

UDC 614.841:536.46

DOI: 10.24025/2306-4412.3.2023.284319

COMBUSTION MODELS FOR BURNING MULTICOMPONENT PYROTECHNIC NITRATE-METALLIZED MIXTURES

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Abstract. The combustion mechanism of compacted mixtures of metallized fuel powders (Mg, Al, etc.), nitrate-containing oxidizers (NaNO_3 , KNO_3 , etc.), organic additives (paraffin, stearin, etc.) and inorganic substances (metal fluorides, metal oxides, etc.) has been established under conditions of external thermal influences, according to which the process of transformation of the initial mixture into combustion products is, on average, stationary and proceeds in three spatially separated zones: condensed phase, where decomposition and high-temperature oxidation of components take place; the interface of phases (burning surface), on which the complete decomposition of components and the ignition of metal particles, which then pass into the flame zone, take place; gas phase (zone of heat release in the flame), in which metal particles burn in a diffusion mode, forming combustion products.

Models of combustion of mixtures that take into account kinetic characteristics of thermal decomposition of the oxidizer, additives of organic and inorganic substances and high-temperature oxidation, ignition and combustion of metal particles in decomposition products, as well as the results of thermodynamic calculations of the temperature of the combustion products of the mixtures and the content of high-temperature condensate (non-oxidized metal) in them have been developed, which allows with a relative error of 8...10% to determine critical ranges of the change in the burning rate of mixtures under conditions of external thermal effects, exceeding which can lead both to the acceleration of the burning process of mixtures and fire-explosive destruction of pyrotechnic products, and to a sharp attenuation of their burning process and failure of products.

Keywords: fire safety, pyrotechnic mixtures, nitrate-containing oxidizers, metal fuels, additives of organic and inorganic substances, thermal influences, combustion processes, combustion models of metallized condensed systems.

Introduction

The purpose of the work: to establish the combustion mechanism of multicomponent compacted mixtures of metal fuel powders and nitrate-containing oxidants with additives of organic and inorganic substances and to develop their combustion models to determine the critical ranges of changes in the burning rate of mixtures, taking into account different values of technological parameters and the influence of external thermal actions.

Research objectives are: analysis of experimental data on the behavior of mixture components under conditions of elevated heating temperatures and external pressures in order to establish the physical and chemical processes occurring in different combustion zones of mixtures; obtaining a system of equations for calculating the dependence of the combustion rate of mixtures on technological parameters (ratio and dispersion of components) and external conditions (elevated heating temperatures and external pressures); determination of critical ranges of changes in technological parameters for a wide range of heating temperatures (up to 800 K) and external pressures (up to 10^7 Pa), exceeding which can lead to a sharp acceleration of the combustion process of mixtures, which leads to the fire-hazardous destruction of products, and to its rapid attenuation and subsequent failure of products under the conditions of their regular use.

Literature review

Currently, compacted mixtures of metal fuel powders (Mg, Al, Ti, Zr etc.), nitrate-containing oxidizers (NaNO_3 , KNO_3 , $\text{Ba}(\text{NO}_3)_2$, $\text{Sr}(\text{NO}_3)_2$ etc.) with organic (paraffin, thiocol, urotropin, stearin, etc.) and inorganic additives (fluorides (NaF , BaF_2 , SiF_2 , AlF_3 etc.), metal oxides (CuO , Cu_2O , NiO , Sb_2O_3 etc.)) substances are widely used in pyrotechnic products of various purposes (firecrackers and stars, lighting and signal cartridges and projectiles, tracers, IR means of thermal protection of various objects, as well as elements of rocket and space technology, etc.) (Akkerman & Penner, 2020; Fang *et al.*, 2019; Marich *et al.*, 2018). The investigated pyrotechnic products in the process of circulation (during storage, transportation and use) may be exposed to extreme external thermal effects (for example, in the event of a fire in warehouses, in the conditions of their transportation, in the event of thermal shock effects, in the conditions of firing and shelling during their launches, in combat zones, etc. (Jia *et al.*, 2021; Mader, 2019; Xu *et al.*, 2018). Systematization and analysis of various cases of fire-explosive activation of products under the specified conditions show that the main reason for this is premature ignition and explosive development of combustion of mixtures in conditions of a sharp increase in heating temperatures and external pressure under their metal shells, which lead to the fire-hazardous destruction of products for surrounding objects (flying sparks, high-temperature combustion products (with a temperature of more than 2000...3000 K), residues unburned mixtures, parts of destructible metal bodies, etc.). Therefore, the prevention of forced fire-hazardous destruction of products in the event of external thermal effects becomes of significant practical importance.

To date, the mechanism and combustion models of two-component pyrotechnic mixtures have been studied in sufficient detail (compacted mixtures of metal fuel powders (Mg, Al, Ti, Zr etc.) and oxygen-containing oxidizers (nitrates of alkaline and alkaline earth metals (NaNO₃, KNO₃, Ba(NO₃)₂, Sr(NO₃)₂ etc.), metal oxides (CuO, Cu₂O NiO, Sb₂O₃ etc.)) (Bai & Yang, 2019; Fateev *et al.*, 2017; Niu & Wang, 2019).

As for three-component mixtures of metallic fuel + nitrate-containing oxidizer + inorganic substance additive and, even more so, four-component mixtures of the above components, currently the above-mentioned studies are not available for them.

Conducted studies of the behavior of the components of mixtures of powders Mg, Al etc., nitrate-containing oxidizers (NaNO₃, Ba(NO₃)₂ etc.), organic (paraffin, stearin) and inorganic additives (NaF, BaF₂ etc.) substances in the conditions of elevated heating temperatures inherent in the process of burning pyrotechnic mixtures based on metal fuels and oxygen-containing oxidizers (Kovalyshyn *et al.*, 2018; Dibrova *et al.*, Improving, 2020), made it possible to establish that the combustion process of the charges of the investigated mixtures is stationary on average and proceeds in the same way as in the case of mixed powders (Kovalyshyn *et al.*, 2018), in the following spatially separated zones: condensed phase; on the burning surface; gas phase (flame zone).

Materials and methods

We will analyze the physical and chemical processes occurring in the specified zones using the following mixtures widely used in pyrotechnic products (Dibrova *et al.*, Fire safety, 2020): Mg + NaNO₃ + paraffin + NaF and Mg + NaNO₃ + stearin + BaF₂.

Condensed phase. The considered inorganic substances begin to melt only at temperatures of 1268...1553 K, and decompose – at temperatures higher than 1700...1800 K. Therefore, these additives within the c-phase can be considered inert, and their contribution to the combustion process of the mixture is carried out already in the flame zone. The change in temperature in the so-called warm-up area of the c-phase of the mixture, which is characterized by a change in temperature from T_0 (the initial temperature of the mixture) to T_m (melting point NaNO₃), is described by the heat conduction equation (Fateev *et al.*, 2017):

$$\lambda_c \frac{\partial^2 T}{\partial \xi^2} - \rho_c u c_c \frac{\partial T}{\partial \xi} = 0, \quad (1)$$

wherein ξ – the direction that coincides with the direction of the coordinate x , opposite to which the combustion front propagates through the mixture sample with speed u , m/s. At the same time, the first term in equation (1) corresponds to the conductive heat flow (λ_c – thermal conductivity coefficient of the mixture, Wt/m·K), and the second (ρ_c and c_c – density (kg/m³) and specific heat capacity (J/kg·K) mixture) – convective heat flow. Integration of this equation leads to the determination of the thickness of the heating layer of the c-phase of the mixture δ_1 :

$$\delta_1 = \frac{a_c^2}{u}, \quad (2)$$

wherein $a_c^2 = \frac{\lambda_c}{\rho_c c_c}$ – coefficient of thermal conductivity of the mixture, m²/s.

It was established that for the considered mixtures the value of the parameter δ_1 (at $\alpha = 0,5...3,0$) vary from 100 to 200 μm to 3000 to 4000 μm . It is also shown that the decomposition NaNO₃, paraffin and stearin begins immediately after their melting and noticeably intensifies as the temperature rises. At higher temperatures ($T = 1100...1200$ K), corresponding to the temperature values of the combustion surface T_n , complete decomposition of the oxidizing agent is already taking place (to O₂, N₂, etc.) and the indicated additives of organic substances (to CO, CO₂, H₂, etc.) according to the following kinetic equations (Kirichenko *et al.*, 2020):

$$\frac{d\varphi_1}{dt} = k_1 e^{-E_1/RT}, \quad (3)$$

$$\frac{d\varphi_2}{dt} = k_2 e^{-E_2/RT}, \quad (4)$$

wherein φ_1, φ_2 – relative degrees of decomposition of the oxidant and organic matter; k_1, k_2, E_1, E_2 – kinetic constants ($E_1, E_2 \gg RT$). The thickness of the reaction layer is much smaller than the thickness of the heated c-phase layer, and in order of magnitude it has the form

$$\delta_2 \cong \frac{2RT_n a_c^2}{uE_1}. \quad (5)$$

In accordance with formula (5) and taking into account the above values δ_1 , we get that the value of the parameter δ_2 (at $\alpha = 0,5 \dots 3,0$) vary from 10 to 15 μm to 350 to 450 μm , i.e. $\delta_2 < \delta_1$ about an order of magnitude. Thus, for the used metal fuel powders, the thickness of the heating layer of the c-phase is much larger than the average size of their particles (d_m), and the thickness of the reaction layer of the c-phase is already commensurate with the size of the particles, i.e. the following conditions are met

$$\delta_2 \ll \delta_1, \delta_1 \gg d_m, \delta_2 \cong d_m. \quad (6)$$

According to experimental data (Kyrychenko *et al.*, 2018), the process of oxidation of metal particles begins in the gasified NaNO_3 melt, together with additives of organic substances, at temperatures $T > 673$ K. The speed of this process is described by the following equation (Kyrychenko, 2022):

$$\frac{d\varphi_3}{dt} = k_3 C_{O_2}^m \bar{\rho}_g^n e^{-E_3/RT} \text{ at } E_3 \gg RT, \quad (7)$$

wherein φ_3 – degree of metal oxidation; k_3 – pre-exponential multiplier (c^{-1}); E_3 – activation energy (kJ/mol); $C_{O_2}, \bar{\rho}_g$ – relative mass content of O_2 in gaseous products, relative density of the gas medium (relative to normal conditions), respectively; m, n – empirical constants. At the same time with the condition $E_3 \gg RT$ it follows that the intense pre-flame oxidation of metal fuel particles in the gaseous products of decomposition of the oxidant and additives of organic substances also occurs in a narrow reaction layer of the c-phase, where complete gasification of the oxidant and additives of organic substances occurs, i.e., the degree of their decomposition $\varphi_1 = \varphi_{1n} \cong 1, \varphi_2 = \varphi_{2n} \cong 1$.

In experimental studies (Kyrychenko *et al.*, Investigation, 2019), it was also established that during the existence of the reaction layer of the c-phase (within $10^{-1} \dots 10^{-3}$ s) according to the kinetic equation (9), the degree of oxidation of the metallic fuel in the c-phase is $\varphi_3 = \varphi_{3n} = 0,2 \dots 0,3$.

Burning surface. Microcine imaging of the combustion surface of mixtures shows that the ignition of metallic fuel particles occurs immediately after they reach the combustion surface (Kyrychenko *et al.*, Study, 2019). At the same time, the interface between the condensed and gas phases is characterized by significant heterogeneity depending on the ratio of components in the mixture: at $0,5 < \alpha < 3,0$ – the burning process in the first approximation can be considered stable and quasi-stationary (the average burning rate throughout the process is constant); at $\alpha < 0,5$ the combustion process becomes unstable and non-stationary; at $\alpha > 3,0$ there are significant pulsations and emissions of significant masses of oxidizer that did not decompose during the burning process, which quickly subsides. At the same time, the combustion surface of the mixture, as in the case of mixed powders (Kyrychenko, Investigation, 2022), can be characterized by an average temperature T_n . Conducted thermocouple measurements T_n made it possible to establish the nature of its dependence on various factors (Kyrychenko *et al.*, Patterns, 2021). When increasing α from 0,5 to 2,0 T_n monotonically decreases from 1200 K to 1080 K ($T_0 = 293$ K; $P = 10^5$ Pa). At the same time, for a stoichiometric mixture in the region of pressure change from 10^5 Pa to 10^7 Pa, T_n increases monotonically from 1100 K to 1350 K ($T_0 = 293$ K).

Gas phase (flame zone). Metal fuel particles dispersed from the combustion surface in the g-phase burn in gaseous products of thermal decomposition of the oxidizer and

additives of organic and inorganic substances (Motrichuk *et al.*, 2020) (in the O₂ flow – the initial stage of the decomposition of the oxidant; in the O₂ + N₂ flow with the addition of CO + CO₂ and water vapor – the decomposition of the oxidant at the temperatures of the combustion surface, taking into account the decomposition of the additives).

From the data presented in (Kyrychenko *et al.*, 2022), it follows that dispersing metal particles burn in the flow of gaseous decomposition products by the diffusion mechanism with the formation of almost spherical glowing zones, where exothermic reactions of the interaction of the diffusing metal vapors with the oxidant of the surrounding gaseous environment take place. At the same time, for large metal particles ($d_m > 200...300 \mu\text{m}$) the combustion process becomes asymmetric and its quasi-stationarity is violated, and for small particles ($d_m < 50...70 \mu\text{m}$) their disintegration (fragmentation) into smaller burning particles is observed in some cases.

The conductive heat flux can be estimated, as in the case of mixed powders (Kyrychenko *et al.*, Study, 2021), using the following approximate formula:

$$q_c \cong \bar{\lambda}_g \frac{T_{max} - T_n}{\delta_3}, \quad (8)$$

wherein T_{max} – the maximum value of the gas temperature in the zone of heat release of the flame, K; δ_3 – thickness of the heat release zone, m; $\bar{\lambda}_g$ – coefficient of thermal conductivity of gas at temperature $\bar{T} = (T_{max} + T_n)/2$. For stoichiometric mixtures of $d_m < 150...200 \mu\text{m}$ according to experimental data ($T_{max} = 2500 \text{ K}$; $T_n = 1100 \text{ K}$; $\delta_3 = 0,5 \cdot 10^{-2} \text{ m}$; $\bar{\lambda}_g = 0,54 \text{ J/m}\cdot\text{K}$ (Dibrova *et al.*, Regularities, 2020), we have $q_c = 3,8 \cdot 10^6 \text{ J/m}^2$.

We will estimate the radiation heat flow under the assumption that the particles of emitting metal oxides have the same temperature T_{max} ; radiation occurs according to the law of an absolutely black body; the degree of blackness of the flame is equal to one; the influence of the dustiness of the outflowing gas flow and the roughness of the combustion surface, as well as the mutual influence of the emitting particles are absent. With the specified assumptions, the radiation heat flux can be estimated using the well-known formula (Fateev *et al.*, 2017):

$$q_R = \sigma(T_{max}^4 - T_n^4), \quad (9)$$

wherein σ – Stefan-Boltzmann constant.

For stoichiometric mixtures and the obtained experimental data (Kyrychenko, 2021), in accordance with formula (9), we obtain that $q_R \ll q_c$ (more than 4...5 times). In fact, the radiation heat flow from the flame zone during the combustion of mixtures is much smaller, since formula (9) gives an overestimated value q_c (assessment from above).

Results and discussion

The mechanism and mathematical model of the combustion process of mixtures of metallic fuel + nitrate-containing oxidizer + addition of organic and inorganic substances under conditions of external thermal influences (elevated heating temperatures and external pressures). The analysis of experimental data on the physic-chemical processes occurring during the combustion of the considered mixtures was carried out (mixtures with $0,5 < \alpha < 3,0$) allows establishing the mechanism of their combustion according to which the process of transformation of the initial mixture into combustion products is stationary, one-dimensional and proceeds in the following three most characteristic zones in the first approximation: condensed phase (c-phase) of the initial mixture, consisting of a heated layer, where chemical transformations can be neglected, and a reaction layer, in which the solid mixture turns into a gas containing metal particles; the combustion surface of the mixture, on which complete decomposition of the oxidizer and organic substance additives occurs, as well as the ignition of metal particles; gas phase (g-phase – zone of heat release in the flame), in which dispersed metal particles burn in the diffusion mode in the flow of decomposition

products of the oxidant and additives of organic substances, forming combustion products with temperature T_c .

With the above-mentioned basic assumptions about the combustion mechanism of the considered mixtures, as well as the assumption of the absence of heat losses to the environment and external forces, the radiation heat flow from the flame zone of the propagation front, the combustion reaction in the c-phase of the initial mixture can be described by the following system of equations known in the theory of the combustion of condensed metallized mixtures (Kirichenko *et al.*, 2020)

$$\frac{d}{dx} \left(\lambda_c \frac{dT}{dx} \right) - \rho_c c_c \frac{dT}{dx} - \frac{H_o \rho_c \alpha}{1+l_c \alpha} u \frac{d\varphi_1}{dx} - H_n \rho_c u \varepsilon_n \frac{d\varphi_2}{dx} + \frac{H_m \rho_c \alpha}{1+l_c \alpha} u \frac{d\varphi_3}{dx} = 0; \quad (10)$$

$$u \frac{d\varphi_1}{dx} = k_1 e^{-E_1/RT}; \quad (11)$$

$$u \frac{d\varphi_2}{dx} = k_2 e^{-E_2/RT}; \quad (12)$$

$$u \frac{d\varphi_3}{dx} = \frac{k_3 C_{O_2}^m}{\bar{\rho}_c^n} e^{-E_3/RT} \quad (13)$$

with boundary conditions:

$$T = T_0, \varphi_1 = \varphi_2 = \varphi_3 = 0 \text{ при } x \rightarrow -\infty; \quad (14)$$

$$T = T_n, \varphi_1 = \varphi_{1n}, \varphi_2 = \varphi_{2n}, \varphi_3 = \varphi_{3n},$$

$$\lambda_c \left(\frac{dT}{dx} \right) = q_c \text{ при } x = 0. \quad (15)$$

Here ρ_c , c_c , λ_c – density (kg/m^3), specific heat capacity ($\text{J/kg}\cdot\text{K}$) and coefficient of thermal conductivity ($\text{Wt/m}\cdot\text{K}$) of the mixture; H_o , H_n , H_m – thermal effects of decomposition reactions of oxidant and organic matter, oxidation of metal fuel particles (J/kg); φ_i , k_i and E_i ($i = \overline{1,3}$) – relative degrees of decomposition of oxidant and organic matter, oxidation of metal fuel (by mass) and their kinetic constants (s^{-1} , kJ/mol), respectively; l_c – stoichiometric coefficient of the mixture.

To obtain expressions for determining the burning rate, it is convenient to reduce the system of equations (10) – (13) to one equation:

$$\left(\rho_c c_c u (T_n - T_0) - q_c \right) q_c = \frac{\rho_c k_3 \lambda_c H_m C_{O_2}^m R T_n^2}{E_3 \bar{\rho}_c^n (1+l_c \alpha)} e^{-E_3/RT_n} \left[1 - e^{-\frac{E_3(T_n-T_0)}{R T_n^2}} \right] -$$

$$- \frac{\rho_c k_2 \lambda_c H_o l_c \alpha R T_n^2}{E_2 (1+l_c \alpha)} e^{-E_2/RT_n} \left[1 - e^{-\frac{E_2(T_n-T_0)}{R T_n^2}} \right] - \frac{\rho_c k_1 \lambda_c H_n \varepsilon_n R T_n^2}{E_1} e^{-E_1/RT_n} \left[1 - e^{-\frac{E_1(T_n-T_0)}{R T_n^2}} \right]. \quad (16)$$

Definition T_n . Experimental determination of T_n by existing methods is difficult and sufficiently approximate (Kyrychenko *et al.*, Patterns, 2021), and studies devoted to the analytical determination of T_n are currently limited. Therefore, using systematic data on the kinetics of thermal decomposition of the oxidant and additives of organic substances (Kyrychenko *et al.*, Study, 2021) at temperatures characteristic of the reaction zone of the c-phase of the mixture, the value of T_n can be determined based on the assumption that at $T = T_n$ complete decomposition of the oxidant and organic substances occurs, and the degree of oxidation of metallic fuels φ_3 during the existence of the reaction layer of the c-phase during the combustion of the considered mixtures (Kyrychenko *et al.*, 2022) lies within 0,2...0,3. At $\varphi_{1n} = \varphi_{2n} \cong 1$ and $\varphi_{3n} = \varphi^*$ (example, $\varphi^* \cong 0,25$) to determine T_n , we get the following equation:

$$\frac{u^2}{\lambda_c R T_n^2} \left[\rho_c c_c (T_n - T_0) \left[\varphi_3^* - \frac{H_o l_c \alpha + H_n \varepsilon_n (1+l_c \alpha)}{H_m} \right] - \frac{H_m \rho_c}{2(1+l_c \alpha)} \left[\varphi_3^* - \frac{H_o l_c \alpha + H_n \varepsilon_n (1+l_c \alpha)}{H_m} \right]^2 \right] =$$

$$= \frac{k_3 C_{O_2}^m}{E_3 \bar{\rho}_c^n} e^{-E_3/RT_n} \left[1 - e^{-\frac{E_3(T_n-T_0)}{R T_n^2}} \right] - \frac{k_2 H_o l_c \alpha}{E_2 H_m} e^{-E_2/RT_n} \left[1 - e^{-\frac{E_2(T_n-T_0)}{R T_n^2}} \right] -$$

$$- \frac{k_1 H_n \varepsilon_n R T_n^2 (1+l_c \alpha)}{E_1 H_m} e^{-E_1/RT_n} \left[1 - e^{-\frac{E_1(T_n-T_0)}{R T_n^2}} \right]. \quad (17)$$

Definition q_c . For an approximate analytical definition q_c we will use the well-known “heat flow method” (Dibrova *et al.*, Fire safety, 2020), namely: replace the true value of the heat flow from the heat release zone of the g-phase with the average value for this zone:

$$q_c = \lambda_g \left. \frac{dT}{dx} \right|_{x=0} = \bar{\lambda}_g \frac{T_c - T_n}{\Delta x}, \quad (18)$$

wherein $\bar{\lambda}_g$ – coefficient of thermal conductivity of gas at temperature $\bar{T} = (T_c + T_n)/2$; Δx – the distance from the combustion surface to the plane on which the temperature is reached T_c (temperature T_c and the corresponding composition of combustion products are the result of thermodynamic calculations (Motrichuk *et al.*, 2020)). At the same time, the value Δx is found from the equation of motion of a spherical metal particle with a radius r :

$$\frac{d^2x}{dt^2} = 0,15 \frac{\bar{\rho}_g V_n^2}{\rho_m r} + 7,52 \frac{\bar{\rho}_g \bar{\nu}_g V_n}{\rho_m r^2}. \quad (19)$$

wherein $\bar{\rho}_g$ – gas flow density at temperature \bar{T} ; Experimental data on the outflow rate of gaseous decomposition products show that the rate of their relative movement within the heat release zone of the g-phase is $V_n = 1 \dots 10$ м/с. $\bar{\nu}_g$ – coefficient of kinematic viscosity of gas at temperature \bar{T} .

Having integrated equation (19) taking into account the initial conditions on the combustion surface (at $t = 0$; $x = 0$, $\dot{x} = u$), we will find the dependence of the displacement of the particle on time:

$$\Delta x = u\tau_c + 0,15 \frac{\bar{\rho}_g V_n^2}{\rho_m} \int_0^{\tau_c} \left[\int_0^t \frac{dt}{r(t)} \right] dt + 7,52 \frac{\bar{\rho}_g \bar{\nu}_g V_n}{\rho_m} \int_0^{\tau_c} \left[\int_0^t \frac{dt}{r^2(t)} \right] dt, \quad (20)$$

wherein τ_c – the burning time of a metal particle when its radius changes from $r_0 = \frac{d_m}{2}$ to $r_k = \frac{d_k}{2}$ (r_k (d_k) – the radius (diameter) of the metal particle at the end of the flame heat generation zone).

Using the well-known equation (Dibrova *et al.*, Regularities, 2020) we find τ_c :

$$\tau_c = \frac{3}{4ab^2} \left[2 \left(\frac{d_m}{2} \right)^{\frac{2}{3}} \left[1 - (1 + l_c \alpha)^{\frac{2}{9}} (g_{me}^c)^{\frac{2}{9}} \right] + b \left(\frac{d_m}{2} \right)^{\frac{4}{3}} \left[1 - (1 + l_c \alpha)^{\frac{4}{9}} (g_{me}^c)^{\frac{4}{9}} \right] - \frac{2}{b} \ln \left[\frac{1 - b \left(\frac{d_m}{2} \right)^{\frac{2}{3}}}{1 - b \left(\frac{d_m}{2} \right)^{\frac{2}{3}} (1 + l_c \alpha)^{\frac{2}{9}} (g_{me}^c)^{\frac{2}{9}}} \right] \right]. \quad (21)$$

Calculations based on formulas (18), (20) and (21) with the involvement of all necessary data on mixtures and their components (Kovalishyn *et al.*, 2018) show that the heat flow q_c depends most strongly on the coefficient of excess oxidizer and dispersity of the metallic fuel: when increasing α from 0,5 to 3,0 values q_c decrease from $5,83 \cdot 10^6$ Вт/м² to $1,34 \cdot 10^6$ Вт/м² (for a mixture of Mg + NaNO₃ + paraffin + NaF) and from $4,12 \cdot 10^6$ Вт/м² to $0,75 \cdot 10^6$ Вт/м² (for a mixture of Mg + NaNO₃ + stearin + BaF₂); when increasing d_m from 74,5 μm to 182 μm values q_c decreases from $4,1 \cdot 10^6$ Вт/м² to $2,3 \cdot 10^6$ Вт/м² (for a mixture of Mg + NaNO₃ + paraffin + NaF) and from $2,9 \cdot 10^6$ Вт/м² to $1,7 \cdot 10^6$ Вт/м² (for a mixture of Mg + NaNO₃ + stearin + BaF₂).

The above equations (16) – (18), (20), (21) allow you to calculate the burning rate of the mixtures under consideration u , the temperature of their burning surface T_n , as well as the burning rate of dispersed metal fuel particles in the decomposition products of these mixtures, their burning times in the zone of heat release of the flame τ_c , degree of their burnout in this zone $\frac{d_k}{d_m}$, the length of this zone Δx and heat flow from the flame heat release zone q_c . Using the necessary values of the physic-chemical parameters of the mixtures and their components (Kovalyshyn *et al.*, 2018) on a PC with the use of standard software (Kyrychenko *et al.*, Study, 2021), calculations were made of the dependences of the burning rate on the technological parameters (the coefficient of excess oxidant and the relative content of additives of organic and inorganic substances), as well as the main parameters of external thermodynamics (heating

temperature, external pressure) according to the equations indicated above (Fig. 1 – 4). From the results of comparing the calculated dependencies with the obtained experimental data, it follows that there is a complete qualitative match between them, which corresponds to the combustion mechanism of the mixtures proposed above. As for the quantitative differences between the calculation and the experiment, they do not exceed 8...12%.

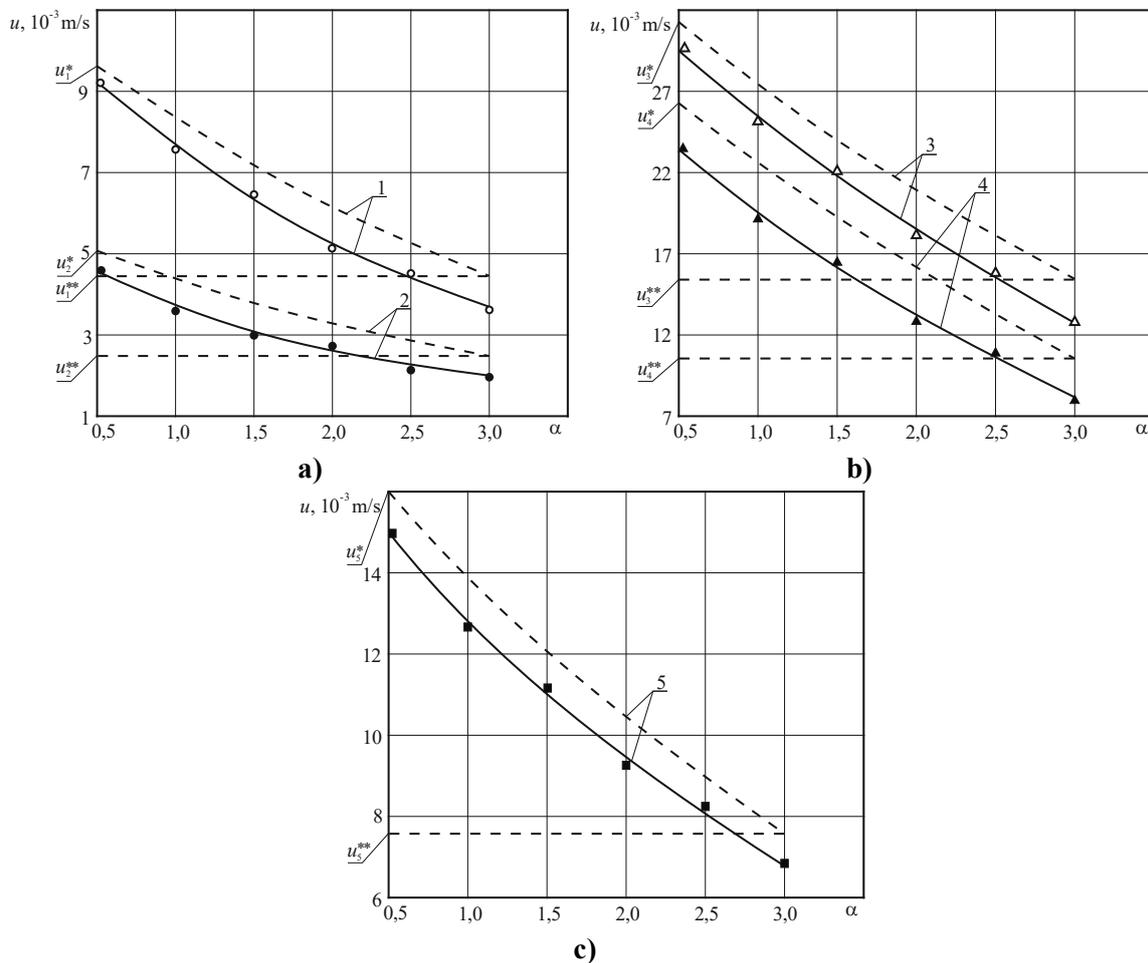


Figure 1. Dependences of the burning rate of mixtures of Mg + NaNO₃ + paraffin + NaF on the coefficient of excess oxidant for different values of the amounts of additives under normal external conditions ($d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$; $T_0 = 293 \text{ K}$; $P = 10^5 \text{ Pa}$):

- a) – $\varepsilon_f = 0$; 1 – $\varepsilon_n = 0,05$; 2 – $\varepsilon_n = 0,2$; b) – $\varepsilon_n = 0$; 3 – $\varepsilon_f = 0,1$; 4 – $\varepsilon_n = 0,05$;
 c) – $\varepsilon_f \neq 0$ and $\varepsilon_n \neq 0$; 5 – $\varepsilon_n = 0,2$; $\varepsilon_f = 0,1$; – – – calculation curve; u_i^* and u_i^{**} ($i = \overline{1,5}$) – critical values of the burning rate of mixtures; \circ , \bullet , Δ , \blacktriangle , \blacksquare – experimental data

In addition, as a result of the calculations, the following critical ranges of changes in the burning rate of mixtures and technological parameters were established (as an example, finely dispersed Mg powders with $d_m = 74,5 \mu\text{m}$ were considered as the most reactive in active gaseous decomposition products (Marich *et al.*, 2018); at the same time, as the results of the calculations showed, in the case of coarsely dispersed Mg powders (with $d_m = 182 \dots 306 \mu\text{m}$), the considered mixtures become less sensitive to external thermal effects), the excess of which leads to the unstable explosive development of their combustion process under the conditions of external thermodynamics (Tables 1, 2):

$$\varepsilon_{ni}^{**} \leq \varepsilon_n \leq \varepsilon_{ni}^*, \quad \varepsilon_{fi}^{**} \leq \varepsilon_f \leq \varepsilon_{fi}^*, \quad u_i^{**} \leq u_i \leq u_i^*, \quad (22)$$

$$i = 1, 2, 3, \dots$$

Table 1. Ranges of change of critical parameters u_i^* and u_i^{**} (10^{-3} m/s), ε_{ni}^* , ε_{ni}^{**} , ε_{fi}^* and ε_{fi}^{**} for mixtures Mg + NaNO₃ + paraffin + NaF and Mg + NaNO₃ + stearin + BaF₂ under normal external conditions ($\alpha = 0,5 \dots 3,0$; $d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$; $T_0 = 293 \text{ K}$; $P = 10^5 \text{ Pa}$), $i = \overline{1,10}$

Mixture Mg + NaNO ₃ + paraffin + NaF	Mixture Mg + NaNO ₃ + stearin + BaF ₂
$u_1^* > 9,6$ and $u_1^{**} < 4,7$ at $\varepsilon_{f1}^* = 0$; $\varepsilon_{n1}^* < 0,05$ and $\varepsilon_{n1}^{**} > 0,05$	$u_6^* > 3,7$ and $u_6^{**} < 1,6$ at $\varepsilon_{f6}^* = 0$; $\varepsilon_{n6}^* < 0,05$ and $\varepsilon_{n6}^{**} > 0,05$
$u_2^* > 5,1$ and $u_2^{**} < 2,5$ at $\varepsilon_{f2}^* = 0$; $\varepsilon_{n2}^* < 0,2$ and $\varepsilon_{n2}^{**} > 0,2$	$u_7^* > 2,1$ and $u_7^{**} < 1,2$ at $\varepsilon_{f7}^* = 0$; $\varepsilon_{n7}^* < 0,05$ and $\varepsilon_{n7}^{**} > 0,05$
$u_3^* > 31,4$ and $u_3^{**} < 16,3$ at $\varepsilon_{n3}^* = 0$; $\varepsilon_{f3}^* > 0,1$ and $\varepsilon_{f3}^{**} < 0,1$	$u_8^* > 25,4$ and $u_8^{**} < 11,9$ at $\varepsilon_{f8}^* = 0$; $\varepsilon_{n8}^* < 0,05$ and $\varepsilon_{n8}^{**} > 0,05$
$u_4^* > 26,1$ and $u_4^{**} < 11,5$ at $\varepsilon_{n4}^* = 0$; $\varepsilon_{f4}^* > 0,05$ and $\varepsilon_{f4}^{**} < 0,05$	$u_9^* > 21,1$ and $u_9^{**} < 8,8$ at $\varepsilon_{f9}^* = 0$; $\varepsilon_{n9}^* < 0,05$ and $\varepsilon_{n9}^{**} > 0,05$
$u_5^* > 16,1$ at $\varepsilon_{n5}^* < 0,2$ and $\varepsilon_{f5}^* > 0,1$; $u_5^{**} < 7,7$ at $\varepsilon_{n5}^{**} > 0,2$ and $\varepsilon_{f5}^{**} < 0,1$	$u_{10}^* > 15,3$ at $\varepsilon_{n10}^* < 0,2$ and $\varepsilon_{f10}^* > 0,1$; $u_{10}^{**} < 5,9$ at $\varepsilon_{n10}^{**} > 0,2$ and $\varepsilon_{f10}^{**} < 0,1$

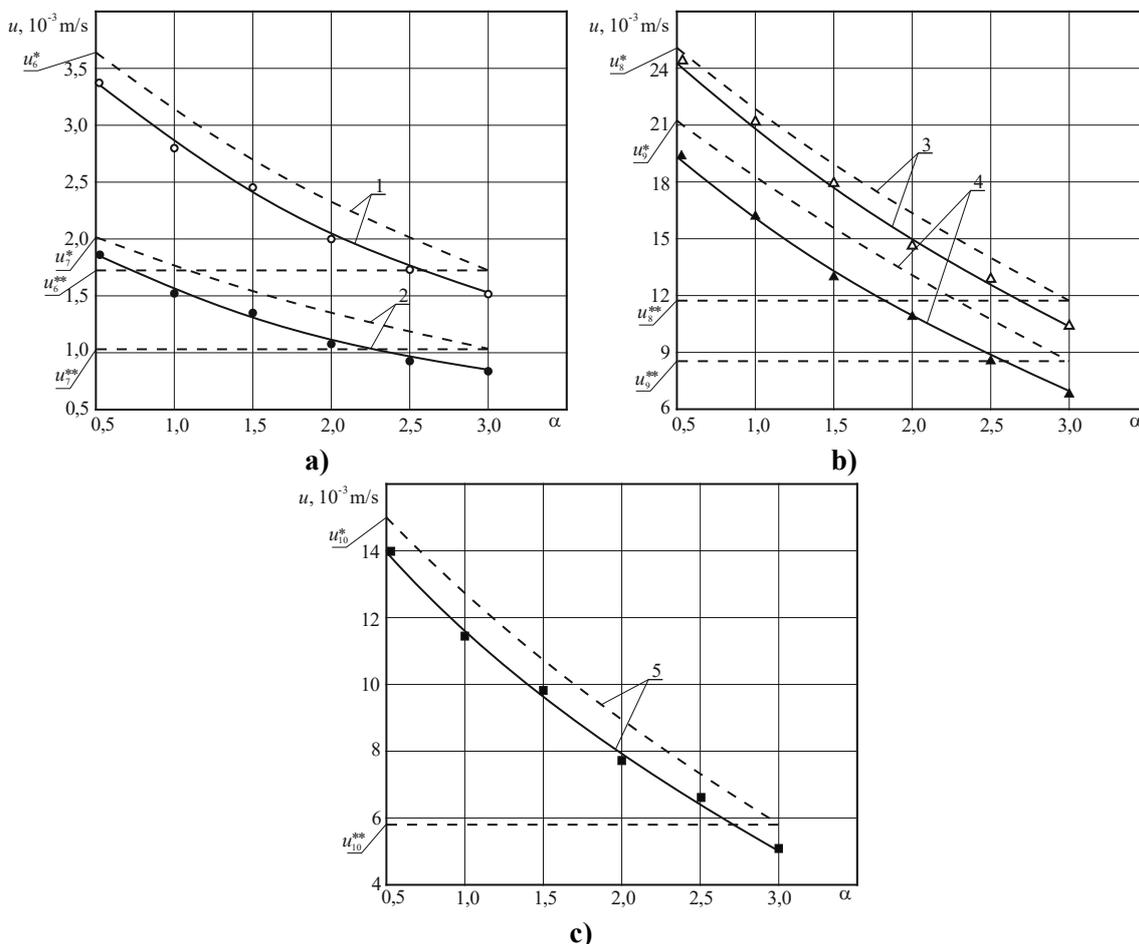


Figure 2. Dependencies of the burning rate of mixtures of Mg + NaNO₃ + stearin + BaF₂ on the coefficient of excess oxidant for different values of the amounts of additives under normal external conditions ($d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$; $T_0 = 293 \text{ K}$; $P = 10^5 \text{ Pa}$):

- a) – $\varepsilon_f = 0$; 1 – $\varepsilon_n = 0,05$; 2 – $\varepsilon_n = 0,2$; b) – $\varepsilon_n = 0$; 3 – $\varepsilon_f = 0,1$; 4 – $\varepsilon_n = 0,05$;
- c) – $\varepsilon_f \neq 0$ and $\varepsilon_n \neq 0$; 5 – $\varepsilon_n = 0,2$; $\varepsilon_f = 0,1$; – – – – calculation curve; u_i^* and u_i^{**} ($i = \overline{6,10}$) – critical values of the burning rate of mixtures; $\circ, \bullet, \Delta, \blacktriangle, \blacksquare$ – experimental data

Table 2. Ranges of change of critical parameters u_i^* and u_i^{**} (10^{-3} m/s), ε_{ni}^* , ε_{ni}^{**} , ε_{fi}^* and ε_{fi}^{**} for Mg + NaNO₃ + paraffin + NaF and Mg + NaNO₃ + stearin + BaF₂ mixtures under conditions of external thermal influences ($\alpha = 0,5 \dots 3,0$; $d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$), $i = \overline{11,40}$

Mixture of Mg + NaNO ₃ + paraffin + NaF	Mixture of Mg + NaNO ₃ + stearin + BaF ₂
At $T_0 = 800 \text{ K}$, $P = 10^5 \text{ Pa}$	
$u_{11}^* > 22,3$ and $u_{11}^{**} < 8,4$ at $\varepsilon_{f11}^* = 0$; $\varepsilon_{n11}^* < 0,05$ and $\varepsilon_{n11}^{**} > 0,05$	$u_{16}^* > 8,4$ and $u_{16}^{**} < 3,7$ at $\varepsilon_{f16}^* = 0$; $\varepsilon_{n16}^* < 0,05$ and $\varepsilon_{n16}^{**} > 0,05$
$u_{12}^* > 12,1$ and $u_{12}^{**} < 4,5$ at $\varepsilon_{f12}^* = 0$; $\varepsilon_{n12}^* < 0,2$ and $\varepsilon_{n12}^{**} > 0,2$	$u_{17}^* > 4,1$ and $u_{17}^{**} < 2,2$ at $\varepsilon_{f17}^* = 0$; $\varepsilon_{n17}^* < 0,05$ and $\varepsilon_{n17}^{**} > 0,05$
$u_{13}^* > 70,2$ and $u_{13}^{**} < 33,5$ at $\varepsilon_{f13}^* = 0$; $\varepsilon_{f13}^* > 0,1$ and $\varepsilon_{f13}^{**} < 0,1$	$u_{18}^* > 62,0$ and $u_{18}^{**} < 11,9$ at $\varepsilon_{f18}^* = 0$; $\varepsilon_{n18}^* < 0,05$ and $\varepsilon_{n18}^{**} > 0,05$
$u_{14}^* > 54,1$ and $u_{14}^{**} < 22,3$ at $\varepsilon_{n14}^* = 0$; $\varepsilon_{f14}^* > 0,05$ and $\varepsilon_{f14}^{**} < 0,05$	$u_{19}^* > 48,1$ and $u_{19}^{**} < 19,2$ at $\varepsilon_{f19}^* = 0$; $\varepsilon_{n19}^* < 0,05$ and $\varepsilon_{n19}^{**} > 0,05$
$u_{15}^* > 52,6$ at $\varepsilon_{n15}^* < 0,2$ and $\varepsilon_{f15}^* > 0,1$; $u_{15}^{**} < 14,7$ at $\varepsilon_{n15}^{**} > 0,2$ and $\varepsilon_{f15}^{**} < 0,1$	$u_{20}^* > 29,3$ at $\varepsilon_{n20}^* < 0,2$ and $\varepsilon_{f20}^* > 0,1$; $u_{20}^{**} < 10,1$ at $\varepsilon_{n20}^{**} > 0,2$ and $\varepsilon_{f20}^{**} < 0,1$

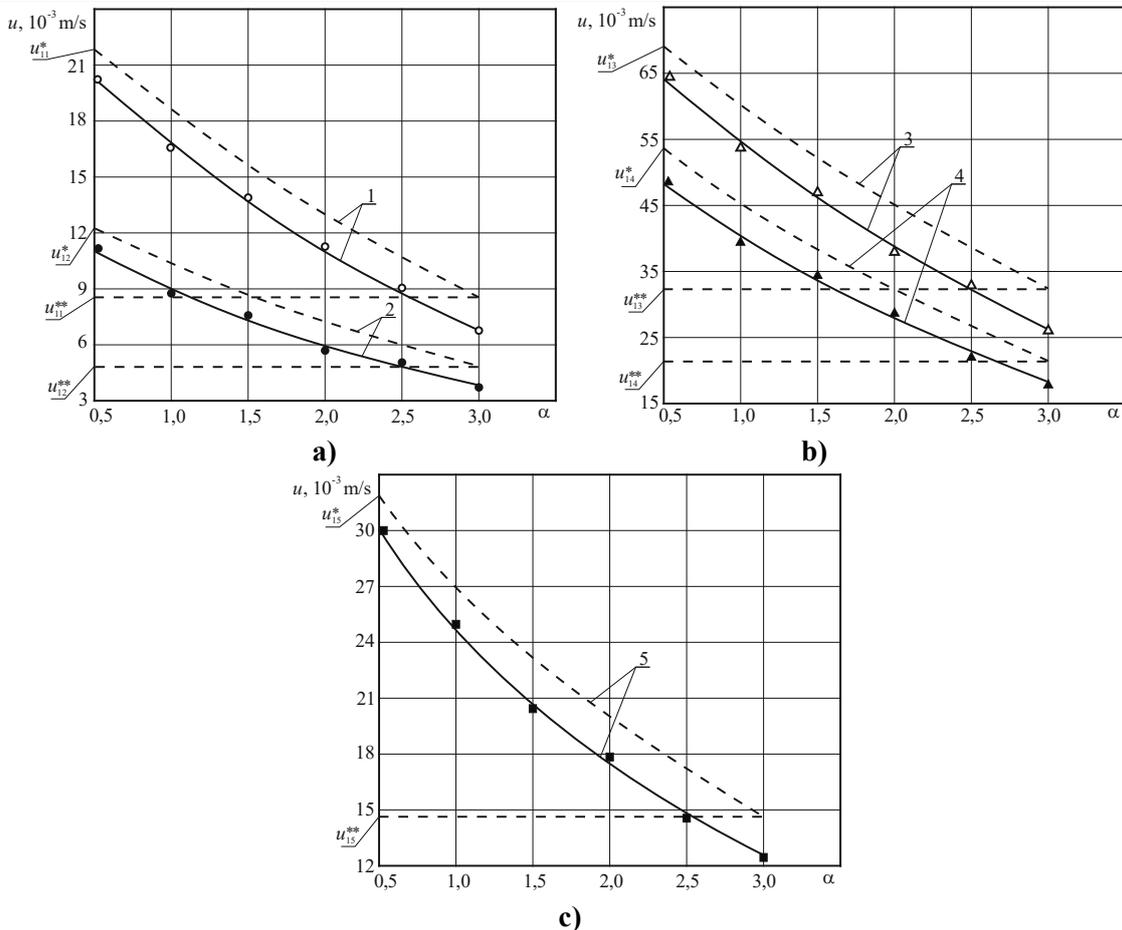


Figure 3. Dependencies of the burning rate of mixtures of Mg + NaNO₃ + paraffin + NaF on the coefficient of excess oxidant for different values of the amounts of additives under conditions of elevated heating temperatures ($d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$; $T_0 = 800 \text{ K}$; $P = 10^5 \text{ Pa}$): a) $\varepsilon_f = 0$; 1 – $\varepsilon_n = 0,05$; 2 – $\varepsilon_n = 0,2$; b) $\varepsilon_n = 0$; 3 – $\varepsilon_f = 0,1$; 4 – $\varepsilon_n = 0,05$; c) $\varepsilon_f \neq 0$ and $\varepsilon_n \neq 0$; 5 – $\varepsilon_n = 0,2$; $\varepsilon_f = 0,1$; – – – calculation curve; u_i^* and u_i^{**} ($i = \overline{11,15}$) – critical values of the mixture burning rate; \circ , \bullet , Δ , \blacktriangle , \blacksquare – experimental data

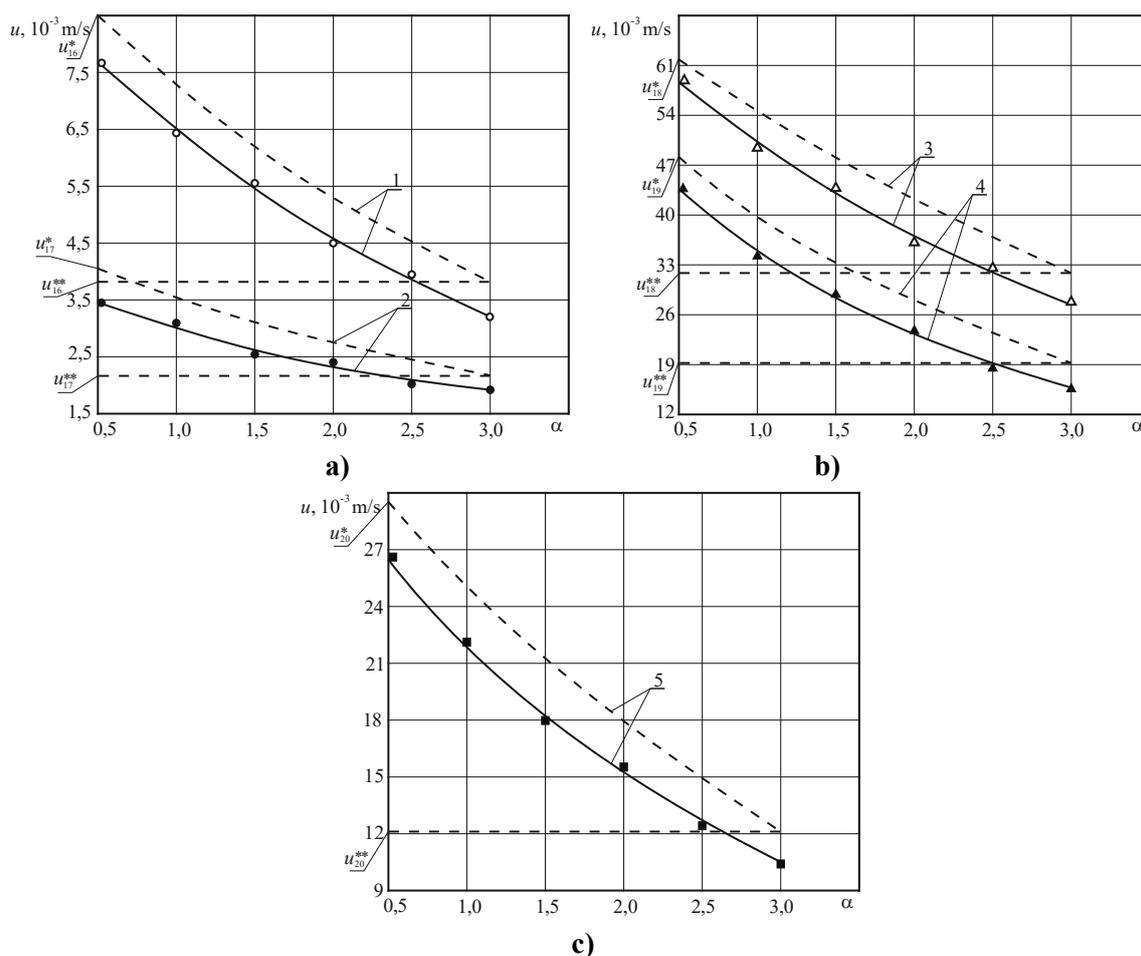


Figure 4. Dependences of the burning rate of mixtures of Mg + NaNO₃ + stearin + BaF₂ on the coefficient of excess oxidant for different values of the amounts of additives under conditions of elevated heating temperatures ($d_m = 74,5 \mu\text{m}$; $d_o = 106 \mu\text{m}$; $C_c = 0,95 \dots 0,96$; $T_0 = 800 \text{ K}$; $P = 10^5 \text{ Pa}$):

- a) – $\varepsilon_f = 0$; 1 – $\varepsilon_n = 0,05$; 2 – $\varepsilon_n = 0,2$; b) – $\varepsilon_n = 0$; 3 – $\varepsilon_f = 0,1$; 4 – $\varepsilon_n = 0,05$;
- c) – $\varepsilon_f \neq 0$ and $\varepsilon_n \neq 0$; 5 – $\varepsilon_n = 0,2$; $\varepsilon_f = 0,1$; – – – calculation curve;
- u_i^* and u_i^{**} ($i = \overline{16,20}$) – critical values of the burning rate of mixtures;
- , ●, Δ, ▲, ■ – experimental data

Thus, mathematical models of the combustion of mixtures of metal + nitrate-containing oxidizer + additives of organic and inorganic substances were developed using standard software provision allows at the design stage and subsequent bench tests of products to predict with an accuracy of 8...12 % the effect of elevated heating temperatures and external pressures (for a wide range of changes in the ratios of components and their dispersion) on the burning rate of mixtures (the duration of action of products equipped with sample mixtures), and as well as critical regimes of sustained combustion propagation under conditions of thermal exposure. This makes it possible to predict various fire-dangerous situations that arise under the conditions of various external thermal effects on the surface of metal casings of products.

Conclusions

The following new results were obtained on the basis of theoretical and experimental studies of the combustion process of multicomponent pyrotechnic nitrate-metallized mixtures under conditions of external thermal effects.

The combustion mechanism of compacted mixtures of metal fuel powders, nitrate-containing oxidizers and additives of organic and inorganic substances under conditions of elevated heating temperatures and external pressures has been established, according to which the processes of transformation of the initial solid mixture into combustion products are stationary and proceed in the following zones: condensed phase (c-phase), where the initial mixture is heated, thermal decomposition of oxidizers and additives, high-temperature oxidation of particles occurs metal fuels in active decomposition products; the phase interface (combustion surface), on which the mixture loses the properties of a condensed medium, while metal particles ignite on it, which are then carried by the flow of gaseous decomposition products into the flame zone; gas phase (g-phase, zone of heat release in the flame), in which transferred metal particles burn in the diffusion mode in gaseous decomposition products, forming combustion plumes.

Mathematical models of the combustion process of four-component mixtures of metal + oxidizer + organic additive + inorganic additive under conditions of external thermal influences were developed, which use the kinetic characteristics of the thermal decomposition of the oxidizer and substance additives, high-temperature oxidation, ignition and combustion of metal fuel particles in different zones, as well as the results of thermodynamic calculations of the temperature of the combustion products of mixtures and the amount of high-temperature condensate (non-oxidized metal) in them, which allows with a relative error of 8...10% to calculate the dependence of the combustion rate of mixtures on technological parameters (the nature and ratio of components, their dispersion) and external conditions (elevated heating temperatures, external pressures). These models make it possible to determine, for a wide range of changes in the heating temperature (up to 800 K) and external pressure (up to 10^7 Pa), the critical ranges of changes in technological parameters, exceeding which can lead to a sharp increase in the rate of combustion of mixtures (with an excess of metallic fuel), which leads to fire-explosive destruction of pyrotechnic products, as well as to a sharp decrease in the burning rate (with an excess of oxidizer) and subsequent failure of the products during their normal use.

Acknowledgments

None.

Conflict of Interest

Authors declare no conflict of interest.

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МОДЕЛІ ГОРІННЯ БАГАТОКОМПОНЕНТНИХ ПРОТЕХНІЧНИХ НІТРАТНО-МЕТАЛІЗОВАНИХ СУМІШЕЙ

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Анотація. Встановлено механізм горіння ущільнених сумішей з порошків металізованих палих (Mg, Al та ін.), нітратовмісних окиснювачів (NaNO₃, KNO₃ та ін.), добавок органічних (парафіну, стеарину та ін.) та неорганічних речовин (фторидів металів, оксидів металів та ін.) в умовах зовнішніх термічних впливів, згідно з яким процес перетворення вихідної суміші на продукти згоряння, у середньому, є стаціонарним та здійснюється у трьох просторово розділених зонах: конденсована фаза, де відбувається розкладання та високотемпературне

окиснення компонентів; поверхня розділу фаз (поверхня горіння), на якій відбувається повне розкладання компонентів та займання частинок металу, які потім переходять у зону полум'я; газова фаза (зона тепловиділення у полум'ї), в якій частинки металу згоряють у дифузійному режимі, утворюючи продукти згоряння. Розроблено моделі горіння сумішей, які враховують кінетичні характеристики термічного розкладання окиснювача, добавок органічних та неорганічних речовин і високотемпературного окиснення, займання та горіння частинок металу у продуктах розкладання, а також результати термодинамічних розрахунків температури продуктів згоряння сумішей та вмісту у них високотемпературного конденсату (неокисненого металу), що дозволяє з відносною похибкою 8...10 % визначати критичні діапазони зміни швидкості горіння сумішей в умовах зовнішніх термічних дій, перевищення яких може призводити як до прискорення процесу горіння сумішей та пожежовибухонебезпечного руйнування піротехнічних виробів, так і до різкого згасання їх процесу горіння та відмови виробів.

Ключові слова: пожежна безпека, піротехнічні суміші, нітратовмісні окиснювачі, металеві пальні, добавки органічних та неорганічних речовин, термічні дії, процеси горіння, моделі горіння металізованих конденсованих систем.

Дата надходження: 11.07.2023

Прийнято: 10.08.2023