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COMBUSTION, EXPLOSION, AND SHOCK WAVES

Convective Burning of Fine Ammonium Nitrate–Aluminum Mixtures in a Closed Volume Bomb

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Abstract—It is commonly assumed that the burning of ammonium nitrate–aluminum mixtures is much less prone to undergo a transition to explosion and detonation than similar mixtures based on ammonium perchlorate. However, this conclusion has been made for mixtures based on commercial-grade ammonium nitrate with large particles. In this study, the combustion of fine loose-packed mixtures of ammonium nitrate and aluminum in a closed-volume bomb has been examined. It has been shown that fine mixtures (ammonium nitrate with a particle size of less than 40 μm and an ASD-4 aluminum powder with spherical particles with a size of about 4 μm) undergo high-intensity combustion; in experiments with a stoichiometric mixture, explosions are observed. The explosions occur in the initial phase of convective combustion and lead to abrupt pressure pulsations with an amplitude of a few kilobars and to the destruction of the cup in which the sample is placed. The dynamics of development of the explosion has been analyzed in detail using numerical simulation. According to the results of experiments with varied parameters—the degree of dispersion of the ammonium nitrate powders, the aluminum content in the mixture, the length and diameter of the charge, and the level of pressure generated by the combustion of the igniter,—threshold conditions have been determined to separate the following modes: the absence of ignition, layer-by-layer combustion, or convective combustion with a transition into an explosion in experiments with a stoichiometric mixture.

Keywords: convective burning, explosion, closed volume bomb, ammonium nitrate, ammonium perchlorate, aluminum

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INTRODUCTION

It is commonly assumed [1–4] that mixtures of ammonium nitrate with combustible additives are much less prone to undergo explosion than similar mixtures based on ammonium perchlorate. An example to support this conclusion is provided by the results of two studies conducted with mixtures of ammonium perchlorate [3] and ammonium nitrate [4] with ASD-4 aluminum in a closed volume bomb under almost the same combustion conditions. Thus, the burning of most of ammonium perchlorate–aluminum mixtures readily transited into an explosion at charge lengths exceeding a critical value (25–30 mm), whereas the combustion of the most active stoichiometric mixture of ammonium nitrate and aluminum at a maximum charge length for the given conditions (80 mm) occurred smoothly, typically in the convective combustion mode, without explosions at a relatively low rate of pressure rise in the bomb. However, it should be noted that, in those experiments, ammonium perchlorate and ammonium nitrate had different degrees of dispersion. The ammonium perchlorate had an average particle size of 20 μm. The ammonium nitrate, as a component of commercial explosives, was used mostly in the form of porous granules with a diameter of 1–2 mm. Several experiments conducted in a closed volume bomb with ground ammonium nitrate with particles of a 250–400 μm fraction have shown that, in this case, the combustion intensity increases [4]. The same tendency has been confirmed by the numerical simulation results [4]. It has become clear that comparison of the mixtures should be conducted using oxidants with the same degree of dispersion.

In this study, it has been shown that, if ammonium nitrate is ground to a particle size of less than 40 μm, then the intensity of the combustion of mixtures of this ammonium nitrate with fine aluminum in a closed volume bomb abruptly increases; in experiments with a stoichiometric mixture at a charge length of 40 and 80 mm, explosions are observed. Qualitatively, the behavior of mixtures of aluminum with ground ammonium nitrate is similar to the behavior of mixtures with ammonium perchlorate particles of the same size, although a certain difference in the explosion hazard in favor of ammonium perchlorate is preserved. Below, we describe the results of experiments on the combustion of mixtures of fine ammonium nitrate and aluminum taken in different amounts in a

Table 1. Summarized data on the combustion of mixtures of ASD-4 aluminum and ammonium nitrate (less than 40 μm) in a closed volume bomb

Diameter/length of charge, mm	Aluminum content in mixture, %	Pressure generated by igniter, MPa	Registered process	Rate of pressure rise during convective combustion, MPa/ms	Total combustion time, ms	Experiment number
16/40	18	Less than 8	No combustion	–	–	187
16/40	18	8–12	Layer-by-layer combustion	–	–	69, 70
16/40	18	12–36	Convective combustion	0.3–0.6	100–320	68, 181–186
16/40	18	More than 36	Explosion	–	6.1	47
10/80	18	Less than 16	No combustion	–	–	58, 59, 60
10/80	18	More than 20	Explosion	–	9, 10	62, 63
10/45	18	36	Convective combustion	3.4	25	55
10/55	18	36	Convective combustion	125	7.6	57
10/65 and 10/80	18	36	Explosion	–	6.2 6.6	54 56
10/80	8	Less than 20	No combustion	–	–	205
10/80	8	20–26	Layer-by-layer combustion	–	–	189, 190, 220, 221
16/40	8	More than 26	Convective combustion	0.3	360, 700	49, 222
16/40	31	Less than 10	No combustion	–	–	191, 192
16/40	31	11–36	Convective combustion	115–190	13, 15	50, 237
16/40	47	Less than 1	No combustion	–	–	218
16/40	47	1–36	Convective combustion	0.2–12	60, 80–3380	52, 193–196

closed volume bomb, which are supplemented by thermodynamic calculations and numerical simulation.

EXPERIMENTAL RESULTS

A loose-packed mixture of ammonium nitrate and aluminum (0.8–0.96 g/cm^3 depending on the aluminum content) was placed in a steel or Textolite cup with a diameter of 10 or 16 mm and a length of about 100 mm. The nominal volume of the bomb was 100 cm^3. Combustion was initiated using a nichrome spiral and a weighed portion of the igniter consisting of a mixture of grated smoky gunpowder and Sokol gunpowder. Aluminum in the form of an ASD-4 powder had spherical particles with an average size of about 4 μm. Commercial-grade ammonium nitrate—the starting material in the form of porous granules with a diameter of 1–2 mm—was ground in a vibrating mill. A fine fraction of ammonium nitrate with a particle size of less than 40 μm and a coarse fraction with a particle size of 250–630 μm were selected for the experiments. The aluminum content in the mixture was varied from 8 to 47 wt %. In the experiments, we also were varied the diameter and length of the charge (from 38 to 80 mm) and the weight of the igniter; the combustion of the igniter provided the generation of a pressure in the bomb in a range of 6–40 MPa within ~5 ms. The combustion process was registered using an AVL pressure sensor (an eigenfrequency of 250 kHz) installed in the bomb chamber. Combustion intensity was estimated from the maximum rate of pressure rise.

The experimental results are shown in Tables 1 and 2 and in Figs. 1–3. Depending on the initial properties, all the experiments can be divided into three groups. The first group includes experiments in which the pressure generated by the igniter was so low that the mixture was not ignited. At a higher pressure, the mixture was ignited; however, it underwent a layer-by-layer combustion or a portion of the charge remained unburned (Fig. 1, curve *1*). At a pressure above a threshold value, the combustion of the charge occurred in a convective mode or ended in an explosion. During explosion (a typical example of pressure

Table 2. Summarized data on the combustion of mixtures of ASD-4 aluminum and coarse-fraction ammonium nitrate (250–630 μm) in a closed volume bomb

Diameter/length of charge, mm	Aluminum content in mixture, %	Pressure generated by igniter, MPa	Registered process	Rate of pressure rise during convective combustion, MPa/ms	Total combustion time, ms	Experiment number
16/48	8	20	Layer-by-layer combustion			209
16/50	8	31	Layer-by-layer combustion			219
16/41	8	36	Convective combustion	0.5	280	43
16/44	18	7	No combustion			210
16/43	18	11	No combustion			211
16/43	18	15	Convective combustion	1.0	744	212
16/39	18	36	Convective combustion	6.0	39	41
16/40	31	2	No combustion			215
16/40	31	4	Convective combustion	0.6	1140	219
16/40	31	6	Convective combustion	9.0	305	213
16/39	31	36	Convective combustion	125	28	39
16/38	47	0.8	No combustion			216
16/37	47	1.4	Convective combustion	22	1550	217
16/35	47	36	Convective combustion	30	29	40

recording is shown in Fig. 1, curve *4*), the initial smooth portion in the diagram is followed by sudden sharp pressure pulsations of a characteristic damped form with an initial signal amplitude of 200–300 MPa. The post-experiment inspection showed that the cup containing the test charge was destroyed to small pieces, most of which burned down.

In the convective combustion mode, the maximum rate of pressure rise in the recorded curve can be used as a measure characterizing the combustion intensity. The respective values are listed in the sixth column of Tables 1 and 2. Low-intensity combustion can be detected if the rate of pressure rise does not exceed 1 MPa/ms. This process occurs for a few hundred milliseconds; the estimate of the average combustion rate yields a value of less than 1 m/s. Moderate-intensity combustion takes place if the rate of pressure rise lies in a range of 1 to a few tens of megapascals per second. In the case of high-intensity combustion, the rate of pressure rise achieves a value of 100 MPa/s and higher. This combustion becomes dangerous and can transit into an explosion. In all cases corresponding to the convective combustion mode, the diagrams of pressure in the bomb chamber have approximately the same shape, which can be considered standard (e.g., curve *3* in Fig. 2). The diagram consists of an initial region corresponding to the combustion of the igniter; the main portion corresponding to the combustion of the test charge at a rate of pressure rise that increases, achieves a maximum value, and then decreases; and a maximum point corresponding to the completion of combustion, after which the pressure in the bomb decays. In the case of low-intensity convective combustion, the beginning of combustion of the charge can be preceded by a region in which the pressure decreases after the completion of combustion of the igniter. An example of this diagram is shown in Fig. 2 (curve *1*).

Consider the effect of the properties of the charge on the threshold pressure of convective combustion. It is evident from the results given in Table 1 that the threshold pressure decreases with increasing charge diameter. Thus, for a mixture with a fine fraction of ammonium nitrate (less than 40 μm) at an aluminum content of 18%, in a charge with a diameter of 10 mm, the threshold pressure is 16 MPa; in a charge with a diameter of 16 mm, the pressure decreases to 12 MPa. With an increase in the aluminum content in a series of 8, 18, 31, and 47%, the threshold pressure monotonically decreases for mixtures with both small and large ammonium nitrate particles.

The experiments in which convective combustion was observed and the experiments that ended in an explosion yielded the following results. For mixtures with a coarse fraction of ammonium nitrate (250–630 μm), only convective combustion took place, explosions did not occur. The intensity of the convective combustion of the mixtures increases with increasing power of the

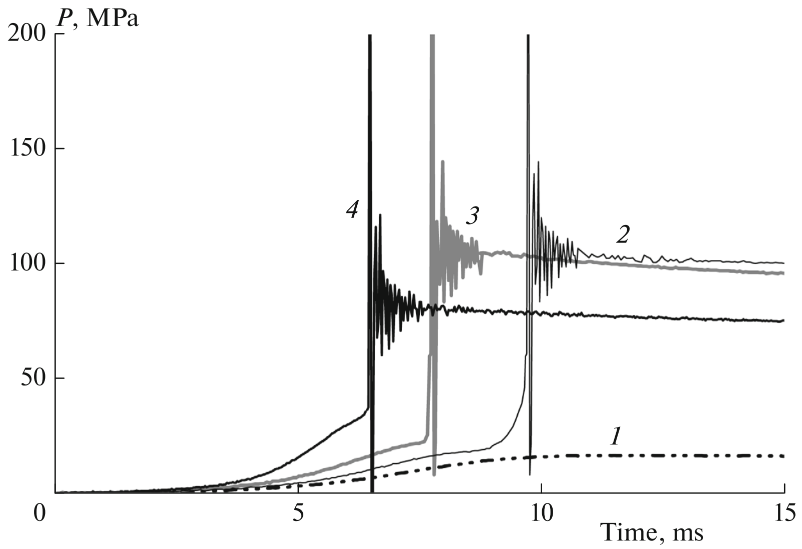

Fig. 1. Effect of the weight of the igniter on the combustion mode of the mixture of fine-fraction ammonium nitrate (less than 40 µm) with 18% of ASD-4 aluminum using charges with a diameter of 10 mm and a length of 80 mm. Weight of the igniter and experiment number: (*1*) 2 g, experiment 60 (no combustion); (*2*) 2.5 g, experiment 63 (explosion); (*3*) 3 g, experiment 62 (explosion); and (*4*) 5 g, experiment 54 (explosion).

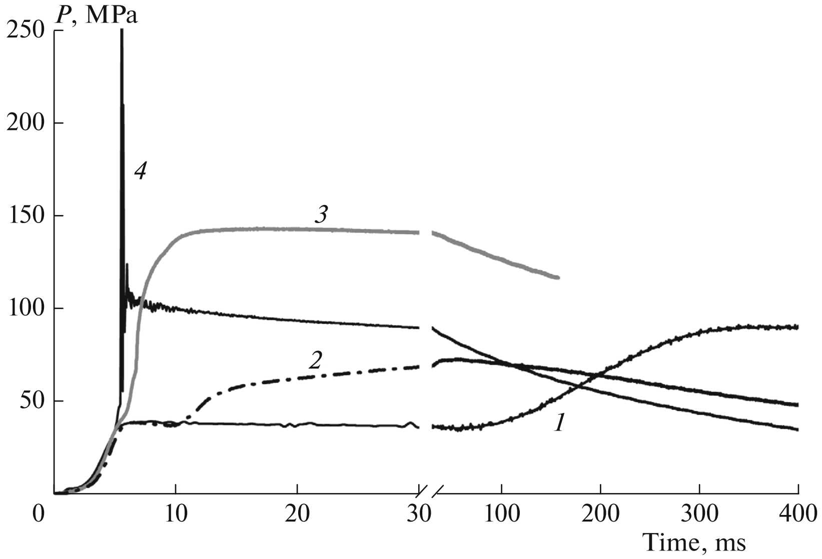

Fig. 2. Effect of aluminum content on the modes of convective combustion and explosion for mixtures based on fine-fraction ammonium nitrate using charges with a diameter of 16 mm and a length of 40 mm at the weight of the igniter of 5 g. Aluminum content and experiment number: (*1*) 8%, experiment 49; (*2*) 47%, experiment 52; (*3*) 31%, experiment 50; and (*4*) 18%, experiment 47 (explosion).

igniter. It is remarkable that the experiments with a stoichiometric mixture with 18% aluminum showed a relatively low rate of pressure rise. For the mixture with 47% Al and particularly the mixture with 31% aluminum, the combustion intensity was significantly higher than that of the mixtures containing 18 and 8% aluminum.

For the mixtures with a fine fraction of ammonium nitrate, explosions were observed only at 18% aluminum. At a charge length of 80 mm, an explosion occurred in all the experiments in which the pressure generated by the igniter was 20 MPa and higher. In the case of charges with a shorter length (40 mm), an explosion took place for the igniter with the greatest weight, which generated a pressure of 36 MPa. At a smaller weight of the igniter, convective combustion without an explosion was observed. For a mixture containing 31% aluminum, high-intensity convective

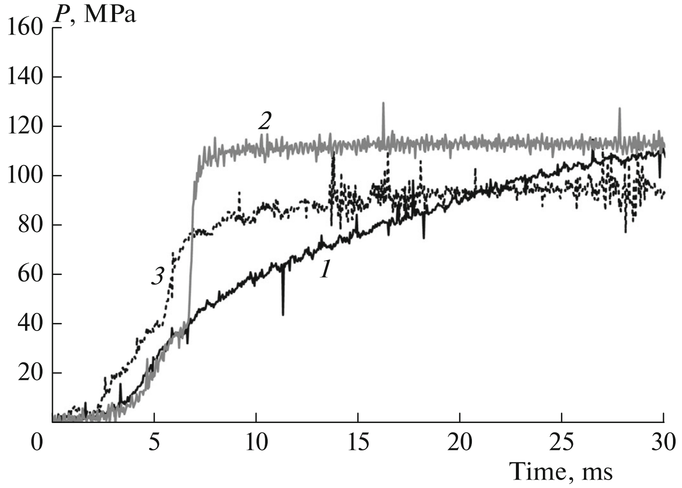

Fig. 3. Effect of aluminum content on the convective combustion modes for mixtures based on coarse-fraction ammonium nitrate (250–630 μm) using charges with a diameter of 16 mm and a length of ~40 mm at the weight of the igniter of 5 g. Aluminum content and experiment number: (*1*) 18%, experiment 41; (*2*) 31%, experiment 39; and (*3*) 47%, experiment 40.

combustion without an explosion occurred. With an increase in the power of the igniter (a pressure range of 11–36 MPa), the rate of pressure rise monotonically increased to values of 115–190 MPa/ms. At an aluminum content in the mixture of 8 and 47%, moderate-intensity convective combustion was observed.

To summarize this section, we can state that an increase in the length and diameter of the charge and the pressure generated by the igniter leads to an intensification of the convective combustion of fine ammonium nitrate–aluminum mixtures via increasing the rate of pressure rise in the bomb and, under certain conditions, results in an explosion. An increase in the ammonium nitrate particle size makes the combustion smoother. An increase in the aluminum content provides an intensification of the combustion and facilitates the occurrence of an explosion. In addition, for fine-fraction ammonium nitrate, the highest combustion intensity is exhibited by the mixture containing 18% aluminum, whereas for coarse-fraction ammonium nitrate, the mixtures with 31 and 47% aluminum undergo combustion at the highest rate of pressure rise.

BRIEF DESCRIPTION OF THE THEORETICAL MODEL

Numerical simulation was conducted using a theoretical model and a computer program that have been previously developed in a one-dimensional approximation in the context of the combustion of oxidizer–aluminum mixtures in a closed volume bomb [5]. The model has been adapted to respond to the intensification of the combustion of ammonium nitrate particles under the action of a heat flow generated during the combustion of aluminum particles. Let us briefly describe the main postulates of the model.

A charge of a given length is placed in the channel of a nondeformable sheath (cup). One of the end faces of the charge is closed; the other end is open and is connected with the bomb chamber. The porous charge consists of spherical oxidizer particles with initial diameter d_{k0}. The aluminum particles have a much smaller size and cover the oxidizer particles with a uniform thin layer. It is contemplated that, at a high content of aluminum, a portion of it can form conglomerates, which are located between the oxidizer particles. In this case, it is necessary to specify the fraction of aluminum in the conglomerates and the conglomerate size and then calculate the effective thickness of the aluminum layer covering the ammonium nitrate particles.

The combustion of the charge is initiated by the triggering of a gas igniter placed in the bomb chamber. The evolution of gases and heat during the combustion of the igniter is determined by time functions; the shape and coefficients of the functions are selected so as to accurately reproduce the experimental pressure–time diagrams in the bomb in the experiments where the igniter did not ignite the charge. After that, in the simulation of experiments in which the pressure generated by the igniter was higher, it is necessary just to change the weight of the igniter, while maintaining the other coefficients constant.

The combustion of ammonium nitrate–aluminum mixtures occurs in two stages. The first stage—the combustion of ammonium nitrate—occurs on the surface of the particles at a layer-by-layer combustion rate taken from the literature. The second stage—the diffusive combustion of aluminum particles in the products of combustion of ammonium nitrate—occurs at a rate determined by the known function of the particle diameter and the content of oxidizing reagents in the gas phase. The combustion of aluminum particles is accompanied by high temperatures. In the basic model, we did not take into account the fact that, under the action of the heat flow coming from the burning aluminum particles into the zone of combustion of ammonium nitrate, the combustion rate of the ammonium nitrate must increase; therefore, in this study, we have introduced a respective correction (see below).

The ignition and combustion of the charge are considered in terms of the mechanics of a two-phase multicomponent medium. The gas phase includes the air that fills the pores of the charge in the initial state, the combustion products of the igniter, and the gas component of the combustion products of the charge. The solid phase includes the initial mixture components and alumina in the form of α-Al_2O_3, which is the dispersed combustion product of aluminum. The phases are governed by different equations of state and exhibit different velocities, temperatures, and pressures. The total number of the components of the medium in the scheme under discussion is six: ammonium nitrate, original aluminum, coarse aluminum (in conglomerates), alumina, a gas phase, and a complex of oxidizing agents in the gas phase. Respective conservation equations are written for each of the components.

The combustion products flow deep into the charge through the pores and thereby cause the heating of the surface of the pores with the subsequent ignition and combustion of ammonium nitrate and aluminum. The amount of the heat that comes from the gas to the surface of the pores is calculated using the known semiempirical formulas that relate the heat transfer coefficient to the flow parameters and the temperatures of the gas and the surface of the pores through the generalized Nusselt and Reynolds numbers [6]. The ignition of ammonium nitrate is governed by the achievement of a given ignition temperature T_{ig} on the particle surface. To calculate the heating and the ignition time, a two-layer unit cell, which is a spherical oxidizer particle coated with a thin aluminum layer, is considered at each point along the length of the charge. The heat-conduction equation with the third-kind boundary condition is solved for each of the particles. Aluminum undergoes ignition after melting at a temperature of 943 K.

The combustion of the igniter and the charge causes an increase in pressure, which acts on the porous layer of the mixture before the flame front and thus leads to the compaction of the layer. The compaction is calculated using the empirical data on the intergranular stress determined in experiments on the static loading of the porous layer of the mixture on a press. These experiments were conducted in a range of the bulk density to a porosity of 0.05.

The bomb chamber space into which the flowing gases and dispersible particles of the mixture penetrate is regarded as a completely mixed reactor. It is assumed that the oxidizer and aluminum particles penetrated into the chamber after the dispersion of the charge undergo further combustion at an initial rate at which they flowed out of the cup.

Consider the formulas for the combustion rate of ammonium nitrate and aluminum used in this study. In the base model [5], the layer-by-layer combustion rate of ammonium nitrate particles is represented by the following empirical formula

$$u_{pk} = bP^{\nu} \exp \beta_T (T_0 - T_{\text{ref}}). \tag{1}$$

This formula includes empirical coefficients b (combustion rate constant), ν (pressure exponent), and β_T (temperature coefficient); pressure P; and the difference between the initial and reference values of the charge temperature. To a first approximation, we take into account that the heat flow from the secondary flame generated by the aluminum particles that burn in the oxidizer conversion products increases the combustion temperature of ammonium nitrate and, as a consequence, the combustion rate thereof. Equation (1) is represented as follows:

$$u_{pk} = bP^{\nu} \exp \beta_T G (T_b - T_{b,\text{ref}}). \tag{2}$$

Here, the difference between the initial temperatures is replaced by the difference between the current combustion temperature of the oxidizer T_b and the reference value of this temperature in the absence of a heat flow from the burning aluminum particles $T_{b,\text{ref}}$; G is a factor on the order of unity, which takes into account the effect of a number of secondary factors owing to which a change in the combustion temperature can have a slightly different effect on the layer-by-layer combustion rate than the effect of a change in the initial temperature. These factors include, for example, the difference between the heat capacity of ammonium nitrate at the initial temperature and the heat capacity of the combustion products of this material.

The combustion temperature of the oxidizer, which appears in Eq. (2), is calculated using the following formula that defines the heat balance for a layer-by-layer combustion wave:

$$c_g (T_b - T_{ig}) + c_k (T_{ig} - T_0) = Q + \frac{q}{\rho_k u_{pk}}. \tag{3}$$

Here, c_g and c_k are the specific heat capacities of the gas and the solid oxidizer, T_{ig} is the temperature on the surface of the oxidizer particle, T_0 is the initial temperature, Q is the heat of combustion of the oxidizer, and ρ_k is the density of the oxidizer. Heat flow q that comes into the zone of combustion of the oxidizer from the high-temperature gas phase that accumulated the heat of combustion of the aluminum particles and flows around the burning oxidizer particles is determined by formula

$$q = \alpha_s (T_g - T_b) + \varepsilon \sigma T_g^4. \tag{4}$$

Table 3. Thermodynamic calculations of the characteristics and the combustion product composition for ammonium nitrate–aluminum mixtures as applied to experiments in a closed volume bomb at a charging density of 85 kg/m^3

Aluminum content, wt %	TDM, g/cm^3	T, K	P, MPa	Q, MJ/kg	S, J/(kg K)	γ	Products (mol %) with a content of more than 0.3%												
							Al–OH	Al–H	Al	Al_2O	H	NO	OH	O	O_2	N_2	H_2	H_2O	Al_2O_3 (cond.)
2	1.73	1930	58	2.5	9.2E+3	1.22	–	–	–	–	–	0.3	–	–	12.9	28.5	–	57.4	0.9
8	1.77	2780	75	4.1	9.3E+3	1.18	–	–	–	–	–	1.4	1.1	–	8	28.3	–	57.3	3.7
18.3	1.84	3840	88	6.3	9.1E+3	1.14	–	–	–	–	0.9	1.5	4	0.4	1.2	27.8	5.8	48.7	9.5
30	1.93	4430	89	7.6	8.7E+3	1.12	–	–	–	–	4.7	0.7	3.3	0.3	–	25.8	23.2	24.3	15.9
47	2.07	4260	74	6.6	7.6E+3	1.12	2.9	1.4	2.0	8.1	5	–	–	–	–	22.3	37.7	1.9	17.8

Here, the first and second terms describe the contribution of convection and radiation, respectively (σ is the Stefan–Boltzmann constant and ε is the coefficient that is less than unity and responsible for the nonideality of the radiation). Finally, the heat transfer coefficient is as follows:

$$\alpha_s = \lambda_g \mathrm{Nu}/d_p, \tag{5}$$

where Nusselt number Nu and pore diameter d_p are determined by the same formulas as in the previous version of the model. Estimates using the derived formulas show that the combustion rate of ammonium nitrate can increase severalfold under the action of the heat from the burning aluminum particles.

The combustion rate of aluminum particles can be found on the basis of experimental data on the burning time of the dispersed aluminum introduced into the composition of a propellant, which is generally defined by the formula

$$t_b = d^n_{\mathrm{Al},0}/K \tag{6}$$

and the generalized Sreznevsky formula

$$d^n_{\mathrm{Al}} = d^n_{\mathrm{Al},0} - Kt. \tag{7}$$

Here, exponent n lies in a range of 1.5–2 depending on the contribution of the convective component and constant K is proportional to the concentration of oxidizing reagents in the gas: $K = C_1 Y_{\mathrm{ox}}$ [7].

Differentiating Eq. (7), for the combustion rate of aluminum particles, we finally obtain

$$u_{\mathrm{Al}} = -d_{\mathrm{Al}}/2 = \frac{C_1 Y_{\mathrm{ox}}}{2n d^{n-1}_{\mathrm{Al}}}. \tag{8}$$

The model and the initial system of first-order partial differential equations have been discussed in detail in [5]. The problem is difficult for numerical approximation and calculations because of the strong nonlinearity associated with an abrupt switching of the sources and the presence of several processes with different time and spatial scales. The initial system of equations was solved numerically using an implicit difference scheme and a matrix sweep technique.

THERMODYNAMIC PROPERTIES OF AMMONIUM NITRATE–ALUMINUM MIXTURES

Thermodynamic calculations were conducted using the TDS program (a problem with a constant internal energy and a constant volume, the equation of state of an ideal gas) [8]. The results of calculations for a loading density of 85 kg/m^3 are shown in Table 3. The table lists the theoretical maximum density of the mixture (TMD), temperature (T), pressure (P), heat of combustion (Q), entropy (S), polytropic index (γ), and the composition of the combustion products. The calculations showed the following. Although the mixture with an aluminum content of about 18.3% formally corresponds to the stoichiometric composition, the maxima of temperature, pressure, and heat of combustion are shifted to the region of rich mixtures. This feature is attributed to the fact that aluminum, which is a more active chemical element, is capable of displacing hydrogen from water vapors, the amount of which during the combustion of a stoichiometric mixture is close to 50%. This capability is exhausted only at an aluminum content of 47%; the gaseous products that appear in this mixture contain aluminum. It is only a further increase in the aluminum content in this the mixture (these data are not shown) that leads to the appearance of liquid aluminum in the combustion products.

INITIAL DATA FOR CALCULATIONS

In accordance with the experimental conditions, the calculations were conducted for charges with a length of 40 mm, an initial porosity of 53%, and four composites with different aluminum content of 8, 30, 18.3, and 47 wt %. The diameter of the ammonium nitrate particles d_{k0} was 40 μm (fine fraction) or 400 μm (coarse fraction). The thickness of the aluminum

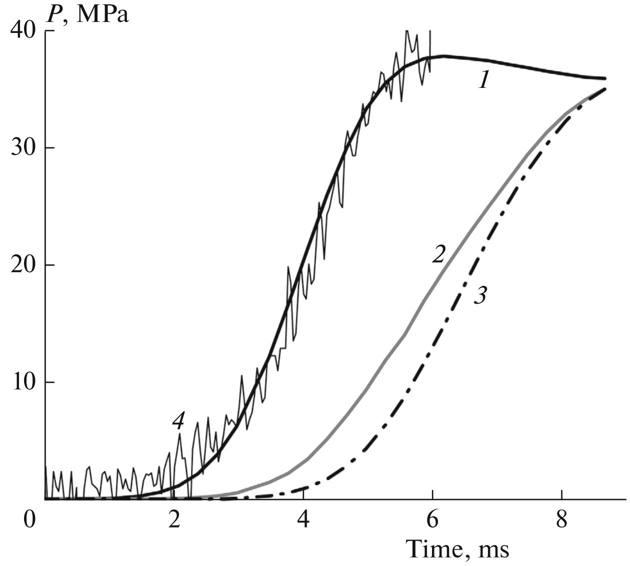

Fig. 4. Propagation of a filtration wave from the combustion of the igniter along an inert charge with a length of 40 mm at $d_0 = 40$ μm. Position of conventional pressure sensors along the charge: (*1*) 0, (*2*) 20, and (*3*) 38 mm; (*4*) experimental diagram of the chamber pressure in the region corresponding to the combustion of the igniter.

coating covering the ammonium nitrate particles (subscript AN) was calculated using the formula

$$\frac{2h}{d_{k0}} = \left[1+\frac{m_{\rm Al}}{\rho_{\rm Al}}\frac{\rho_{AN}}{m_{AN}}\right]^{1/3} - 1 \approx \frac{m_{\rm Al}\,\rho_{AN}}{3\rho_{\rm Al}\,m_{AN}}. \qquad (9)$$

It was assumed that the entire aluminum is located in the layer and no conglomerates are formed between the ammonium nitrate particles.

The data on the layer-by-layer combustion rate of ammonium nitrate differ depending on measurement conditions. The calculations were conducted at a pressure exponent of $n = 0.53$ and a rate constant corresponding to the combustion rate of 4 mm/s at a pressure of 10 MPa [9]. The temperature coefficient of the combustion rate was assumed to be $5.8 \cdot 10^{-3}$ K^{-1} [10]; heat of combustion, 1.5 MJ/kg; combustion temperature, $T_{b,\,\rm ref} = 1460$ K; ignition temperature, 800 K; and the concentration of oxidizing components in the combustion products of ammonium nitrate (oxygen and water), 0.49. Here, we took into account the recommendation given in [7], which reads that, owing to the lower activity of water, the water content in the combustion products should be multiplied by 0.6.

The diameter of the aluminum particles was taken to be 4 μm; the exponent in Eq. (8) was assumed to be 1.5. Since we failed to find experimental data on the combustion time of so small particles in the literature, the constant of $C_1 = 0.2$ cm$^{1.5}$/s was selected according to the reference point taken from [11], according to which the combustion time of an aluminum particle with a size of 10 μm in a medium with $Y_{\rm ox} = 0.1$–0.2 is about 1 ms. The ignition temperature of aluminum particles was taken to be 943 K; the heat of melting, 0.8 MJ/kg; and the heat of combustion in the decomposition products of ammonium nitrate, 17.8 MJ/kg. The empirical formula for the calculation of intergranular stresses (in MPa) was $\sigma = 200(1-\phi/0.53)^3$, the polytropic index of the gas was 1.25; the coefficients of viscosity of the gas and the specific heat capacity and thermal conductivity of both phases were assumed to be the same as in [5].

SIMULATION RESULTS

The propagation of a filtration wave across the charge under the pressure of the combustion products of the igniter of 36 MPa within 5.5 ms was simulated under the assumption that the studied porous material is inert. The simulation results are shown in Figs. 4 and 5. For particles with a size of 40 μm, the travel of the filtration wave to the closed end of the charge takes about 3 ms. By the end of the combustion of the igniter, the pressure on the closed end of the charge effectively increases. For particles with a size of 400 μm, the pressure on the face end increases simultaneously with the pressure in the volume of the bomb with a delay that does not exceed 0.5 ms at a pressure drop along the length of the charge of no more than 6 MPa. The combustion of the igniter causes the compaction of the charge from an initial porosity of 53% to a value of 38%; the compaction is quite uniformly distributed along the length of the charge.

Consider the results of simulation of the combustion process for mixtures containing ammonium nitrate with a particle size of 40 μm. If the aluminum content in the mixture and the length of the charge are maintained constant (e.g., 30% and 40 mm), while the pressure generated by the igniter is gradually increased, then three different situations will take place; they can be divided by two pressure thresholds. The first threshold provides the lower limit for the region in which the combustion products of the igniter ignite the face end of the charge; however, convective combustion does not occur. For the used input data, this threshold is ~6 MPa. At lower pressures, the mixture is not ignited. To provide the occurrence of convective combustion under the action of the igniter, the pressure generated by the igniter should exceed 20 MPa. In actual fact, the situation in the region below the second threshold can be interpreted as a layer-by-layer combustion of a porous charge.

An example of results of the numerical simulation above the convective-combustion initiation threshold for a 40-μm-long charge from a mixture containing 30% aluminum is shown in Figs. 6–8. The calculated diagram of the bomb chamber pressure is shown in Fig. 6. For comparison, the same figure shows an experimental diagram derived in experiment no. 50.

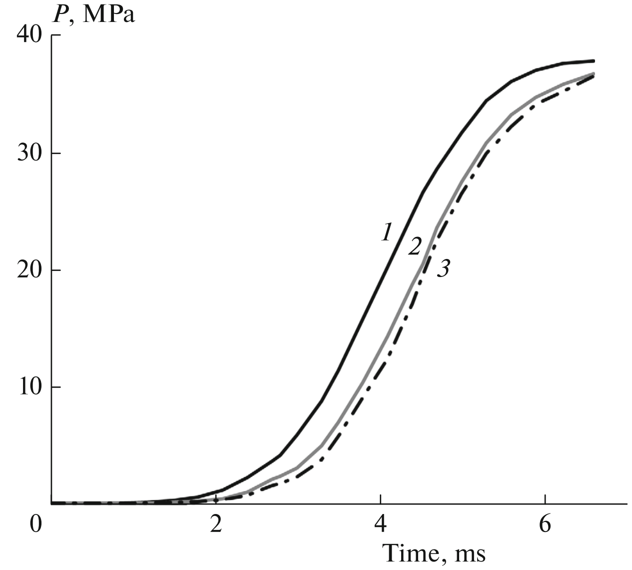

Fig. 5. Propagation of a filtration wave from the combustion of the igniter along an inert charge with a length of 40 mm at d_0 = 400 μm. Position of conventional pressure sensors along the charge: (*1*) 0, (*2*) 20, and (*3*) 38 mm.

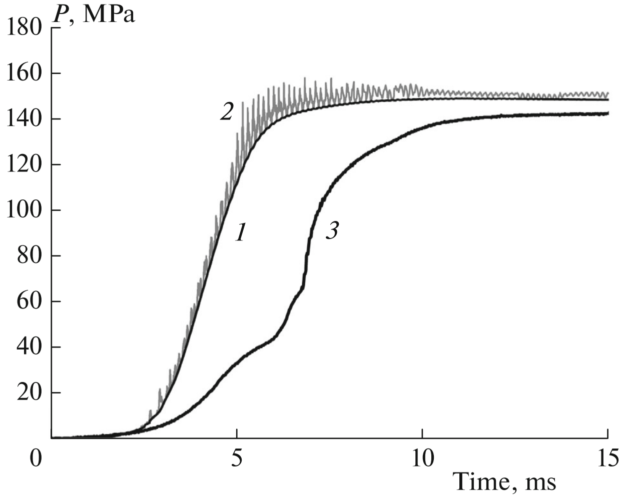

Fig. 6. Example of the calculation results for a mixture composed of 30% aluminum and ammonium nitrate with a particle size of 40 μm using a 40-mm-long charge; (*1*) calculated bomb chamber pressure, (*2*) maximum pressure in the combustion wave front, and (*3*) experimental diagram of the bomb chamber pressure (experiment no. 50).

The calculated diagram has a smoother shape; the maximum rate of pressure rise is about 60 MPa/ms; the diagram does not exhibit sharp changes in the slope, which are observed in the experimental curve; the combustion of the charge begins at a lower pressure. However, the two diagrams have almost identical maximum pressures (140–150 MPa) and approximately the same combustion time (a total combustion time of 11–13 ms at the duration of the active phase of the process of 3–3.5 ms); this fact suggests that the theory and experiment are in good qualitative agreement.

Figure 7 shows the trajectories of the ignition fronts of ammonium nitrate and aluminum for the same version of calculations. The combustion of aluminum begins with almost a millisecond delay with respect to ammonium nitrate; however, having passed a distance of about 14 mm along the charge, the fronts actually merge. In a region with a length of 10–30 mm, the ignition fronts have an approximately constant velocity of ~9.3 m/s; in the initial region with a length of up to 10 mm, the velocity is higher. This feature is attributed to the effect of compaction, which is most pronounced in the initial phase of the process and causes the displacement of the upper end of the charge. The respective velocity component is superimposed on the velocity of motion of the combustion front across the mixture particles and thus provides a total increase in the convective combustion rate. In a region with a length of more than 30 mm, the fronts are abruptly slowed down because the combustion gases fill the pores of the charge and actually equalize the gas pressure along the length of the charge before the combustion front. This process has two consequences. First, the pressure equalization hinders the inflow of new portions of the gas generated in the combustion front; accordingly, the convective combustion rate decreases in proportion to the decrease in the gas velocity. Second, the effect of intergranular stresses leads to the formation of a stable flow of the c-phase along the entire length of the charge in the direction of the bomb chamber. The result is that the unburned portion of the charge is moved into the chamber and burns out there.

The structure of the flow in the form of spatial profiles of the main variables (in a dimensionless form) in the vicinity of the combustion front at a certain instant of time in the phase of uniform convective combustion is shown in Fig. 8. The thickness of the heated zone before the combustion front does not exceed 1 mm; the maximum gas flow velocity is 34 m/s. The concentration of oxidizing reagents in the gas and the volume fraction of the c-phase, which have maximum values immediately before the flame front, rapidly decrease behind the combustion front. In this zone, the flow velocities of the gas and the c-phase also decrease and become negative; the burning particles, along with the gas, are intensively moved into the bomb chamber.

Simulation of the combustion of a charge of the same length with 47% aluminum showed that all of the basic properties of combustion remain the same as the properties of the compound containing 30% aluminum; the only difference was a lower combustion intensity. The maximum rate of pressure rise was 45 MPa/ms versus 60 MPa/ms for the mixture containing 30% aluminum; the maximum pressure was found to be 130 MPa;

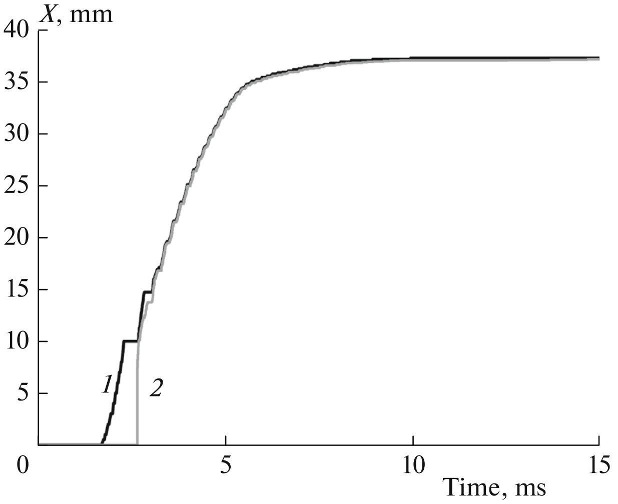

Fig. 7. Diagrams of propagation of the ignition fronts of (*1*) ammonium nitrate and (*2*) aluminum for the calculation version shown in Fig. 6. The average velocity before the beginning of an active flattening of the trajectory is 9.3 m/s.

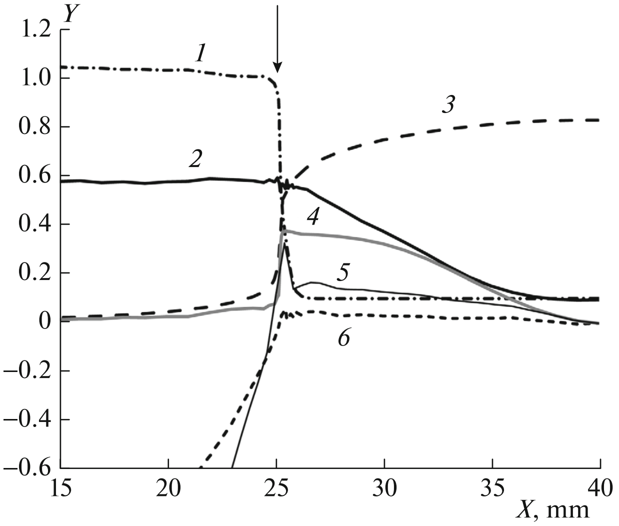

Fig. 8. Spatial profiles of the main characteristics of the flow at the time of 4.9 ms for the calculation version shown in Fig. 6. The position of the flame front at the point of 25.4 mm is indicated by an arrow; (*1*) the gas temperature of 1000 K, (*2*) the pressure of 100 MPa, (*3*) the volume fraction of the c-phase, (*4*) the content of oxidizing reagents in the gas, (*5*) the gas flow velocity of 100 m/s, and (*6*) the flow velocity of the c-phase of 100 m/s.

the convective combustion rate in the region of 10–28 mm was ~6 m/s.

Quite another process is observed for the mixture containing 18% aluminum. The ignition of the charge results in the occurrence of convective combustion, which undergoes a very rapid development. The process is illustrated in Figs. 9 and 10, where the calculation results are shown as spatial profiles of the pressure and the volume fraction of the c-phase at different instants of time separated only by a few tens of microseconds. The pressure profiles show that, as early as by the time corresponding to curve *1*, when the combustion front is at the point with a coordinate of ~12 mm, a sharp pressure peak is formed in the vicinity of this front; the pressure amplitude is more than 4 times higher than the pressure in the bomb chamber.

This significant pressure drop causes an intense outflow of the gases and the c-phase in a direction opposite to the propagation of the combustion front, which achieves sonic velocities at the outlet of the bomb chamber. However, despite these high flow velocities, the outflow is still insufficient to provide the unloading of the combustion zone, and the peak pressure amplitude continues to increase (curve *2*). The combustion wave moves across the charge at a velocity of ~100 m/s and initiates a plastic wave, which runs before the combustion front at a velocity of ~330 m/s and leads to the compaction of the c-phase to a relative density of 0.75–0.80. The mass velocity of the c-phase in the plastic wave is 70–80 m/s. The plastic wave causes the compression of the gas in the pores, which is evident in the gas pressure profiles as a small-amplitude front. This front is visible in pressure profiles *1* and *2* at points $X = 18$ and 28 mm (Figs. 9, 10). At the next instant of time (profiles *3*), the plastic wave front reaches the closed end of the charge and is reflected from it to cause a surge of gas pressure and a further increase in the bulk density of the c-phase to 0.92–0.95. These factors give rise to the formation of a powerful reverse-traveling wave with a high peak gas pressure that runs toward the combustion front (profiles *4*). The meeting of the reverse-traveling wave with the combustion front (profiles *5*) leads to the following: the peak pressure increases twofold, the motion of the combustion wave front ceases, the flow velocities of the gas and the c-phase become negative along the entire length of the charge, and the burning and nonburning particles of the mixture are discharged into the bomb chamber.

A process with these properties should apparently be regarded as an explosion. Simplifications of the model associated with the fact that the bomb chamber is simulated as a volume with homogeneous characteristics do not provide calculations of the exit of the explosion wave from the charge into the bomb chamber. In addition, these significant pressure drops will lead to the destruction of the cup giving rise to the formation of additional waves in the bomb chamber. This rapid development of the process is attributed to the high combustion rate of aluminum particles; the rate is maintained owing to the high concentration of oxidizing reagents in the gas; despite the consumption of the gas caused by the combustion of the aluminum particles, the concentration remains in the combustion zone at a level of 0.2. Recall that, in the calculations conducted for a mixture with an aluminum content of 30%, which are shown in Fig. 8, the concentration of

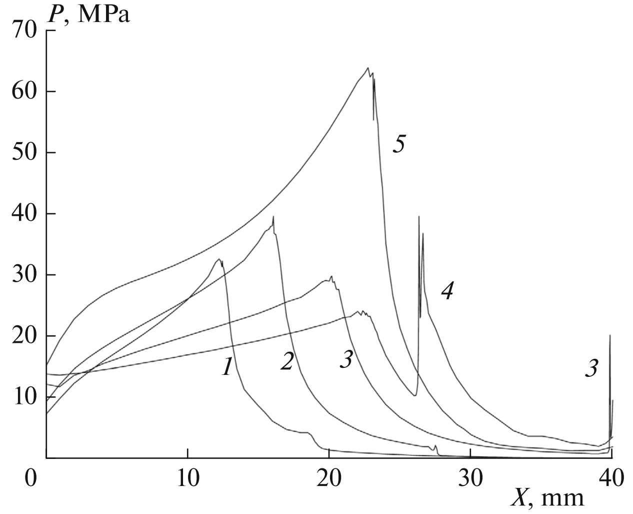

Fig. 9. Calculation results for a mixture composed of 18% aluminum and ammonium nitrate with a particle size of 40 μm using a 40-mm-long charge. Spatial profiles of pressure at the different instants of time: (*1*) 2.88, (*2*) 2.91, (*3*) 2.95, (*4*) 2.97, and (*5*) 3.01 ms.

oxidizing reagents behind the combustion front rapidly decreases to values that are 5–6 times lower than the above figure.

With a decrease in the charge length to 16 mm, the process loses some explosive properties; in particular, owing to the slower increase in pressure in the wave front, a plastic wave is not excited by the reflection from the face end. The calculated diagrams of pressure in the bomb chamber and in the flame front for this version are shown in Fig. 11. It is evident that, in the initial phase of the process, a significant pressure drop between the combustion wave front and the bomb chamber, which causes sonic velocities of the outflow of the burning aerosuspension at the outlet of the chamber, still takes place. Within a short time, the pressure in the wave front increases to 70 MPa, the velocity of the combustion front approaches 100 m/s, and the rate of pressure rise in the chamber achieves 75 MPa/ms. However, after that, with the propagation of the combustion front and the increase in the chamber pressure, the pressure drop, along with the outflow velocity, decreases. It is only a decrease in the charge length to 10 mm that leads to the combustion that is completely deprived of explosive properties and occurs in a smooth convective mode at a maximum rate of pressure rise at a level of 17 MPa/ms and a combustion front velocity of about 7 m/s.

For the mixture containing 8% aluminum, a fairly vigorous combustion takes place only for long charges (40 mm and longer). In appearance, the process is similar to the combustion that was "observed" in the calculation for the mixture with 18% aluminum and a short charge (16 mm). After the beginning of the convective combustion, the peak pressure rapidly increases to 40–50 MPa. However, the outflow achieves sonic velocities and thereby causes the unloading of the combustion zone; the amplitude of the peak, along with the pressure drop between the wave front and the bomb chamber, rapidly decreases.

Calculations for mixtures with coarse-fraction ammonium nitrate (400 μm) show the possibility of the occurrence of a quiet moderate-intensity convective combustion at all aluminum concentrations. With an increase in the aluminum content, the maximum

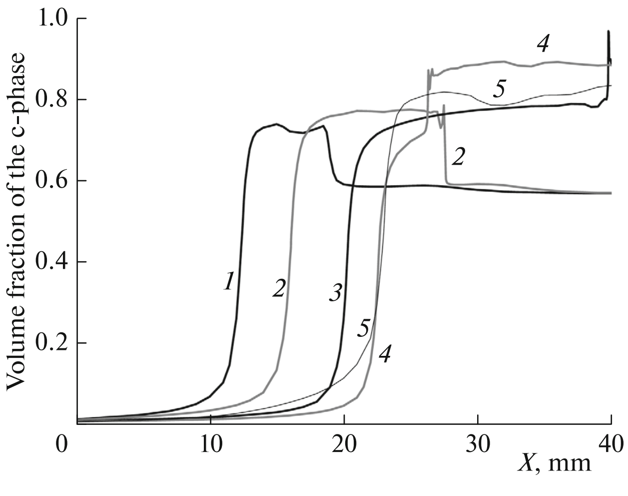

Fig. 10. Calculation results for a mixture composed of 18% aluminum and ammonium nitrate with a particle size of 40 μm using a 40-mm-long charge. Spatial profiles of the volume fraction of the c-phase at the different instants of time: (*1*) 2.88, (*2*) 2.91, (*3*) 2.95, (*4*) 2.97, and (*5*) 3.01 ms.

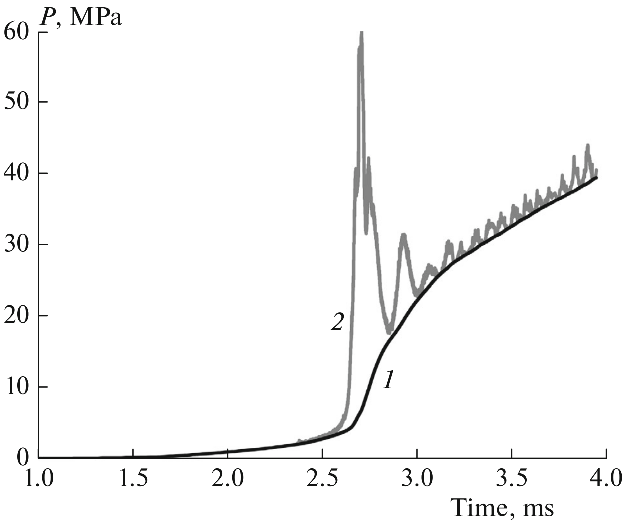

Fig. 11. Calculated diagrams of pressure (*1*) in the bomb chamber and (*2*) in the flame front for a 16-mm-long charge from a mixture of fine-fraction ammonium nitrate and 18% aluminum.

rate of pressure rise and the convective combustion rate monotonically decrease from 56 MPa/ms and 18 m/s, respectively, at 18% aluminum to 35 MPa/ms and 5 m/s, respectively, at 47% aluminum. The properties of the combustion process are close to those of the process shown in Figs. 7–9 for the mixture of fine-fraction ammonium nitrate with 30% aluminum. The main differences are as follows. The large particle size of ammonium nitrate facilitates the filtration of gases and thus leads to a higher convective combustion rate than that of the mixtures based on fine-fraction ammonium nitrate. At the same time, in the combustion zone, the rate of evolution of gaseous products of combustion of ammonium nitrate per unit volume significantly decreases. As a consequence, the combustion of the aluminum particles occurs at an extremely low concentration of oxidizing gases, which hardly achieves 1–2%. It is this concentration value that really limits the combustion rate of the aluminum particles and provides the absence of explosive events in the case of coarse-fraction ammonium nitrate.

In general, despite the qualitative agreement between the calculations and experiment for mixtures with coarse-fraction ammonium nitrate, there are some obvious differences. Thus, with an increase in the aluminum content, the combustion intensity increases in the experiments and decreases in the calculations. The discrepancy is most probably attributed to the fact that, in the case of coarse-fraction ammonium nitrate and a high aluminum content, a substantial portion of the aluminum is united in conglomerates located between the ammonium nitrate particles. Although this probability is included in the theoretical model, in view of the lack of explicit data, the calculations were conducted on the assumption that the entire aluminum is located on the surface of the ammonium nitrate particles. It is only natural that, in this case, the consumption of heat for heating and ignition is higher and the convective combustion rate decreases.

CONCLUSIONS

The combustion of loose-packed mixtures of ammonium nitrate and aluminum in a closed-volume bomb has been studied. A fine ASD-4 aluminum powder and ammonium nitrate powders of coarse (250–630 μm) and fine fractions (less than 400 μm), which were prepared by grinding and sieving the initial commercial-grade product, were used as the mixture components. The following parameters were varied in the experiments: the aluminum content in the mixture (from 8 to 47 wt %), the charge length (from 10 to 80 mm), the charge diameter (10 and 16 mm), and the pressure generated during the combustion of the igniter (from 3 to 36 MPa at a combustion time of ~5 ms). The combustion process was registered using an AVL high-frequency pressure sensor installed in the bomb chamber. The combustion intensity was estimated from the maximum rate of pressure rise. It was shown that a decrease in the ammonium nitrate particle size leads to an abrupt intensification of the combustion of mixtures with aluminum; in experiments with fine-fraction ammonium nitrate (a particle size of less than 40 μm) at an aluminum content of 18%, explosions accompanied by sharp pressure pulsations with an amplitude of a few kilobars and by the destruction of the cup containing the initial mixture were observed. Numerical simulation clarifies the details of the development of the explosive process. The explosion begins with a drastic rise in pressure in the convective combustion wave front, during which a peak with an amplitude several times exceeding the pressure in the bomb chamber is formed. The velocity of the outflow of the combustion products and the burning particles of the mixture, which is caused by this pressure drop, rapidly achieves sonic velocities; however, the combustion of the aluminum particles is dominant over the effect of unloading and the peak pressure continues to increase. The combustion wave moves along the charge at a velocity of ~100 m/s and thereby initiates a plastic wave, which runs before the combustion front at a velocity of ~330 m/s and leads to the compaction of the c-phase to a relative density of 0.75–0.80. Having reached the closed end of the charge, the plastic wave front is reflected to cause a surge of gas pressure and a further increase in the volume fraction of the c-phase to 0.90–0.93 giving rise to the formation of a powerful reverse-traveling wave with a high peak gas pressure that runs toward the combustion front. The meeting of the reverse-traveling wave with the combustion front leads to a twofold increase in the peak pressure, which achieves kilobar values, and the cessation of the combustion wave front propagation. After that, a depression wave is formed; it moves the burning and nonburning particles of the mixture and the combustion products into the bomb chamber.

An important role in the development of an explosion is played by the combustion rate of ammonium nitrate per unit volume of the mixture. In the case of fine-fraction ammonium nitrate, this value is sufficient to maintain a high concentration of oxidizing gases during the combustion of aluminum particles. In the case of coarse-fraction ammonium nitrate, the concentration of oxidizing gases decreases severalfold. Owing to this feature, the combustion rate of aluminum particles is insufficient to withstand the action of the unloading caused by the outflow of combustion products and burning particles, the peak pressure begins to decrease, and no explosion is developed.

In the experiments in which the pressure generated by the igniter and the charge length were varied, three different modes were observed for each of the studied mixtures: the absence of combustion (the mixture was not ignited because of the low amount of the igniter); a layer-by-layer combustion of the mixture, and a convective combustion with the transition into an explosion in experiments with a stoichiometric mixture.

The threshold pressures of initiation of convective combustion, which were about 20 MPa for a stoichiometric mixture with fine-fraction ammonium nitrate, decreased with an increase in the aluminum content and the sodium nitrate particle size.

Prior to our experiments, it has been assumed that ammonium nitrate–aluminum mixtures undergo smooth combustion and are significantly superior in explosion safety to similar mixtures based on ammonium perchlorate. However, once ammonium nitrate was ground to sizes typically used for ammonium perchlorate (less than 40 μm), the intensity of combustion of mixtures thereof with fine aluminum increased; in experiments with a stoichiometric mixture, there occurred explosions similar to those previously observed for mixtures with ammonium perchlorate. Although these explosions in a closed volume bomb are observed at smaller sizes of the charge and in a broader range of aluminum content, it can be stated that fine mixtures of ammonium nitrate and aluminum are also highly explosive and require appropriate attention.

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