

In conclusion, we note that the above analysis makes it possible so to vary the parameters of the process as to modify its characteristics in the required direction.

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MODEL OF ALUMINUM AGGLOMERATION DURING THE COMBUSTION OF A COMPOSITE PROPELLANT

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A distinctive characteristic of the combustion of heterogeneous metal-containing compositions is agglomeration, i.e., the enlargement of the metal particles due to coalescence as the combustion wave passes through the condensed material, as a result of which the size of the particles leaving the combustion surface is much greater than that of the starting metal particles [1, 2]. In spite of much research, we still lack an experimentally confirmed mechanism of aluminum agglomeration application to the combustion of ammonium perchlorate-based composite propellants [2].

In [3], on the basis of the experimental data, it is suggested that the agglomeration of aluminum involves the coalescence of the metal particles within cells between the oxidizer grains, leading to the formation of agglomerations with dimensions determined by the size of the cells and their aluminum content. The possibility of agglomeration as a result of the accumulation of metal particles in closed "pockets" between the oxidizer particles was previously suggested in [4], but no quantitative data on the size of the agglomerations were given. The literature does not contain any systematic quantitative data on the effect of the packing structure of the dispersed components on the size of the agglomerations formed during combustion. This led to the experimental investigation of the dependence of aluminum agglomeration on the particle size and concentration of the components in the mixture [3] and the ambient pressure [5]. The object of the present study is to give a theoretical foundation to the experimental results obtained, on the basis of a statistical examination of the packing of the dispersed components of the composition.

From a study of thin sections it was established [5] that at a relatively low volume oxidizer content the composite propellant may be regarded as a random packing consisting of hard particles (oxidizer and metal) and a continuous matrix. Since the ammonium perchlorate (AP) particles are much bigger than the aluminum particles, accumulations of metal particles, randomly dispersed in the matrix and in mutual contact, are formed in the spaces between oxidizer grains. For the compositions considered in [5] (Table 1) the cells containing the metal "clusters" are generally closed (bounded by AP particles). Figure 1 shows a thin section ob-

Novosibirsk. Translated from *Fizika Goreniya i Vzryva*, Vol. 17, No. 4, pp. 9-17, July-August, 1981. Original article submitted August 11, 1980.

TABLE 1. Characteristics of the Compositions Investigated [5]

No. of composition	β_{Al}	β_{AP}	D_{AP} , μm	\bar{v}_{AP}	No. of composition	β_{Al}	β_{AP}	D_{AP} , μm	\bar{v}_{AP}
1	0,12	0,37	50	0,4	7	0,12	0,37	220	0,70
2	0,12	0,37	75	0,3	8	0,17	0,37	280	0,16
3	0,12	0,37	110	0,25	9	0,27	0,33	75	0,30
4	0,12	0,37	200	0,18	10	0,27	0,33	110	0,25
5	0,12	0,37	280	0,16	11	0,27	0,33	200	0,18
6	0,12	0,37	360	0,16	12	0,27	0,33	220	0,70

Remark. β , volume fraction; D , mean particle diameter; and \bar{v} , coefficient of variation of the particle-size distribution.

tained from a composition with $\beta_{Al} = 0.12$, $\beta_{AP} = 0.37$ and $D_{AP} = 110 \mu m$. It should be kept in mind that in a three-dimensional packing each cell is also bounded in a direction perpendicular to the plane of the figure.

As a result of the considerable difference in geometric scale between the hard components, it is possible to study the packing of the particles successively, i.e., in studying the distribution of the AP particles we can lump the metal particles with the continuous phase, since owing to their smallness and low concentration the metal particles should not affect the packing of the much larger oxidizer grains. The next step is to consider the metal/matrix system and find the packing characteristics of the aluminum particles. According to [3], in the first step it is necessary to determine the size of the cells between oxidizer particles, and in the second the distribution of the metal particles, since contact between the particles is a necessary condition of agglomeration during the passage of the combustion wave.

Let us consider a two-phase metal/matrix system. For simplicity, we assume that the metal particles are monodisperse and spherical in shape. Then, the basic criterion determining the arrangement of the particles in the packing will be the coordination number N_c , i.e., the average number of contacts per particle. The N_c of ordered packings is given by the geometry and can easily be determined [6]. In [7] it was shown that such symmetrical packings may be regarded as particular cases of more general irregular arrangements. If as the initial coordination of the spherical particles in a random packing we take the coordination of the particles about some selected particle, then we will obtain the same coordination about any other particle within the first coordination sphere. These new coordination spheres will include the particle selected as the initial particle (located in the center of the sphere), likewise the outer particles of the first coordination sphere and, finally, the outer particles of the second coordination sphere. As a result there is formed an aggregation of ever-increasing size [7]. The only condition that must be satisfied by such aggregations is that for any point within the aggregation there exists some finite number of neighbors not greater than 14 [7].

The coordination number serves [8] as a special parameter of the radial distribution function $f(R)$ which is [7] the probability of finding particles of a random ensemble at a distance R from the center of a given particle. It is not yet possible to calculate the radial distribution function theoretically for any heterogeneous system. Only for statistical mixtures modeled by means of Poisson packings is it possible to calculate this function for the case of monodisperse spheres [8].

The compositions considered in [5] are characterized by relatively low metal contents and correspondingly small values of N_c . With such mixtures, for determining N_c it is possible to use a relation [9] derived on the assumption that the number of contacts is distributed according to Poisson's law

$$N_c = -8 \ln(1 - \beta), \quad (1)$$

where $\beta = \beta_{Al}/(\beta_{Al} + \beta_m) = \beta_{Al}/(1 - \beta_{AP})$ is the volume fraction of aluminum particles in the system, β_{Al} , β_m , β_{AP} are the volume contents of the components; the volume fractions of the heterogeneous components (AP and Al) β_i are related with the particle concentrations (number of particles per unit volume N_i) by the obvious expressions $\beta_i = (\pi/6)D_i^3 N_i$ (D_i are the particle diameters). In view of the monodispersity of the particles, N_c (1) does not depend on their size. Calculation of the average number of contacts from Eq. (1) gives $N_c = 1.7$ for

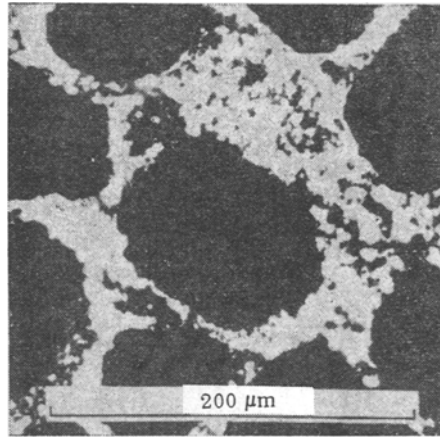


Fig. 1

compositions 1-7, $N_c = 2.5$ for composition 8, and $N_c = 4.1$ for compositions 9-12. Clearly, as the volume fraction of the particles decreases, so do the number of contacts per particle and the probability of the structure being connected, i.e., the probability that between any two particles in a two-phase system there exists a continuous (from particle to particle) path.

In [10] a critical estimate was obtained for the minimum number of contacts $N_c^* = 2$ for which the structure can still be considered completely connected. At $N_{c \min} < N_c < N_c^*$ the possibility of one or more continuous chains of particles extending from one edge of the composition to the other is preserved. At $N_c < N_{c \min}$ the probability of the existence of infinitely extended particle chains is close to zero. The relation between $N_{c \min}$ and the connectivity criterion N_c^* takes the form [10]

$$N_{c \min} = [1 - f(0)] N_c^* \quad (2)$$

where $f(0)$ is the fraction of particles not having contacts with other particles. For a Poisson particle contact probability distribution we have $f(0) \approx 0.2$ and $N_{c \min} \approx 1.6$. Thus, in the compositions investigated the aluminum particles are connected, and the necessary condition of agglomeration (contact) is satisfied.

The specimen may also be regarded as a metal/matrix system into which AP particles have been randomly introduced. For a random packing the cells differ with respect to both shape and size even when the particles are monodisperse spheres. Accordingly, the cell size distribution can be obtained only by statistical methods [11]. In accordance with the principles of stereology [12], the volume of a cell of arbitrary shape enclosed between AP particles can be found as the volume of the equivalent sphere with diameter equal to the mean free distance \bar{l} (between particle surfaces)

$$\bar{l} = 4(1 - \beta_{AP}) / S_{AP}, \quad (3)$$

where S_{AP} is the specific surface of the oxidizer powder (total particle surface per unit volume). For a system of monodisperse spherical particles ($S_{AP} = 6/D_{AP}$) relation (3) takes the form

$$\bar{l} = (2/3)(1 - \beta_{AP}) D_{AP}. \quad (4)$$

Knowing \bar{l} we can find the expected size of the agglomerations for a known cell metal content

$$D_{agg}^{cal} = [\beta_{Al} / (1 - \beta_{AP})]^{1/3} \bar{l} = A(\beta_{AP}, \beta_{Al}) D_{AP}, \quad (5)$$

where $A = (2/3) [\beta_{Al} / (1 - \beta_{AP})^2]^{1/3}$ is the proportionality factor introduced in [5]. In Fig. 2 the agglomeration diameters calculated from (3)-(5) are compared with the experimental values for the compositions investigated in [5]. The axis of abscissas corresponds to the experimental values obtained in [5] for the volume-average diameters of the agglomerations D_{agg}^{exp} , and the ordinate axis to the values of D_{agg}^{cal} . The 20% relative error limits are also indica-

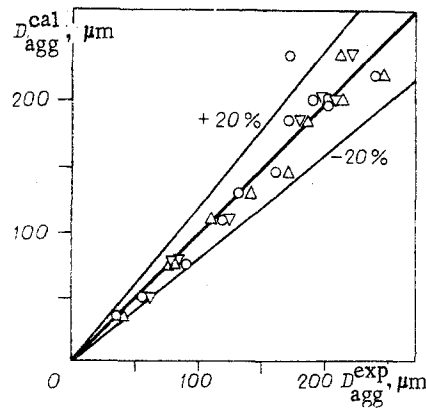


Fig. 2

ted. Clearly, there is good agreement between the experimental and calculated values for most of the investigated compositions on the pressure interval 1-40 atm.

A study of Fig. 2 leads to a number of conclusions. Firstly, a discrepancy between D_{agg}^{cal} and D_{agg}^{exp} is observed only for compositions with a low volume fraction of metal containing very coarse ($>300 \mu m$) oxidizer. The fact that the values of D_{agg}^{exp} are lower than the theoretical estimate is easy to explain if account is taken of the fact that, as noted above, for compositions 1-7 [5] the average number of contacts between particles N_c is close to $N_{c min}$. Accordingly, for these compositions, when the diameter of the AP grains is large, it is possible for more than one agglomeration to be formed within each cell. In the end this leads to a fall in the average size of the agglomerations. Secondly, with these compositions, as the pressure increases, the agreement between the experimental and the calculated data improves, which is probably associated with the more favorable conditions for particle contact in the case of specimens burning at elevated pressures owing to the rise in the temperature of the reacting surface and the more intense gasification of the matrix,

It should be noted that relation (3) for the mean cell size is based on the laws of geometric probability and does not involve any arbitrary assumptions. Accordingly, in principle, (3) is also valid for a packing consisting of a polydisperse set of particles; however, it makes it possible to obtain only the average cell size [12]. At the same time, since the experiments give the size distribution of the agglomerations, it is interesting to try to find a relation between the distribution functions for the agglomerations and the intergranular cells. One possibility for determining the sizes of the cells is to find the distribution of the distances between particles (particle centers) in the packing. The solution of the problem of the distances between random points in space is known [13]. It can be extended to the case where the random points are replaced by a system of monodisperse spheres and the distribution of these spheres in space is considered [14]. Under these conditions the mean intercenter distance is found from the expression

$$\bar{R} = 0.277(4\pi/3\beta_{AP})^{1/3} D_{AP}. \quad (6)$$

However, the applicability of expression (6) is limited to low particle concentrations. The solution becomes physically meaningless at $\beta_{AP} \geq \beta_1 = 0.0884$, since at $\beta_{AP} > \beta_1$ the particles interpenetrate. This is because in deriving (6) the limiting condition $\bar{R} \geq D_{AP}$ ($\bar{R} = D_{AP}$ is the particle contact condition) was not introduced.

Taking this into account, let us consider a packing consisting of a set of polydisperse particles. We will assume that, as a first approximation, the system of particles can be characterized by the mean dimension D and that the mean free distance is related to the mean intercenter distance by the simple dependence $\bar{l} = R - D$. Then as a basis for examining the problem we can take the distribution of the centers of the spherical particles as random points in space and assume that the number of points appearing in any region of volume V is a Poisson random quantity with mean NV [13].

If R_1, R_2, \dots, R_n are the distances from a certain fixed point to the nearest random points 1, 2, ..., n, respectively, then the function of the radial distance to the n-th nearest neighbor is written [13]

$$f_n(R_n) dR_n = \frac{3 \left(\frac{4}{3} \pi N\right)^n}{\Gamma(n, \alpha)} \exp\left(-\frac{4}{3} \pi N R_n^3\right) R_n^{3n-1} dR_n, \quad (7)$$

where α is the distribution function truncation parameter; N , number of particles (particle centers) per unit volume; $\Gamma(n, \alpha) = \int_{\alpha}^{\infty} t^{n-1} e^{-t} dt$, incomplete gamma function [15]. Here it has been

taken into account that as the lower limit of integration it is necessary to take $R_n = \bar{D}$ and not $R_n = 0$, which is manifested in the substitution of the incomplete gamma function $\Gamma(n, \alpha)$ for the function $\Gamma(n)$ [13].

Dimensional analysis suggests that $f_n(R_n)$ is a function of the single composite parameter $\lambda = NR_n^3$; accordingly, making the substitution $x = (4/3)\pi\lambda$, we rewrite (7) in the form

$$f_n(x) dx = x^{n-1} e^{-x} dx / \Gamma(n, \alpha), \quad (8)$$

After transformations, we obtain the obvious relation

$$x = R_n^3 \bar{V} N / (\bar{D})^3 = 8\beta_{AP} R_n^3 / (\bar{D})^3,$$

where $\beta_{AP} = \bar{N}\bar{V}$ is the volume fraction of AP particles in the composition; $\bar{V} = (\pi/6)\bar{D}^3$ is the mean particle volume.

As may be seen from (8), the intercenter distance distribution function has the form of a gamma distribution [15], in which the gamma function is replaced by an incomplete gamma function. When integrating (8) for the purpose of finding \bar{R} and the variance of \bar{R} it is necessary to take the quantity $x = (4/3)\pi N(\bar{D})^3 = 8\beta_{AP}(\bar{D})^3/\bar{D}^3$ as the lower limit. Computation of the moments of the function $f_n(x)$ [15] gives the values of the dimensionless (normalized to \bar{D}) mathematical expectation

$$E_n = \frac{1}{2} [\bar{D}^3 / (\beta_{AP} (\bar{D})^3)]^{1/3} \frac{\Gamma\left(n + \frac{1}{3}, \alpha\right)}{\Gamma(n, \alpha)}, \quad (9)$$

the variance

$$\sigma_n^2 = \frac{1}{4} \left[\frac{\bar{D}^3}{\beta_{AP} (\bar{D})^3} \right]^{2/3} \frac{1}{[\Gamma(n, \alpha)]} \{ \Gamma(n, \alpha) \Gamma(n + 2/3, \alpha) - [\Gamma(n + 1/3, \alpha)]^2 \}^{1/2} \quad (10)$$

and the coefficient of variation

$$\tilde{V}_n = \frac{\sigma_n}{E_n} = \left\{ \frac{\Gamma(n + 2/3, \alpha) \Gamma(n, \alpha)}{[\Gamma(n + 1/3, \alpha)]^2} - 1 \right\}^{1/2}. \quad (11)$$

It is clear from (9)-(11) that the moments of the distribution depend on the particle concentration through β_{AP} , on the particle-size distribution through $\bar{D}^3/(\bar{D})^3$, and on the ordinal number of the neighbor n , whereas the coefficient of variation depends only on n . The oxidizer fractions used in [5] had Gaussian particle-size distributions. It is easy to show [15] that in this case the condition of symmetry of the AP particle diameter distribution function leads to the third central moment of the distribution function vanishing, and we then have the relation $\bar{D}^3/(\bar{D})^3 = 3\tilde{V}_{AP}^2 + 1$, where \tilde{V}_{AP} is the coefficient of variation of the AP grain-size distribution of the corresponding functions.

The quantities $E_n(R_n)$, $\sigma_n^2(R_n)$ and $\tilde{V}_n(R_n)$ were calculated for a series of values of β_{AP} and V_{AP} . The incomplete gamma functions were calculated in accordance with standard subprograms on a BESM-6 computer for the values $n = 1, 2, \dots, 14$ ($n_{\max} = N_c^{\max} = 14$ [7]). As an example, Table 2 gives the results of the calculations for $\beta_{AP} = 0.33$ ($V_{AP} = 0; 0.3$) and $\beta_{AP} = 0.37$ ($V_{AP} = 0$). Note that $\tilde{V}_n(R_n)$ increases with increase in V_{AP} and decrease in β_{AP} . It is interesting to observe that, starting from a certain n ($n = 10$ in Table 2), \tilde{V}_n ceases to depend on V_{AP} . This indicates that the polydispersity of the oxidizer does not affect the "long-range" order in the packing [8]. Moreover, the coefficient of variation has a maximum for a value of n corresponding to the mean coordination number for a packing with a

TABLE 2. Results of Computer Calculations of the Packing Parameters

n	$\beta_{AP} = 0.33$ $\bar{D}^3 / (\bar{D})^3 = 1$		$\beta_{AP} = 0.33$ $\bar{D}^3 / (\bar{D})^3 = 1.27$		$\beta_{AP} = 0.37$ $\bar{D}^3 / (\bar{D})^3 = 1$	
	E_n	$10 \cdot \tilde{V}_n$	E_n	$10 \cdot \tilde{V}_n$	E_n	$10 \cdot \tilde{V}_n$
1	1,105	0,815	1,130	0,988	1,095	0,762
2	1,130	0,955	1,164	1,137	1,116	0,851
3	1,163	1,062	1,210	1,228	1,144	0,960
4	1,204	1,134	1,268	1,297	1,175	1,073
5	1,245	1,197	1,335	1,338	1,221	1,120
6	1,310	1,247	1,406	1,332	1,272	1,180
7	1,370	1,222	1,478	1,234	1,325	1,182
8	1,430	1,159	1,546	1,172	1,380	1,033
9	1,488	1,104	1,610	1,121	1,436	1,080
10	1,542	1,074	1,670	1,075	1,484	1,039
11	1,593	1,048	1,725	1,019	1,535	1,021
12	1,641	0,975	1,777	0,975	1,979	0,972
13	1,687	0,902	1,827	0,902	1,620	0,920
14	1,730	0,895	1,873	0,895	1,667	0,894
E_n	1,440	—	1,501	—	1,360	—

given oxidizer content β_{AP} . At the same time, there is a certain increase in the mean inter-center distance $\bar{E}_n = (1/14) \sum_{n=1}^{14} E_n(R_n)$ with increase in \tilde{V}_{AP} .

Using the properties of the moments of the functions [15], we obtain relations for the parameters of the agglomeration diameter distribution

$$\sigma(D_{agg}) = \sigma(\bar{L}) = \sigma(\bar{R}), \quad (12)$$

$$E(D_{agg}) = [\beta_{A1} / (1 - \beta_{AP})]^{1/3} \bar{L}, \quad (13)$$

$$\tilde{V}_{agg} = \frac{\sigma(D_{agg})}{E(D_{agg})} = [(1 - \beta_{AP}) / \beta_{A1}]^{1/3} \frac{\sigma(\bar{R})}{\bar{L}}. \quad (14)$$

Relations (12)-(14) make it possible to determine the mean size of the agglomerations and the width of the spectrum, starting from the packing parameters of the particles in the composition. The curves in Fig. 3 represent the calculated dependences of the coefficients of variation \tilde{V}_{agg} on \tilde{V}_{AP} for compositions with $\beta_{AP} = 0.33$ and $\beta_{AP} = 0.37$ [volume content of dispersed components in compositions: 1) 1-7, 2) 8, 3) 9-12]. The points represent the experimental values of \tilde{V}_{agg}^{exp} from [5]. The agreement between the theoretical values and the experimental values \tilde{V}_{agg}^{exp} is satisfactory. The use of broader oxidizer fractions leads to a broadening of the agglomeration size spectrum, as predicted by the theoretical model.

The values of D_{agg} for a monodisperse packing ($\tilde{V}_{AP} = 0$) calculated from the Kendall-Moran model (Eq. (13)) can be compared with the values D_{agg}^{cal} calculated in accordance with the simplified scheme (Eq. (5)). The comparison indicates that the values of D_{agg} calculated by these two different methods lie close together. Thus, the relative error $\delta = (D_{agg} - D_{agg}^{cal}) / D_{agg}$ is 9 and 12% for compositions 1-8 and 9-12, respectively. Our terms of reference did not include the question of the limits of applicability of the calculation method described; however, we assume that it can be used for determining the expected size of the agglomerations for compositions with $\beta_{AB} < 0.5$. Mathematical modeling methods [9] can be used for calculating the parameters of denser random packings of the oxidizer grains.

Our results and those obtained in [3, 5] make it possible to formulate the following model of the agglomeration of aluminum during the combustion of AP-based composite propellants. The aluminum particles in the specimen are in contact with each other. The time spent by the particles in the heated layer of the condensed phase $\tau_1 \sim \kappa / u^2 \sim 10^{-3}$ sec (κ is the thermal diffusivity) at $u \approx 10^{-2}$ m/sec considerably exceeds the time τ_2 ($\tau_2 = 10^{-4} - 10^{-5}$ sec [2]) needed for the formation of bridges between the particles capable of holding them together under the action of the gas flow. Accordingly, the passage of the combustion wave is accompanied

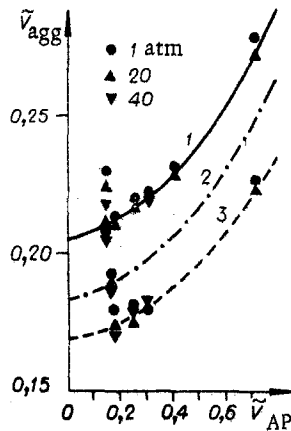


Fig. 3

by the progressive coalescence of the particles, and the rate of propagation of the thermal wave in the spaces between AP grains will be greater than the average for the specimen as a whole owing to the high thermal conductivity of the aluminum.

Initially, when the particle contact areas are small, the rate of heat transfer from the combustion surface is limited by the contact resistance of the particles [16]. At temperatures close to the melting point of aluminum, when the oxide shell starts to crack as a result of the difference between the thermal expansion coefficients for Al and Al_2O_3 [1] and "bridgelets" of molten metal are formed between the particles [2], the rate of heat transfer will be determined by the thermal conductivity of the metal. The progressive heating of the framework of aluminum particles and the matrix occupying the spaces between them results in the intense gasification of the latter. This leads to a sharp increase in capillary pressure and a corresponding increase in the tensile strength of the particle aggregations [17].

After reaching a maximum at a certain value of the liquid content of the aggregations, the strength gradually falls and is determined by the net effect of the capillary forces governing the formation of liquid "bridgelets" between particles and the capillary pressure of the liquid in the aggregation [17]. It is to be assumed that by the time the strength falls sharply due to the presence of liquid, the contact bridges will already have had time to form. The presence of capillary forces should also lower the requirements with respect to the relation $\tau_2 \ll \tau_1$. The possible oxidizing agents for Al are the AP decomposition products, and the length L of gaseous diffusion through the liquid matrix for characteristic process time τ is $(D\tau)^{1/2} \sim 10^{-5}-10^{-6}$ m; accordingly, only metal particles situated close to AP grains can be oxidized. The burnup rate is maximal at the contact boundary of the matrix/metal system [18]. This leads to the isolation of each cell, and hence the agglomeration is the product of the coalescence of all the metal particles present in a given cell.

On the basis of these results we may draw the following conclusions. The proposed model of the agglomeration of aluminum satisfactorily describes the laws of variation of the size of the agglomerations with variation of the oxidizer particle size and the content of the components of the composition, and moreover with variation of the width of the size spectrum of the oxidizer particles employed. In order to determine the mean size of the agglomerations formed during the combustion of compositions containing narrow oxidizer fractions, it is possible to use a simplified scheme for the calculation of the agglomeration diameter. Variation of the external pressure and the linear burning rate of the specimens has almost no effect on the size of the agglomerations over the investigated interval of variation of these parameters. It is to be expected that this agglomeration model will also be applicable to compositions with other nonmelting oxidizers in cases where the corresponding temperature and time conditions for the coalescence of the metal particles in the combustion wave are satisfied.

The authors wish to thank E. G. Klimova for carrying out the computer calculations.

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DYNAMICS OF GAS COMBUSTION IN A CONSTANT VOLUME IN THE
PRESENCE OF EXHAUST

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The urgency of the question of protecting closed volumes from explosion by the method of depressurization requires the production of physicomathematical models which would permit obtaining dependences of the fundamental parameters of gas mixture combustion (pressure, rate of pressure rise, temperature of the fresh mixture and the combustion products, etc.) on the time before and after the opening of the faulty areas. While the dynamics of gas combustion in a closed volume has been studied sufficiently well [1-5], models describing the combustion process after depressurization of the volume have been developed slightly and possess a number of weaknesses.

Approximate methods which do not take account of the dynamics of the development of an explosion after the beginning of the gas run-off [6, 7] are used most often to determine the areas of the faulty holes and openings. This can result in groundlessly exaggerated or dangerously reduced values of the faulty hole areas. Moreover, such methods do not afford the possibility of investigating the influence of important factors as, e.g., the level of depressurization pressure at the maximum explosion pressure. The pressure change during an explosion was studied in [8] only for the subcritical exhaust mode and in a narrow band of excess pressures ($\Delta p < 50$ kPa). A turbulization factor is introduced in [9, 10] to explain the pressure peaks experimentally observed during combustion. The assumption about the exhaust of just fresh mixture [9] and utilization of just the law of mass conservation and the adiabaticity condition [10] did not permit the authors of these papers to describe the change in pressure with time $p(t)$ in a constant volume with gas exhaust; only values were estimated. A more developed mathematical model, proposed in [11], assumes the exhaust of either the fresh mixture or of the combustion products.

The purpose of this paper is to obtain a physicomathematical model that will describe the dynamical characteristics of gas combustion in a constant volume in the presence of ex-

Balashikha. Translated from Fizika Goreniya i Vzryva, Vol. 17, No. 4, pp. 17-24, July-August, 1981. Original article submitted July 14, 1980.