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IGNITION OF A METALLIZED COMPOSITE PROPELLANT BY A HOT PARTICLE

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Abstract: A theoretical study is executed for the process of the solid-phase ignition of a metallized composite propellant by a heated to a high temperatures particle. The process characteristics are calculated by taking into account in an explicit form the structural heterogeneity of the propellant due to finely dispersed metal particles because of the significant difference in their thermal conductivity from the thermal conductivity of the polymer matrix. Besides the process characteristics are calculated by averaging the thermophysical properties of the propellant according to known equation. The ignition delay time of the propellant are established in a wide variation range of the initial temperature (800–1500 K) of the local heat source. It was found that in the case when the structure of the metallized composite propellant is described by a homogeneous medium with effective values of thermophysical properties, the difference between the calculated ignition delay times and the known experimental data is about 75 %. The difference between the calculated ignition delay times and the known experimental data does not exceed the measurement error of 10–15 %

when mathematical model describes the real heterogeneous structure of propellant. It is shown that taking into account the structural heterogeneity of metallized propellant has a greater influence on the reliability of the results of mathematical modeling, and the influence of the distribution of finely dispersed metal particles is negligible.

Keywords: metallized composite propellant, hot particle, ignition, mathematical modeling.

INTRODUCTION

Investigation of the conditions and characteristics of the processes of heating and ignition of high-energy materials is an urgent task in connection with the widespread use of such materials as a fuel [1–6] in engines and gas generators for various purposes.

The addition of a metallic component to the composite propellant affects the conditions and characteristics of the combustion initiation. It is connected with the dependence of the thermophysical properties of the heated near-surface layer of composite propellant on the concentration of the dispersed metal.

Mathematical modeling of physicochemical processes occurring during ignition can be performed when implementing two approaches for description the thermophysical properties of metallized composite propellant:

1. Accounting for a real heterogeneous structure.

2. Representation of a heterogeneous structure by a homogeneous medium with average values of thermophysical properties.

The following expressions are widely known for averaging the thermophysical properties [7–10]:

$$\rho_e = \rho_c \varphi_c + \rho_d \varphi_d ; \tag{1}$$

$$C_e = C_c \mu_c + C_d \mu_d , \tag{2}$$

where the subscripts *e*, *c*, *d* is effective value, continuous phase, and discontinuous phase, respectively, φ is the volume fraction, and μ is the mass fraction.

Thermal conductivity is one of the most significant factors when calculating the ignition delay time of fuel. Metallized composite propellant can be represented as a polymer matrix (combustible component and oxidizer) with inclusions of finely divided metal particles. There are several well-known equations [8–21] for calculating the thermal conductivity:

$$\lambda_e = \lambda_c \mu_c + \lambda_d \mu_d ; \tag{3}$$

$$\lambda_e = \lambda_c \varphi_c + \lambda_d \varphi_d ; \tag{4}$$

$$\frac{1}{\lambda_e} = \frac{\varphi_c}{\lambda_c} + \frac{\varphi_d}{\lambda_d} ; \tag{5}$$

$$\lambda_e = \lambda_c \left[\frac{(\lambda_d / \lambda_c + 2) + 2(\lambda_d / \lambda_c - 1)\varphi_d}{(\lambda_d / \lambda_c + 2) - (\lambda_d / \lambda_c - 1)\varphi_d} \right]. \tag{6}$$

$$\lambda_e = \frac{1}{4}\left[\gamma + \sqrt{\gamma^2 + 8\lambda_c \lambda_d} \right], \tag{7}$$

where $\gamma = (3\varphi_d - 1)\lambda_d + (3\varphi_c - 1)\lambda_c$;

$$\lambda_e = \lambda_c \frac{(2 + 2\gamma\varphi_d)(2 + (2\gamma - 1)\varphi_d)}{(2 - \gamma\varphi_d)(2 - (\gamma + 1)\varphi_d)}, \tag{8}$$

where $\gamma = \dfrac{\lambda_d - \lambda_c}{2\lambda_c + \lambda_d}$;

$$\frac{1}{\lambda_e} = \frac{1 - B}{\lambda_c} + \frac{1}{B'\sqrt{CC'}} \ln\left[\frac{B' + B\sqrt{C}C'/2}{B' - B\sqrt{C}C'/2} \right], \tag{9}$$

where $B=\sqrt{3\varphi_d/2}$, $C=4\sqrt{2/3\varphi_d}$, $B'=\sqrt{\lambda_c+B(\lambda_d-\lambda_c)}$, $C'=\sqrt{\lambda_d-\lambda_c}$;

$$\lambda_e=\lambda_c\left[1+3\gamma\varphi_d+3\gamma^2\left(1+\frac{\gamma}{4}+\frac{\gamma^2}{256}+\ldots\right)\varphi_d^2\right], \tag{10}$$

where $\gamma=\dfrac{\lambda_d-\lambda_c}{2\lambda_c+\lambda_d}$;

$$\lambda_e=\lambda_c\left[\frac{1+AB\varphi_d}{1-B\psi\varphi_d}\right], \tag{11}$$

where A=1–k_e, $\psi=1+\dfrac{1-\varphi_{d\max}}{\varphi_{d\max}^2}\varphi_d$, $B=\dfrac{(\lambda_d/\lambda_c-1)}{(\lambda_d/\lambda_c+A)}$.

The purpose of the present work is analysis of the influence of a mathematical description of the thermophysical properties of a metallized composite propellant on the reliability of the prediction of ignition characteristics.

FORMULATION OF THE PROBLEM

The following scheme was proposed for the solid-phase ignition of a metallized composite propellant. The hot particle is situated on the surface of a composite propellant at the initial time (Fig. 1). As a result of conductive heat transfer from the local heat source, the near-surface layer of composite propellant is heated. The temperature rising in the heated region of the fuel initiates exothermic interaction of the combustible component (butyl rubber) and oxidant (ammonium perchlorate). The reaction rate increases according to Arrhenius dependence, and ignition occurs.

Ignition criteria are following:

1. The heat released as a result of the exothermic reaction exceeds the heat outflow to the condensed phase from the local heat source.

2. The temperature in the oxidation reaction zone exceeds the temperature of the hot particle.

The numerical simulations is performed for a composite propellant consisting of a mixture of ammonium perchlorate (80 wt %), butyl rubber (15 wt %), and aluminum powder (5 wt %). The ignition source is a heated to a high temperatures (T_p=800–1500 K) steel particle in the shape of parallelepiped.

MATHEMATICAL MODEL

The ignition problem is solved in a rectangular coordinate system (Fig. 1). The processes occurring during the induction period are described by the following system of partial differential equations:

the heat conduction equation for the inert gas:

$$\rho_1 C_1 \frac{\partial T_1}{\partial t} = \lambda_1 \left(\frac{\partial^2 T_1}{\partial x^2} + \frac{\partial^2 T_1}{\partial y^2} \right); \tag{12}$$

the heat conduction equation for the hot particle:

$$\rho_2 C_2 \frac{\partial T_2}{\partial t} = \lambda_2 \left(\frac{\partial^2 T_2}{\partial x^2} + \frac{\partial^2 T_2}{\partial y^2} \right); \tag{13}$$

the heat conduction equation for the polymer matrix with account of the exothermic reaction:

$$\rho_3 C_3 \frac{\partial T_3}{\partial t} = \lambda_3 \left(\frac{\partial^2 T_3}{\partial x^2} + \frac{\partial^2 T_3}{\partial y^2} \right) + Q_3 W_3; \tag{14}$$

the heat conduction equation for the metal particles in composite propellant:

$$\rho_4 C_4 \frac{\partial T_4}{\partial t} = \lambda_4 \left(\frac{\partial^2 T_4}{\partial x^2} + \frac{\partial^2 T_4}{\partial y^2} \right); \tag{15}$$

The rate of the exothermic reaction between the combustible component and oxidizer in the heated area of composite propellant:

$$W_3 = \rho_3 k_3^0 \exp\left(-\frac{E_3}{R_t T_3}\right). \quad (16)$$

Here: t is the time, s; t_d is the ignition delay time, s; x, y are the coordinates, m; x_l, y_h are the sizes of the solution domain, m; x_p, y_p are the sizes of the hot particle, m; T is the temperature, K; λ is the thermal conductivity, W/(m K); ρ is the density, kg/m^3; C is the specific heat, J/(kg K); Q_3 is the fuel enthalpy of oxidation, J/kg; W_3 is the reaction rate of fuel oxidation, kg/(m^3·s); k_3^0 is the pre-exponential factor, s^{-1}; E_3 is activation energy, J/mole; R_t is the perfect gas constant, J/(mole·K); subscripts 1, 2, 3, and 4 denote, respectively, inert gas, hot particle, polymer matrix, aluminum particles.

Initial conditions:

initial temperature distribution is uniform in the solution area of the ignition problem (Fig. 1): in inert gas and metallized composite propellant $T=T_0$, in local heat source $T=T_g$.

Boundary conditions:

at the interfaces between the components with different thermophysical properties (polymer fuel–oxidizer, metal particles, inert gas, local heat source), perfect thermal-contact conditions are set:

$$\lambda_i \frac{\partial T_i}{\partial x} = \lambda_j \frac{\partial T_j}{\partial x},\ T_i = T_j;\ \lambda_i \frac{\partial T_i}{\partial y} = \lambda_j \frac{\partial T_j}{\partial y},\ T_i = T_j;$$

at the external boundary of the solution domain (Fig. 1), the conditions of absence of temperature gradients are set:

$$\frac{\partial T_i}{\partial x} = 0;\ \frac{\partial T_i}{\partial y} = 0.$$

The algorithm of numerical solution of ignition problem (Eqs. (12)–(16) with initial and boundary conditions) is based on the finite-difference method, locally one-dimensional method, iteration method, and sweep method. Ignition characteristics is calculated at the mesh points of the finite-difference grid at coordinate and time steps of up to 10 μm and 0.1 μs, respectively.

The conservativeness of the finite-difference scheme is carried out. The error within which the heat conservation law held in the ignition problem area (Fig. 1) is not exceed 1.5 %.

RESULTS AND DISCUSSION

The numerical simulation of the ignition process is carried out at the following parameters: the initial temperature of the metallized composite propellant and inert gas T_0=293 K, the initial temperature of the hot particle T_p=800–1500 K; the sizes of the hot particle x_p=2.5 mm, y_p=2 mm; the sizes of the aluminum particle x_m=y_m=0.08 mm; the sizes of the solution area x_l=y_h=8 mm.

The thermophysical properties [22–29] of the components (Fig. 1):

λ_1=0.026 W/(m·K); ρ_1=1.161 kg/m^3; C_1=1190 J/(kg·K);

λ_2=36 W/(m·K); ρ_2=8100 kg/m^3; C_2=545 J/(kg·K);

λ_3=0.472 W/(m·K); ρ_3=1776 kg/m^3; C_3=1260 J/(kg·K);

λ_4=343 W/(m·K); ρ_4=2700 kg/m^3; C_4=930 J/(kg·K).

The kinetic parameters of the exothermic reaction between the combustible component and oxidizer in the composite propellant [22–25]: E_3=50·10^3 J/mole, $Q_3k_3^0$=1.69·10^9 J/(kg·s).

The main task of the theoretical investigation is to establish the influence of the description method of the heterogeneous structure of metallized composite propellant on the ignition delay time (t_d) and on the minimum initial temperature (T_p) of the local heat source necessary to initiate the combustion process. The analysis is carried out by two approaches:

1. The real heterogeneity of the propellant is taken into account due to the finely dispersed metal particles.

2. The heterogeneous structure of propellant is interpreted as a homogeneous medium with effective values of thermophysical properties, which is calculated by Eqs. (1)–(11).

Fig. 2 shows the dependences $\lambda_e=f(\mu_d)$ of the thermal conductivity of metallized propellant on the mass fraction of finely divided aluminum particles calculated by Eqs. (3)–(11). It was found that the variation of the concentration of metal powder (μ_d=5–25 %) in the propellant has a significant effect on the value of λ_e (Fig. 2*a*), especially if λ_e is calculated by Eqs. (3) and (4). It is found that the effective value of the thermal conductivity of a metallized propellant (e.g. λ_e=86.1 W/(m·K) at μ_d=25 %), calculated by Eqs. (3)–(11), differs by a factor of tens from the thermal conductivity λ_3=0.472 W/(m·K) of the main fuel component – the polymer matrix. This fact allows formulating a hypothesis about a possible difference in the conditions and characteristics of the ignition of a metallized composite propellant

with effective thermophysical properties calculated using the λ_e values corresponding to Fig. 2*a* and 2*b*.

Fig. 3 shows the dependence of the ignition delay time (t_d) of the metallized propellant on the initial temperature (T_p=800–1500 K) of the local heat source. Region *3* (see Fig. 3) corresponds to the results of the numerical simulations in which the thermal conductivity of the metallized propellant is calculated by Eqs. (5)–(11) (see the values of λ_e at μ_d=5 % in Fig. 2*b*). Curves *4* (see Fig. 3) correspond to the conditions of the ignition of the metallized propellant with λ_e values calculated by Eqs. (3) and (4) (see the values of λ_e at μ_d=5 % in Fig. 2*a*). The higher thermal conductivity of the metallized propellant the large depth of heat-up of the propellant near-surface layer. As a result of the intense cooling of the local heat source and heat sink from the exothermic reaction zone into the bulk of the propellant (relative to region *3* in Fig. 3), the minimum value of the initial temperature of the heat source required to initiate combustion increases by 200–300 K (from 750 to 1000 K), whereas the ignition delay times increase 5–7 times at the corresponding values of T_p.

Besides Fig. 3 illustrates results of the experimental research [30] of metallized composite propellant ignition by steel hot particle in conditions of various initial temperatures T_p=850–1000 K of local heat source. The difference between the values of the ignition delay time (see curve *2* in Fig. 3) calculated within the framework of the mathematical model (Eqs. (12)–(16) with the initial and boundary conditions) explicitly taking into account of the structural heterogeneity of the propellant (aluminum particles) and the corresponding averaged values of experimental data does not exceed 7 %. It allows concluding that when the heterogeneity of the

substance is taken into account explicitly in the mathematical model of the ignition process of a metallized composite propellant at the local heating, a satisfactory agreement of the results of theoretical and experimental studies is achieved.

Mathematical model with a homogeneous structure of the propellant and with effective values of thermophysical properties can be used at relatively high (T_p>1250 K) initial temperatures of the local heat source. In this case the thermal conductivity can be calculated by any of Eqs. (5)–(11).

The structure of metallized propellant shown schematically in Fig. 1 is one of the possible versions which are a priority for a practical application. The authors [31] pay great attention to modeling the structure of metallized propellant. As alternative versions of the propellant structure (Fig. 1) several other variants of the structure (Fig. 4) are considered under the condition of a constant concentration of the metal powder. Steel particles with sizes x_p=y_p=2 mm heated to temperatures of 850–1500 K is considered as local heat sources.

Table and Fig. 5 show the results of numerical simulation. The maximum difference between the ignition delay times (for limit values) does not exceed 4.5 % for different configurations of the propellant structure. Minimum values of ignition delay time are typical for structures with relatively small (2–3 aluminum particles) agglomerates of metal powder in the near-surface layer of propellant. In this case, micro regions with relatively high thermal conductivity take place in the near-surface layer of metallized composite propellant. The rate of heating of the polymer matrix increases as a result of heat transfer along the direction of x coordinate. The exothermic interaction of butyl rubber and ammonium perchlorate is intensified. As a

result the ignition delay time decreases in comparison with analog characteristic to the propellant structure with a uniform distribution of aluminum particles.

The worst variant of the induction process in conditions of the technology of mixing the components of metallized composite propellant that is not satisfactory for practice is the interaction of the local heat source with the agglomerate of aluminum particles in the propellant structure, if the size of agglomerate is comparable with the sizes of the hot particle. In this case, it is difficult to state the stability of the ignition process without further investigation. Therefore, for practical applications, it is possible to formulate a recommendation on the use of a group (10–15) of hot particles for initiation of combustion process.

CONCLUSIONS

As a result of the theoretical investigation, it is established that at close to the limiting ignition conditions (at T_p<1250 K) it is necessary to take into account the structural heterogeneity of the metallized composite propellant for a reliable prediction of the process characteristics. The results of mathematical modeling differ from the experimental data of the ignition delay time by 75 %, if the heterogeneous structure of composite propellant is interpreted as a homogeneous medium with effective values of thermophysical characteristics. Different models of the heterogeneous structure of metallized composite propellant have a negligible effect on the reliability of the calculated ignition delay time.

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Table. Ignition delay time of metallized composite propellant

T_p, K	t_d^*, s	t_d^{**}, s	t_d^{***}, s	t_d^{****}, s	ε_{max}, %
850	4.862	4.635	4.669	4.986	4.5
900	2.902	2.799	2.870	2.882	3.5
1100	0.571	0.557	0.567	0.562	2.4
1300	0.193	0.189	0.189	0.192	2.3
1500	0.092	0.092	0.089	0.093	0.6

t_d^* – ignition delay time for propellant structure in fig. 1;

t_d^{**} – ignition delay time for propellant structure in fig. 4*a*;

t_d^{***} – ignition delay time for propellant structure in fig. 4*b*;

t_d^{****} – ignition delay time for propellant structure in fig. 4*c*.

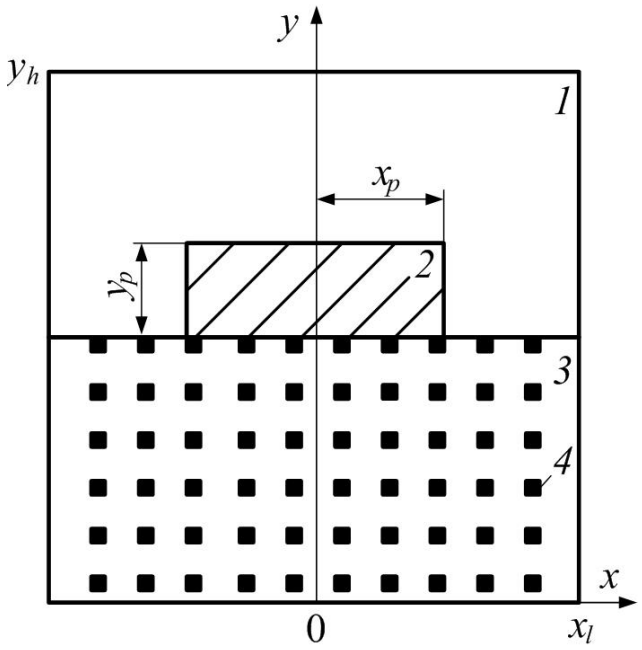

Fig. 1

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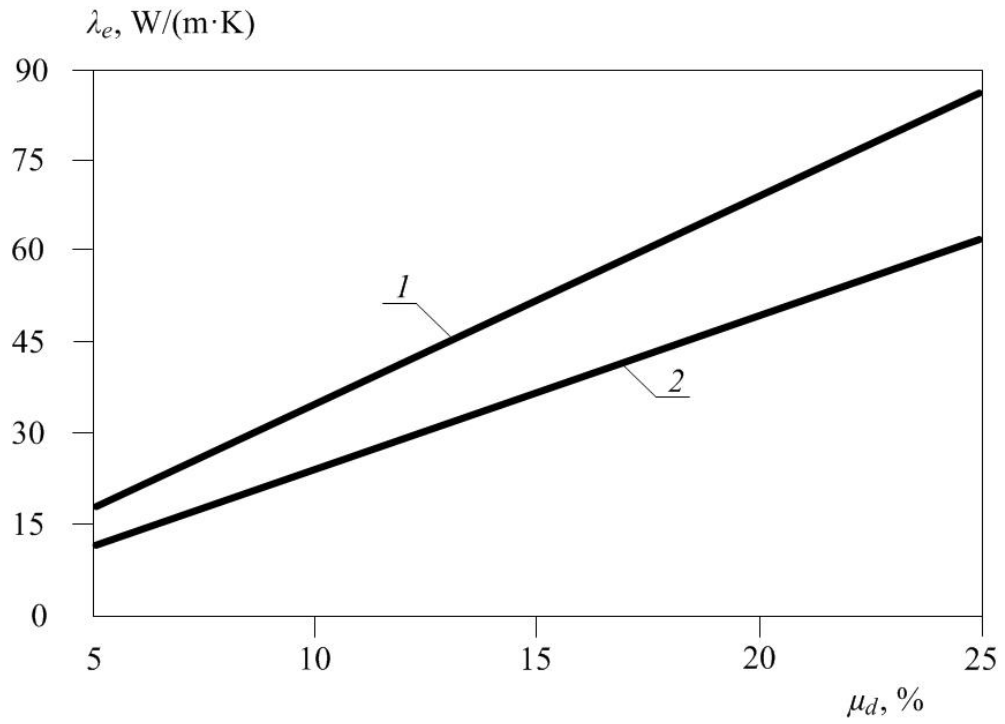

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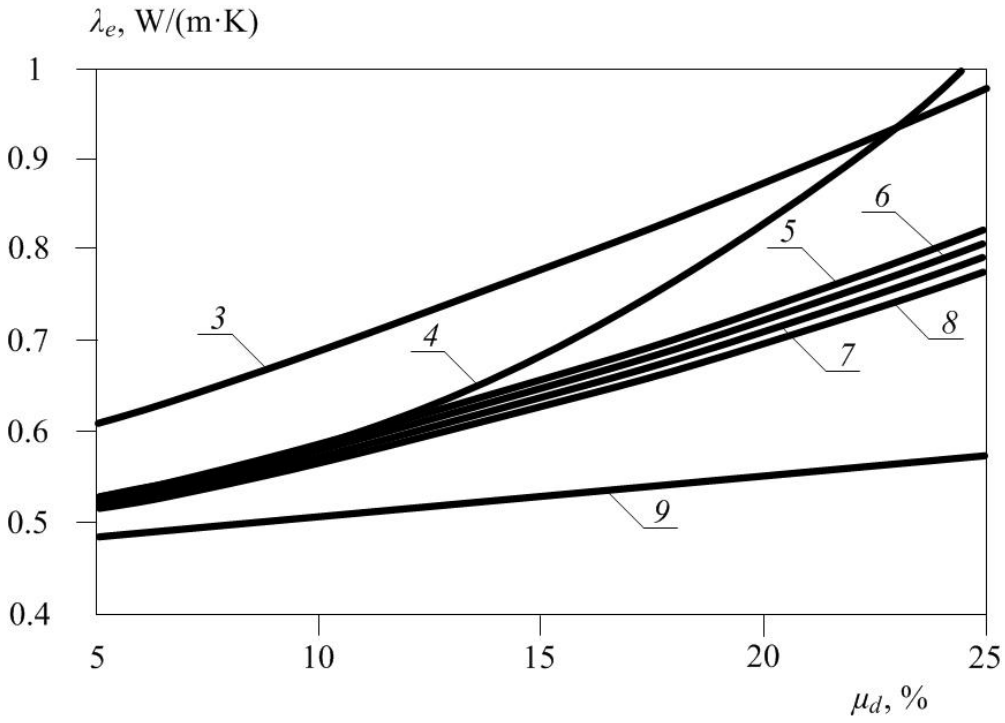

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Fig. 2

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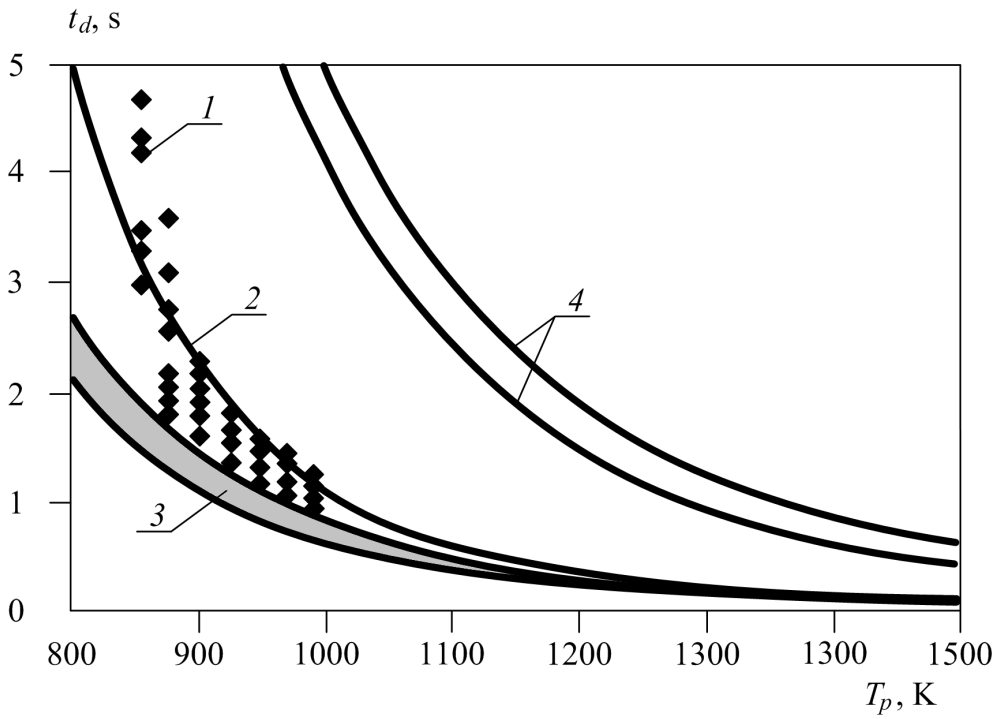

Fig. 3

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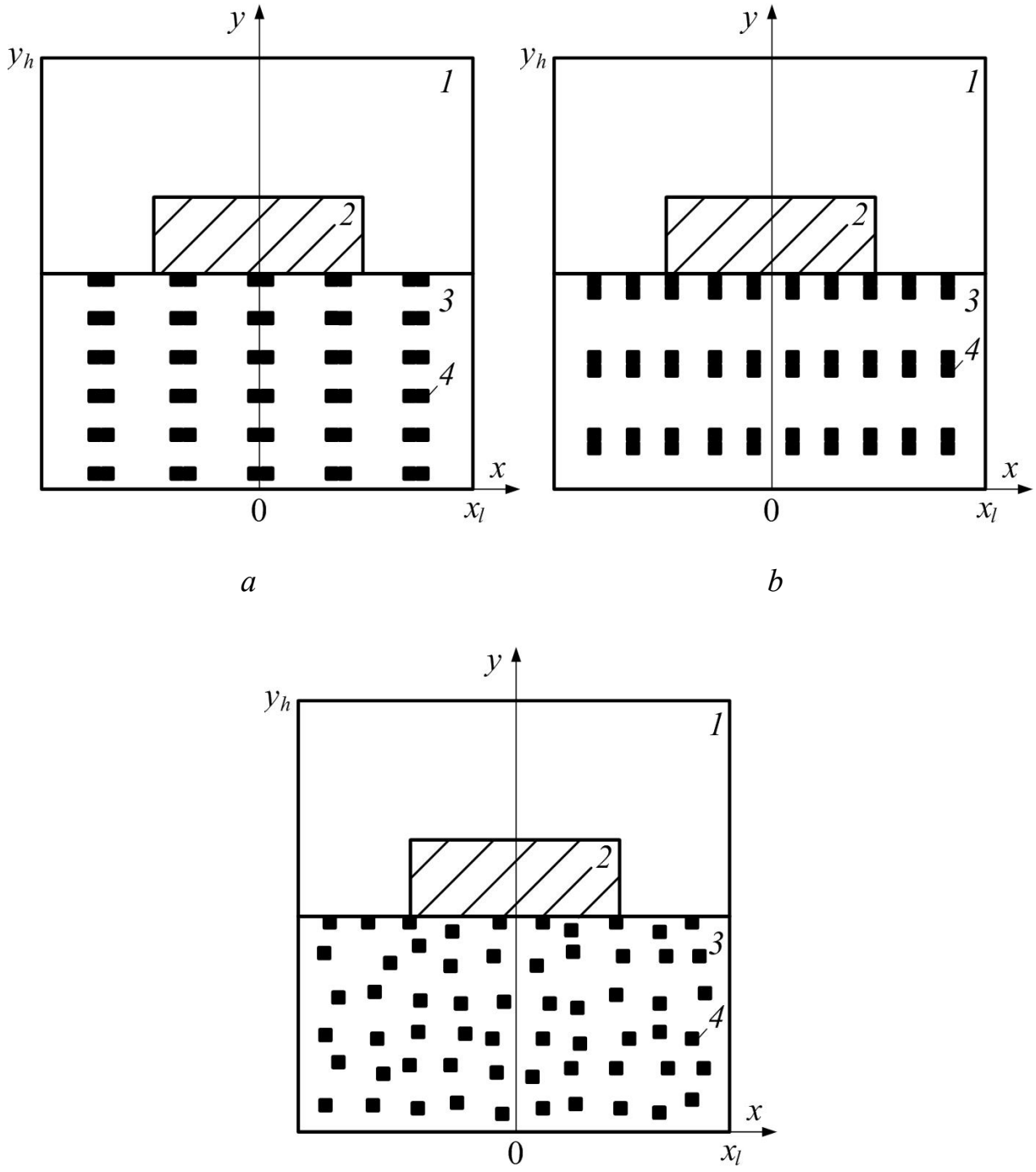

Fig. 4

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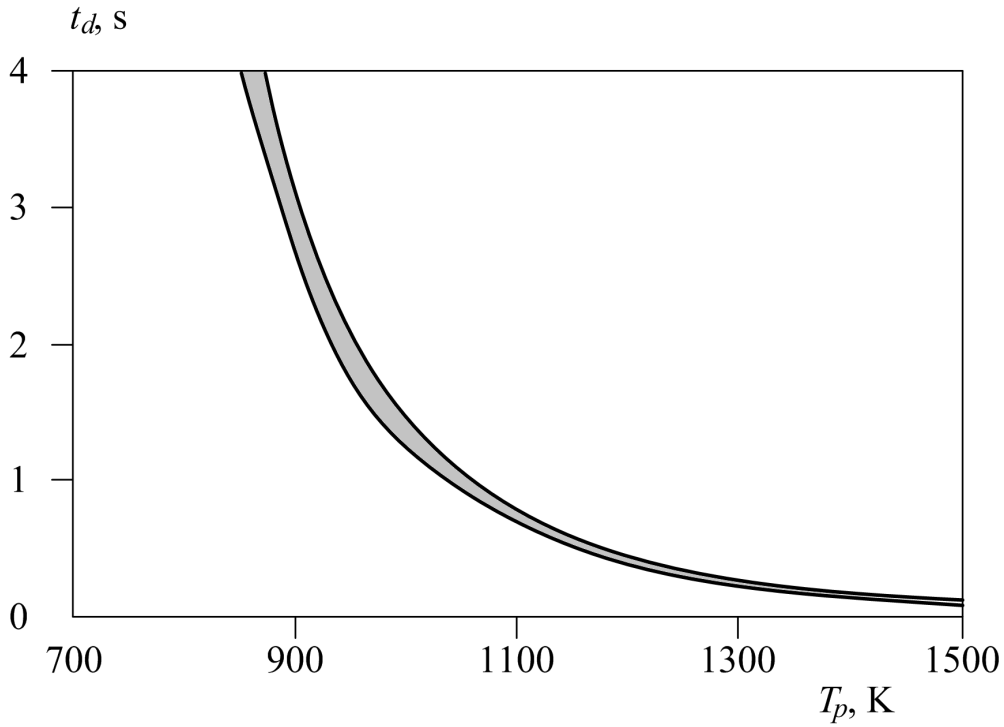

Fig. 5

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CAPTIONS TO ILLUSTRATIONS

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Fig. 1. Schematic of the solution domain of the ignition problem: *1* – inert gas; *2* – local heat source; *3* – matrix of combustible component and oxidizer; *4* – aluminum particle.

Fig. 2. The thermal conductivity of the propellant as a function of the mass fraction of metal particles, calculated by equations (3)–(11): *1* – (3); *2* – (4); *3* – (9); *4* – (7); *5* – (8); *6* – (11); *7* – (6); *8* – (10); *9* – (5).

Fig. 3. Ignition delay time of a metallized composite propellant versus initial temperature of hot particle: *1* – experimental data [30]; *2–4* – results of numerical simulation: *2* – with consideration of the structural heterogeneity of the propellant; *3* (colored area) – the thermal conductivity of the propellant calculated by equations (5)–(11); *4* – the thermal conductivity of the propellant calculated by equations (3) and (4).

Fig. 4. Schematic of the solution domain of the ignition problem: *a* – agglomeration of metal particles in the direction of the axis *x*; *b* – agglomeration of metal particles in the direction of the axis *y*; *c* – random distribution of metal particles in the composite propellant structure.

Fig. 5. Area (colored) of ignition delay time at various distribution of metal particles in composite propellant structure and at variation of initial temperature of local heat source.