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MATHEMATICAL MODELLING OF THE COMBUSTION PROCESS OF PARTICLES OF TWO-COMPONENT METAL ALLOYS IN THE DECOMPOSITION PRODUCTS OF PYROTECHNIC MIXTURES

Purpose. *The purpose of this work is to establish the mechanism and develop a mathematical model of the combustion process of AMC particles in gaseous decomposition products of nitrate-containing oxidants and organic additives to determine the dependence of their combustion time on technological parameters and external conditions.*

Methods. *Modern methods of physical and chemical analysis: thermocouple methods of temperature measurement (tungsten-rhenium thermocouples were used); non-contact methods of recording the beginning and end of the burning of metal particles (temperature indicators, photosensitive elements, etc); film shooting methods (film camera "Konvas-auto" (shooting speed 30 frames/s)) and methods of micro-filming SKS-1M film camera (shooting speed 3000...5000 frames/s)) of the burning process of individual metal particles; methods of X-ray structural and micro-X-ray spectral analysis for conducting chemical analysis.*

Results. *As a result of studies of the combustion process of aluminium-magnesium alloy (AMA) particles in products of thermal decomposition of pyrotechnic mixtures based on nitrate-containing oxidizers and organic additives, it was established that the process of combustion of alloy particles in gaseous decomposition products of mixtures that are formed (oxygen, oxygen + nitrogen or air in the presence of no more than 10 % $CO + CO_2$) with an alloy content of no more than 80 % Al flows stably through the diffusion mechanism; with a content of Mg in the alloy of at least 20 %, the combustion process of the particles becomes unstable and becomes explosive as a result of their spontaneous fragmentation into smaller particles, the combustion of which becomes intermittent and accelerates sharply. Two points of view on the mechanism of diffusion combustion of alloy particles are also formulated: according to one of them, the particle combustion process takes place in two stages (first there is vapour phase combustion of Mg from the alloy, as the most volatile component, and then vapour phase combustion of the remaining molten drop of Al), and according to the second, simultaneous evaporation and combustion of both particles takes place. A mathematical model of diffusion combustion of particles of two-component metal alloys in gaseous products of thermal decomposition of pyrotechnic mixtures based on nitrate-containing oxidants and additives of organic substances has been developed. Using the example of AMA particles, it is shown that the developed combustion model allows, with an accuracy of 5...10 %, to determine the dependence of its combustion times on the technological parameters of the alloy powders (the content of the components in the alloy, the radius of the particle) and external conditions (the composition of the environment, the concentration of oxygen in it). As a result of calculations carried out on a PC in dialog and real-time mode, the following dependencies were established: an increase in the Mg content in the alloy and the concentration of O_2 in the gaseous medium, as well as a decrease in the radius of the particle leads to a decrease in the burning time of the particle and a sharp increase in its burning rate, which contributes to destabilization and explosive development of the combustion process of pyrotechnic mixtures under external thermal conditions.*

Scientific novelty. *As a result of studying of the process of combustion of AMC particles in products of thermal decomposition of pyrotechnic mixtures based on nitrate-containing oxidizers and additives of organic substances (in the amount of 5...10%), two points of view on the mechanism of diffusion combustion of alloy particles were formulated: according to one of them, the process of particle combustion proceeds in two stages (first there is a vapour-phase burning of Mg from the alloy, as the most volatile component, and then there is a vapour-phase burning of the remaining molten drop of Al), and according to the second, simultaneous evaporation and burning of both particles takes place; at the same time, spinel is the basis of combustion products in both cases $MgAl_2O_4$.*

Practical significance. *The results of theoretical and experimental studies, which are obtained in the form of mathematical models and a database of experimental data on the influence of technological parameters of mixture charges on critical modes of explosive development of their combustion process under conditions of external thermal influences (elevated heating temperatures, external pressures, etc.), allow at the stage of manufacturing products by optimization of technological parameters (ratio of components, dispersion of metallic fuel and its nature) to increase their ignition temperature and reduce the probability of fire-explosive destruction of products in the conditions of their storage and transportation, taking into account the influence of external thermal actions; in the conditions of launch of products by optimising the speeds and angles at which they are fired during their application, to reduce the heating temperature of the metal shells of mixture charges and to reduce the number of premature explosive destruction of products during firing and flight.*

Introduction.

Currently, pyrotechnic mixtures of metal fuels (two-component alloys of metals (aluminium-magnesium alloys (AMA), etc.), nitrate-containing oxidizers (nitrates of alkali and alkaline earth metals, etc.) and additives of various organic substances (paraffin, stearin, iditol, etc.) are widely used are used in various branches of the national economy and military equipment (incendiary mixtures for signalling, lighting and tracers, pyrotechnic products of IR technology, elements of rocket and space technology, etc.). At present, due to the limitations of experimental research on the combustion of particles of two-component metal alloys, including AMA etc., in active gaseous environments (Akkerman & Penner, 2020; Leontyev *et al.*, 2021; Sergeev *et al.*, 2020), it is impossible through their analysis to reveal the main regularities of the combustion process of these particles and to establish the mechanism of its occurrence in the indicated environments. Therefore, the purpose of this work is to establish the mechanism and develop a mathematical model of the combustion process of AMA particles in gaseous decomposition products of nitrate-containing oxidants and organic additives to determine the dependence of their combustion time on technological parameters and external conditions.

Literature review

The use of pyrotechnic mixtures based on AMA, nitrate-containing oxidizers and additives of various organic substances in various sectors of the national economy and military equipment is reflected in works (Bai & Yang, 2019; Berezhnaya *et al.*, 2020; Fang *et al.*, 2019). In conditions of forced external heating (for example, during the ignition of warehouses where products equipped with charges from the mixtures in question are stored, in conditions of transportation with intense convective heating of their surfaces, or in case of shock thermal effects on the metal cases of products in conditions of shot and flight) pyrotechnic mixtures can ignite prematurely with a sharp acceleration of the further process of their combustion, destruction of products due to the formation of high-temperature combustion products that fly at high speeds in different directions, which are fire-hazardous for surrounding objects (buildings,

fuel and lubricants, launchers with service personnel etc.) (Jia *et al.*, 2021; Syromyatnikov *et al.*, 2020; Shchukin *et al.*, 2019). Therefore, in order to prevent premature fire-explosive destruction of products under conditions of external thermal influences, it is necessary, first of all, to have data on the regularities of the process of combustion of metal fuel particles in the decomposition products of mixtures when they are heated. At the moment, theoretical and experimental studies of the combustion of metal particles (Mg, Al, Ti, Zr, etc.) in gaseous oxidising media (oxygen, mixtures of oxygen and nitrogen, air, etc.) have been sufficiently completed. It was shown that the process of burning of metal particles proceeds according to the diffusion mechanism with the formation of bright luminescence zones (Dibrova *et al.*, 2020). The influence of the nature of the oxidising medium and metallic fuel, as well as the dispersion of components and external conditions (elevated heating temperatures and external pressures, environmental composition, etc.) on the combustion characteristics of metallic fuel particles has been established (Kirichenko *et al.*, 2020). Mathematical models of the combustion process of metal particles have been developed, which allow to calculate the speed and time of their combustion depending on the characteristics of the powders and external conditions (Kyrychenko, Dibrova, Motrichuk, Vashchenko, Kolinko, 2019).

Research methods

As two-component (binary) metal alloys in pyrotechnic mixtures based on nitrate-containing oxidizers ($NaNO_3$, KNO_3, $Ba(NO_3)_2$, $Sr(NO_3)_2$, etc.) and additives of organic substances (paraffin, stearin, thiocol, etc.) in currently, aluminium-magnesium alloy (AMA) is most widely used (Kyrychenko, Dibrova, Motrychuk, Vashchenko, Kolinko, Tsybulin, 2019; Kyrychenko, Gvozd *et al.*, 2021). As a result of research (Kyrychenko, 2021) on the behaviour of mixture components under external heating conditions within the main combustion zone of the considered mixtures (k-phase reaction zone characterised by a range of temperature changes of 1100...1300 K (under normal conditions)) (Kyrychenko, Kovalyshyn *et al.*, 2021) such organic substances as paraffin, stearin and thiocol completely decompose to

gaseous products (the main ones are CO and CO_2). At the same time, gaseous CO and CO_2 products in the main decomposition products of nitrate-containing oxidizers ($O_2 + N_2$ stream) are present in small quantities (the amount of organic additives in the mixture does not exceed 5...10 %), and the rate of oxidation of metal fuel particles into CO and CO_2 is an order of magnitude lower , than in the decomposition products of the oxidant. Therefore, the results of studies of combustion processes of AMA in the specified gaseous products, including air, which is based on a mixture of $O_2 + N_2$ gases, are considered below.

All studies were conducted on AMA powders produced by the pyrotechnic industry on standard pyrotechnic equipment using modern methods of physical and chemical analysis (Kyrychenko, Investigation, 2022): thermocouple methods of temperature measurement (tungsten-rhenium thermocouples were used); non-contact methods of recording the beginning and end of the burning of metal particles (temperature indicators, photosensitive elements, etc.); methods of film shooting (film camera "Konvas-automat" (shooting speed 30 frames/s)) and methods of micro-filming film camera CKC-1M (shooting speed 3000...5000 frames/s)) of the burning process of individual metal particles; methods of X-ray structural and micro-X-ray spectral analysis for chemical analysis. The relative measurement error of the main characteristics of the burning process of metal particles (burning speed, burning time) did not exceed 5...7 %.

Study of the mechanism of the combustion process of AMA particles in gaseous decomposition products of mixtures. The process of burning aluminium-magnesium particles is quite complicated. During studies of the combustion process of individual immobile particles of aluminium-magnesium alloys using high-speed cinematography and low-inertia thermocouples, it was established that the combustion mechanism of these particles in oxygen-argon mixtures, in air and carbon dioxide is very similar to the combustion mechanism of pure magnesium, i.e., the beginning of the ignition process of particles is preceded by oxidation reactions occurring on their surface, and the combustion process proceeds according to the diffusion mechanism. At the same time, it is possible to formulate, at most, two points of view on the combustion of

alloy particles. According to one of them, the process of combustion of an aluminium-magnesium alloy particle proceeds in two stages: the burning of magnesium from the alloy particle and the burning of the remaining molten drop of aluminium. Both phases of combustion take place in the vapour phase (fig. 1, 2).

According to the results of spectroscopic studies, the burning of both components of the Al – Mg alloy begins simultaneously, but magnesium burns out faster than aluminium. In contrast to this, there is a point of view that during the burning of an aluminium-magnesium alloy, the more volatile magnesium first burns out (in a vapour phase flame), and the non-volatile aluminium is then dispersed in the form of small particles that burn out.

According to the second point of view, there is simultaneous evaporation and burning of both particles.

The alternate evaporation of magnesium and aluminium from the particle and the complete absence of complex molecules based on Mg and Al in the pair were established, while the combustion products consist mainly of spinel $MgAl_2O_4$ (table 1).

The dependence of the ratio of the radius of the luminescence zone (r_l, μm) to the initial radius of the particle (r_0, μm) on the relative burning time ($\bar{\theta} = \frac{\tau_c}{\tau_{0c}}$, where τ_c and τ_{0c} – respectively, the current burning time of the particle and the time of its complete combustion, ms) particles of the Al – Mg alloy is more complex than that of pure aluminium and magnesium (fig. 3). Thus, when burning particles of an alloy of 30 % Al + 70 % Mg in air, the ratio is r_l/r_0 in a very short period of time ($\approx 10^{-4}$ s) reaches the maximum value (ab). Then, for a considerable time, this ratio practically does not change (plateau bc). When the process of burning Mg from the particle ends, the ratio r_l/r_0 begins to decrease to a minimum (point d). Further, at the moment of the beginning of the burning of aluminium, it increases sharply again (section de), reaching a maximum (point e). As the aluminium burns out, the ratio r_l/r_0 decreases monotonically (ef), approaching in magnitude to the size of the glowing oxide particle that remained after the combustion process was completed. The nature of the relationship change $r_l/r_0 = f(\bar{\theta})$ when the oxygen concentration in the medium changes, it is preserved for particles of alloys containing no

more than 80 % Al. At the stage of steady-state combustion of magnesium from the particles of the ratio r_l/r_0 decreases with a decrease in the concentration of oxygen in the environment and the content in the alloy. If at the end of burning Mg from the particle ratio r_l/r_0 decreases to unity (the glow zone disappears), then the particle splits with the subsequent explosive development of the combustion process.

Thus, the combustion process of AMA particles in the gaseous products of thermal decomposition of the considered mixtures (oxygen, oxygen + nitrogen or air in the presence of no more than 40 % CO + CO$_2$) with an alloy content of no more than 80 % Al proceeds stably by the diffusion mechanism.

Discussion results

**Mathematical model of diffusion combustion of a particle of a two-component metal alloy in gaseous decomposition products of mixtures.** The model for calculating the rate of diffusion burning of a single particle of an alloy of two metals was considered in a standard setting (Kyrychenko *et al.*, 2022): heat and mass transfer processes proceed quasi-stationarity within the limits of the reduced pellicle with an outer radius $r_1 = r_0 \frac{Nu}{Nu\text{-}2}$ (r_0 – the initial radius of the particle, Nu – the Nusselt criterion (Kyrychenko, Methodology, 2022); physical constants are constant and the same for all components of the gas mixture; metal oxides completely condense on the combustion surface. It is believed that the alloy is an ideal solution, that is, it obeys Raoult's law (Kyrychenko, Methodology, 2022). The temperature and composition are assumed to be constant over the entire volume of the particle.

**Table 1** - Radiography of products of combustion of alloys

Alloy composition	Basic oxides	Secondary oxides
Al	α-Al$_2$O$_3$	γ-Al$_2$O$_3$
Al – 5 % Mg	γ-Al$_2$O$_3$	MgAl$_2$O$_4$
Al – 12 % Mg	MgAl$_2$O$_4$	γ-Al$_2$O$_3$
Al – 36 % Mg	MgAl$_2$O$_4$	α-Al$_2$O$_3$, γ-Al$_2$O$_3$
Al – 43 % Mg	MgAl$_2$O$_4$	γ-Al$_2$O$_3$, MgO
Al – 50 % Mg	MgAl$_2$O$_4$, MgO	γ-Al$_2$O$_3$
Al – 55 % Mg	MgAl$_2$O$_4$, MgO	α-Al$_2$O$_3$, γ-Al$_2$O$_3$
Al – 65 % Mg	MgAl$_2$O$_4$, MgO	–
Al – 80 % Mg	MgO	MgAl$_2$O$_4$
Al – 90 % Mg	MgO	MgAl$_2$O$_4$
Mg	MgO	–
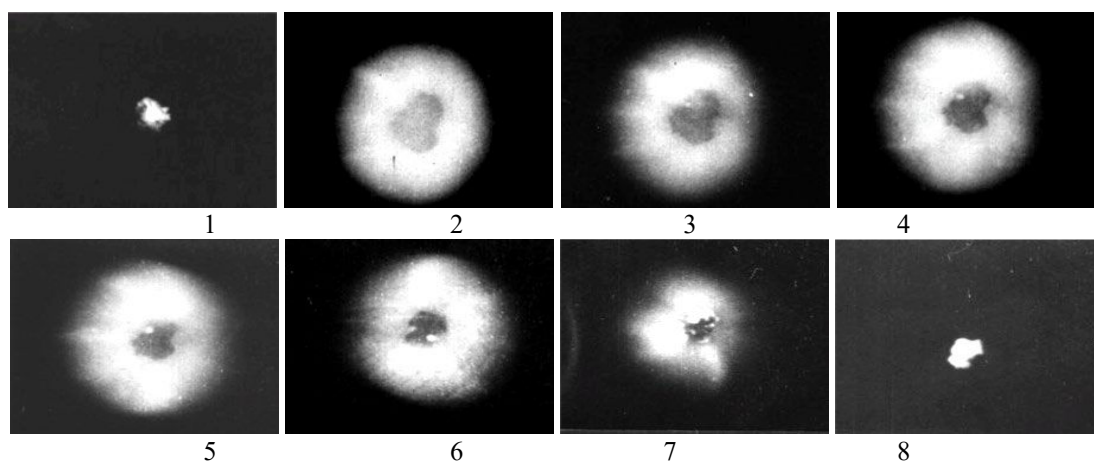
Fig. 1. Microfilms of the combustion process of a particle of an alloy of 50 % aluminium + 50 % magnesium in the environment of gaseous decomposition products of nitrate-containing oxidants (75 % oxygen + 25% nitrogen at P = 10^5 Pa (complete decomposition)): 1 – a picture of an alloy particle before its ignition; 2, 3, 4 – pictures characterising the burning of magnesium from the particle; 5, 6, 7 – pictures characterising the burning of aluminium from the particle; 8 is a photograph of an oxide particle formed after the combustion of an alloy particle
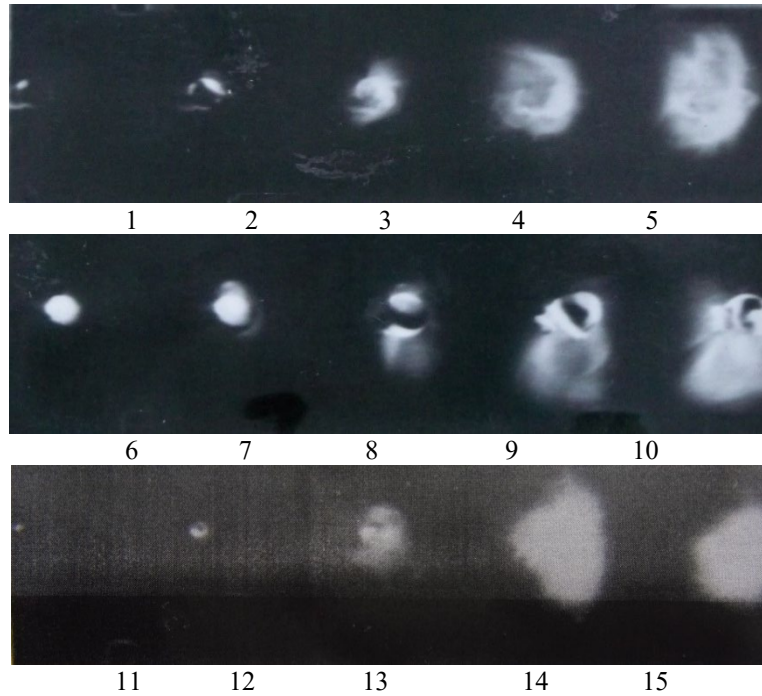
Fig. 2. Microfilms of the combustion process of a particle of an alloy of 40 % aluminium + 60 % magnesium in an environment of gaseous decomposition products of mixtures (P = 10^5 Pa): 1, 2, 3, 4, 5 – images characterising the combustion of a particle in an environment of 60 % O_2 + 30 % N_2 + 10 % CO + CO_2; 6, 7, 8, 9, 10 – pictures characterising the combustion of a particle in an environment of 65 % O_2 + 25 % N_2 + 10 % CO + CO_2; 11, 12, 13, 14, 15 are pictures characterising the combustion of a particle in an environment of 55 % O_2 + 35 % N_2 + 10 % CO + CO_2
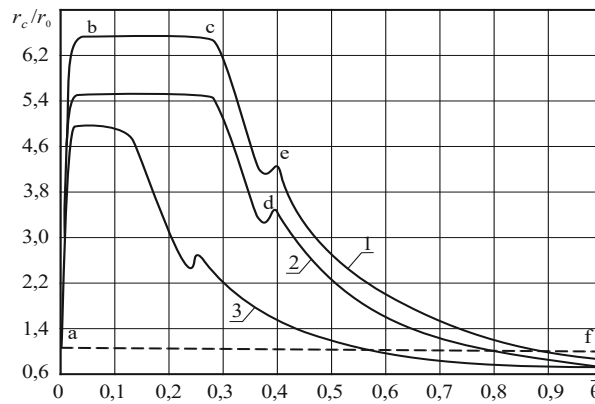
Fig. 3. Dependence of the ratio of the radius of the glow zone to the initial radius of the aluminium-magnesium alloy particle on the relative time of its burning: 1 – burning of the 30 % Al + 70 % Mg alloy in air; 2 – combustion of an alloy of 30 % Al + 70 % Mg in a mixture of 15 % O_2 + 85 % Ar; 3 – combustion of 50 % Al + 50 % Mg alloy in air

In the accepted assumptions, the processes of heat and mass transfer inside the reduced pellicle are described by a known system of differential equations (Motrichuk *et al.*, 2020):

1) in the inner zone of the reduced pellicle ($r_0 \le r \le r_c$), on which exothermic reactions of the interaction of a pair of metals and an environmental oxidant take place (the so-called glow zone):

$$\rho D\left(\frac{d^2 n_1}{dr^2} + \frac{2}{r} \cdot \frac{dn_1}{dr}\right) - \rho V \cdot \frac{dn_1}{dr} = 0, \qquad (1)$$

$$\rho D\left(\frac{d^2 n_2}{dr^2} + \frac{2}{r} \cdot \frac{dn_2}{dr}\right) - \rho V \cdot \frac{dn_2}{dr} = 0, \qquad (2)$$

$$\lambda\left(\frac{d^2 T}{dr^2} + \frac{2}{r} \cdot \frac{dT}{dr}\right) - \rho V c_P \cdot \frac{dT}{dr} = 0; \qquad (3)$$

2) in the outer zone of the reduced pellicle ($r_c \le r \le r_1$):

$$\rho D\left(\frac{d^2 n_{ox}}{dr^2} + \frac{2}{r} \cdot \frac{dn_{ox}}{dr}\right) - \rho V' \cdot \frac{dn_{ox}}{dr} = 0, \qquad (4)$$

$$\lambda\left(\frac{d^2 T}{dr^2} + \frac{2}{r} \cdot \frac{dT}{dr}\right) - \rho V' c_P \cdot \frac{dT}{dr} = 0. \qquad (5)$$

This system of equations is solved under the following boundary conditions:

1) the concentration of the metal vapour near the surface of the particle corresponds to the saturating one for the temperature T_0 and is determined in accordance with Raoult's law by the formulas:

$$n_1|_{r_0}= n_{10}=\left(\left(1+\frac{\mu_3}{\mu_1}\right)+\left(\frac{\mu_2+\mu_3}{\mu_1}\right)\cdot\frac{P_2^*}{P_1^*}\cdot\frac{1-N_1}{N_1}\cdot exp\left(\frac{l_1-l_2}{RT_0}\right)+\frac{\mu_3}{\mu_1}\cdot\frac{P}{P_1^*\cdot N_1}\cdot exp\left(\frac{l_1}{RT_0}\right)\right)^{-1}, \quad (6)$$

$$n_2|_{r_0}= n_{20}=\left(\left(1-\frac{\mu_3}{\mu_1}\right)+\left(\frac{\mu_2+\mu_3}{\mu_1}\right)\cdot\frac{P_1^*}{P_2^*}\cdot\frac{N_2}{1-N_2}\cdot exp\left(\frac{l_2-l_1}{RT_0}\right)+\frac{\mu_3}{\mu_2}\cdot\frac{P}{P_2^*\cdot(1-N_2)}\cdot exp\left(\frac{l_2}{RT_0}\right)\right)^{-1}, \quad (7)$$

where P – full gas pressure; μ_1, μ_2, μ_3 – molecular weights of metal vapours and inert gas, respectively; l_1, l_2 – heat of vaporisation of metals; P_1^*, P_2^* – steel for these metals; N_1, N_2 – mole fractions of metals in a particle: $N_1 + N_2 = 1$;

2) on the burning surface, the reaction proceeds instantaneously, and the concentration of the oxidizer and vapours of both metals are considered to be zero:

$$n_1|_{r_c}= n_2|_{r_c}= n_{ox}|_{r_c}= 0; \quad (8)$$

3) temperature of the particle and the combustion surface:

$$T|_{r_0}=T_0; \quad T|_{r_c}= T_r; \quad (9)$$

4) the temperature and oxidant content are specified on the outer boundary of the reduced pellicle:

$$T|_{r_1}=T_1; \quad n_{ox}|_{r_1}= n_{ox_1}; \quad (10)$$

5) from the equation of discontinuity and the ratio of stoichiometry, the relationship of gas flow velocities in the inner and outer zones of the reduced pellicle is expressed as:

$$\rho V' r^2 = \beta(\gamma-1)\rho_0\cdot V_0\cdot r_0^2, \quad (11)$$

where $\beta =\frac{\beta_1 n_{10}+\beta_2 n_{20}}{n_{10}+n_{20}}$; β_1, β_2 – the ratio of mass flows of the oxidizer and steam of the given metal; γ – the ratio of the mass flow of the gaseous reaction product to the mass flow of the oxidizer; index "0" means that the parameters are taken at $r = r_0$;

6) metal vapours and oxidizer are brought to the combustion surface in a stoichiometric ratio:

$$\frac{dn_{ox}}{dr}\bigg|_{r_c} = -\left(\beta_1\frac{dn_1}{dr}+\beta_2\frac{dn_2}{dr}\right)_{r_c}; \quad (12)$$

7) the mass flow of gases on the particle surface is equal to the flow of metal vapours:

$$\rho_0 V_0 = -\rho_0 D_0\left(\frac{dn_1}{dr}+\frac{dn_2}{dr}\right)_{r_0} +\rho_0 V_0(n_{10}+ n_{20}); \quad (13)$$

8) since as the more volatile component burns out, the composition of the particle changes (and, therefore, the temperature of the particle will also change), the expression for the heat flow to the particle also takes into account the heat that goes into its heating:

$$\lambda\frac{dT}{dr}\bigg|_{r_0} = -\rho_0 D_0\left(l_1\frac{dn_1}{dr}+ l_2\frac{dn_2}{dr}\right)_{r_0} + \rho_0 V_0(l_1 n_{10}+ l_2 n_{20})+\frac{G_c\cdot\rho_c\cdot r_0}{3}\cdot\frac{dT_0}{d\tau}; \quad (14)$$

where $\rho_c=\dfrac{1-\left(1\frac{\mu_1}{\mu_2}\right)\cdot N_1}{\frac{1-N_1}{\rho_{1c}}+\frac{\mu_1}{\mu_2}\cdot\frac{N_2}{\rho_{2c}}}$; ρ_{1c}, ρ_{2c} – values of the densities of the pure metals that make up the particle;

9) heat release on the combustion surface (to condensed oxide):

$$\lambda\left(\frac{dT^+}{dr}-\frac{dT}{dr}\right)_{r_c}= \rho_c D_c\left(\beta_1\cdot q_1\cdot\frac{dn_1}{dr}+\beta_2\cdot q_2\cdot\frac{dn_2}{dr}\right)_{r_c}, \quad (15)$$

where the indices "+" and "−" refer to the outer and inner zones of the reduced pellicle, respectively.

The change in the composition of the particle as it burns out is related to the flow of components as follows:

$$-\frac{d\mu_1}{d\tau}= -4\pi\left(\rho_0\cdot D_0\cdot r_0^2\cdot\frac{dn_1}{dr}\bigg|_{r_0} - \rho_0\cdot V_0\cdot n_{10}\right), \quad (16)$$

$$-\frac{d\mu_2}{d\tau}= -4\pi\rho_0\cdot r_0^2\cdot\left(D_0\cdot\frac{dn_1}{dr}-V\cdot n_1\right)_{r_0}. \quad (17)$$

Current values of the masses of metals μ_1 and μ_2 in the particle are connected by a ratio:

$$\frac{\mu_1}{\mu_1+\mu_2}=\left(1+\frac{\mu_2}{\mu_1}\cdot\left(\frac{1}{N_1}+1\right)\right)^{-1}. \quad (18)$$

In the accepted notation, the burning time of a particle is found from equation:

$$-\rho_c\cdot\frac{dr_0}{d\tau}= \rho_0\cdot V_0. \quad (19)$$

If we consider that the Lewis criterion $Le =\frac{\lambda}{\rho c_P D}$ is equal to one, which corresponds to the states of the considered gas mixtures under

normal conditions ($T_0 = 293$ K, $P = 10^5$ Pa) and introduce a new variable

$$\eta = exp\left(\frac{\rho_0 \cdot V_0 \cdot r_0 \cdot c_P}{\lambda} \cdot (1 - \frac{r_0}{r})\right), \qquad (20)$$

then the system of equations (1) – (5) will be simplified.

Then for the inner and outer zones of the reduced pellicle we have:

$$\frac{d^2T}{d\eta^2} = \frac{d^2n_1}{d\eta^2} = \frac{d^2n_2}{d\eta^2} = 0, \qquad (21)$$

$$\eta \cdot \frac{d^2T}{d\eta^2} + (1 - \beta(\gamma-1))\frac{dT}{d\eta} = \eta \cdot \frac{d^2n_{ox}}{d\eta^2} + (1 - \beta(\gamma-1))\frac{dn_{ox}}{d\eta} = 0, \qquad (22)$$

and boundary conditions (12) – (15) will be written as:

$$\frac{dn_{ox}}{d\eta}\bigg|_{\eta_c} = -\left(\beta_1 \frac{dn_1}{d\eta} + \beta_2 \frac{dn_2}{d\eta}\right)_{\eta_c}, \qquad (23)$$

$$1 = -\left(\frac{dn_1}{d\eta} + \frac{dn_2}{d\eta}\right)_{\eta=1} + n_{10} + n_{20}, \qquad (24)$$

$$c_P \cdot \frac{dT}{d\eta}\bigg|_{\eta=1} = -\left(l_1 \frac{dn_1}{d\eta} + l_2 \frac{dn_2}{d\eta}\right)_{\eta=1} + (l_1 n_{10} + l_2 n_{20}) + \frac{G_c \cdot \rho_c \cdot r_0}{3\rho_0 \cdot V_0} \cdot \frac{dT_0}{d\tau}, \qquad (25)$$

$$c_P \cdot \left(\frac{dT^+}{d\eta} - \frac{dT^-}{d\eta}\right)_{\eta_c} = \beta_1 \cdot q_1 \cdot \frac{dn_1}{d\eta}\bigg|_{\eta_c} + \beta_2 \cdot q_2 \cdot \frac{dn_2}{d\eta}\bigg|_{\eta_c}. \qquad (26)$$

Equations (16) and (17) will have the form:

$$-\frac{d\mu_1}{d\tau} = -4\pi r_0^2 \cdot \rho_0 \cdot V_0 \cdot \left(n_{10} - \frac{dn_1}{d\eta}\bigg|_{\eta=1}\right), \qquad (27)$$

$$-\frac{d\mu_2}{d\tau} = -4\pi r_0^2 \cdot \rho_0 \cdot V_0 \cdot \left(n_{20} - \frac{dn_2}{d\eta}\bigg|_{\eta=1}\right). \qquad (28)$$

Using the conditions (23) – (26) it is possible to obtain the expressions:

$$\rho_0 \cdot V_0 = \frac{\lambda Nu}{2c_P \cdot r_0} \cdot ln\eta_1, \qquad (29)$$

$$\eta_1 = \frac{\left(1 + (\gamma-1) \cdot n_{ox_1}\right)^{\frac{1}{\beta \cdot (\gamma-1)}}}{1 - n_{10} - n_{20}}, \qquad (30)$$

$$\frac{G_r \cdot \rho_c \cdot r_0}{3\rho_0 \cdot V_0} \cdot \frac{dT_0}{d\tau} = \left((\beta_1 \cdot n_{10} \cdot q_1 + \beta_2 \cdot n_{20} \cdot q_2) \cdot n_{ox_1} + \beta \cdot (n_{10} + n_{20}) \cdot (T_r - T_0)\right) \cdot$$
$$\cdot (1 - n_{10} - n_{20}) \cdot \left((n_{10} + n_{20}) \cdot \left(n_{ox_1} + \beta \cdot (n_{10} + n_{20})\right)\right)^{-1} - \frac{l_1 n_{10} + l_2 n_{20}}{n_{10} + n_{20}}. \qquad (31)$$

Thus, to calculate the time of complete combustion (τ_c) size of alloy particles r_0, which determines the fire-explosive modes of development of the combustion process of the considered mixtures under the conditions of external thermal actions (elevated heating temperatures, external pressures, the composition of the environment, etc.), we have a system of equations (6), (7), (18), (19), (27) – (31).

As an oxidizer, the gas mixture $O_2 + N_2$ was considered, which is the main active decomposition products within the k-phase of the considered pyrotechnic mixtures under conditions of external thermal influences (Motrichuk *et al.*, 2020).

With the involvement of all necessary physical and chemical constants (Kuznetsov *et al.*, 2019; Niu & Wang, 2019) calculations were carried out on a PC using standard application program packages in dialog and real-time mode according to the indicated system of equations for the AMA particle dependences of burning time τ_c from the ratio of components in the particle (magnesium concentration w_1), its medium size r_0 and oxygen concentrations n_{ox} in the gas mixture $O_2 + N_2$, which forms the basis of the gaseous decomposition products of the mixtures (fig. 4 – 6). It follows from the data presented in fig. 4 – 6: with increasing Mg content in the alloy from 0,2 to 1,0 values τ_c decreases by 1,8 times (for $n_{ox} = 0,2$) and 2,7 times (for $n_{ox} = 0,8$), that is, when increasing n_{ox} dependence $\tau_c(w_1)$ strengthens; when the radius of the particle increases r_0 from $r_0 = 15$ μm to $r_0 = 190$ μm size τ_c increases by 5,7 times (for $n_{ox} = 0,2$) and in 4,3 times (for $n_{ox} = 0,2$), that is, when growing n_{ox} dependence $\tau_c(r_0)$ is already weakening; during growth n_{ox} from $n_{ox} = 0,1$ to $n_{ox} = 1,0$ τ_c also decreases by 2,3 times (for $r_0 = 190$ μm) and 2,7 times (for $r_0 = 155$ μm), that is, when decreasing r_0 dependence $\tau_c(n_{ox})$ gets stronger. Comparing the results of calculations with experimental data shows that the difference between them does not exceed 5...10 %.
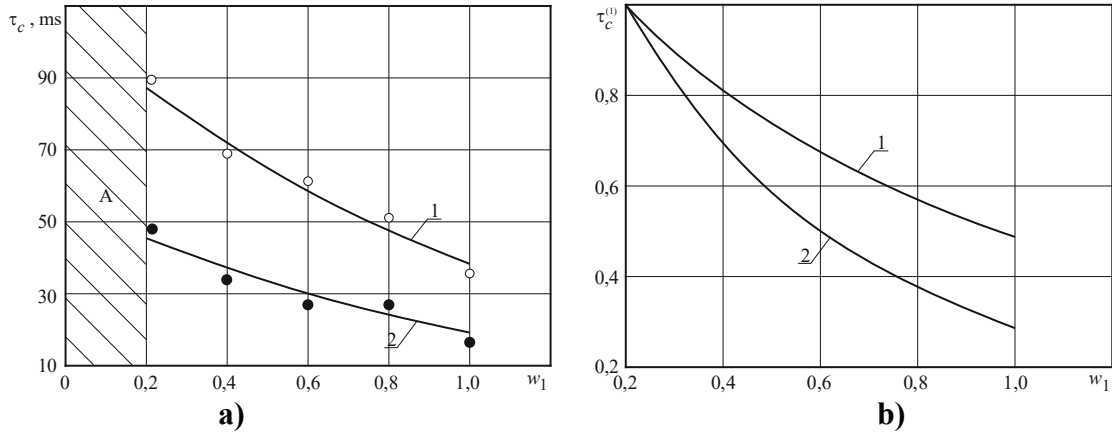
Fig 4. Dependencies of combustion times τ_c (a) and $\tau_c^{(1)}$ (b) AMA particles from the Mg content in the alloy for different values of the O_2 concentration in the gas medium ($r_0 = 155$ μm; $\tau_c^{(1)} = \frac{\tau_c}{\tau_c|_{w_1 = 0,2}}$, where $\tau_c|_{w_1 = 0,2}$ – value τ_c at $w_1 = 0,2$): A – the region of unstable and explosive combustion of AMA particles; 1 – $n_{ox} = 0,2$; 2 – $n_{ox} = 0,8$;—— – calculation curve; ○, □ – experimental data
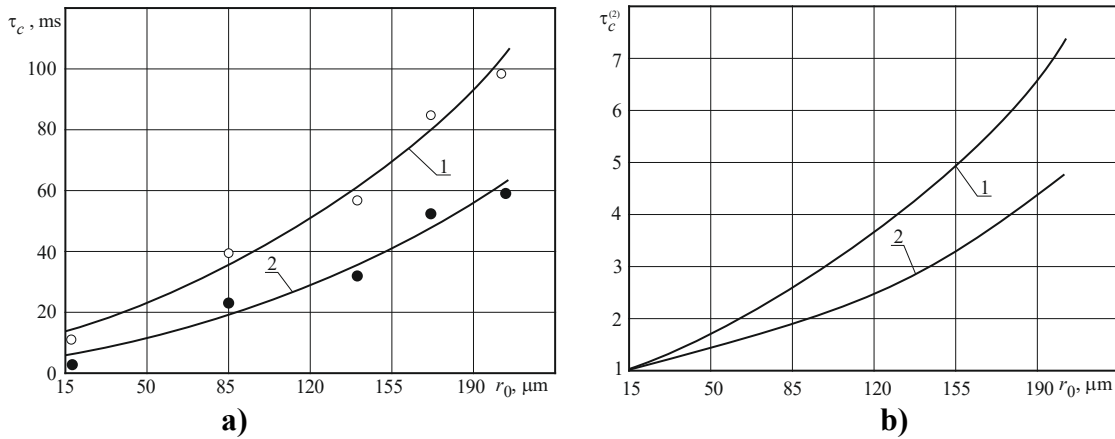
Fig. 5. Dependencies of combustion times τ_c (a) and $\tau_c^{(2)}$ (b) of AMA particles from its radius for different values of O_2 concentration in a single medium ($w_1 = 0,4$; $\tau_c^{(2)} = \frac{\tau_c}{\tau_c|_{r_0 = 15\ \mu m}}$, where $\tau_c|_{r_0 = 15\ \mu m}$ – value τ_c at $r_0 = 15\ \mu m$): 1 – $n_{ox} = 0,2$; 2 – $n_{ox} = 0,8$; —— – calculation curve; ○, □ – experimental data
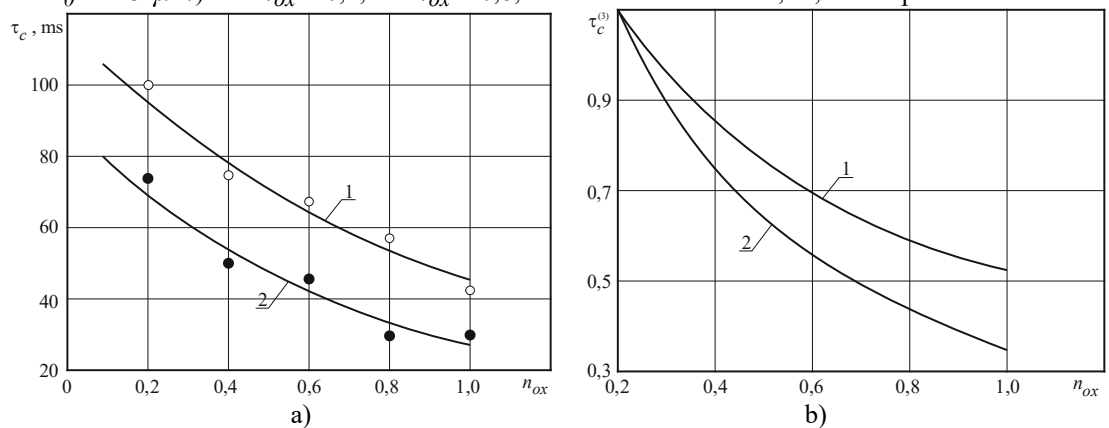
Fig. 6. Dependencies of combustion times τ_c (a) and $\tau_c^{(3)}$ (b) AMA particles from the O_2 concentration in the gas medium for different values of its radius ($w_1 = 0,4$; $\tau_c^{(3)} = \frac{\tau_c}{\tau_c|_{n_{ox} = 0,2}}$, where $\tau_c|_{n_{ox} = 0,2}$ – value τ_c at $n_{ox} = 0,2$): 1 – $r_0 = 190\ \mu m$; 2 – $r_0 = 155\ \mu m$; —— – calculation curve; ○, □ – experimental data

Thus, the developed method of calculating the combustion time of AMA particles in the products of thermal decomposition of pyrotechnic nitrate mixtures allows, at the stage of designing and manufacturing products based on them, to predict with an accuracy of 5...10 % fire-explosive modes of their premature activation under conditions of external thermal actions.

Conclusions

1. As a result of studies of the combustion process of AMA particles in products of thermal decomposition of pyrotechnic mixtures based on nitrate-containing oxidizers and additives of organic substances (in the amount of 5...10 %) it was established:

− dependence of the radius of the glowing zone (burning front) r_0 near the alloy particle from the time of its burning τ_{ϑ} is more complex than near the particles of pure Al and Mg (there are areas of sharp growth of the plateau and rapid decrease, which alternate) and is determined by the ratio of Al and Mg in the alloy, as well as the concentration of O_2 in the gas medium;

− based on dependency analysis $\tau_c(r_0)$ it is shown that the process of combustion of alloy particles in the gaseous decomposition products of the resulting mixtures (oxygen, oxygen + nitrogen or air in the presence of no more than 10 % $CO + CO_2$) with an alloy content of no more than 80 % Al proceeds stably by the diffusion mechanism; with a content of Mg in the alloy of at least 20 %, the combustion process of the particles becomes unstable and becomes explosive as a result of their spontaneous fragmentation into smaller particles, the combustion of which becomes intermittent and accelerates sharply;

− two points of view on the mechanism of diffusion combustion of alloy particles are formulated: according to one of them, the particle combustion process takes place in two stages (first there is vapour phase combustion of Mg from the alloy, as the most volatile component, and then vapour phase combustion of the remaining molten drop of Al), and according to the second, simultaneous evaporation and combustion of both particles takes place; at the same time, spinel is the basis of combustion products in both cases $MgAl_2O_4$.

2. On the basis of the conducted research, a mechanism was established and a mathematical model of the combustion of AMA particles in gaseous products of thermal decomposition of pyrotechnic mixtures based on nitrate-containing oxidizers and organic substance additives was established, which allows to determine with a relative error of 5...10 % the dependence of its combustion times on the technological parameters of the alloy powders (the content of components in the alloy, the radius of the particle) and external conditions (the composition of the environment, the concentration of oxygen in it).

3. As a result of calculations carried out on a PC in dialog and real-time mode, the following dependencies were established: an increase in the Mg content in the alloy from 0,2 to 1,0 and the concentration of O_2 in the gas medium from 0,1 to 1,0, as well as a decrease in the particle radius from 190 μm to 15 μm leads to a decrease in the value of τ_c by 1,8...5,7 times and a sharp increase in its burning speed, which contributes to the destabilisation and explosive development of the combustion process of pyrotechnic mixtures under conditions of external thermal effects.

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МАТЕМАТИЧНЕ МОДЕЛЮВАННЯ ПРОЦЕСУ ГОРІННЯ ЧАСТИНОК ДВОКОМПОНЕНТНИХ МЕТАЛЕВИХ СПЛАВІВ У ПРОДУКТАХ РОЗКЛАДАННЯ ПІРОТЕХНІЧНИХ СУМІШЕЙ

__Мета.__ Тому метою даної роботи є встановлення механізму та розробку математичної моделі процесу горіння частинок АМС у газоподібних продуктах розкладання нітратовмісних окиснювачів та добавок органічних речовин для визначення залежностей часу їх згоряння від технологічних параметрів та зовнішніх умов.

__Методи дослідження.__ Сучасні методи фізико-хімічного аналізу: термопарні методи вимірювання температури (використовувались вольфрам-ренієві термопари); безконтактні методи фіксування початку та кінця горіння частинок металів (температурні індикатори, фоточутливі елементи тощо); методи кінозйомки (кінокамера "Конвас-автомат" (швидкість зйомки 30 кадр/с)) та методи мікрозйомки кінокамера СКС-1М (швидкість зйомки 3000…5000 кадр/с)) процесу горіння окремих металевих частинок; методи рентгеноструктурного та мікрорентгеноспектрального аналізу (МРСА) для проведення хімічного аналізу.

__Результати.__ В результаті проведених досліджень процесу горіння частинок алюмінієво-магнієвого сплаву (АМС) у продуктах термічного розкладання піротехнічних сумішей на основі нітратовмісних окиснювачів та добавок органічних речовин встановлено, що процес горіння частинок сплаву у газоподібних продуктах розкладання сумішей, які утворюються (кисень, кисень + азот або повітря у присутності не більше 10 % CO + CO_2) при вмісті у сплаві не більше 80 % Al протікає стабільно по дифузійному механізму; при вмісті у сплаві Mg не менше 20 % процес горіння частинок стає нестійким та набуває вибухонебезпечного характеру в результаті їх спонтанної фрагментації на більш дрібні частинки, горіння яких стає уривчастим та різко прискорюється. Сформульовано також дві точки зору на механізм протікання процесу дифузійного горіння частинок сплаву: згідно однієї з них процес горіння частинки протікає у дві стадії (спочатку відбувається парофазне вигоряння Mg з сплаву, як найбільш летучого компоненту, а потім парофазне горіння розплавленої краплі Al, що залишилася), а згідно другої має місце сумісне випаровування та вигоряння обох частинок. Розроблено математичну модель дифузійного горіння частинок двокомпонентних металевих сплавів у газоподібних продуктах термічного розкладання піротехнічних сумішей на основі нітратовмісних окиснювачів та добавок органічних речовин. На прикладі частинок АМС показано, що розроблена модель горіння дозволяє з точністю 5…10 % визначати залежності часів її згоряння від технологічних параметрів порошків сплаву (вмісту компонентів у сплаві, радіусу частинки) та зовнішніх умов (складу навколишнього середовища, концентрації у ньому кисню).

В результаті проведених розрахунків на ПК у режимі діалогу та реального часу встановлено наступні залежності: збільшення вмісту Mg у сплаві та концентрації O_2 у газовому середовищі, а також зменшення радіусу частинки призводить до зменшення часу горіння частинки та різкого зростання швидкості її горіння, що сприяє дестабілізації та вибухонебезпечному розвитку процесу горіння піротехнічних сумішей в умовах зовнішніх термічних дій.

__Наукова новизна.__ В результаті проведених досліджень процесу горіння частинок АМС у продуктах термічного розкладання піротехнічних сумішей на основі нітратовмісних окиснювачів та добавок органічних речовин (у кількості 5…10 %) сформульовано дві точки зору на механізм протікання процесу дифузійного горіння частинок сплаву: згідно однієї з них процес горіння частинки протікає у дві стадії (спочатку відбувається парофазне вигоряння Mg з сплаву, як найбільш летучого компоненту, а потім парофазне горіння розплавленої краплі Al, що залишилася), а згідно другої має місце сумісне випаровування та вигоряння обох частинок; при цьому основу продуктів згоряння у обох випадках складає шпінель MgAl_2O_4.

__Практичне значення.__ Результати теоретичних та експериментальних досліджень, які отримані в вигляді математичних моделей та бази експериментальних даних по впливу технологічних параметрів зарядів сумішей на критичні режими вибухонебезпечного розвитку процесу їх горіння в умовах зовнішніх термічних впливах (підвищені температури нагріву, зовнішні тиски тощо) дозволяють на стадії виготовлення виробів шляхом оптимізації технологічних параметрів (співвідношення компонентів, дисперсності металевого пального та його природи) збільшувати температуру їх займання та знизити вірогідність пожежовибухонебезпечних руйнувань виробів в умовах їх зберігання та транспортування з врахуванням впливу зовнішніх термічних дій; в умовах запуску виробів шляхом оптимізації швидкостей і кутів під якими вони вистрелюються при їх застосуванні знижувати температуру нагріву металевих оболонок зарядів сумішей та зменшувати кількість передчасних вибухонебезпечних руйнувань виробів під час пострілу і польоту.

__Ключові слова:__ піротехнічні суміші, нітратовмісні окиснювачі, металеві пальні, органічні речовини, процеси горіння, математичне моделювання.

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