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AGGLOMERATION IN COMBUSTION OF ALUMINISED SOLID PROPELLANTS WITH VARIED FORMULATION

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ABSTRACT

The paper generalises the author's experimental data on characteristics of agglomerates sampled in combustion of model solid propellants. The data were obtained at pressures 0.6÷8.5 MPa. The propellant formulations consisted of AP, HMX, RDX, Al and different type inert and energetic binders. The content of Al in the propellants varied in the range 16.8÷23%. In some formulations the commercial grade aluminium (~15 µm) was partially replaced by ultra fine one (~0.1 µm). Based on the results of particle size distribution and chemical analyses two different types (scenarios) of aluminium combustion behaviour are described with **high** and **weak** agglomeration trends, respectively. Experimental approach for objective determining the burning law for individual agglomerates in solid propellant flame is discussed.

INTRODUCTION

Using the metal as an efficient rocket propellant fuel was suggested in the eve of the rocket era [1]. Unfortunately, a potential effectiveness of metallized solid propellant could not be totally realised in practice due to the metal agglomeration, which is characterised by enlargement of initial size of aluminium particles in the combustion wave.

Agglomeration of metal particles may cause the losses of specific impulse of the rocket motor due to incomplete combustion of metal and formation of slag in the combustion chamber. Despite the relatively long history (~ 50 years) of developing metallized solid propellants the mechanism of agglomeration is not studied in detail and there is no ways to predict size distribution and combustion completeness for any new propellant formulation.

Depending on the propellant formulation and combustion conditions, the strong or weak metal agglomeration trends can be manifested. It happened that the first solid propellants (at least in Russia) exhibited strong agglomeration trends. In 1970s-1980s a number of experimental work was performed on studying the agglomeration on propellants with inert binder and first mathematical (pocket-type) models were formulated. Development of new solid propellants based on novel components and characterised by complicated geometrical structure revealed shortcomings of simple representations of agglomeration process and gave

impact for detailed studying the influence of different factors on combustion of metallized compositions.

Actually, because of the absence of detailed physico-mathematical model of agglomeration the direction of experimental research is chosen on the basis of common physical considerations. In attempt to determine the key factors affecting the agglomeration of Al particles, we performed series of experiments on combustion of aluminised propellants of variable formulation. The results of these experiments along with brief description of experimental approach are presented below. Some suggestions on specially designed experiments directed to better understanding the Al combustion behaviour are formulated for the future work.

EXPERIMENTAL APPROACH

The essence of the approach proposed is collecting practically total amount of condensed combustion products (CCP) with their subsequent detailed size distribution and chemical analyses. As a result, one obtains data on realistic size distribution of CCP that allows, in particular, getting a reliable estimate for contributions of coarse and fine particles into total CCP mass. In addition, the detailed data can be obtained on the extent of metal conversion by determining the content of unburned aluminium within given size fractions of CCP particles. Using more sophisticated chemical analysis, the data on formation of carbon and nitrogen containing compounds in CCP (for instance, aluminium nitride) can be also obtained.

To get needed experimental information the flow through bomb was designed [2] for sampling CCP in wide size range, Fig. 1. The propellant specimen commonly is about 12 mm in diameter and 8-12 mm long. It burns in plexiglas protective tube with the flame directed downward. Inner diameter of tube equals to the specimen diameter. After leaving the tube the combustion products mix with co-current inert gas (usually nitrogen) and the CCP particles are trapped by a set of metallic wire mesh screens and by an aerosol analytical filter. Using tubes of different lengths allows a variation in residence time for the CCP particles in flame environment. A few samples with total propellant mass of 3-4 g were combusted at identical pressure and generated 1-2 g of CCP. Sampled CCP particles are fractionated via wet and dry sieving and then are subjected to particle size analysis using proper method for each size range (optical microscope for coarse particles and automatic sizer Malvern-3600E for fine particles). Chemical analysis is performed for each CCP fraction to determine free (metallic) aluminium content using permanganatometric method [3]. The following characteristics of CCP were calculated for general population of sampled particles by computer code that uses the mass contribution, size distribution function, and unburned aluminium content for each fraction as input data.

Size distribution function for CCP particles, in the form of histograms of the relative mass of CCP, $f_i(D) = m_i / (m_p \cdot \Delta D_i)$, where m_i is the mass of CCP in the i -th size interval, m_p is the total mass of propellant burned, ΔD_i is the width of i -th histogram size interval.

Size distribution function for aluminium in CCP, $f_i^{Al}(D) = f_i(D) \cdot \varepsilon_j^{Al}$, where $f_i(D)$ is the histogram of relative mass of CCP, ε_j^{Al} is the mass content of aluminium in the j -th sieve fraction which contains i -th size range.

Mean sizes D_{mn} , calculated by formula $D_{mn} = \sqrt[m-n]{\left(\sum_{i=1}^k D_i^m \cdot N_i \right) / \left(\sum_{i=1}^k D_i^n \cdot N_i \right)}$, where D_i is the midrange and N_i is the number of particles in i -th size range.

When treating the experimental data, it is convenient to divide sampled CCP onto “coarse”, i. e. agglomerate particles, and “fine” (smoke), i. e. oxide particles. Based on the own experience and the literature data, the boundary between those particle fractions can be conventionally defined as 60 μm , approximately. This usually corresponds to the local minimum on the curve $f(D)$ of the total size distribution. However, sometimes there is no distinctive minimum in this range of particle sizes and it becomes useful to examine data on aluminium content in different particle size fractions of CCP. In this case the local minimum of function $f^{Al}(D)$ may give a value of the boundary size. The particles with size exceeding the boundary one are called agglomerates.

The following *dimensionless parameters of CCP* are used below. These parameters were calculated for fine and agglomerate particles on the basis of experimental mass size distribution functions $f(D)$ and $f^{Al}(D)$. The parameters are scaled by the total mass of propellant burned, M_{prop} (for example, $m_{\text{ag}} = M_{\text{ag}}/M_{\text{prop}}$, where M_{ag} is the mass of agglomerate particles).

m_{ag} – dimensionless mass of agglomerates, m_{f} – dimensionless mass of fine particles,

$m_{\text{CCP}} = m_{\text{f}} + m_{\text{ag}}$ – total dimensionless mass of CCP,

$m_{\text{ag}}/m_{\text{CCP}}$ – relative mass fraction of agglomerates in CCP,

m_{ag}^{Al} – dimensionless mass of free aluminium in agglomerates.

EXPERIMENTAL RESULTS

The combustion experiments were conducted with a number of propellants based on inert or energetic binder and containing AP, HMX, RDX, and Al. As mentioned above, each firing run generates data on the properties of CCP formed in combustion of particular propellant upon given conditions. Generalised map of experimental results for propellants of varied formulation is shown in Fig. 2. Main features of studied propellant formulations are presented in Table 1.

In co-ordinates $\{D_{30}\}$; $\{m_{\text{ag}}/m_{\text{CCP}}\}$ the domain in the left bottom edge corresponds to the “weak” agglomeration trend whereas the opposite corner corresponds to the “strong” agglomeration trend. Qualitatively, weak agglomeration trend is characterised by a relatively small mean size of agglomerates and their small contribution to the total mass of CCP. The opposite trend is realised in the case of strong agglomeration.

Visual observations accomplished by high-speed shadow photography under flush illumination of a xenon lamp revealed at low pressures some specific features of the propellant combustion behaviour (Fig. 3). In the case of weak agglomeration the numerous relatively small (sometimes non-burning) particles are recorded above the burning surface and only rarely large coral shape aggregates appear on the surface. The latter can be ignited from the top and then leave the burning surface. Usually, for “weakly” agglomerating propellants there is no distinctive boundary between the fine and coarse particles in total particle size distribution. In the case of strong agglomeration the round shape agglomerate may start burning within the propellant surface layer with formation of typical “halo” and smoke tail. In addition, sometimes the ejected crystals of oxidiser are recorded in the gas zone.

The combustion behaviour of studied propellants can be assigned to the weak or strong agglomeration scenarios, see Table 2. This assignment is conventional because characteristic parameter values are overlapping. All data reported in Fig. 2 and Tables 1 and 2 were obtained in experiments without protective tube (i. e. with freezing the particles at close

distance from the burning surface) in the pressure range 0.6-8.5 MPa. Most propellant formulations are based on heterogeneous components (Al, AP, HMX, RDX) with ordinary size of grains, see Table 1. In the propellant formulations EM1, EM2, EM3 the ultra fine AP designated as "UFAP" was used. In the propellant formulations A1, A2, A3 and AN1, AN2, AN3 the ordinary Al was partially replaced by ultra-fine aluminium powder "Alex", with the ratio (Alex/Al) being varied. Other abbreviations are: DEGDN - diethylene glycol dinitrate, TO - transformer oil, FO - ferrocene oil.

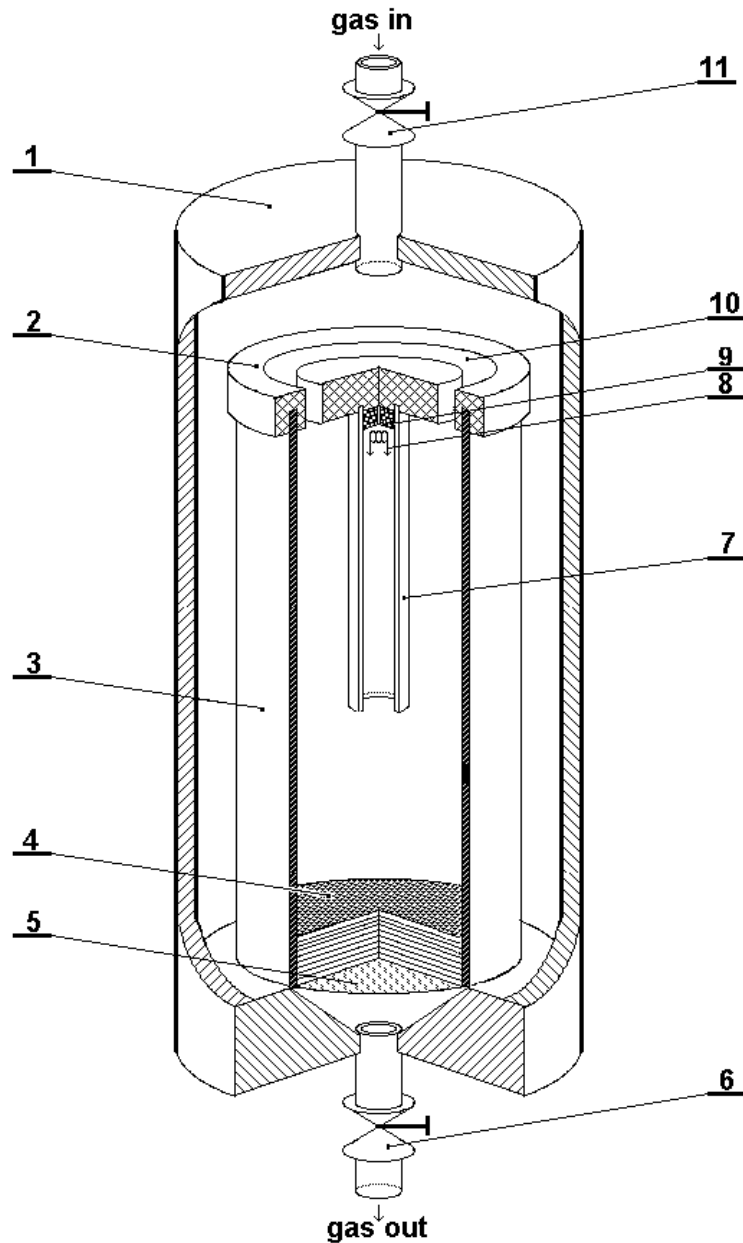


Fig. 1. Chart of the bomb for CCP sampling:

- | | |
|----------------------------------|--|
| 1 - bomb body, | 7 - protective tube, |
| 2 - top cover of the cylinder 3, | 8 - ignition wire, |
| 3 - thin wall cylinder, | 9 - propellant specimen, |
| 4 - stack of wire mesh screens, | 10 - ring slot for blowing the cylinder 3, |
| 5 - filter, | 11 - gas inlet valve. |
| 6 - gas outlet valve, | |

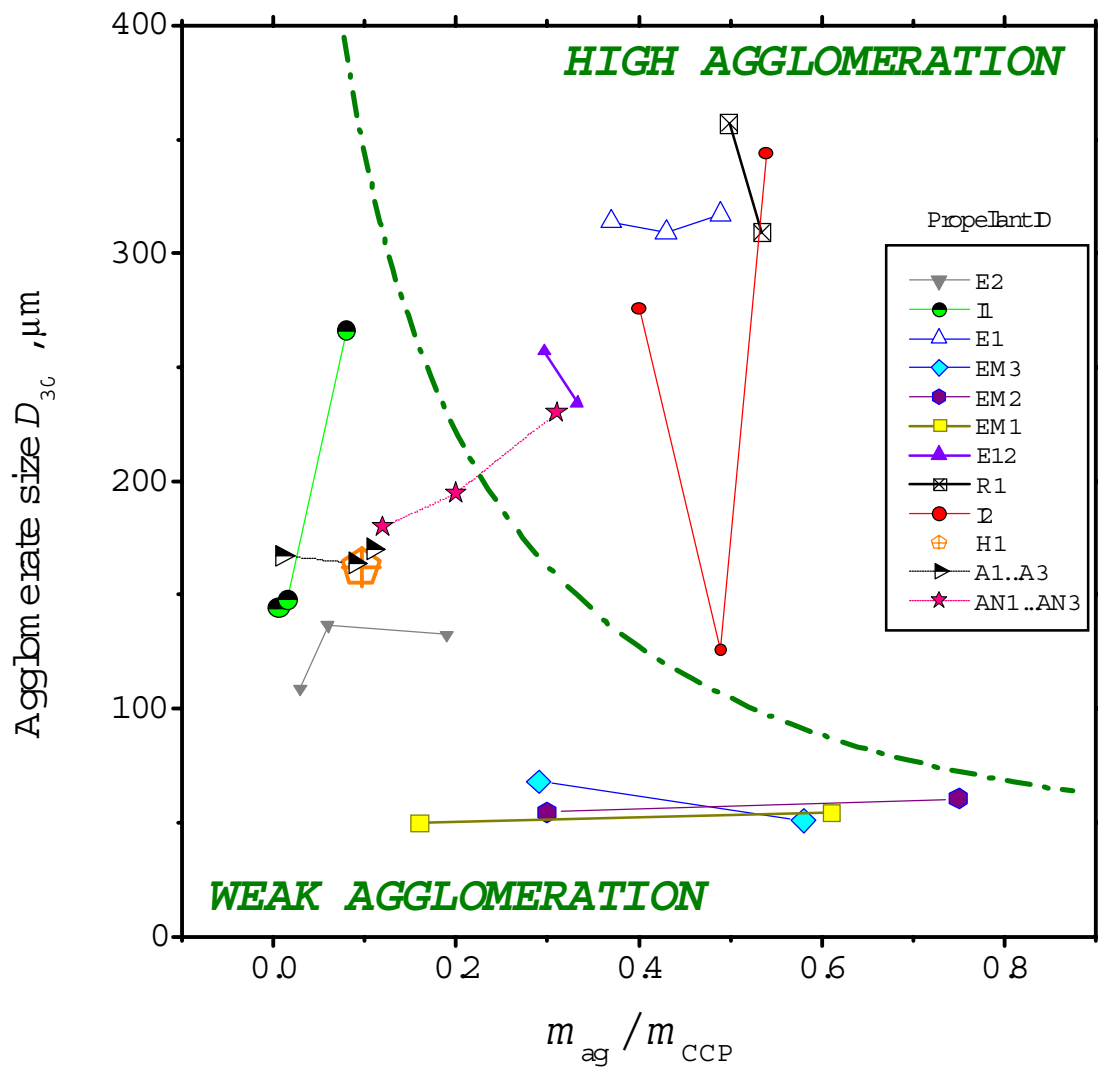


Fig. 2. Map of agglomeration trends.

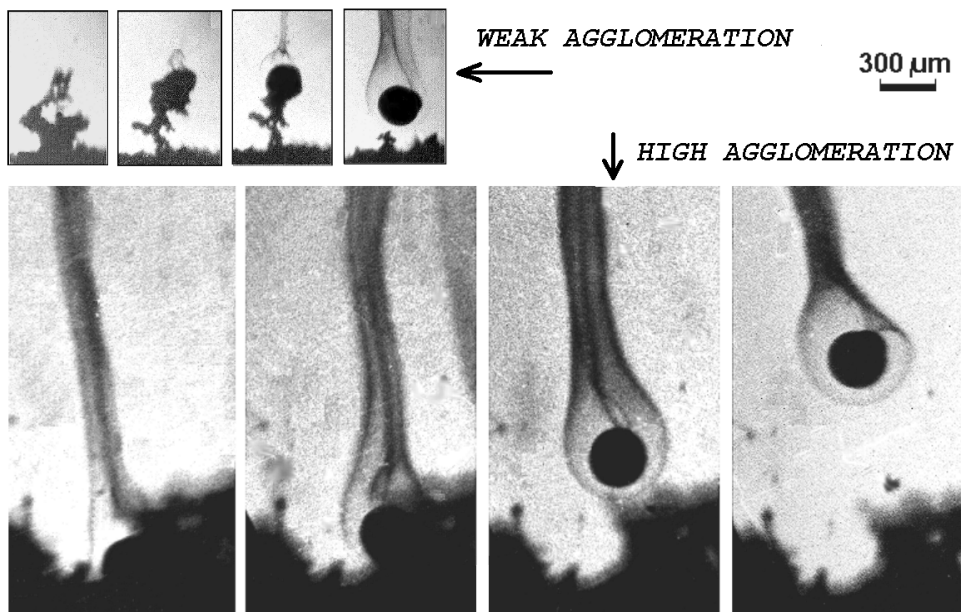


Fig. 3. High speed photography of weak and high agglomeration, 2000 f/s, 1 atm, identical scale.

Table 1. Propellant formulations (% mass) corresponding to curves in Fig. 1.

Propt ID	Main ingredients	Tested pressures P , MPa; Burning rate r , mm/s
E2	18% Al: $D_{30} = 14 \mu\text{m}$ 30% binder: buradiene-nitryl/DEGDN 15% AP: $S=6000 \text{ cm}^2/\text{g}$ 37% AP: 200-315 μm	$P = 2.2, 4.4, 8.5$; $r = 6.12(P \cdot 10)^{0.24}$ $r(4.4) = 16 \text{ mm/s}$
I1	16.8% Al: $D_{30} = 14 \mu\text{m}$ 15% binder: butadiene/TO 31.8% AP: $S=6000 \text{ cm}^2/\text{g}$ 13.1% AP: 200-315 μm 23.3% HMX: 315-1000 μm	$P = 0.6, 2.2, 6.4$; $r = 1.13(P \cdot 10)^{0.65}$ $r(4.4) = 13 \text{ mm/s}$
E1	20% Al: $D_{30} = 14 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 7% AP: $S=6000 \text{ cm}^2/\text{g}$ 18% AP: 200-315 μm 35% HMX: 315-1000 μm	$P = 0.6, 2.2, 6.2$; $r = 1.88(P \cdot 10)^{0.52}$ $r(4.4) = 13 \text{ mm/s}$
E12	20% Al: $D_{30} = 14 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 7% AP: $S=6000 \text{ cm}^2/\text{g}$ 18% AP: 200-315 μm 35% HMX: 250-700 μm	$P = 2.4, 6.6$; $r = 1.88(P \cdot 10)^{0.45}$ $r(4.4) = 10 \text{ mm/s}$
R1	20% Al: $D_{30} = 14 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 7% AP: $S=6000 \text{ cm}^2/\text{g}$ 18% AP: 200-315 μm 35% RDX: 315-1000 μm	$P = 2.4, 6.5$; $r = 0.88(P \cdot 10)^{0.62}$ $r(4.4) = 9 \text{ mm/s}$
EM1	22% Al: $D_{30} = 14 \mu\text{m}$ 14.9% binder: isoprene/FO/TO 37.1% UFAP: $D < 1 \mu\text{m}$ 26% HMX: 160-315 μm	$P = 0.6, 4.1$; $r(4.1) = 63 \text{ mm/s}$
EM2	23% Al: $D_{30} = 14 \mu\text{m}$ 15.2% binder: isoprene/FO/TO 37.8% UFAP: $D < 1 \mu\text{m}$ 24% AP: $S=2700 \text{ cm}^2/\text{g}$	$P = 0.6, 4.1$; $r(4.1) = 60 \text{ mm/s}$
EM3	23% Al: $D_{30} = 14 \mu\text{m}$ 15.2% binder: isoprene/FO/TO 37.8% UFAP: $D < 1 \mu\text{m}$ 24% AP: 200-315 μm	$P = 0.6, 4.1$; $r(4.1) = 63 \text{ mm/s}$
I2	18% Al: $D_{30} = 14 \mu\text{m}$ 18% binder: isoprene/TO/catalyst 27% AP: $S=6000 \text{ cm}^2/\text{g}$ 37% AP: 200-315 μm	$P = 2.2, 4.3, 8.5$ $r = 5.21(P \cdot 10)^{0.21}$ $r(4.3) = 11 \text{ mm/s}$

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Table 1 (continued). Propellant formulations (% mass) corresponding to curves in Fig. 1.

Propt ID	Main ingredients	Tested pressures P , MPa; Burning rate r , mm/s
H1	20% Al: $D_{30} = 6 \mu\text{m}$ 15% binder: HTPB 26% AP: $S=6700 \text{ cm}^2/\text{g}$ 39% AP: 160-315 μm	$P = 4.4$; $r(4.4) = 6 \text{ mm/s}$
A1	18% Alex: $D \sim 0.1 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 21% AP: $S=6700 \text{ cm}^2/\text{g}$ 41% AP: 160-315 μm	$P = 4.6$; $r(4.6) = 290 \text{ mm/s}$
A2	5.4% Alex: $D \sim 0.1 \mu\text{m}$ 12.6% Al: $D_{30} = 6 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 21% AP: $S=6700 \text{ cm}^2/\text{g}$ 41% AP: 160-315 μm	$P = 4.4$; $r(4.4) = 60 \text{ mm/s}$
A3	18% Al: $D_{30} = 6 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 21% AP: $S=6700 \text{ cm}^2/\text{g}$ 41% AP: 160-315 μm	$P = 4.6$; $r(4.6) = 48 \text{ mm/s}$
AN1	5.4% Alex: $D \sim 0.1 \mu\text{m}$ 12.6% Al: $D_{30} = 6 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 9% AP: $S=6700 \text{ cm}^2/\text{g}$ 18% AP: 160-315 μm 35% HMX: 250-1000 μm	$P = 4.6$; $r(4.6) = 46 \text{ mm/s}$
AN2	1.5% Alex: $D \sim 0.1 \mu\text{m}$ 16.5% Al: $D_{30} = 6 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 9% AP: $S=6700 \text{ cm}^2/\text{g}$ 18% AP: 160-315 μm 35% HMX: 250-1000 μm	$P = 4.6$; $r(4.6) = 35 \text{ mm/s}$
AN3	18% Al: $D_{30} = 6 \mu\text{m}$ 20% binder: buradiene-nitryl/DEGDN 9% AP: $S=6700 \text{ cm}^2/\text{g}$ 18% AP: 160-315 μm 35% HMX: 250-1000 μm	$P = 4.6$; $r(4.6) = 22 \text{ mm/s}$

DISCUSSION

Analysis of the results presented in Fig 2 and Tables 1 and 2 shows that there is no simple correlation between the propellant formulation and the agglomeration trend observed. It is known that the agglomeration phenomenon is the function on multiple factors. They include the burning rate, or residence time, geometrical structure of the propellant, physico-chemical properties of the components, heating rate and heat exchange in the combustion wave, etc. Therefore, it is a difficult technical task to conduct experiments for revealing the role of a chosen fixed factor. To illustrate, one may examine the results of experiments [4, 5]

corresponding to the lines 1 and 4 in Table 2. The propellants tested I2 and E2 were identical in terms of the mass content of coarse AP particles (identical geometrical structure) and had close in value the burning rates. In experiments, the propellant I2 with inert binder exhibited higher agglomeration trend than the propellant E2 with energetic binder. However, the propellants with the same energetic binder manifested strong agglomeration trend when part of AP was replaced with HMX or RDX (Table 2, lines 2 and 3, propellants E1, E12, AN3 and R1). Note, that the effect was more pronounced in the case of RDX.

Table 2. Characteristic parameters of agglomeration scenarios and types of propellant formulation

No	Propellant ID	HIGH AGGLOMERATION TREND $m_{ag}/m_{CCP}=0.3\div 0.54, D_{30}=130\div 430 \mu\text{m}$	Nitramine	Reference
1	I2	isoprene rubber +TO + catalyst	-	[4, 5]
2	E1, E12, AN3	buradiene-nitril rubber +DEGDN	HMX	[4, 5, 6]
3	R1	buradiene-nitril rubber +DEGDN	RDX	present work
		WEAK AGGLOMERATION TREND $m_{ag}/m_{CCP} =0.01\div 0.20, D_{30}=140\div 270 \mu\text{m}$	Nitramine	Reference
4	E2	buradiene-nitril rubber +DEGDN	-	[4, 5]
5	I1	buradiene rubber +TO	HMX	[4, 5]
6	AN2	buradiene-nitril rubber +DEGDN, Alex	HMX	[6]
7	H1	□□□□	-	present work
		Weak agglomeration, high-burning-rate propellants* $m_{ag}/m_{CCP} =0.01\div 0.30, D_{30}=50\div 230 \mu\text{m}$	Nitramine	Reference
8	EM1	isoprene rubber +TO + FO + UFAP	HMX	[7, 8]
9	EM2, EM3	isoprene rubber +TO + FO + UFAP	-	[7, 8]
10	AN1	buradiene-nitril rubber +DEGDN, Alex	HMX	[6]
11	A1, A2, A3	buradiene-nitril rubber +DEGDN, Alex	-	[6]

*) High-burning-rate propellants conventionally have $r > 40$ mm/s at $P = 4$ MPa.

Another illustration comes from experiments with the earliest version of solid propellants based on the butyl rubber [9]. The propellants based on the butyl rubber binder cured with paraquinon dioxim demonstrated strong agglomeration trend but those based on the same binder cured with quinol ester exhibited weak agglomeration trend. Thus, the above examples show that an interaction of different unknown factors may play a role in developing the agglomeration trends.

One may suppose that agglomeration process takes special conditions for merging and sticking together the virgin Al particles that implies finite residence time on the burning surface and sufficient heat feedback from the gas phase. In addition, the physical state and properties of decomposed binder as well as a presence of the oxidising gases in the pocket volume may play significant role. Probably, the most trivial is the effect of the residence time. For fast burning propellants the residence time becomes too short for creating the bridges between the Al particles while they are keeping on the burning surface. Replacement of part of AP by nitramines usually leads to slight decrease in the burning rate and significantly lowers the heat feedback from the gas phase due to smaller reactivity of nitramine decomposition products. It can be a possible cause of strengthening the agglomeration trend for formulations containing nitramines. Another example of the burning rate effect gives

examination of the combustion behaviour of propellants AN3, AN2, AN1 (Table 2, lines 2, 6, 10; Al/AI ratio equals 0/100, 8.3/91.7, 30/70, correspondingly). It is seen that increase in the burning rate results for the propellants with given formulation in transition from high to weak agglomeration trend.

There is a very little known about the properties of decomposed polymers at the burning surface temperatures. The attempt of studying the effect of binder decomposition characteristics on Al agglomeration was undertaken in [10]. The data on thermal behaviour of 3 different (2 inert and 1 energetic) binders at 50 K/min heating rate were obtained. Some interesting observations on liquefying the polymer and interaction with AP grains were made. Qualitative conclusion has been drawn on essential influence of the binder nature on the character of metallized propellant burning. However, there is not sufficient justification for using the data at low heating rate to explain the combustion behaviour characterised by much higher heating rate (ca. 500 K/s and more).

Obviously, for better understanding the agglomeration phenomenon the behaviour of polymer material under fast heating rates has to be explored. For this end in our laboratory the technique for preparing pure and loaded with heterogeneous components thin (100-150 μm) films of polymer has been elaborated and some preliminary experiments were conducted. The heating rate of 500-600 K/s was achieved and temperature of softening for several polymer materials was determined. It should be mentioned that a number of binder formulations include inert or energetic plasticisers whose content may amount up to 70-80% by mass. Upon heating such binders, the first stage usually involves evaporation and/or decomposition of plasticizer with subsequent degradation of polymer matrix.

Evolution of individual agglomerates

The difficulties in studying the agglomeration in combustion of metallized propellants are caused by the great variety of dimension and residence time for agglomerates and by uncertainty of the combustion conditions (variation of local temperature and concentration profiles). An important role in developing the mechanism of Al combustion in the solid propellant flame belongs to the experiments with specially designed propellant formulations that provide generation of approximately equal size agglomerates burning under well-defined conditions.

In accordance with original idea [11] the propellant under study consists of non-metallized matrix filled with a finite number of heavy metallized macro inclusions. The matrix can be treated as homogenised material because only small grain size ingredients ($D_{30} < 7 \mu\text{m}$ for all) are used for its manufacturing. Use of homogenised matrix provides uniformly distributed and clearly specified gas environment for combustion of Al particles. Besides, by varying the formulation of matrix one may vary the content of gas oxidising species, which can be estimated by an equilibrium code.

The material for inserted macro elements designated as metallized inclusions (MI) was intentionally chosen to be similar to the pocket matter in metallized propellant, i. e. it closely corresponds to local composition in space between coarse oxidiser grains in real propellant. In particular, the material for MI includes fine AP. The composition of the propellant components is presented in Table 3. The energetic binder is based on butadiene nitril rubber plastisized with DEGDN. The MI elements were beforehand prepared by cutting the cured heavy metallized propellant. When specimen burns, each MI transforms in the combustion wave into single agglomerate whose size is determined by the initial size and composition of

macro element (i. e. MI). Fortunately, the fine AP facilitates an ignition of MI in a very effective way so that most part of agglomerates is ignited on the burning surface. It should be underlined that the better uniformity of initial MI, the more unimodal agglomerates are formed during the combustion of specimen.

Table 3
Matrix and Inclusion Formulations

Ingredient, % mass.	Matrix	MI
Energetic binder	27	42.6
Al ($D_{30}=6 \mu\text{m}$)		42.6
AP ($S=6700 \text{ cm}^2/\text{g}$)	34	14.8
HMX ($D_{30}=3.5 \mu\text{m}$)	39	-

In preliminary experiments [12, 13], each propellant specimen for sampling test included carefully selected and counted MIs (usually 60÷120 in number), i. e. "agglomerate seeds", having cubic shape with size of rib about 500 μm and weight about 0.2 mg each. Summary mass fraction of MI in the propellant specimen was 1.2÷3% that meant that the composition of the gaseous combustion products was not practically changed as a result of the agglomerate combustion. To guarantee the ignition of each MIs, they were placed in the matrix bulk no closer than 1 mm from the sample holder cup walls and bottom and from the open butt-end of specimen. The residence time for agglomerate in flame is calculated as the time of motion from starting point in the bulk of matrix to the cone surface corresponding to the mixing of combustion products with environmental inert gas, see Fig. 4.

The experimental data obtained has been treated from the point of view of the metal consumption kinetics that is important for calculation of total heat release in the propellant combustion. Experimentally determined mass of unburned aluminium in sampled agglomerates, $m_{\text{ag}}^{\text{Al}}$, gives a measure for the metal combustion incompleteness: $\eta = m_{\text{ag}}^{\text{Al}}/m_0^{\text{Al}}$. Here m_0^{Al} is the initial mass of aluminium in MI. In accordance with the basic idea the agglomerate size is constant and only pressure and agglomerate residence time may vary the η -value. Analysis of experimental data allowed us to construct the following approximate expression for η in dependence of t and P : $\eta(t, P) = 2.86 \cdot t^{-0.28} \cdot P^{-0.20}$, where $20 < t < 90$ ms, $10 < P < 64$ atm, $D = 400\text{--}540 \mu\text{m}$ (Fig. 5). The 3D-plot in Fig. 5 demonstrates that function η monotonously decreases with both the residence time and pressure. This empirical fact permits to characterise the Al conversion degree unambiguously by t or P (in the range of parameters under study).

For validating the agglomerate combustion model it is important to know how the oxide mass is distributed between the agglomerates and "smoke". By processing the experimental data we determined the dependence (Fig. 6) of the mass fraction of oxide accumulated on agglomerate, $\varphi = m_{\text{ox}}^{\text{ag}} / m_{\text{ox}}^{\text{exp}}$, on the metal combustion incompleteness. Here $m_{\text{ox}}^{\text{ag}}$ is the measured mass of oxide in agglomerate and $m_{\text{ox}}^{\text{exp}}$ is the expected (calculated) mass of oxide forming in the course of the combustion of aluminium mass consumed. This dependence has a form $\varphi = 0.54 + 0.21 (1-\eta)$ that means that a higher extent of conversion of metal corresponds to a higher mass fraction of Al oxide accumulated on the burning agglomerate.

In addition, the dependence of the dimensionless agglomerate mass m/m_0 on the extent of aluminium conversion has been determined in the form: $m/m_0 = 0.46 + 0.18 (1-\eta)$. It shows (Fig. 7) that the mass of the burning agglomerate increases with Al conversion extent. Note that this dependence, when extrapolated to the minimal degree of the metal conversion ($1-\eta = 0$), gives the value $m/m_0=0.46$ which is close in magnitude to initial aluminium content in the material used for preparing MIs (42.6%). In other words, the agglomerate mass evolution starts from $m=0.46 \cdot m_0$ but not from $m=m_0$. Consequently, one may expect that original MIs

are subjected in the combustion wave first to pyrolysis of binder and AP particles (similar to the process of volatile components release from heated coal) and then aluminium particles start to merge, melt and ignite.

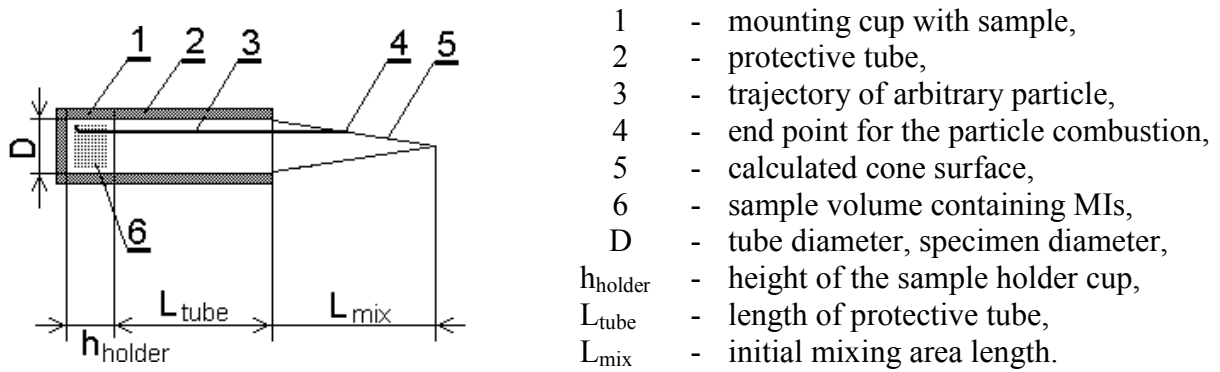


Fig. 4. Firing test geometry.

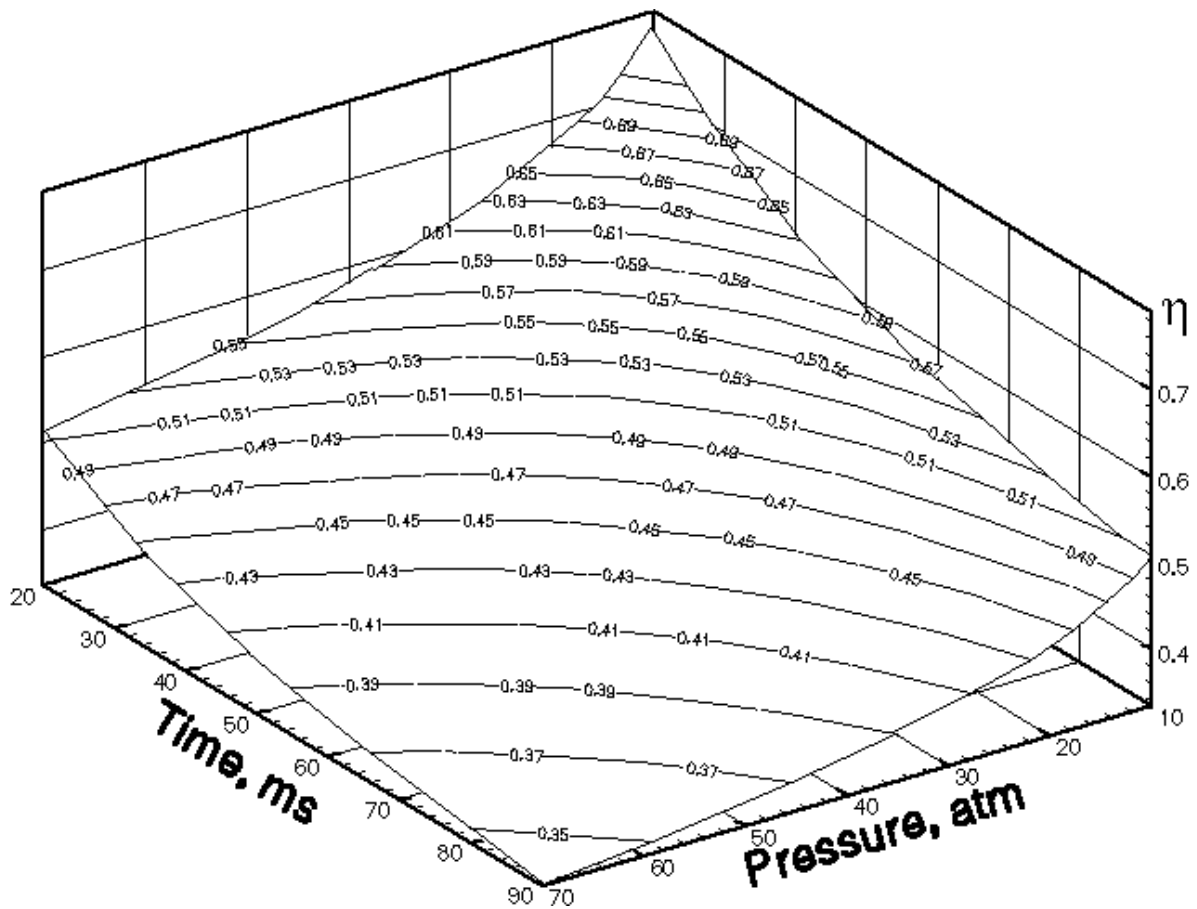


Fig. 5. Dependence of incompleteness of aluminium combustion η on pressure and residence time: 3D representation of matching function $\eta(t, P) = 2.86 \cdot t^{0.28} \cdot P^{-0.20}$.

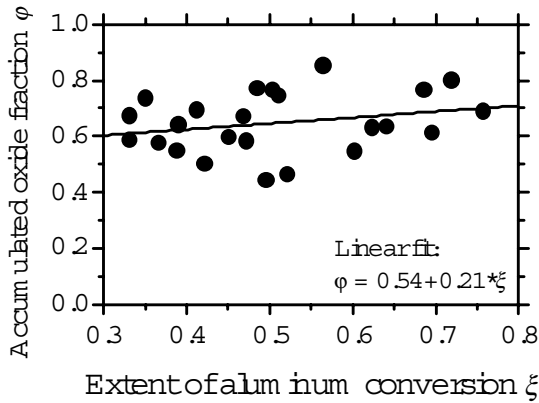


Fig. 6. Mass fraction of oxide accumulated on the burning agglomerate *versus* extent of aluminium conversion $\xi = (1-\eta)$.

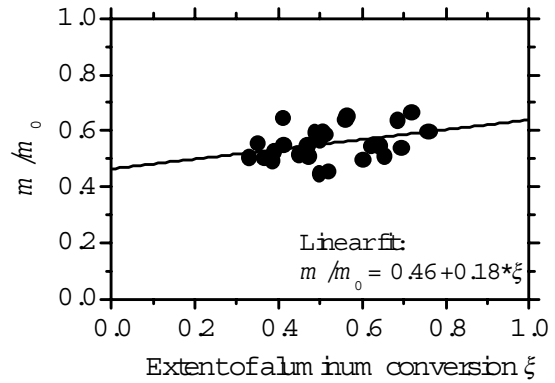


Fig. 7. Agglomerate mass *versus* extent of aluminium conversion $\xi = (1-\eta)$.

CONCLUSIONS AND FUTURE PLANS

The results of this work show that the experimental approach based on detailed examination of practically total amount of condensed combustion products of metallized propellant provides an effective tool for studying common features of Al combustion in the propellant flame. Some useful information on the effects of the combustion conditions and propellant formulation on the Al combustion behaviour has been obtained and generalised. In particular, the role of nitramines in strengthening the agglomeration trends has been revealed, the dependencies of the mean agglomerate size and metal conversion degree on estimated residence time in the propellant flame have been explored for different combustion conditions and propellant formulations.

At the same time the analysis of experimental data available shows a shortage of information on specific conditions of Al combustion in the propellant flame as well as on physico-chemical properties of the propellant components under the burning surface temperature. In fact, there is no realistic information on the local values of the burning surface temperature and heat feedback from the gas phase, on the residence time for given size agglomerate within the subsurface layer and in the propellant flame, on the agglomerate burning law and evolution of the metal oxide particles. This means that specially designed experiments have to be planned and conducted in order to get insight into intrinsic mechanism of Al combustion behaviour.

It has to be mentioned that the results of studying the combustion of the metal drops in air or another gas media give some complementary information which cannot be entirely applied to the analysis of the real agglomerates (aggregates of sintered individual particles) combustion in the flame of solid propellant [14]. The first attempts of studying the combustion of individual agglomerates in the propellant flame and decomposition of polymer films have been undertaken in our laboratory. It is planning in the nearest future to continue work on studying the combustion behaviour of “super agglomerates” (heavy metallized insertions within a non-metallized propellant matrix) as well as the decomposition and ignition behaviour of thin films of pure and particle loaded binder. The goal of the future research is getting information on different size individual agglomerate behaviour in the flame of propellant of varied formulation and on thermal decomposition behaviour of different types of polymers (pure and mixed with heterogeneous components) under fast heating rates and relatively high temperatures.

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