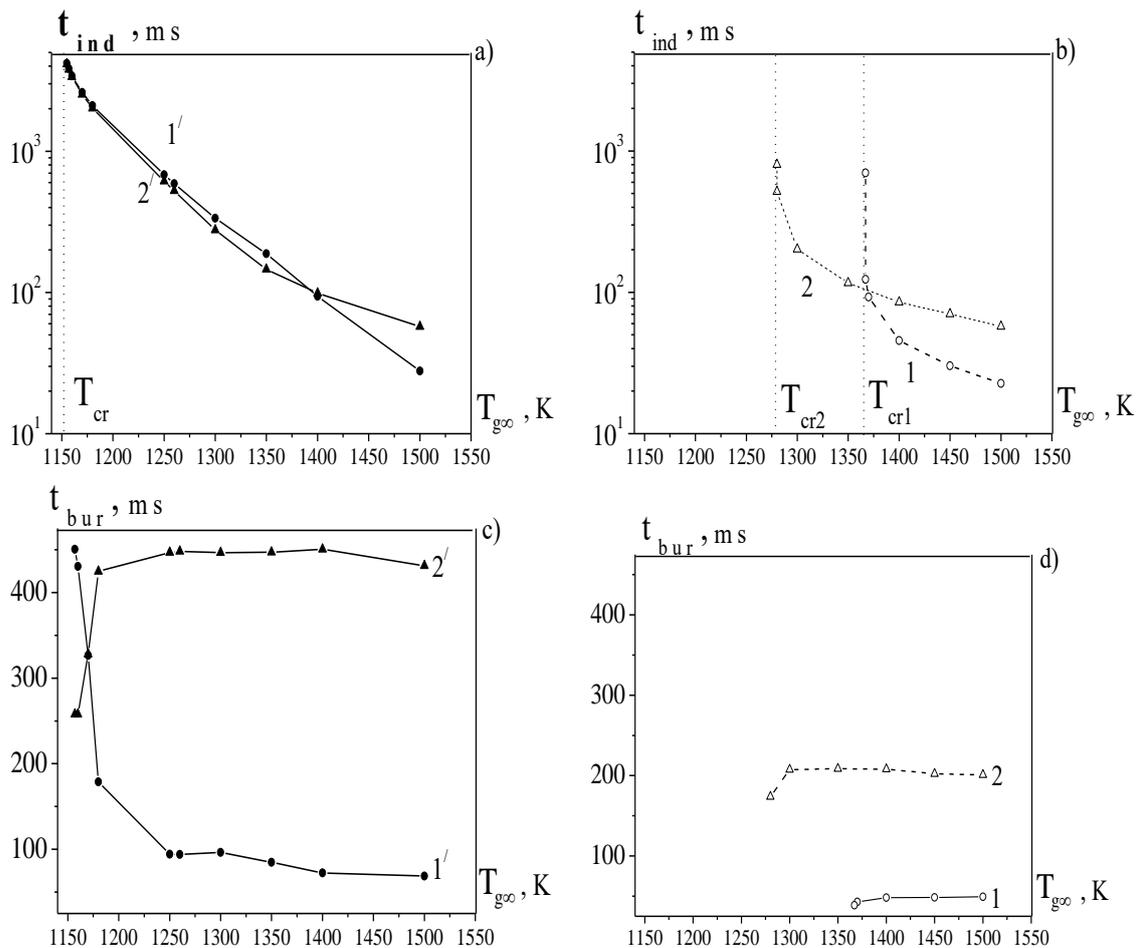


Fig. 1. Dependencies of the induction period (t_{ind}) and burning time (t_{bur}) on the initial gas temperature ($T_{g\infty}$) for: a), c) – a two-fraction gas curtain (curves 1' – $\bullet\bullet\bullet\bullet$ – $d_{b1}=60 \mu\text{m}$, 2' – $\blacktriangle\blacktriangle\blacktriangle\blacktriangle$ – $d_{b2}=120 \mu\text{m}$, $C_{mb}=0.0244 \text{ kg/m}^3$) and b), d) – subunit part (curves 1 – $\circ\circ\circ\circ$ – $d_b = 60 \mu\text{m}$, 2 – $\triangle\triangle\triangle\triangle$ – $d_b = 120 \mu\text{m}$). d_b is the initial diameter of the particle, C_{mb} is the initial mass concentration of the fuel.

longer than the burning time of the small fraction, with the ratio of their initial diameters equal to 2, which confirms the diffusion mode of burning of these particles. The combustion times of particles of the gas suspension of both fractions are approximately 2 times longer than the combustion time of single particles of the corresponding diameters, which is due to a significant decrease in the coefficient of excess oxygen in the volume of the gas suspension.

It should be noted that near the critical gas temperature T_{cr} , the ignition delay time of a single particle is much shorter than the ignition delay time of particles of identical sizes in the conditions of a gas suspension (Fig. 2).

It follows from Figure 2a, b that the critical ignition conditions for a single particle are clearly expressed. Namely, when the gas temperature decreases by just one degree, the particle does not ignite, but warms up to a temperature slightly higher than the gas temperature (curve 2, Fig. 2a). The critical extinction conditions for a single burning particle are also clearly defined, namely: when the diameter of the particle reaches a critical value, its temperature and oxidation rate decrease sharply (curves 1, Fig. 2a, b). The extinction of particles in the conditions of a gas suspension occurs in a «degenerate» mode [19] due to the proximity of the temperatures of particles and gas (Fig. 2c, d). There is no clear break on the $d(t)$

dependences, which would characterize a sharp change in the oxidation regime.

Figure 3 shows the characteristics of combustion and extinction of a two-fraction gas suspension at different initial temperatures of the heated air. The maximum combustion temperature of particles of the fine fraction in the region of low gas temperatures is lower than that of large ones (Fig. 3, a) due to large heat losses to the gas and the lack of oxidant during the combustion stage. The concentration of the oxidant at the moment of ignition and burning of small particles is significantly reduced as a result of oxygen consumption during the reaction with large particles that are ignited earlier.

In fig. 3, b shows the dependences of the extinction diameters d_E of the gas suspension particles on the initial temperature of the heated gas in which they are located. Extinction of the particles of the gas suspension occurs in a degenerate mode, since at the final stage of combustion the temperatures of the particles and the gas differ slightly. When the diameter of the particle reaches a critical value, there is a slight decrease in the rate of oxidation. After the extinction moment, the particles continue to oxidize slowly in the kinetic mode [19].

From fig. 3b shows that in the conditions of a gas suspension, the critical attenuation diameters d_E of the large and small fractions differ, since the attenuation of the fractions occurs at different gas temperatures. Large

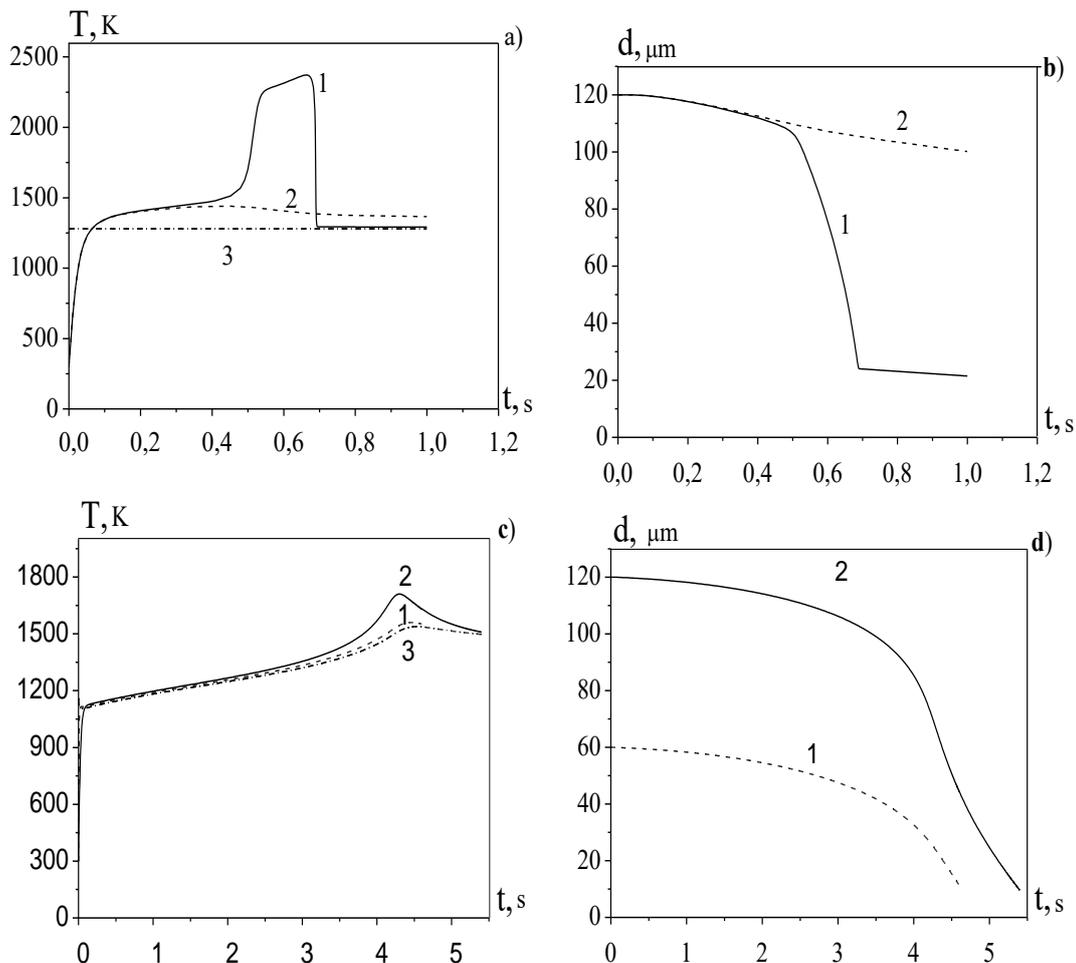


Fig. 2. Time dependences of temperature and diameter of particles near critical ignition conditions: a, b) single particle $d_b = 120 \mu\text{m}$, 1 – $T_{g\infty} = 1280 \text{ K}$; 2 – $T_{g\infty} = 1279 \text{ K}$; 3 – level $T_{g\infty}$ c, d) two-fraction gas suspension: 1 – $d_{b1} = 60 \mu\text{m}$, 2 – $d_{b2} = 120 \mu\text{m}$, 3 – level T_g ; $T_{g\infty} = 1155 \text{ K}$.

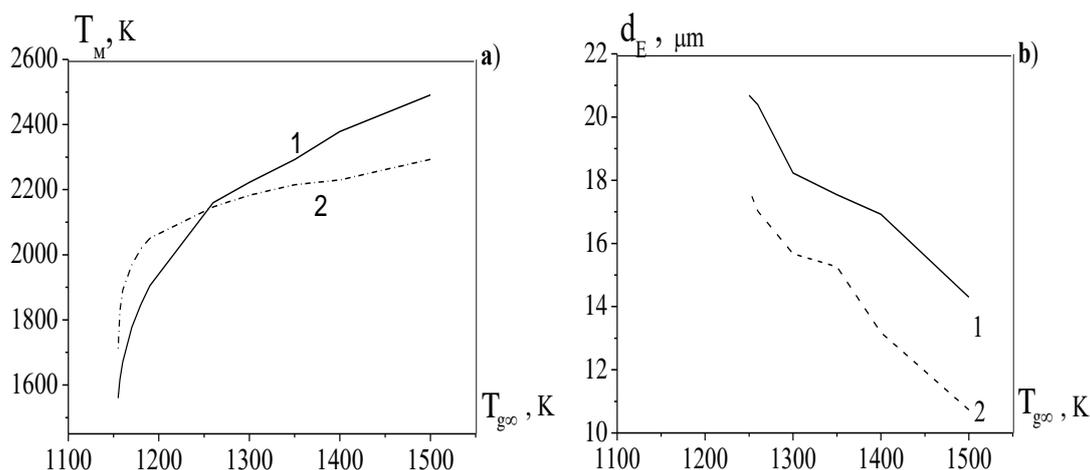


Fig. 3. Dependencies of the maximum combustion temperature T_M (a) and the extinction diameter d_E (b) of the particles of the two-fraction gas suspension on the initial gas temperature, 1 – $d_{b1}=60 \mu\text{m}$, 2 – $d_{b2}=120 \mu\text{m}$.

particles are quenched at a higher gas temperature and are characterized by a smaller quenching diameter compared to small particles. For a single particle, the extinction diameter does not depend on the initial diameter of the particle and is determined only by the temperature of the surrounding gas.

Conclusions

Thus, it was found that at high gas temperatures, the period of induction of the small fraction of gas suspension is less than that of the large fraction, and at low temperatures, on the contrary, it is longer. This is due to the growth of molecular-convective heat losses to the surrounding gas for the small fraction. It is shown that the ignition temperatures of small and large particles under the conditions of a two-fraction gas suspension practically coincide, while those of single carbon particles of the same diameters differ.

It was established that in the region of low gas temperatures, the combustion temperature of particles of the small fraction is lower than that of the large fraction. This is explained by the large heat removal from small particles by molecular-convective means and the lack of oxygen for a stoichiometric ratio with the fuel at the stage

of combustion. Oxygen was consumed during the combustion of large particles, which ignite earlier at low gas temperatures.

The duration of the stable high-temperature stage of heat and mass transfer (burning time) of carbon particles in the conditions of a two-fraction gas suspension was calculated, as well as the critical diameters of the particles, reaching which they extinguish. It is proved that, unlike a single particle, in a gas suspension the critical diameter of extinction of particles of large and small fractions is different, which is due to the unequal temperature of the surrounding gas at the moment of extinction of particles of different initial diameters. At the same time, the extinction of gas suspension particles, unlike a single particle, has a "degenerate" character.

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Вивчення високотемпературного тепломасообміну двофракційних газозависів і поодиноких вуглецевих частинок в нагрітому повітрі

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В роботі проведено дослідження закономірностей високотемпературного тепломасообміну в двофракційних газозависах вуглецевих частинок в нагрітому повітрі. Ці дослідження актуальні для прогнозування високотемпературних процесів в камерах згорання та визначення основних характеристик цих процесів: періоду індукції, часу та температури горіння, критичних параметрів займання і згасання. Фізико-математичне моделювання задачі проведено на основі рівнянь тепломасообміну та хімічної кінетики для компонент газозавису з урахуванням молекулярно-конвективного і променевого механізмів переносу тепла. Вивчено характеристики займання, горіння та згасання двофракційного газозавису вуглецевих частинок з рівними масовими концентраціями дрібної (60 мкм) і великої (120 мкм) фракцій в інтервалі температур 1100 ÷ 1500 К; проведено порівняння з характеристиками для поодиноких частинок відповідних діаметрів. Доказано, що при зниженні температури газу період індукції дрібної фракції може перевищувати період індукції великої фракції, при цьому температура горіння дрібних частинок стає нижчою за температуру горіння великих частинок. При високих температурах газу (1400К, 1500К) ситуація змінюється на протилежну. Знайдено критичні параметри займання і згасання двофракційних газозависів (температури газу, діаметри частинок).

Ключові слова: тепломасообмін, займання, горіння, згасання, газозавис, вуглецеві частинки.