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PRINCIPAL INVESTIGATOR: **VALERY A. BABUK**

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PERSONNEL,
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**VALERY A. BABUK,
PRINCIPAL INVESTIGATOR**

**VYACHESLAV A. VASILYEV,
SENIOR SCIENTIST**

**VITALY V. SVIRIDOV,
JUNIOR RESEARCHER**

**MAXIM S. MALAKHOV,
POST GRADUATE STUDENT**

**PETR A. NASLEDNIKOV,
POST GRADUATE STUDENT**



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14. ABSTRACT

This report results from a contract tasking Baltic State Technical University as follows: The contractor will investigate the evolution of condensed phase combustion products (CPCP) in the gas phase and will obtain the following experimental data for various distances from the burning propellant surface: 1) proportion of highly dispersed oxide (HDO) particles and agglomerates 2) parameters of dispersity for HDO particles and agglomerates and 3) agglomerate structure and chemical composition. The following data will be obtained in the study of agglomeration for propellants based on ammonium nitrate (AN): 1) portion of metal taking part in the agglomeration 2) parameters of dispersity for CPCP leaving burning propellant surface and 3) agglomerate structure and chemical composition.

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ABSTRACT

The report includes 83 pages, 57 figures, 9 tables.

Key words: SRP, CCP, AGGLOMERATE, HDO, STRUCTURE, DISPERSITY, EVOLUTION, MODEL, COATINGS.

The report describes the results of investigating the evolution of CCP in the gas phase and how the use of coating of initial MF influences the agglomeration process.

The investigation has involved the experimental study of the following:

- morphological and chemical properties of agglomerates and HDO particles at various stages of evolution process at two levels of pressure;
- characteristics of agglomerate evolution in the medium of cold oxidizer;
- characteristics of CCP (agglomerates and HDO particles) depending on properties of initial MF coatings at two pressure levels.

The obtained data have been analyzed and the results of analysis have made it possible to refine the physical concepts of the investigated phenomena. A feasibility of controlling the agglomeration process has been shown.

The previously developed model of agglomerates evolution has been refined, and a feasibility to use it for predicting the agglomerate characteristics in evolution has been demonstrated.

ABBREVIATIONS

SRP - solid rocket propellant
 CCP - condensed combustion products
 HDO - highly dispersed oxide
 FCOS - fluorine-containing organic substances
 CVB - constant volume bomb
 MF - metal fuel

NOMENCLATURE

P - pressure
 T - temperature
 a_k - oxidizing potential of gas phase (mole fraction of oxidizing components in gas mixture)
 D - agglomerate diameter
 d - HDO particle diameter
 D_{43} - mass medium diameter of agglomerates
 d_{43} - mass medium diameter of HDO particles
 f_m - mass function of size distribution density
 η - mass share of oxide in agglomerate
 Z_m - share of unburned metal in agglomerates relative to initial metal in propellant
 Z_m^{ox} - share of initial metal in propellant used to form oxide in agglomerates
 Z_m^{HDO} - share of initial metal in propellant used to form HDO particles
 Z_m^A - share of initial metal in propellant used to form agglomerates
 R, r - radius
 F - force
 U - propellant burn rate
 M - mass
 J - mole flow of substance
 Q_C - conductive heat flow
 Q_R - radiant heat flow
 Ω - area
 ρ - density
 H - molar enthalpy
 L - molar evaporation heat
 V - volume or velocity
 ΔV - by-pass flow rate
 λ - thermal conductivity
 C_p - isobaric heat capacity
 Nu - Nusselt number
 χ - share of metal drop covered by oxide «cap» in agglomerate
 $\pi = 3.14$
 $R_0 = 8.314 \text{ J}/(\text{mol}\cdot\text{K})$, universal gas constant

Subscripts

M - metal

ox - oxide

o - oxidizer

ox.d - dissociated oxide

a - agglomerate

b - bubble

p - HDO particle

A, B, E, F, C - boundaries of characteristic zones
in agglomerate combustion model

AE, EB, BF, FC - characteristic zones in agglomerate combustion model

Superscripts

in - in-flow

out - out-flow

c - condensed

g - gaseous

d - dissociated

A, B, E, F, C - boundaries of characteristic zones
in agglomerate combustion model

max - maximal value

INTRODUCTION.

The use of metal fuel (MF) as a component of SRP (Al, as a rule) generates a whole spectrum of problems [30]. Their solution is possible through studying and description of MF evolution process which is understood as a set of physical and chemical transformations of Al and its compounds during their residence in the engine chamber.

The present work is devoted to investigation of various aspects of the evolution process and comprises the stages of «life» of Al and its compounds in the condensed phase of burning propellant, and as a component of two-phase flow in the gas phase.

The work contents have been defined more accurately in the course of discussion of the investigation results in Reno, Nevada (January 16, 1998) and in the process of consultations with Prof. M. W. Beckstead.

1. INVESTIGATION OF CCP EVOLUTION IN TWO-PHASE FLOW

Efficiency of MF use in the propellant is considerably determined by realization of CCP evolution process when they are traveling incorporated in two-phase flow. Just the given process essentially determines the CCP properties which, in turn, produce decisive effect on realization of phenomena governing the quality of propellant-engine system: formation of losses in specific impulse, slag formation in the engine chamber, impact of combustion product flow on construction elements, stability of engine operation.

Below the results of investigation of this process are given which included obtaining experimental information, its generalization, creating a number of descriptions which make it possible to solve the problem of predicting the CCP characteristics.

1.1. CCP EVOLUTION IN THE FLOW OF SRP COMBUSTION PRODUCTS.

1.1.1. ANALYSIS OF LITERATURE.

The works on studying the given process started as soon as metalized SRP emerged. They comprised the problems related to obtaining experimental data on various manifestations of evolution process and to its modeling.

Methods of experimental research.

To solve the problem of obtaining experimental data on properties of condensed combustion products (CCP) in the process of their evolution in the gas phase, two approaches can be used:

- 1 - direct determination of CCP particles parameters;
- 2 - determination of gas phase or engine characteristics governed by CCP properties.

The first approach can be realized when contact and non contact methods are used. The contact methods of research consist in sampling of particles out of combustion products flow with subsequent analysis of their dispersity, chemical composition, structure and other properties. By now a number of methods for sampling CCP both directly in the engine and in various experimental setups [1-5 et al.] has been developed.

In contrast to contact methods the non contact ones produce, as a rule, lesser disturbances in the investigated field. They allow one to obtain unique data due to a possibility of direct CCP diagnosis in the evolution process. The above-stated regards, first of all, optical methods with image formation - methods of visualizations including impulse and shadow photography, high speed filming, etc. [1].

Visualization methods possess high obviousness, allow one to observe CCP particles directly in the process of propellant combustion. Under certain conditions these methods make it possible to reliably register the true size of particles in the gas phase of burning propellant. It is possible to combine photometry methods with CCP particles' temperature measurement [6]. However, it is worth noting that representativeness of

analysis in visualization is not high and its application becomes difficult at high pressure due to increase in background radiation.

A number of works used the second approach in the experimental study of CCP properties in evolution process. Data obtained in these works were used to determine the combustion efficiency of metal fuel depending on conditions in combustion chamber. The temperature of gas phase and overall pressure impulse were used as a parameters characterizing the combustion efficiency. The attractive features of these methods are the non contact nature of analysis and possibility to analyze the whole set of CCP particles. At the same time the obtained data on the evolution process are limited and indirect in character. Besides, the application of the mentioned techniques requires to substantiate the correctness of measuring precisely the gas phase temperature in the first case, and the lack of effect which the character of two-phase flow in the nozzle unit can produce on the magnitude of overall pressure impulse in the second case.

Characteristics of the evolution process.

With account for the principal differences in the nature of formation and properties of CCP we can distinguish two fractions of CCP supplied from the burning propellant surface into gas phase: agglomerates and HDO particles. The CCP properties determine the possibility of interphase and interfraction interaction in the two-phase flow which are, in essence, the evolution process. The most prominent and obvious interaction is the agglomerate metal combustion which results in changing composition (both chemical and dispersive) and temperature of the flow. The agglomerate properties (considerable size, complex structure, presence of substances capable of chemical transformations) make them the principal factor of the evolution process. In general case, the data on both the single particle burning in active media and transformations of the whole set of particles in propellant burning can be used for analysis of the given process.

Qualitative characteristics.

The metal particles combustion in active gas environments displays bright luminous area saturated with products of metal particles burning which is identified as burning area [1, 7-9 et al.]. The blowing of gas around burning particle results in formation of a peculiar «tail» of combustion products extending to considerable distances in the direction of blow-out [6-7 et al.]. The metal is in a liquid state at a temperature close to its melting point at the given pressure [6]. The particle form is close to equilibrium one [10]. The temperature of burning area around particle exceeds both the particle surface temperature and that of surrounding gas phase. For the case of metal combustion as part of SRP the temperature of the particle burning area in the pressure range of 2.0 - 4.0 MPa is 3500 - 3700 K [11] which is 300 - 400 K higher than the equilibrium temperature of combustion products.

Condensed products of agglomerate metal combustion are, as a rule, oxide compact particles of relatively small size (about 1 μm and less) [4-5 et al.]. However, some hollow spheres commensurable with initial metal particle size may be also formed [1]. Very often aluminum drops are observed on the inside surface of the hollow spheres.

In metal (Al) combustion in various laboratory setups [1, 12 - 14] a number of specific effects are noted which consist in particles rotation, their trajectory curving due

to combustion products discharge from the particle surface, particles fragmentation. Summarization of results yielded a conclusion that these effects are encouraged by pressure decrease, ambient temperature rise and an increase in oxidizing potential of environment (especially due to oxygen concentration rise).

Metal combustion in SRP combustion products environment occurs under conditions when H_2O and CO_2 (O_2 is practically absent) are predominant oxidizing gases. As a rule, these gases concentration is considerably lower than corresponding one characteristic of gas burners and that factor apparently determines low probability of such effects occurrence in combustion of metal as a component of SRP. A number of works assume that CO_2 and H_2O are equivalent oxidizers. At the same time one work [15] shows that H_2O is a more effective oxidizer.

Generally accepted is the statement that the burning metal particle surface may have a formation representing an oxide drop which various authors call differently (oxide "cap"). At the same time there isn't a conception about the nature of the oxide "cap" and its evolution in the combustion process which is shared by everybody.

The CCP particles in the two-phase flow are in liquid state and in principle capable of gasodynamical deformation and breaking down.

Presence of Al and Al_2O_3 in agglomerate raises the question of such formations existence stability. The work [16] establishes that at temperature $1900^\circ C$ and atmospheric pressure (this temperature is lower than Al boiling point at the given pressure) the mixture of condensed Al_2O_3 and Al in inert environment can completely evaporate from the heating element surface, and Al_2O_3 volatility in Al presence is higher by about a factor of 100. Thus, interphase interaction within the agglomerate is possible.

Kinetics of metal particles combustion.

Quantitative data on single particles evolution are obtained in as full measure as possible for characteristics of their combustion time (as applied to agglomerates - combustion time of agglomerate metal). The given circumstance is a direct consequence of the fact that combustion kinetics study is undoubtedly expedient for resolving practical problems.

Experimental works on kinetics study was carried out for metal particles combustion both in specially created active environments (mixtures of H_2O , CO_2 , O_2 with inert gases) and in the environment of propellant combustion gaseous products [8, 15, 17-26]. As a rule, the experimental results were presented by means of approximating the dependence of particles burn time (t_b) on their initial diameter (D_0) as follows:

$$t_b = \frac{D_0^n}{k}$$

where: k - coefficient depending on oxidizing gases concentration;
 n - parameter taking on values in the range of 1.5 to 2.0.

It was established that the ambient temperature doesn't practically affect the t_b magnitude. The experiments with changing the temperature within the range of 2000 - 3000 K show that the burn time remains practically invariable [1].

It is assumed that dependence of t_b on pressure exists. This dependence has the following character. Burn time t_b does not depend on pressure when $P > 2.5 MPa$; in the region of lower pressures t_b tends to grow with pressure fall. However, the fall isn't big

and at 1.0 - 1.5 MPa comes to 15-20 % of t_b magnitude at the high pressure (4.0 - 5.0 MPa). Work [18] registers a similar general character of dependence $t_b = f(P)$. But the low pressure region is more extended and within its range the more essential increase in t_b takes place.

The use of empirical dependencies obtained by various authors under the similar conditions allows one to discover an essential scattering of t_b values (in relative values it may reach 400% !). The presence of a significant difference in metal particles burn rate determined with help of these dependencies is illustrated in work [27]. A number of obvious factors can be pointed out which should affect the kinetics of metal particles combustion but aren't taken in account in the papers considered. They include blowing out the particles with external gas flow, and burning particles (agglomerates) structure which may differ under specific conditions of an experiment. Besides, in the opinion of authors in works [15,18] the effectiveness of gases as oxidizers for Al is different. Introduction of effective concentration C_{eff} [15] makes it possible to cut down the scattering of experimental data on combustion time, especially in the range of small t_b values. C_{eff} value is determined in the following way:

$$C_{eff} = \sum C_j X_j$$

where: X_j - mole share of j-th oxidizing gas;

C_j - constants which according to the data of work [28] for H_2O , CO_2 , and O_2 come to 0.53, 0.135, and 1.0.

Evolution of the entire population of CCP particles in the combustion chamber.

Authors of the present report have carried out systematic investigation of agglomerates evolution [29] which was based on CCP quench - collection at various distances from the burning propellant surface and their comprehensive analysis. A totality of phenomena taking place in agglomerates evolution has been revealed. They are as follows:

- combustion of agglomerate metal in gas-phase mode;
- chemical reaction of Al and Al_2O_3 containing in agglomerate and formation of gaseous products;
- changing structure of agglomerate;
- deposition of HDO particles on agglomerates from external gas flow;
- agglomerate movement in the two-phase flow.

On the basis of the obtained experimental data a mathematical model of single agglomerate evolution has been developed, then on the basic of the latter - a model of agglomerate population evolution. In constructing the model a principle of decomposition and synthesis of separate phenomena descriptions has been used.

The use of this model for agglomerate population if a description of carrier gas phase is available allows us to carry out modeling evolution for the entire agglomerates population. The conducted comparison of modeling results and experimental data on evolution of agglomerates, which are formed in combustion of a number of propellant compositions, has shown their satisfactory agreement and, first of all, regarding the phenomena of metal combustion and oxide carrying away. The lack of data on HDO

evolution characteristics hasn't allowed us to clarify the mechanism of HDO particles and agglomerates interaction.

On the whole, the carried out investigation has achieved a considerable progress in understanding the physical nature of the processes in agglomerates evolution and obtaining the practically significant results which make possible to predict the agglomerates characteristics. But the lack of studies on HDO evolution makes it incomplete and limited and doesn't allow us to describe the process under consideration as applied to the entire population of CCP particles. Besides, the study results have been obtained for a limited number of propellants and therefore degree of their universal usefulness is still a question.

1.1.2. FORMULATING THE STUDY GOALS.

With account for results of literature analysis the goals of studying the process of CCP evolution in the gas phase can be formulated as follows:

1. To obtain as much as possible comprehensive experimental data (quantitative and qualitative in nature) on the properties of entire CCP population at various stages of the evolution process with propellants ensuring various initial properties of CCP (at the burning propellant surface).

2. To analyze the efficiency of the previously developed model of the agglomerates evolution for prediction of its characteristics.

As an object of investigation it is expedient to use the propellants investigated at the first stage of the work [30] for which the CCP characteristics near the burning propellant surface are obtained.

1.1.3. METHOD OF EXPERIMENTAL INVESTIGATION.

To resolve the problems of the present investigation, namely obtaining representative and reliable experimental data on the CCP evolution process in the flow of combustion products of aluminized SRP a method of CCP particles quench-collection by means of their «freezing» in the constant volume bomb has been used as the main «tool» of the research. This method has been used in research of metal fuel agglomeration in the process of SRP combustion carried out at the first phase of the project [30]. It is distinguished by a high degree of informativeness and representativeness of the obtained results, as well as by the simplicity of realization. Besides, high speed filming was used for obtaining qualitative characteristics when the propellant samples were burned in constant pressure bomb.

Research technique.

Studying the CCP evolution suggests obtaining experimental data on CCP parameters depending on time of their residence in the flow of propellant combustion products. The essence of technique used for investigating the CCP evolution consists in the following.

To create under experimental conditions a flow of propellant combustion products the samples of propellants are burned in cylindrical tubes (Fig. 1.1, 1.2). The present

method is the simplest and convenient for investigation. Thus, the CCP evolution under conditions of unidirectional flow of solid propellant combustion products is being studied. Variation the CCP residence time in the combustion products flow is ensured by changing the tube length. The CCP evolution has also been studied under conditions of high velocity flow created by means of narrowing the tube (Fig. 1.3).

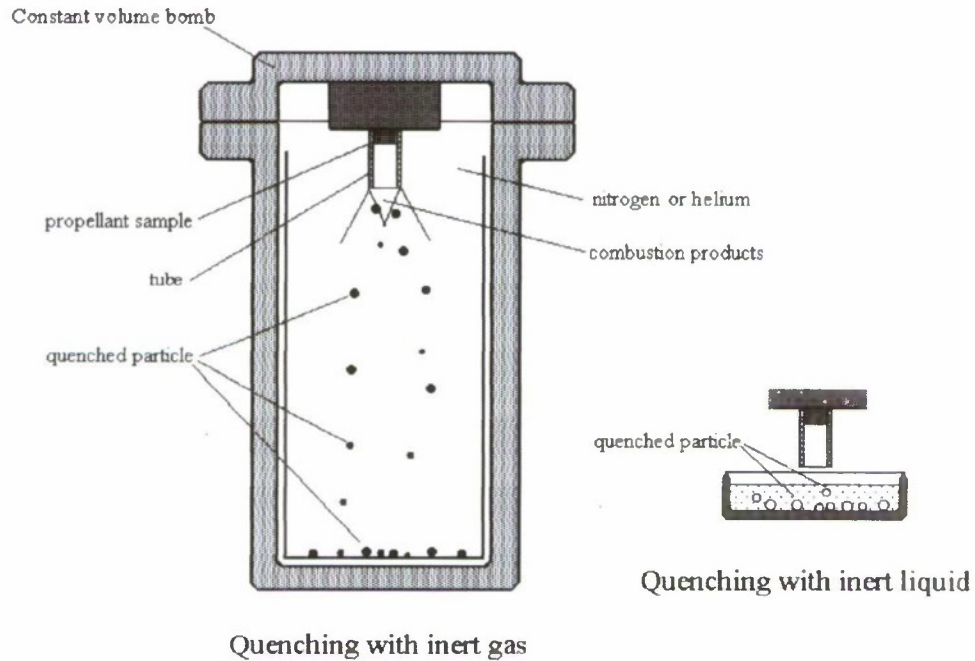


Figure 1.1. Experimental setup for studying agglomerates evolution.

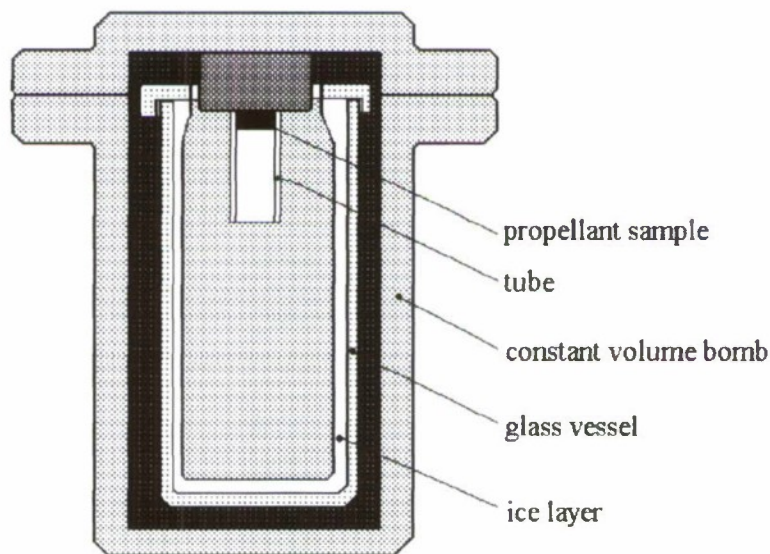


Figure 1.2. Experimental setup for studying HDO particles evolution.

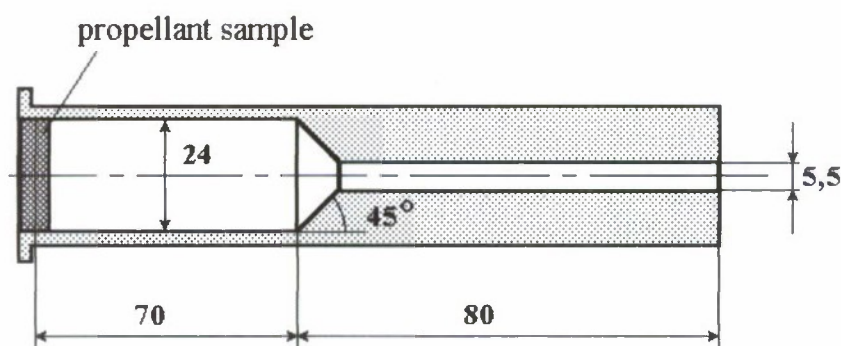


Figure 1.3. Narrowing tube providing creation of high velocity flow of combustion products.

Freezing the CCP particles (agglomerates and HDO particles) at the required cross section of the combustion products flow is made with help of inert gas (nitrogen or helium) in the constant volume bomb (CVB) or with help of inert liquid in a special container. With the first method of freezing (with help of inert gas) CCP particles sampling is made with help of a special container-collector (Fig. 1.1) or with help of a glass vessel covered inside with an ice layer (Fig. 1.2).

Sampling technique with help of special container-collector presented in Fig.1.1 is convenient for collecting agglomerates, but it doesn't allow one to sample HDO particles. Therefore, the given technique has been used as a basic one in studying the agglomerate evolution. More labor-consuming technique using the glass vessel with ice layer inside (presented in Fig. 1.2) has been applied for studying the HDO evolution. Essence of this technique was described in the report on the first phase of project [30].

The samples of solid propellants have been made in cylindrical form. Their diameter corresponded to the inside diameter of the tube. The side surfaces of samples were jacketed. The samples burned from one end. The ignition of samples was made with electric current through a filament. The samples of aluminized propellants were made with non metalized solid propellant base layer. It ensured removal of last agglomerate portions from the burning surface into the gas phase and created conditions for their evolution. The tubes, as a rule, were made of sublimating materials: plexiglass ($C_5H_8O_2$) or fluoroplastic-4 (C_2F_4)_n. Gasification of tube material ensures the absence of CCP particles depositing on the channel inside surface. The use of precisely this technique makes it possible to avoid intensive depositing of CCP particles on the inside surface of tube channel. But the use of gasifying tube material results in some distortions in the properties of the combustion products flow. The degree of distortion depends on the tube material and the tube's length and diameter ratio.

The previously conducted experiments have shown that the intensity of plexiglass and fluoroplastic gasification under the effect of combustion products flow is about the same. The distortion degree decreases with an increase in the tube inner diameter and a decrease in the tube length. Taking into account the limitations for the propellant sample mass [30] the tube inner diameter is taken equal to 24 mm. At the same time the thickness of solid propellant sample shouldn't exceed 3-4 mm. As a result of previously conducted special experiments it was established that with the tube inner diameter of 24

mm we can ignore the distortions in properties of the propellant combustion products flow under a condition that the tube's length doesn't exceed 70 mm.

The area of CCP particles sampling by «freezing» them with inert gas is defined in accordance with the previously proposed approach [30]: sampling conditions for all CCP particles are averaged and the sampling area is presented as flat surface A-A, located at L_s distance (average) from the propellant sample burning surface (Fig. 1.4.).

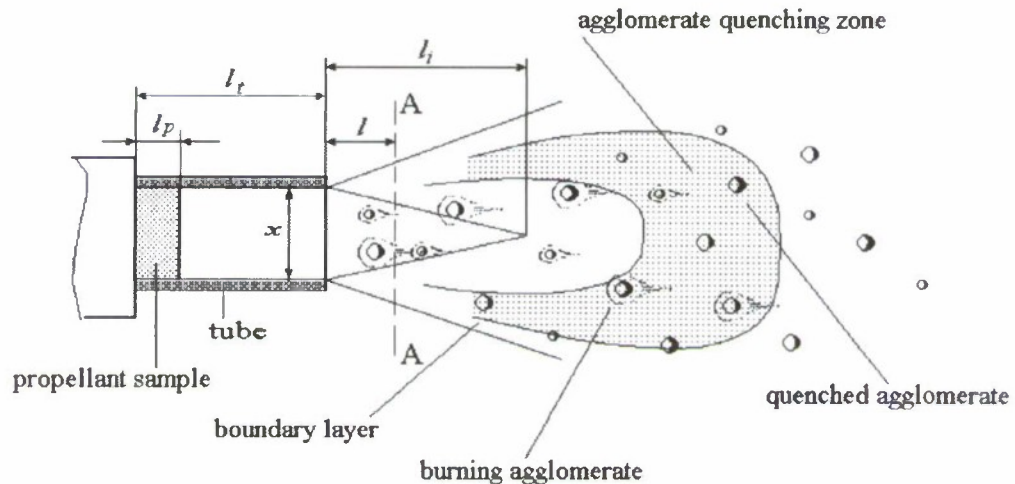


Figure 1.4. General layout of CCP sampling.

According to Fig. 1.4.: $L_s = l_t - 0,5l_p + l$

where: $l = 1,2x$, for helium,

$l = 0,15x$, for nitrogen.

The sampled CCP particles undergo dispersity, chemical, and structural analysis. The techniques of those analyses are described in the previous report [30].

The dispersity analysis of agglomerates is carried out with help of diffraction laser analyzer «Analyzette-22» of Frich make. The problem of distortions in the dispersity characteristics of agglomerates in their freezing and storage was examined in [30], where it was concluded that results of the agglomerates dispersity analysis quite well correspond to the conditions of the combustion products flow. The HDO particles dispersity analysis is carried out with help of analyzer «Coulter 4» according to the technique described in [30]. The correctness of employing the given technique has been confirmed by a special study in which electronic microscope has been used for study of the HDO particles sampled. Besides, within the framework of the present investigation the effect of tube material properties on the registered dispersity of HDO particles has been studied.

In defining the characteristics of agglomerates chemical composition, the additional oxidation of agglomerates metal has been taken into account and the true characteristics (without distortions caused by the additional oxidation) have been found. The agglomerates' structure has been studied on their microsections.

The conducted analyses yield the following main characteristics of sampled CCP:

1. Size distributions of CCP particles;
2. Parameters Z_m , Z_m^{ox} , Z_m^{HDO} , D_{43} , d_{43} , and η .

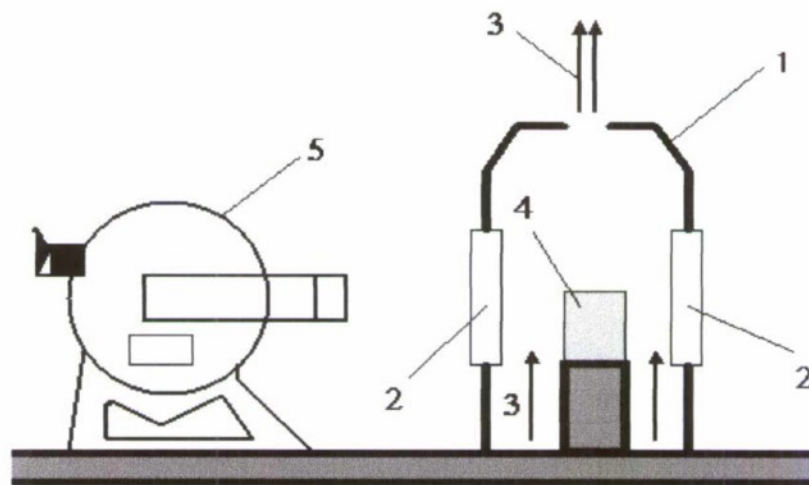
Original information about size distribution of CCP particles is histogram [30]. Approximating the histogram allows us to made up mass function of size distribution density for CCP particles (f_m). This information (function f_m) is more visual and useful for analyzing particle size distribution.

Parameters Z_m and η are defined directly by chemical analysis. Then Z_m^{ox} and Z_m^{HDO} values are found with help of proportions:

$$Z_m^{ox} = [54/102] [\eta/(1-\eta)] Z_m$$

$$Z_m^{HDO} = 1 - Z_m - Z_m^{ox}$$

Qualitative data have been obtained with help of high speed filming in the own light of CCP particles in the gas phase (Fig.1.5). The used movie camera is «CKC-1M» of Russian make, enlargement on the film is 1,5-4,5, and speed of filming is 2000-4000 frame/sec. Due to the problems caused by the gas phase background radiation, these experiments were mainly conducted at atmospheric pressure.



1 - constant pressure bomb, 2-optic windows, 3- inert gas, 4 -propellant sample, 5- camera.

Figure 1.5. Layout of experimental study with high speed filming.

1.1.4. RESULTS OF RESEARCH AND THEIR ANALYSIS.

Quantitative experimental study of CCP particles evolution has been carried out at two pressure levels: 1 and 6 MPa. In the process the same propellants have been used as those studied at the first stage of the project. The propellants' properties are given in the previous report [30]. In the same report the data on CCP parameters close to propellant burning surface, at the distance of $L \approx 1\text{mm}$ from it are presented. To determine the changes in CCP characteristics in their evolution, the agglomerates sampling was carried out at two cross sections of combustion products flow at $\sim 30\text{ mm}$ and $\sim 70\text{ mm}$ from the propellant sample burning surface.

Regarding HDO particles, the investigation strategy was as follows. Experiments were carried out for those propellants and conditions of their combustion where expected influence of agglomerates evolution on evolution of the whole CCP either take place or

not. In the first case, the propellants 1 and 5 at low pressure were used, and in the second case, the propellant 1 at high pressure was used ($Z_m^a (L_s \approx 1\text{mm}) = 0.013$). For these cases, volume of experimental data was increased.

The experiments have yielded dependencies of the main CCP characteristics above listed on the distance L_s . The results are given in Fig. 1.6 - 1.14.

The obtained experimental results have quite a high degree of representativeness: coverage of CCP particles sampling (agglomerates and HDO particles) is not less than 93 - 95%. The reliability of experimental data was ensured by their quite great volume: for the same conditions, experiments were repeated at least ten times for agglomerates, and five times for HDO particles. In Fig. 1.6 - 1.14 the parameters' mean values and their confidence intervals with confidence probability of 0.997 are shown. Conducted analysis allows to suggest that scattering in obtained data is the result of scattering in SRP parameters.

Let's examine the results of studying the technique for determination of dispersity of HDO particles. As soon as the particles are sampled it is natural to expect their coagulation. To prevent this phenomenon in conducting dispersity analysis, the sampled HDO particles are processed with ultrasound (dispergation). But dispergation can also lead to coagulation intensification (appearance of new coagulates). Besides, in the application of the given technique there is a theoretical possibility of the initial HDO particles destruction.

To find out the peculiarities in realization of the mentioned phenomena the results obtained in the course of samples analysis with help of analyzer "Coulter 4" and electronic microscope have been compared at various dispergation duration (t_d). It allowed us to establish the following:

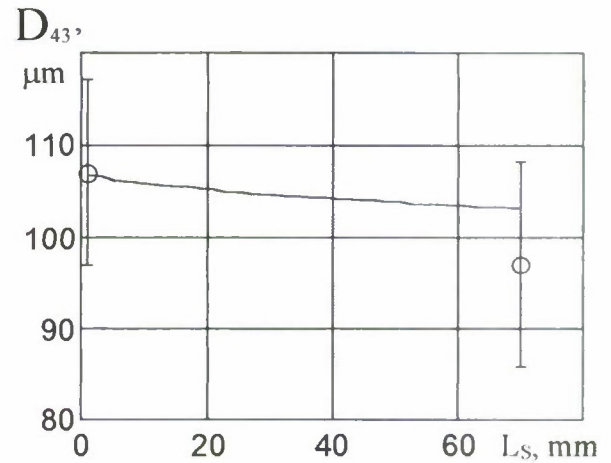
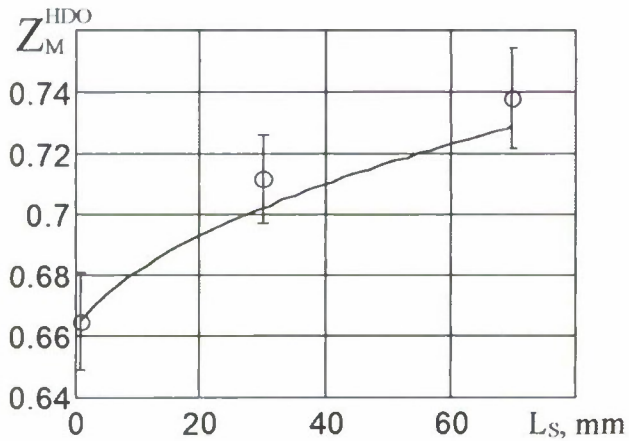
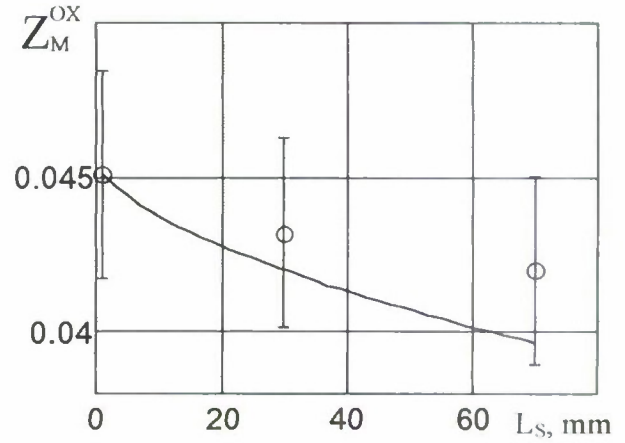
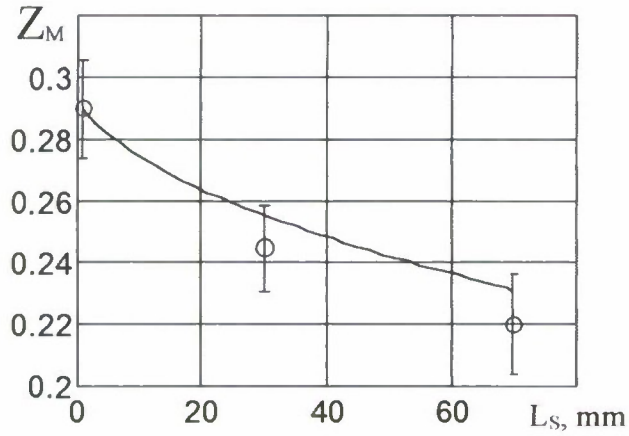
1. Coagulates formation is an objective reality (Fig.1.15). With an increase of t_d their number first decreases and then grows (Fig.1.16). When the registered dispersity is maximum (when $t_d = t_d^{\text{opt}}$) the coagulates are actually not observed.

2. Crushing of initial particles (availability of irregular shape particles) is not registered. The particles forming the first mode in the HDO particle size distribution function (f_m) near the burning propellant surface [30] have regular spherical shapes (Fig.1.17).

The given results confirm the validity of the previously accepted approach according to which we assume that the true dispersity parameters are those determined at $t_d = t_d^{\text{opt}}$ condition. It should be noted that high labor expenditures in experiments on finding out the dispersity parameters hampers accurate measurements of t_d^{opt} and results in an error in parameters' measurements. The given error may have sufficient influence on accuracy of measured dispersity of HDO particles.

The problem of coagulation of the sampled HDO particles raises also a question of tube material properties. The use of tubes made of plexiglass and fluoroplastic results in an essential increase in the soot content in the sampled CCP. Thus in case of using plexiglass tubes that content grows by nearly an order of magnitude (mass share increases from ~0.5% to 3%). The soot is a substance rather actively contributing to coagulation. In presence of a factor ensuring intensification of coagulation process a situation is possible when with $t_d = t_d^{\text{opt}}$ coagulates survive. To check the nature of additional soot formation influence on the registered HDO particles dispersity special

L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm	d_{43} , μm
1	0.290	0.045	0.665	0.227	107	0.35 ± 0.07
30	0.245	0.043	0.712	0.250	-	-
70	0.220	0.042	0.738	0.265	97	0.535 ± 0.09



— model, \circ - experiment

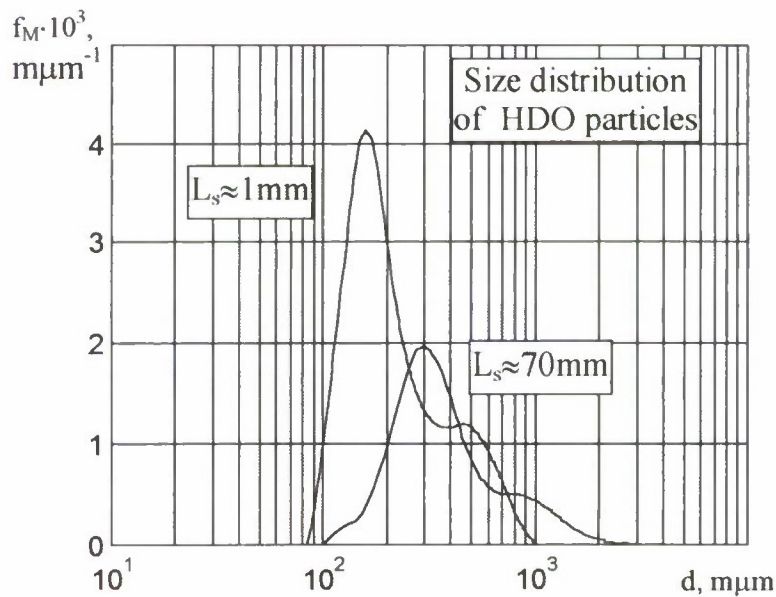


Fig. 1.6. Experimental data for propellant 1, pressure 1MPa.

L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
1	0.210	0.021	0.769	0.159	68
30	0.150	0.018	0.832	0.186	-
70	0.110	0.016	0.874	0.215	57

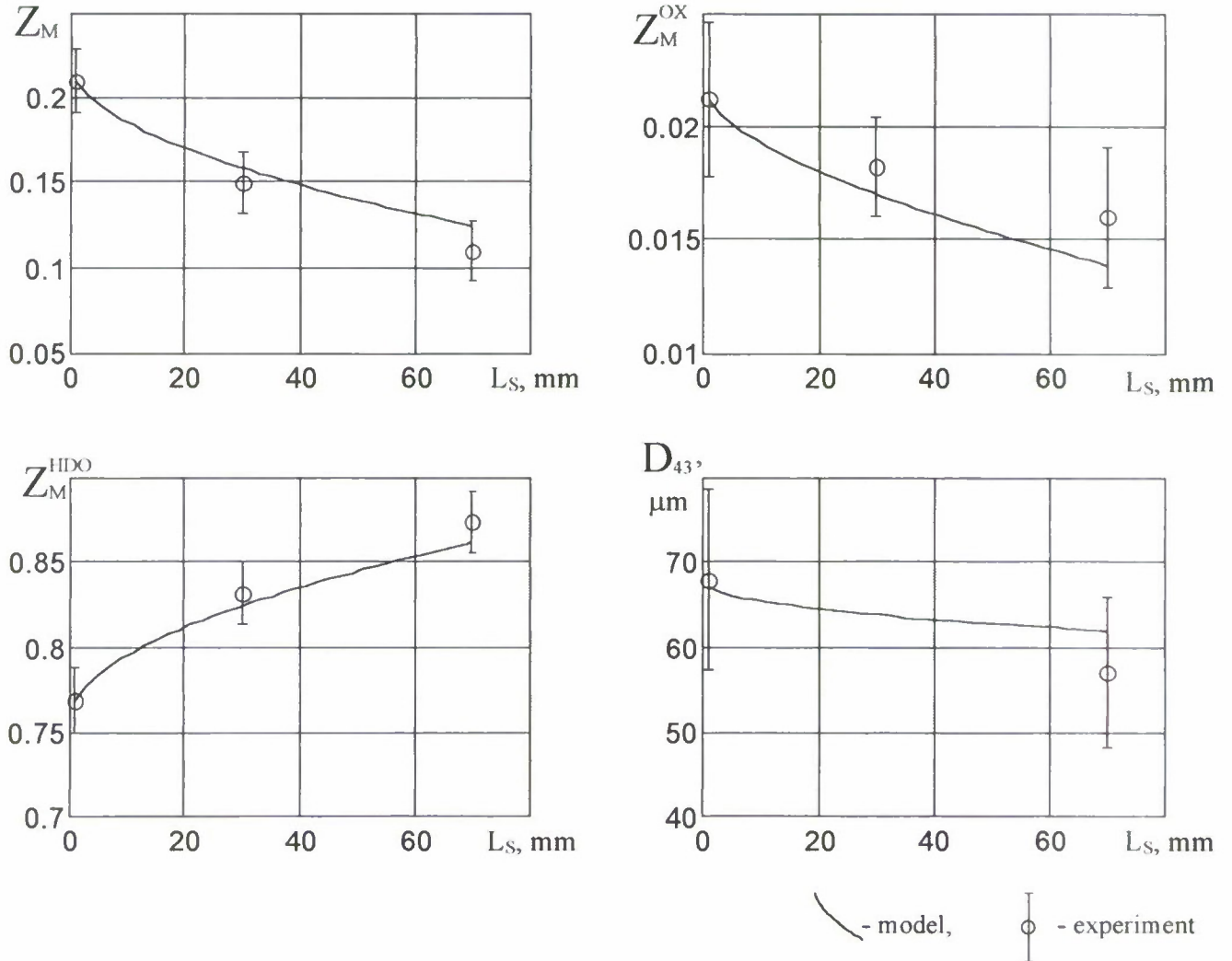


Fig. 1.7. Experimental data for propellant 2, pressure 1MPa.

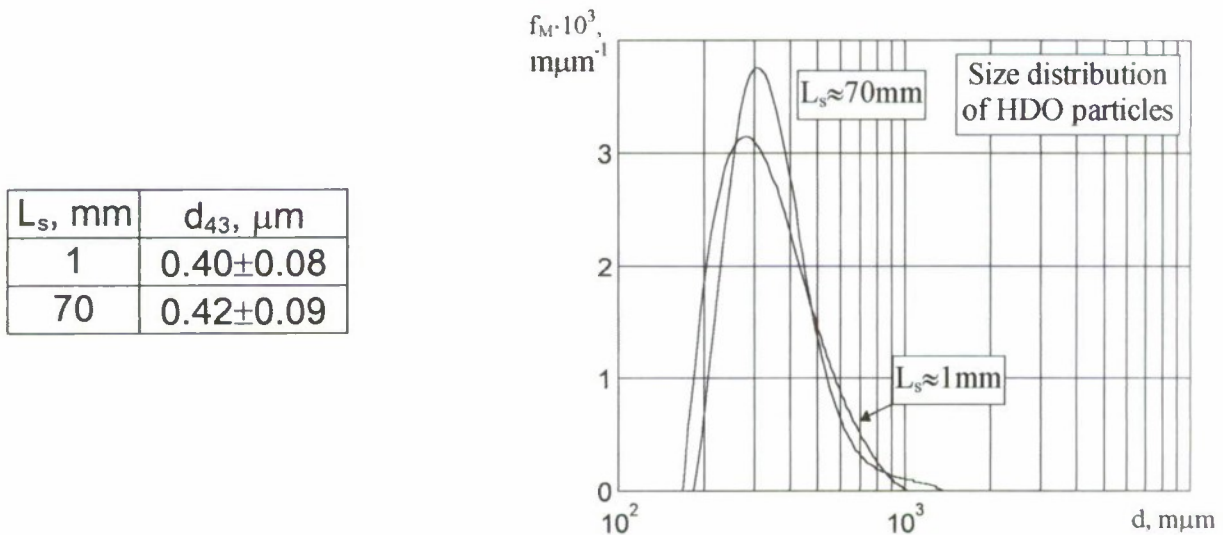
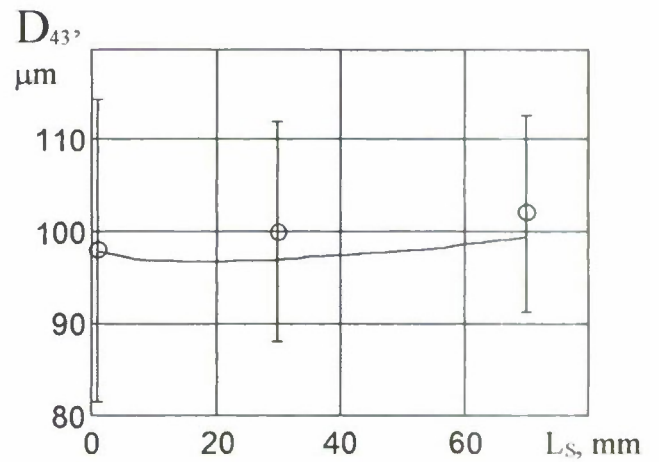
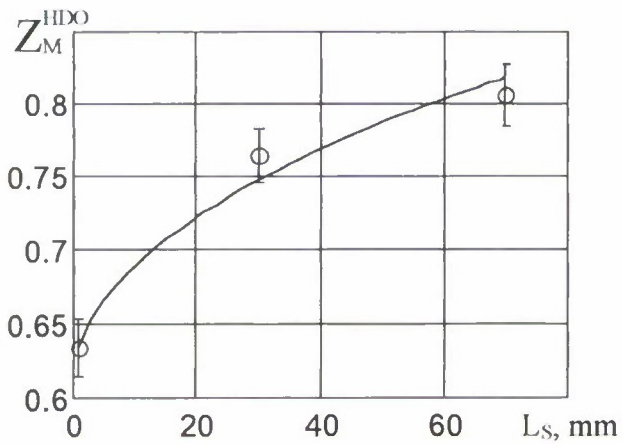
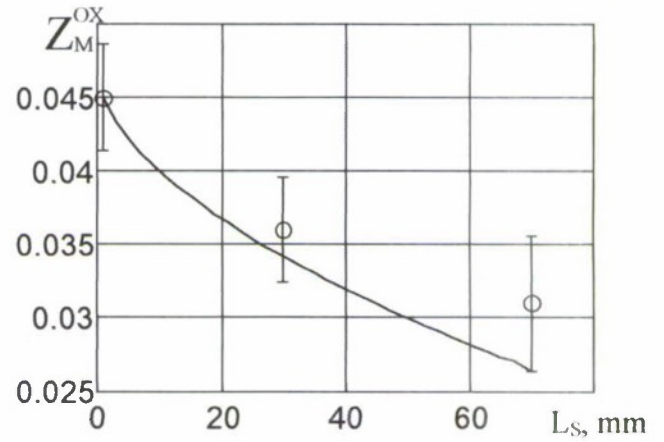
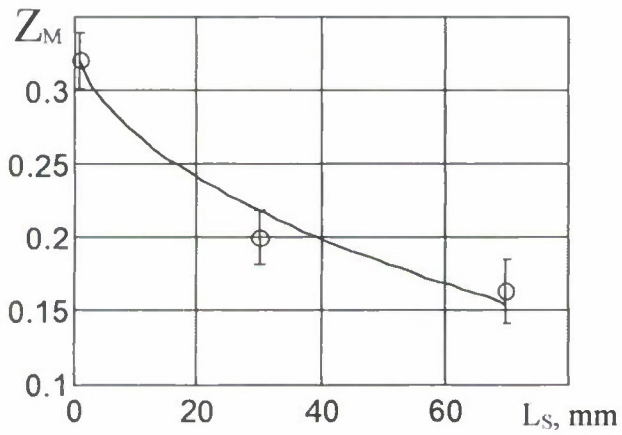


Fig. 1.8. Experimental data for propellant 1, pressure 6MPa.

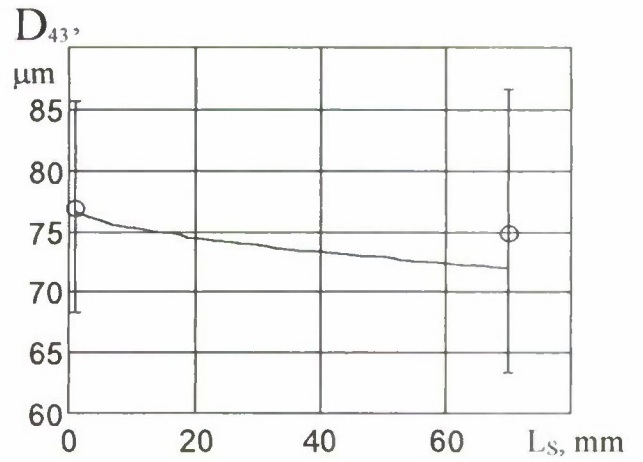
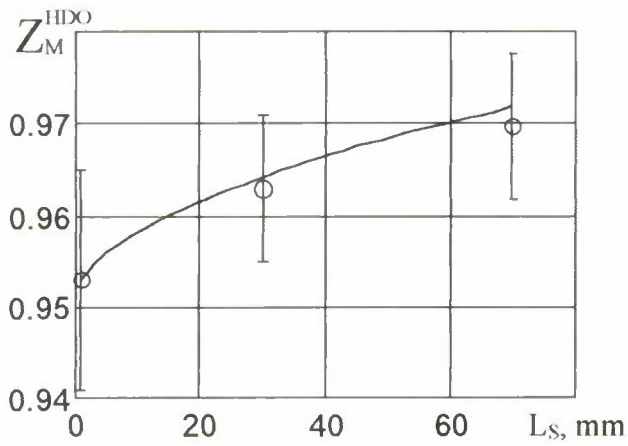
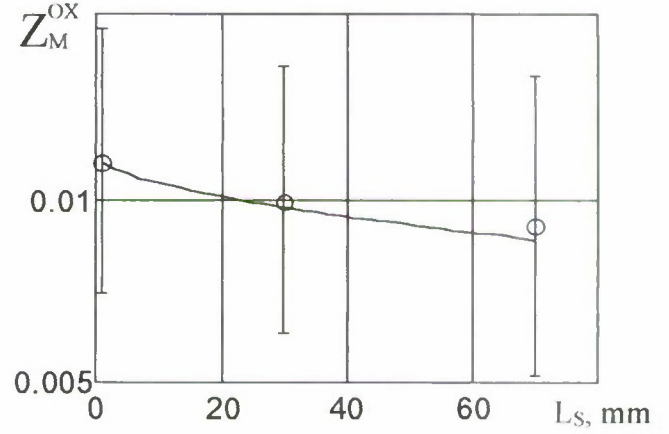
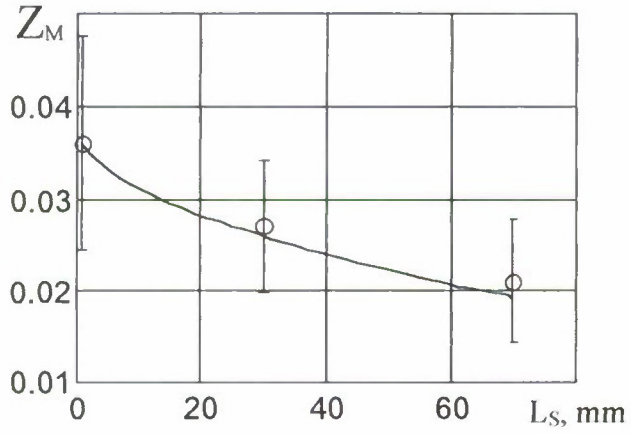
L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
1	0.320	0.045	0.635	0.210	98
30	0.2	0.036	0.764	0.254	100
70	0.163	0.031	0.806	0.264	102



— model, \circ - experiment

Fig. 1.9. Experimental data for propellant 3, pressure 1MPa.

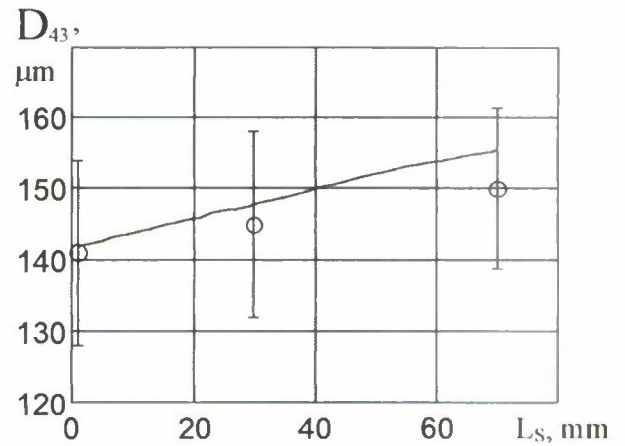
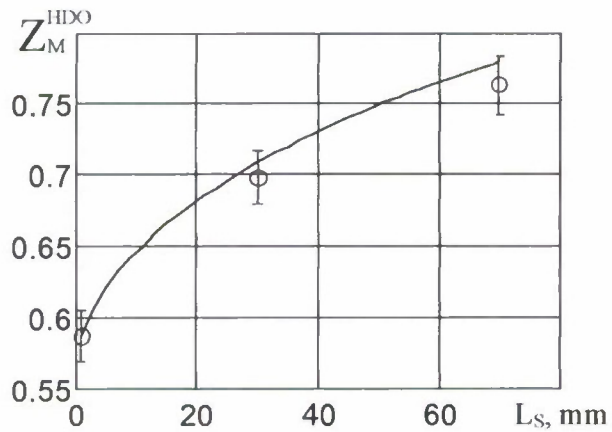
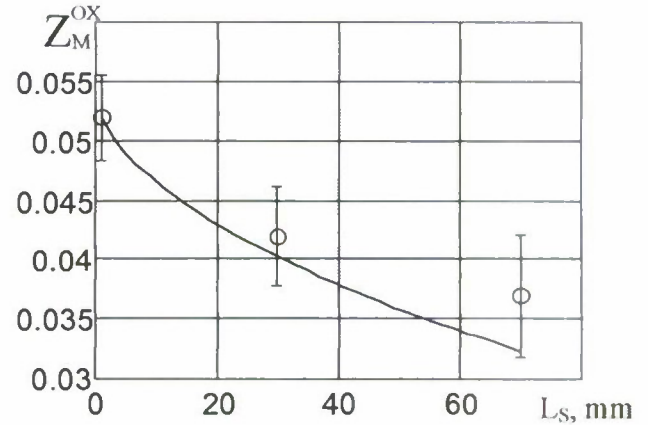
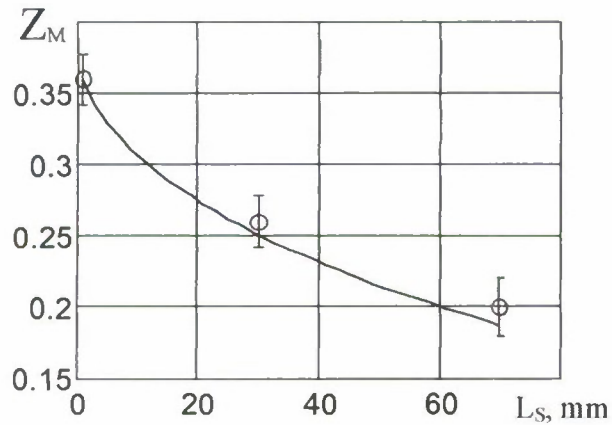
L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
1	0.036	0.011	0.953	0.366	77
30	0.027	0.0995	0.963	0.410	-
70	0.021	0.0925	0.970	0.454	75



— - model, \circ - experiment

Fig. 1.10. Experimental data for propellant 3, pressure 6MPa.

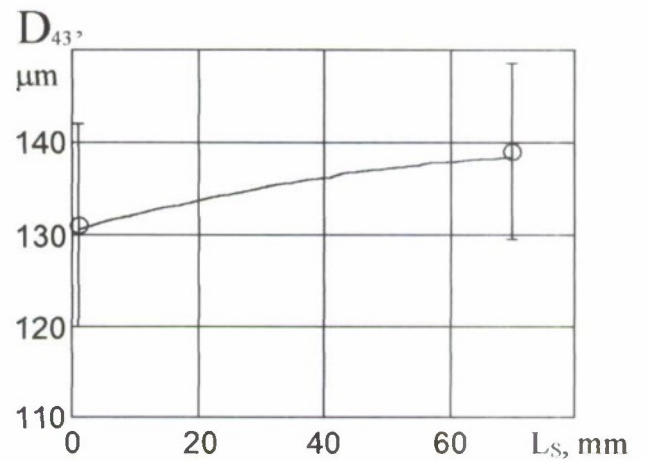
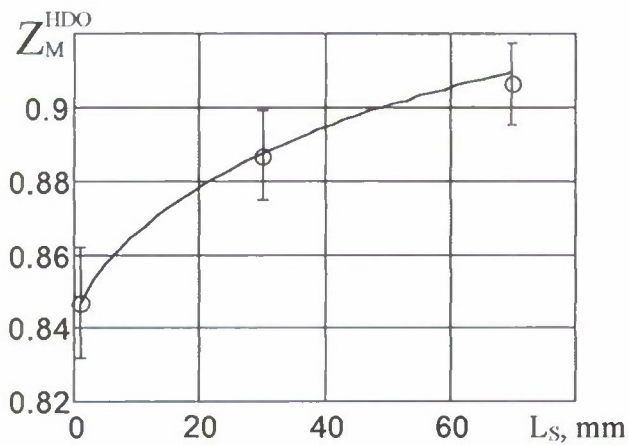
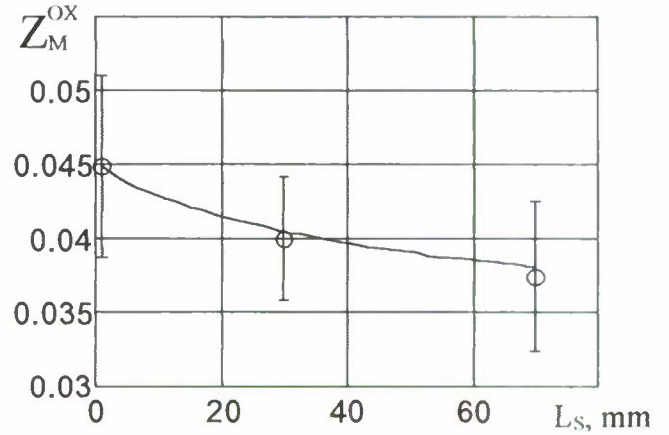
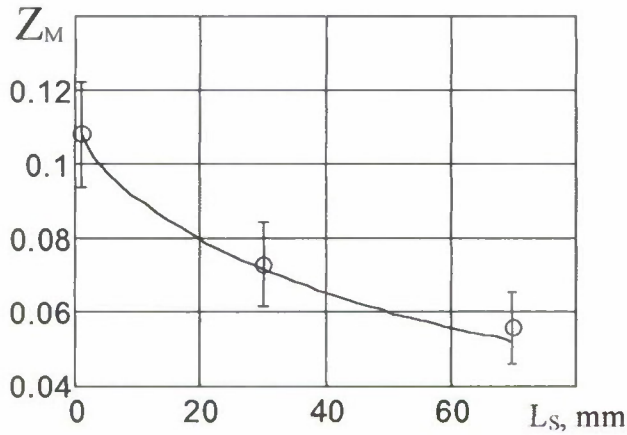
$L_s, \text{ mm}$	Z_m	Z_m^{ox}	Z_m^{HDO}	η	$D_{43}, \mu\text{m}$
1	0.360	0.052	0.588	0.214	141
30	0.260	0.042	0.698	0.234	145
70	0.200	0.037	0.763	0.259	150



— - model, \circ - experiment

Fig. 1.11. Experimental data for propellant 4, pressure 1MPa.

L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
1	0.108	0.045	0.847	0.440	131
30	0.073	0.040	0.887	0.509	-
70	0.056	0.038	0.906	0.558	139



— - model, \circ - experiment

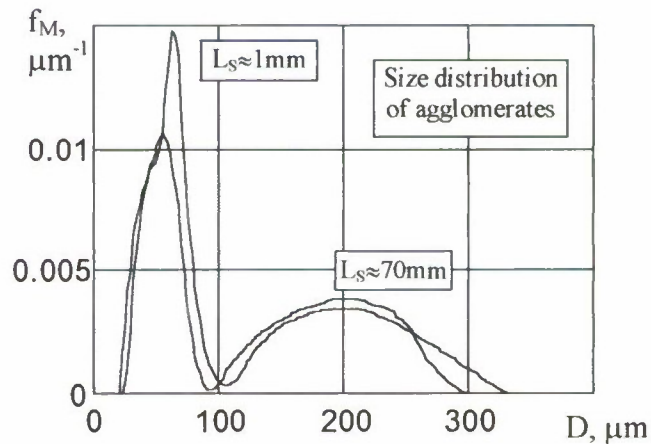


Fig. 1.12. Experimental data for propellant 4, pressure 6MPa.

L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
1	0.390	0.063	0.547	0.234	504
30	0.365	0.060	0.575	0.234	-
70	0.343	0.059	0.598	0.259	514

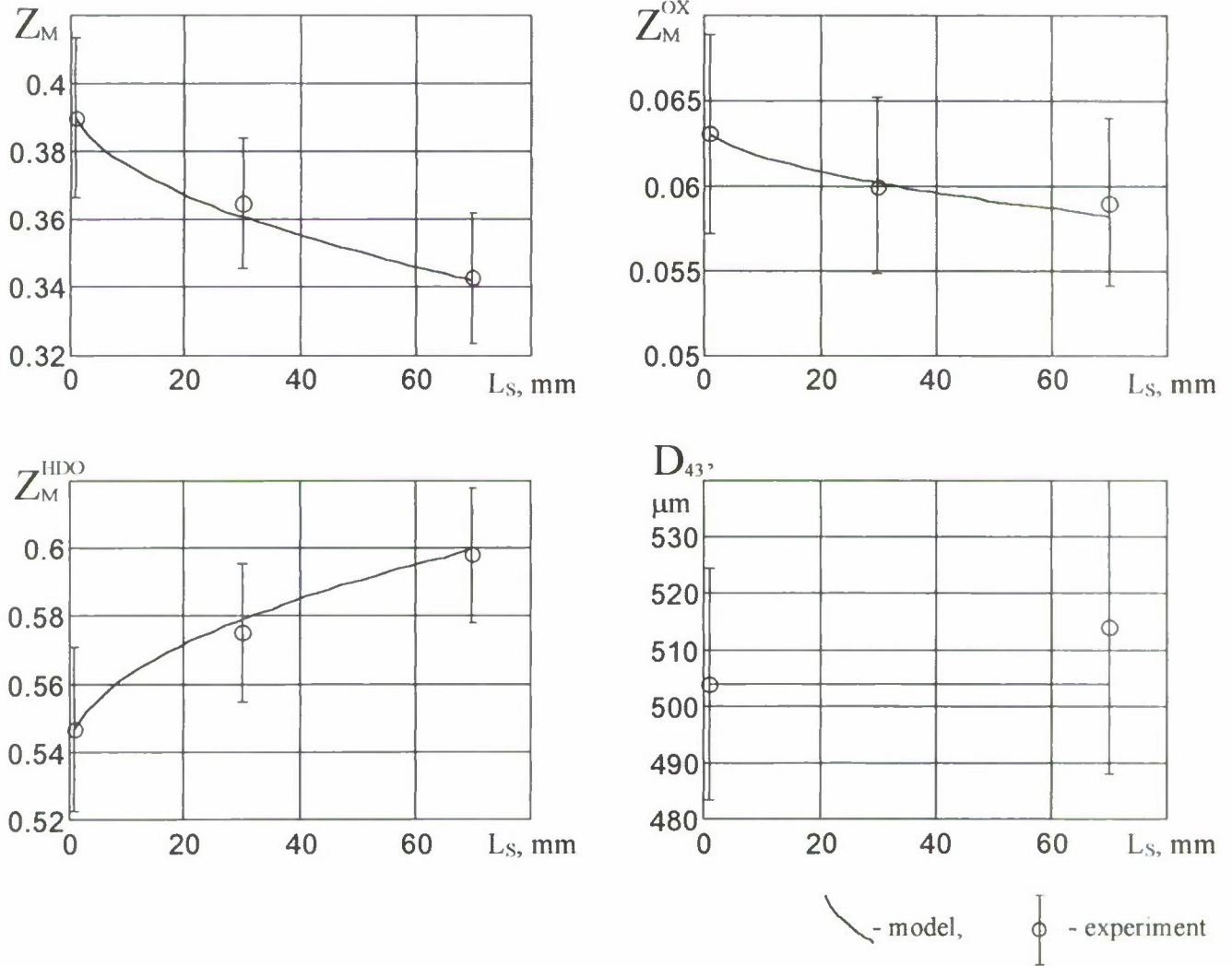


Fig. 1.13. Experimental data for propellant 5, pressure 1 MPa.

L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm	d_{43} , μm
1	0.254	0.090	0.656	0.494	250	0.80 ± 0.16
30	0.198	0.081	0.721	0.434	-	-
70	0.168	0.075	0.757	0.457	285	1.35 ± 0.2

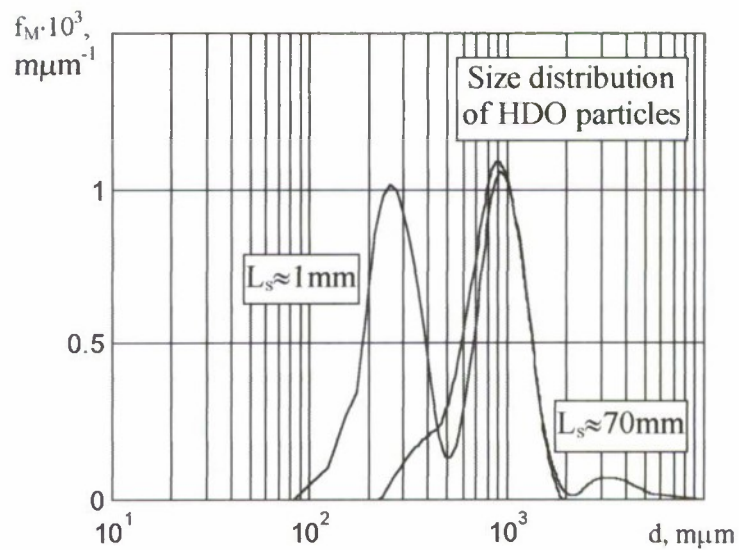
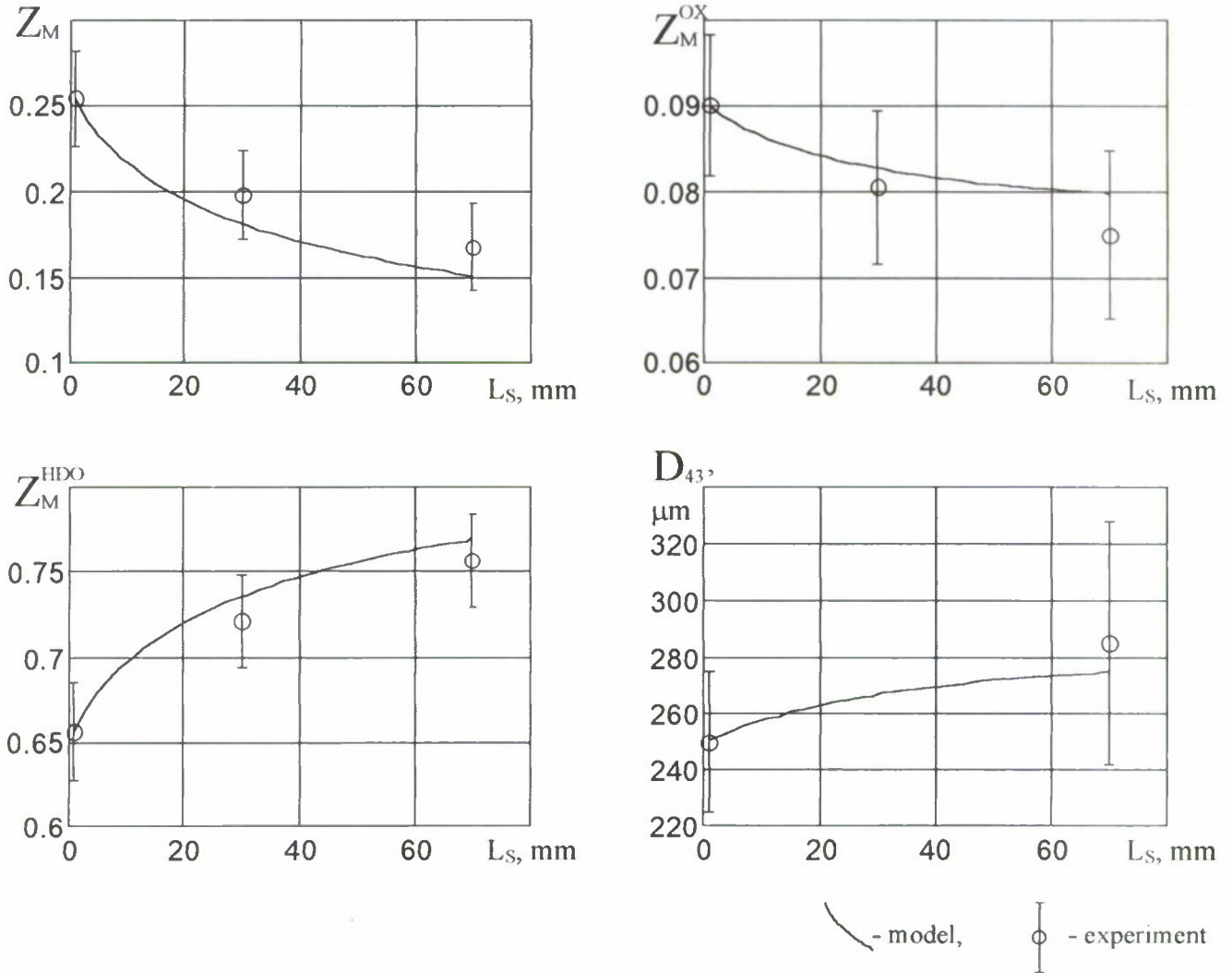


Fig. 1.14. Experimental data for propellant 5, pressure 6MPa.

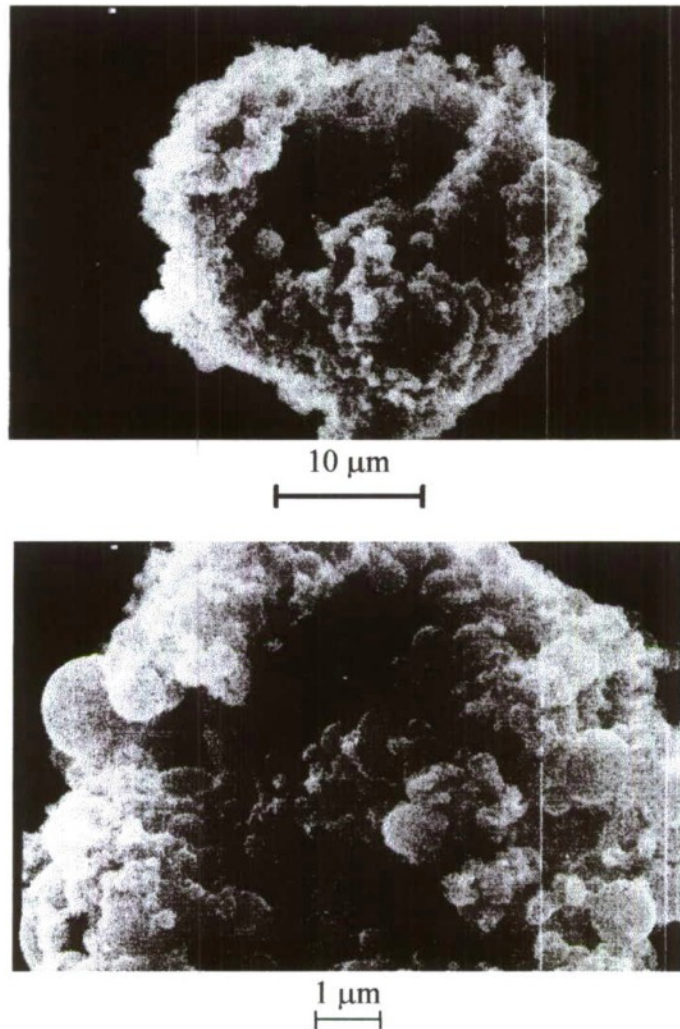
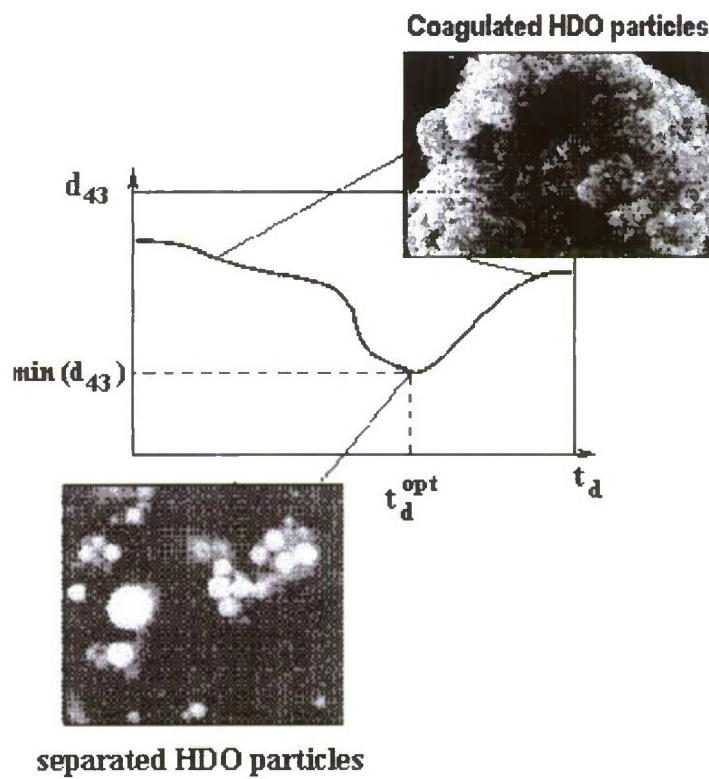


Figure 1.15. Coagulates of HDO particles

Figure 1.16. Change in d_{43} during processing HDO particles with Coulter 4

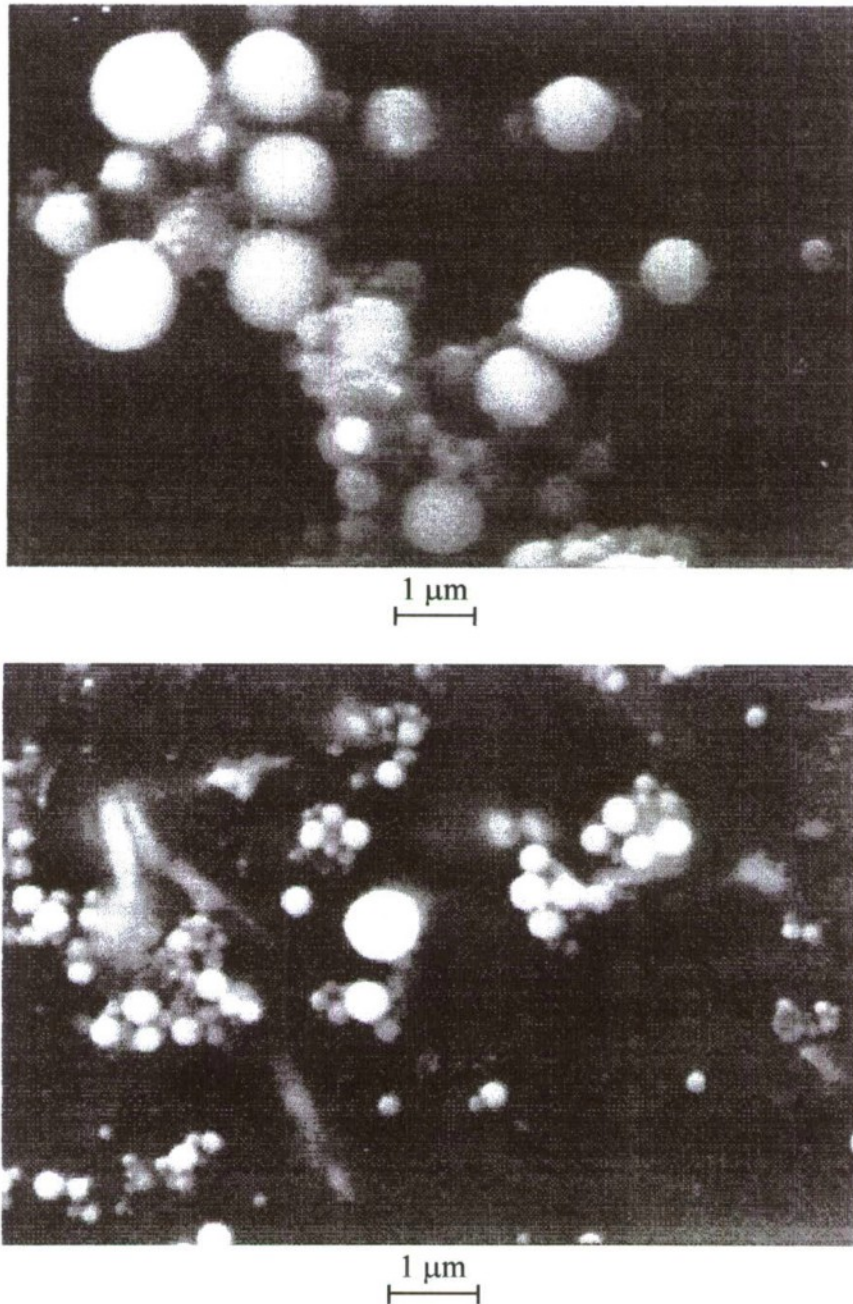


Figure 1.17. HDO particles.

experiments have been carried out in which tubes made of quartz glass and tubes with water frozen on the inner walls («iced» tubes) have been used. The use of tubes of both the first and the second type can result in changing the properties of combustion product flow, and consequently HDO properties. Using the tubes made of quartz glass results in CCP deposition on their walls. (Mass share of depositing agglomerated can reach 40-50%). The «iced» tubes are able to dilute combustion products with water vapors to such extent that it changes the mechanism of the agglomerate evolution (see item 1.2).

The results of experiments give evidence that under conditions of the «iced» tubes the agglomerate evolution parameters are close to the corresponding parameters for conditions of plexiglass and fluoroplastic tubes. Thus, the «iced» tubes can be used to a full extent for estimating the parameters of HDO particles evolution. The use of quartz

tubes is expedient for estimating the character of agglomerate influence on HDO evolution.

The results of the carried out investigation have confirmed the hypothesis about influence of the additional soot formation on the coagulation of the sampled HDO particles. The registered size of HDO particles in case of using quartz and «iced» tubes has proved to be smaller than in case of using plexiglass and fluoroplastic ones (Figure 1.18). In so doing the given effect, as a rule, is more pronounced when quartz tubes are used. It can be supposed that the experimental data obtained with help of «iced» tubes allow one to describe more accurately the HDO evolution process. The information on HDO evolution was given in Figures 1.6 - 1.14 as applied to just those conditions.

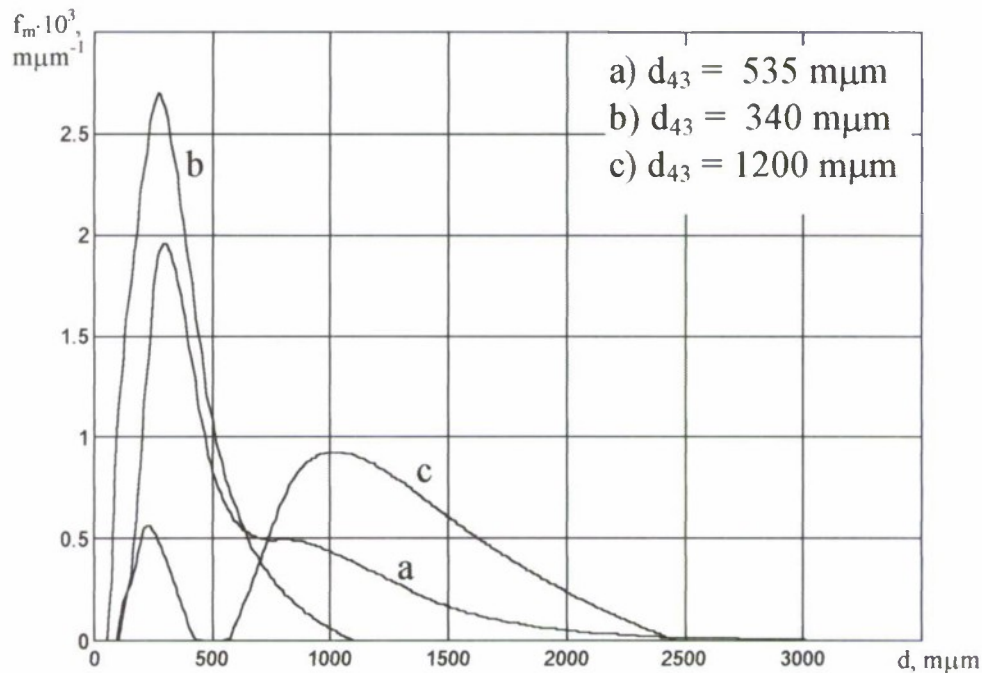


Figure 1.18. HDO particle size distributions at $L_s = 70\text{mm}$ for different tubes used in the experiments: **a** - ice tube, **b** - glass tube, and **c** - plexiglas tube.

The presented results reveal the following regularities of the evolution process:

1. In the process of evolution the quantity of active metal in the flow (Z_m) monotonically decreases, and the quantity of HDO particles (Z_m^{HDO}) grows.
2. A monotonic decrease of oxide content in agglomerates (Z_m^{ox}), that is oxide carrying away, takes place.
3. Mass share (η) of oxide in agglomerates increases.
4. Mass-medium diameter of agglomerates (D_{43}) changes in the evolution slightly.
5. In the process of evolution HDO particles either grow in size or their size practically doesn't change.

The agglomerate structure analysis has made it possible to establish that agglomerates preserve the properties of equilibrium dispersive systems in their evolution. Their internal structure is determined by the surface tension of the substances: aluminum and oxide. The values of contact wetting angles don't practically change and are equal to those given in the previous report [30]. Many investigated agglomerates contain gaseous cavities (bubbles) located at the interface of the substances. The photograph of one of the

agglomerate microsections is shown in Fig 1.19. Besides, in the oxide “caps” of some agglomerates there have been discovered solidified bubbles representing cavities open on the outside of oxide “caps” (Fig 1.20).

On the surface of agglomerate metal facing the gas phase, the tiny oxide particles are found which can be identified as HDO particles of the carrier gas phase (Figure 1.21).

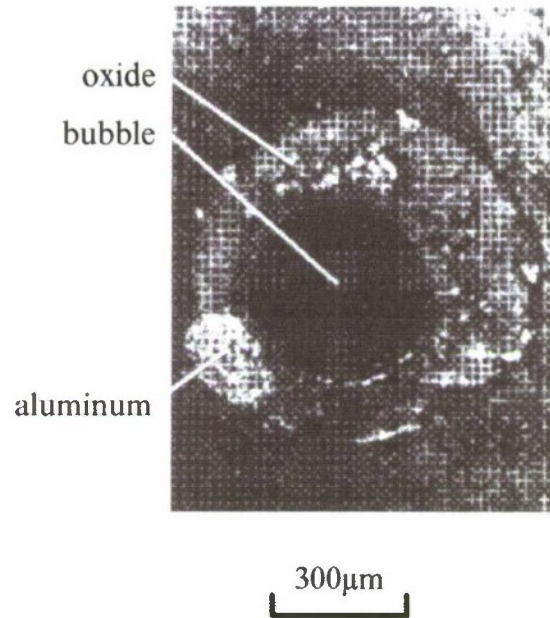


Figure 1.19. Agglomerate microsection.

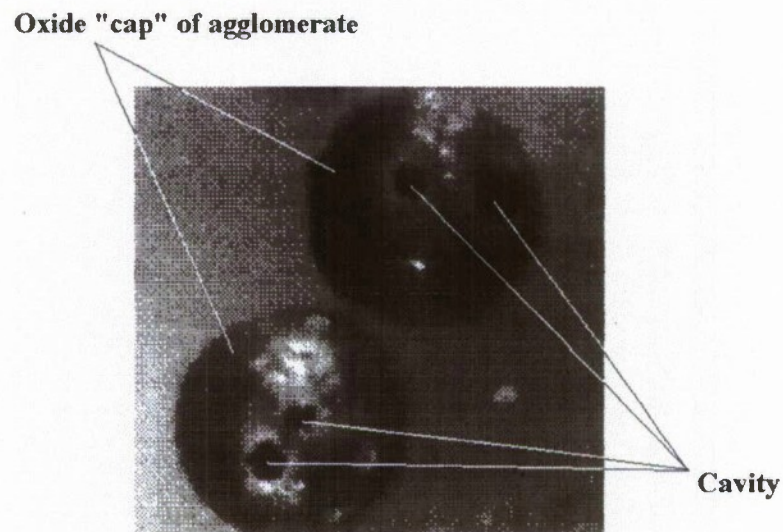


Figure 1.20. Solidified bubbles in oxide «caps».

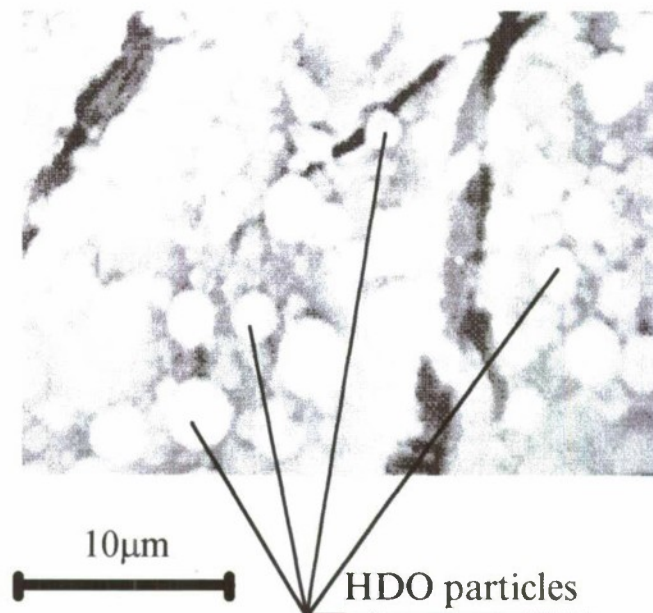


Figure 1.21. HDO particles on the surface of agglomerate metal.

Figures 1.6, 1.8, and 1.14 show character of transformation of the function $f_m(d)$ in the evolution process. In CCP evolution the first mode of HDO particles disappears. Function $f_m(d)$ at $L_s \approx 70\text{mm}$ is either unimodal or bimodal. The second situation is realized in the case when particles formed due to the combustion of metal incorporated in agglomerates of comparatively large size essentially contribute to formation of the entire HDO population. In the case when quantity of agglomerates in the flow is very small (propellant 1, pressure 6MPa), dispersity of HDO particles does not change in the CCP evolution. Availability of HDO particles of rather considerable size (up to $\sim 10\mu\text{m}$) is an objective reality which is confirmed by the results of particles study with help of electronic microscope (Fig. 1.22).

Special experiments to study CCP evolution in high velocity turbulent flow were carried out with the help of the narrowing tubes (Fig 1.3). The conditions most favorable for the process of HDO particles deposition on agglomerates were made up in these experiments. Narrowing of the tube channel begins at the 70 mm distance from the propellant burning surface. At the given cross section of the flow the HDO particles dispersity is less than at the propellant burning surface. The channel geometrical dimensions ensured the gas flow velocity growth up to 110 m/sec. In so doing, the velocities of agglomerates blow-out (ΔV) reach the value of 90 m/sec. Higher magnitudes of these values may result in the gasodynamic breaking down of agglomerates. Thus, the given experiments provide conditions close to speed nonequilibrium limit of the flow which is characteristic of the rocket engine combustion chamber. It should be noted that the use of quite a long tube ($l_1 > 70\text{ mm}$) must result in a perceptible distortion of combustion products flow parameters: changes in composition and temperature of the flow gas phase. But the changes in the indicated properties of the flow are not expected to considerably affect the process of HDO particles deposition on agglomerates.

The above experiments were carried out with propellant 4 at pressure 6 MPa. Their results are shown in Fig 1.23. The experiments (Fig.1.23) show that increase in oxide mass in agglomerates does not take place.

The results of filming of the CCP particles in the gas phase, the same as the data of a number of works [6, 7, 9 et al] give grounds for speaking about so-called "tails" which agglomerates have and which are formed by the products of agglomerates' metal combustion (Fig.1.24). Parameters of a "tail" depend on agglomerate size and properties of the flow which blowing it around. There are some reasons to believe that these parameters determine, to a considerable extent, the properties of HDO particles formed in combustion of agglomerates' metal and then supplied into the carrier gas phase (Fig.1.25).

With account for the fact that many phenomena taking place in the course of the evolution in the gas phase also occur at the burning propellant surface, the results of visualization of particles on this surface are of interest. It should be noted that in a number of cases periodical changes in the size of agglomerating particles (Fig.1.26.) and availability of significant quantity of oxide in their composition have been registered.

The carried out research has yielded a lot of experimental data which allow us to essentially clarify the concept of the CCP evolution process in the rocket engine combustion chamber and to develop more reliable description of the given process.

1.1.5. PHYSICAL CONCEPTS OF CCP EVOLUTION IN THE FLOW OF SRP COMBUSTION PRODUCTS.

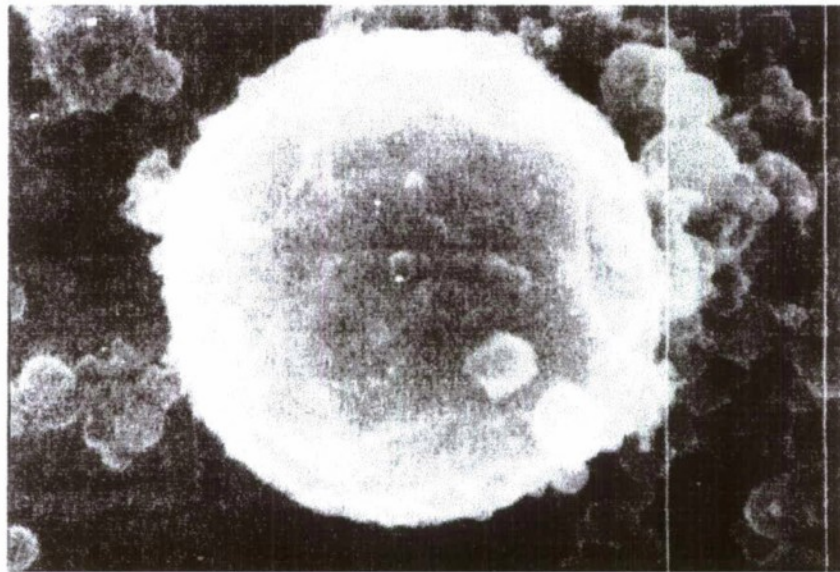
The obtained experimental material makes it possible to present (formulate) a general picture of CCP evolution process when CCP move within the two-phase flow. Both agglomerates and HDO particles take part in the evolution and this process is determined by interphase and interfraction interactions.

The agglomerate metal burns in the gas-phase mode. The combustion process results in the following:

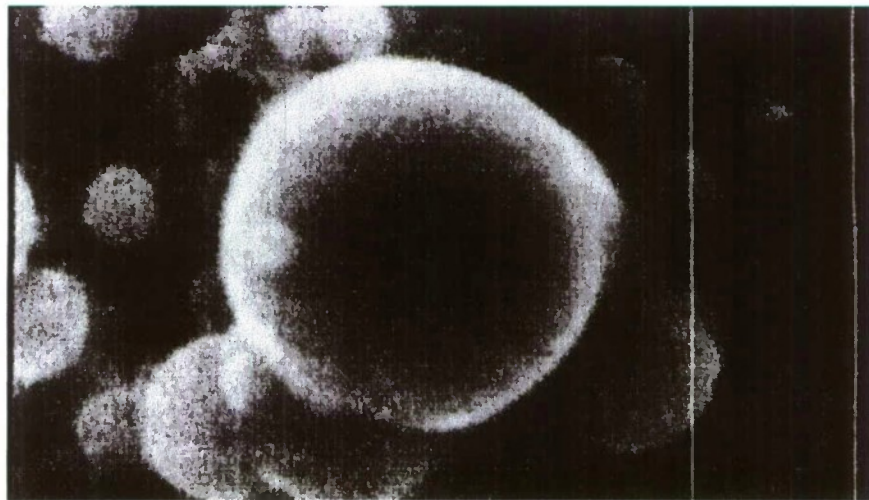
- a certain thermal state of the agglomerate;
- formation of a combustion area where condensed combustion products, HDO particles, are "born".

In interaction of the combustion area with the carrier gas-phase (circumvent flow) a "tail" of combustion products is formed which presents a spatial region within which the intensive transfer of matter, momentum, and energy is going on. The "tail" parameters determine the character of interaction between agglomerate and environment. Within the given region the processes of condensation and coagulation are realized. As the "tail" is breaking down the HDO particles which formed in it become included in the carrier gas-phase. The dispersity of these particles in an essential degree depends on the characteristics of the "tail".

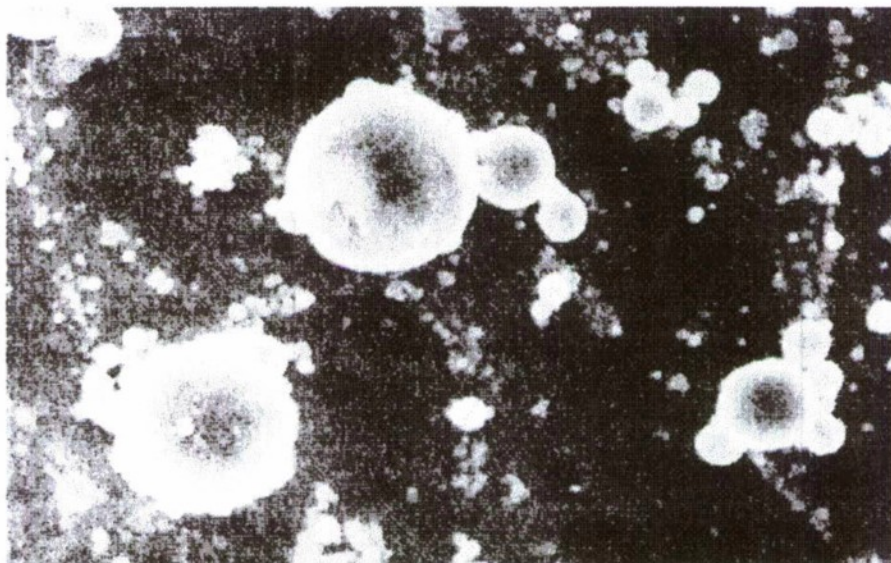
At the temperatures of agglomerates (these temperatures are determined by chemical and thermal interactions) chemical reaction between Al and Al_2O_3 of agglomerates is thermodynamically possible with formation of gaseous products. The given reaction results in formation of gas bubbles on the Al- Al_2O_3 interface, agglomerates blowing up, and, if the conditions are favorable, the break of bubbles through external surface of the "cap" and a release of gaseous products into environment, and, as a consequence, carrying away of agglomerate oxide. The indicated phenomena



1 μm



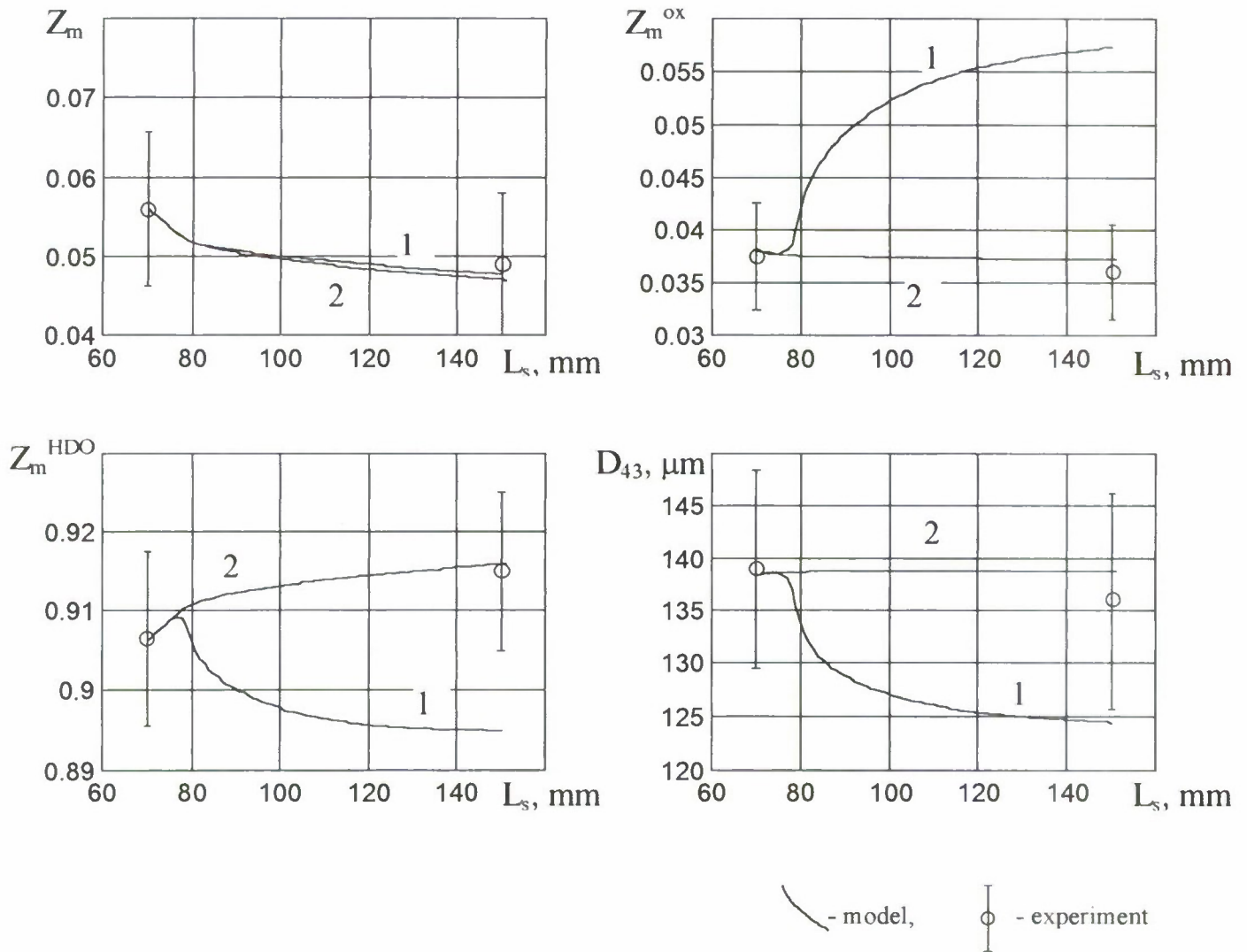
1 μm



10 μm

Figure 1.22. Quite big HDO particles.

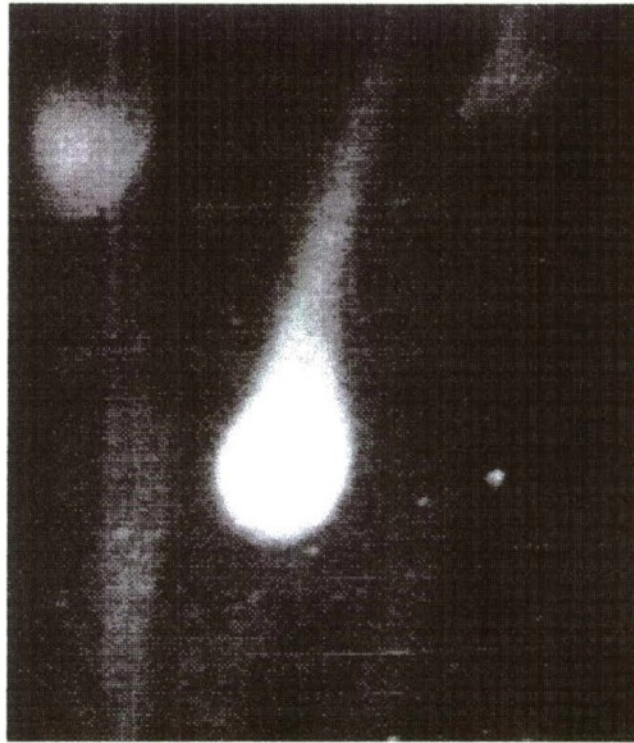
L_s , mm	Z_m	Z_m^{ox}	Z_m^{HDO}	η	D_{43} , μm
70	0.056	0.038	0.906	0.558	139
150	0.049	0.036	0.915	0.580	136



Comments: The calculations are made for two cases:

1. The HDO deposition proceeds in accordance with the deposition model presented in item 1.1.6;
2. The HDO deposition on agglomerates doesn't take place at all.

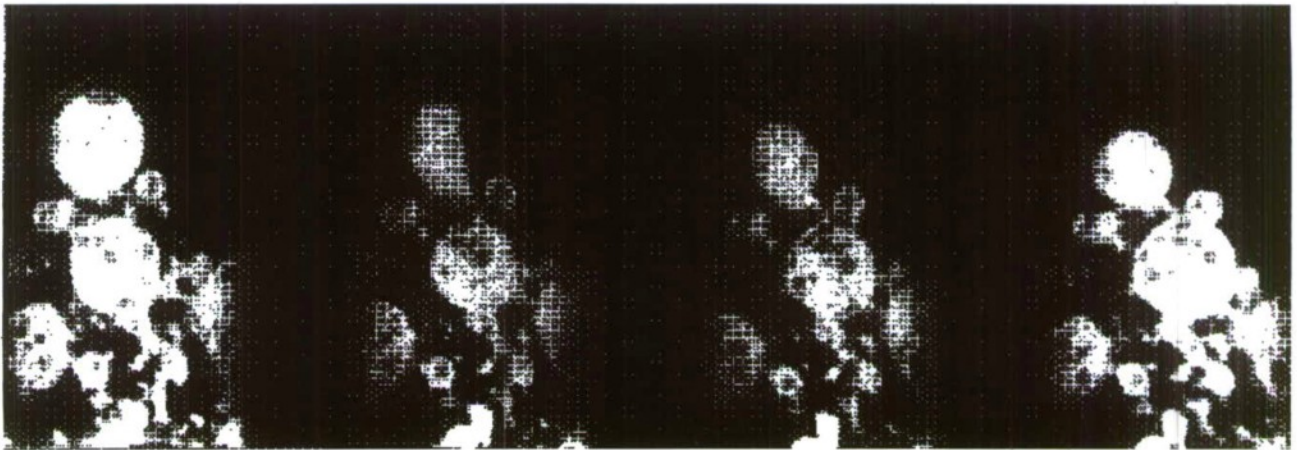
Figure 1.23. Experimental data for propellant 4, pressure 6MPa, narrowing tube.



200 μm

$P = 1.2 \text{ MPa}$

Figure 1.24. Burning agglomerate.



1000 μm

$\Delta\tau = 0.65 \text{ msec}$

Figure 1.26. Agglomerate behavior on propellant burning surface.

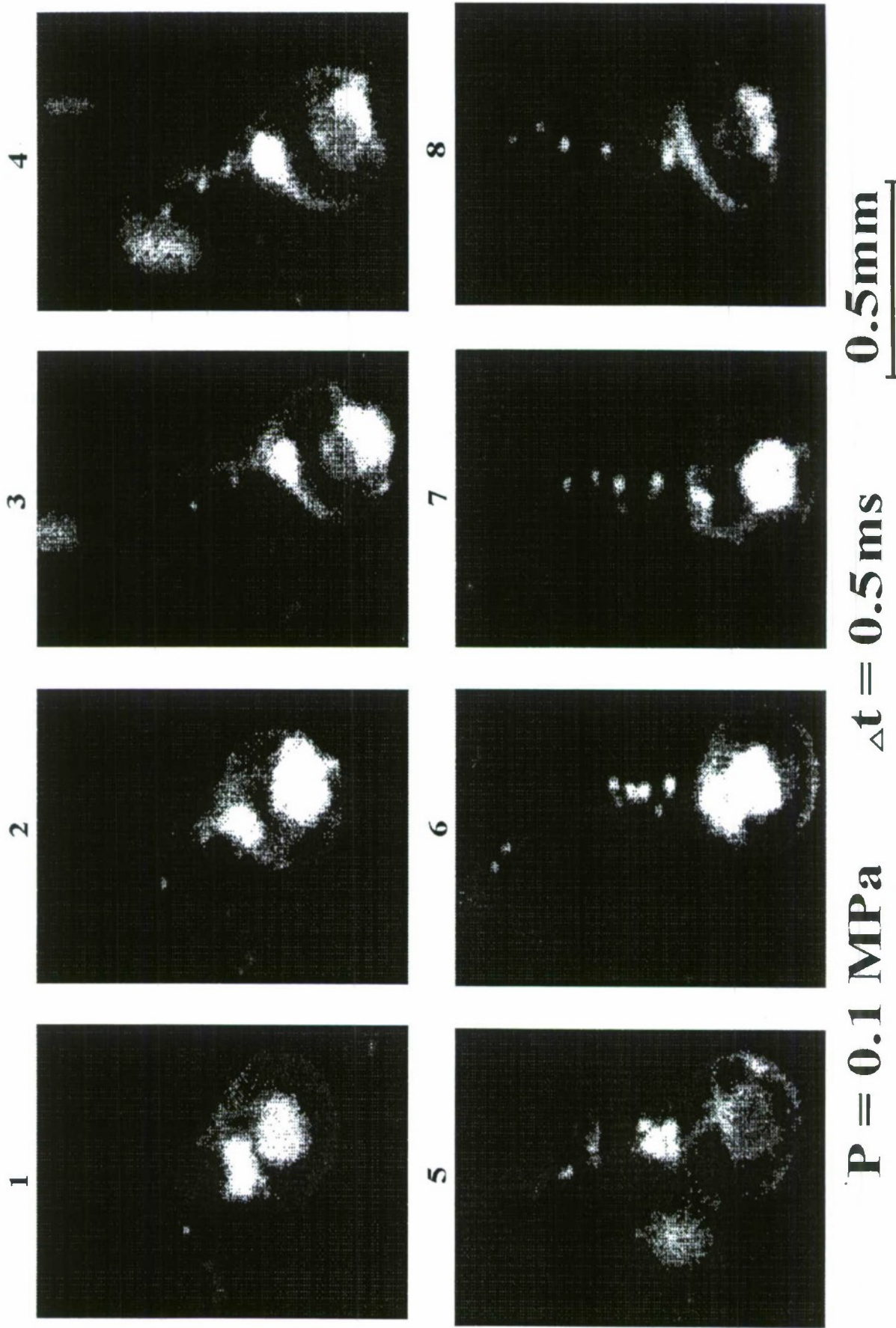


Figure 1.25. Burning of agglomerate when velocity of blowing it around by gas flow is quite low.

can be illustrated by the results of conducted investigation the evolution of both single agglomerates and their populations presented in item 1.1.4.

HDO particles of the carrier gas-phase might deposit on agglomerates, influencing their thermal state. But at the stages of the evolution process, which is the subject of the present research, realization of the given phenomenon doesn't lead to accumulation of oxide in agglomerates.

The internal structure of agglomerates is determined by the presence of metal, oxide, and gas occlusions and by a possibility to use equilibrium approximation.

At the studied stages of the evolution process the agglomerates are oriented in the flow in such a way that the "cap" is turned into direction opposite to the incoming flow. (Realization of that condition is determined by the fact that it is the most stable).

HDO particles at the burning propellant surface are mainly formed as a result of metal combustion which doesn't participate in agglomeration. In so doing, it is most probable that the function of HDO particles size distribution density (f_m) has bimodal character determined by combustion of this metal in two modes [30]. In the evolution process the coagulation of those particles takes place in which the particles forming the first mode participate most actively. With a decrease in countable concentration of particles the intensity of the given process decreases and, beginning at some moment, the effect of that process on HDO dispersity becomes negligible. A decrease in dispersity of HDO particles of the carrier gas-phase also happens due to particles generated during agglomerate metal combustion. These particles size essentially depends on gas dynamic phenomena in the agglomerates and carried gas phase interaction, that is, on agglomerate size, its combustion rate, circumvent flow parameters. There is also a process yielding an increase of dispersity - deposition of the largest HDO particles on the agglomerates. Thus, changes in dispersity of HDO particles of carrier gas phase in the evolution process take place due to the following processes (Fig.1.27):

- particles coagulation apparently caused by Brownian movement (when the flow undergoes turbulization the collisions of turbulent fluctuatory origin become also considerably important);
- deposition of the largest HDO particles on the agglomerates due to their inertness;
- generation of particles during agglomerates metal combustion (each agglomerate is a «local source» of HDO particle generation).

It is obvious that these processes are mutually interacting. The last process is probable to have considerable influence on change in HDO dispersity.

It is interesting to analyze the quality of the earlier developed model of CCP evolution (briefly described in the item 1.1.1) in the light of the obtained experimental material which comprises the information about characteristics of both agglomerates and HDO particles. Comparison of the model analysis results and experimental results allows us to conclude that the developed model describes the phenomena of agglomerate metal combustion, chemical reaction of Al and Al_2O_3 of agglomerates, formation of agglomerate structure and agglomerates' movements with satisfactory accuracy (Fig.1.6-1.14).

It is worth to discuss the problem of modeling the deposition of HDO particles on the agglomerates. The model uses the layout according to which the HDO particles, falling on the surface of agglomerate metal rapidly establish bonds between themselves

and oxide «cap», and then become entrapped by it. The obtained results don't allow us to consider such a layout justified.

Fig. 1.23 presents the results of experiments and of calculations in accordance with the model of evolution mentioned in item 1.1.1. The calculations are made for two cases:

1. For conditions when HDO particles deposition proceeds according to the deposition model; in so doing, it was accepted that dispersity of HDO particles does not change in the evolution (parameter d_{43} is equal to 820nm).
2. For conditions when HDO particles deposition on agglomerates doesn't take place at all.

According to the deposition model parameter Z_m^{ox} should show considerable increase (by 1.5 times approximately) during the evolution in the tube narrow channel. But the experimental results indicate that it doesn't take place. The experimental data are in quite a good agreement with the results of calculations made for the case when HDO deposition on agglomerates doesn't take place.

The analysis of sampled agglomerates surfaces, the results of resolving the problem of flowing around the agglomerate of a gas suspension with HDO particles of the carrier gas phase in the presence of blowing out from its surface - all these give some reasons to believe that in many experimental situations the HDO particles may reach the agglomerate surface (metal drops in the agglomerates). Thus, the HDO particles fall on the agglomerate surface but don't increase the oxide mass in it. As reasons of the given phenomenon might be the following:

- chemical reaction of metal with deposited oxide particles resulting in generation of gaseous products;
- failure of HDO particles to overcome the gas layer between those particles and the agglomerate, and, as a result, an absence of the established contact;
- breaking down of the bridge between the HDO particle and the agglomerate under the effect of tangential component of the HDO particle velocity.

The carried out estimates prove that the most probable reason for lack of oxide accumulation in agglomerates is a chemical reaction between the metal and oxide.

We can believe that at the later stages of the evolution process when the agglomerates change their orientation in the flow (with "cap" facing the incoming flow) the HDO particles deposition will yield the oxide mass growth in agglomerates. In connection with that it becomes necessary to carry out further experimental study of the CCP evolution, including the evolution at these stages.

It is worth to note that the available chemical reaction between the depositing HDO particles and the metal should lead to some changes in thermal state of the agglomerate and appearance of distributed sources of substance and energy near the surface of metal drop, that is, to the phenomena which are not accounted for within the framework of the given model (items 1.1.1.). However, these phenomena apparently don't essentially influence the processes which determine the kinetics of metal combustion and oxide carrying away in the agglomerates, and formation of agglomerates structure and their movement. The consequence of the given circumstance is a quite high degree of agreement between experimental and calculated data on metal combustion and oxide carrying away (mean error in predicting metal combustion is 7% and mean error in predicting oxide carrying away is 8,5%). On the whole, the developed model can be

offered as a "tool" for description of agglomerates properties at those states of the evolution process when the bulk of the agglomerate metal burns out. It should be noted that use of the present study results may lead to more accurate determination of the agreement coefficients in the model of CCP evolution and, as consequence, to increase in accuracy of the prediction (item 1.1.6).

The established laws of the CCP evolution, the obtained experimental material might serve as a basis for development of a model describing the evolution of two-phase flow in the engine chamber, and aimed at obtaining various information on the properties of all the phases and fractions of the CCP.

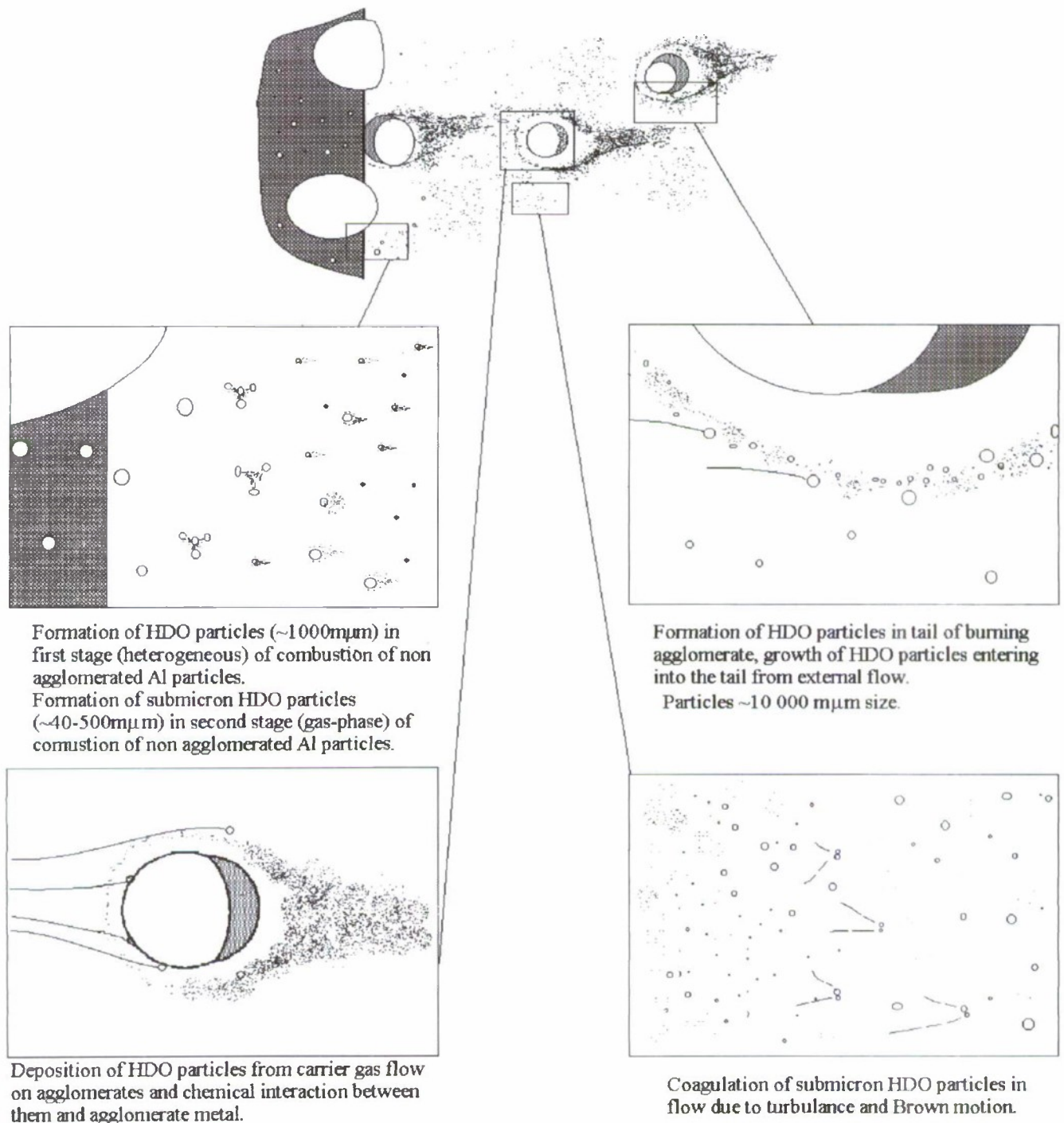


Figure 1.27. Formation and evolution of HDO particles.

1.1.6. IMPROVEMENT OF THE AGGLOMERATES EVOLUTION MODEL.

The developed model for agglomerates evolution [29] ensures predicting various characteristics of the two-phase flow. It is quite natural that this model following accumulation of experimental material should undergo analysis and probably improvement. Below there are described the principal aspects of the given model and the elements of its improvement based on the experimental data obtained within the framework of the present investigation.

In development of the model the principle of decomposition and synthesis of descriptions of separate phenomena has been used. It is based on description of evolution of a single agglomerate. The descriptions of evolution for single agglomerates then synthesized as applied to all agglomerates in combination with description of the environment properties, and that makes it possible to develop the evolution model for population of agglomerates.

Evolution model for single agglomerate.

Single agglomerate evolution model includes the following mathematical descriptions of the following main phenomena taking place in the evolution:

- metal burning;
- chemical interaction between metal and oxide;
- HDO particles deposition on agglomerate;
- changing in agglomerate structure;
- agglomerate movement.

A. Combustion of agglomerate metal.

A great number of experimental data on combustion process of aluminum drops in the environment of solid propellant combustion products have been obtained to date. Their analysis supports the hypothesis of diffusion mode of combustion process running with formation of combustion area of finite dimensions.

The scheme of the model is shown in Figure 1.28.

Modeling of this process is based on the following assumptions:

- The effect of external flow is taken into account in approximation of "reduced film" (zone AC in the Figure 1.28). That is, agglomerate is surrounded by motionless gas AC where heat transfer is carried out by conductivity and mass transfer is carried out by diffusion.
- Metal evaporation takes place in equilibrium mode from the metal drop surface which is free of oxide "cap".
- Burning process is quasi stationary, isobaric, and symmetrical.
- The process is controlled by diffusion, i.e., the chemical reaction rate is much higher than the rate of components diffusion. The result of the given assumption is that all chemical reactions run in a state of equilibrium. In so doing the following schematization of the process becomes justified. Interaction of metal vapors and oxidizing components of environment occurs within an infinitely narrow area B (Fig.1.28). During the given interaction such big amount of energy releases that a part of the

formed oxide is in the gas state and diffuses into environment where its condensation accompanied by H₂O particles formation takes place. The oxide condensation occurs within the area EF (Fig. 1.28). Thus within the area EF there are formed final products of reaction between Al vapors and external gaseous oxidizer, namely: gaseous products of reaction and condensed aluminum oxide in form of H₂O particles. That's why the area EF represents an area of combustion of finite dimensions.

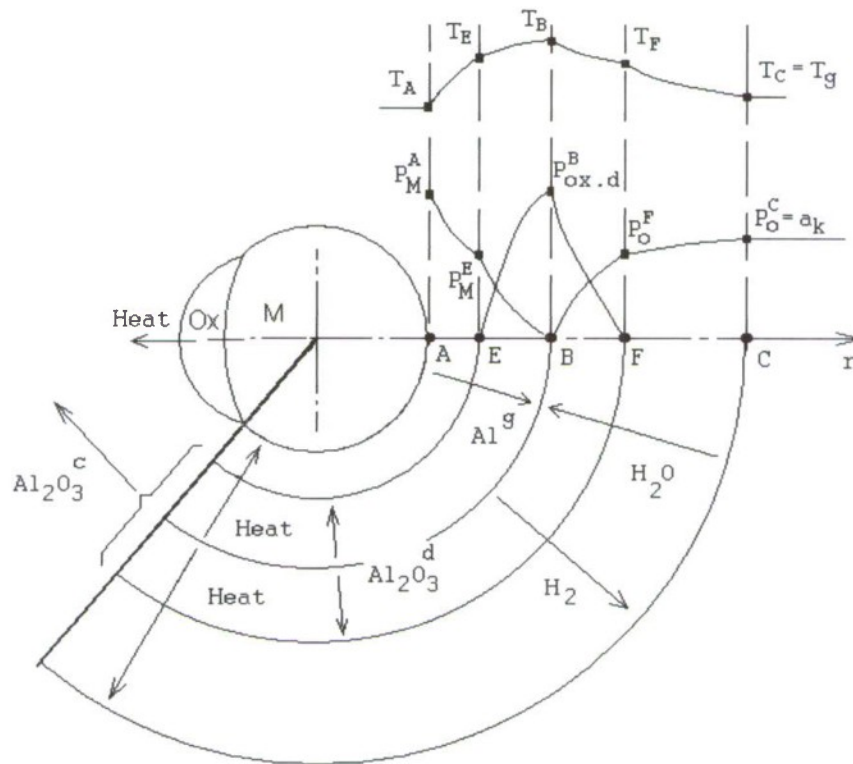
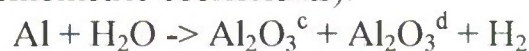


Figure 1.28. Scheme of aluminum combustion model.

- The oxidizing component of environment is water vapors. In the combustion products of SRP the water vapors represent the main oxidizing component as the content of carbon dioxide in them is comparatively low [30]. According to [1] the mixture of CO₂ and H₂O is formally equivalent to the same amount of pure water vapors. In this case the equation of chemical reaction of combustion in area B can be written in form (without account for stoichiometric coefficients):



where Al_2O_3^c and Al_2O_3^d are condensed and dissociated aluminum oxide.

- The equilibrium dissociation of aluminum oxide is described with the following equation (without account for stoichiometric coefficients):



According to the results of thermodynamic estimate it is assumed that from one mole of Al_2O_3 three moles of gaseous substances are formed.

- The products of aluminum oxide evaporation are pseudosubstance - pseudovapor (Al_2O_3^d) which has dualistic properties:

on the one hand, evaporation of one Al_2O_3^c molecule occurs with formation of a few molecules of gaseous substances (i.e., pseudovapor - mixture of different substances);

on the other hand, the products of one molecule evaporation can't be divided and present an integral unity, i.e., the components, which form the mixture of dissociation products, are deprived of any individuality and exist only as components of Al_2O_3^d pseudosubstance.

Pseudovapor is neutral in relation to other components of gas mixture and is capable only of undergoing phase transformation - to condense with formation of Al_2O_3^c . The given assumption allows one to describe quite simply the diffusion and equilibrium condensation of dissociated oxide in area EF (Fig. 1.28).

- Coefficients of transfer in characteristic areas are determined by averaged values of composition and temperature parameters.
- Highly dispersed oxide doesn't influence the diffusion of gaseous substances.
- Binary coefficients of diffusion of all gaseous components are equal in each characteristic area, temperature diffusion does not take place.
- Lewis number is equal to 1. [55].
- The presence of distributed sources of matter and energy in condensation area EF is accounted for by introduction of effective flows.
- HDO particles formed in combustion area EF under effect of gasodynamic forces exerted by the environment are carried out into the «tail» (Fig. 1.28). The present assumption is due to a wish to take into account the real nature of the process, namely, formation of the «tail» saturated with condensed products of agglomerate metal combustion.

The mathematical model is a set of ordinary differential equations describing heat and mass transfer within the «reduced film».

The equation of mass transfer for any i -th component of gas mixture has the following general form:

$$J_i = -\Omega \cdot \left(\frac{\lambda}{c_p} \right) \cdot \left(1 - P_i \frac{\sum_{j=1}^N J_j}{J_i} \right)^{-1} \frac{d\bar{P}_i}{dr} \quad (1.1)$$

where $\bar{P}_i = \frac{P_i}{P}$;

P_i - partial pressure of i -th component;

P - total pressure;

N - number of components for which the mass transfer is carried out through cross-section Ω , including i -th component.

Integration of equations (1.1) with account for boundary conditions:

$$r = R_A : \quad \bar{P}_M = \bar{P}_M^A$$

$$r = R_E : \quad \bar{P}_M = \bar{P}_M^E ; \quad \bar{P}_{\text{ox.d}} = \bar{P}_{\text{ox.d}}^E$$

$$r = R_B : \quad \bar{P}_M = 0 ; \quad \bar{P}_0 = 0 ; \quad \bar{P}_{\text{ox.d}} = \bar{P}_{\text{ox.d}}^B$$

$$r = R_F : \quad \overline{P}_0 = \overline{P}_0^F ; \quad \overline{P}_{\text{ox.d}} = \overline{P}_{\text{ox.d}}^F$$

$$r = R_C : \quad \overline{P}_0 = \overline{P}_0^C = a_k$$

makes it possible to write down 6 equations of components diffusion in characteristic areas, namely:

- two equations for diffusion of metal vapors (in areas AE and EF),
- two equations for diffusion of water vapor (in areas BF and FC),
- two equations for diffusion of dissociated oxide (in areas BE and BF).

The heat situation in characteristic areas is described by four equations of heat balance (conservation of energy) which have the following general form:

$$\sum_{i=1}^n J_i^{\text{in}} H_i - \sum_{j=1}^m J_j^{\text{out}} H_j + Q_c^{\text{in}} - Q_c^{\text{out}} + Q_R^{\text{in}} - Q_R^{\text{out}} = 0 \quad (1.2)$$

Integration of equations (1.2) with the following boundary conditions:

$$r = R_A : \quad T = T_A,$$

$$r = R_E : \quad T = T_E,$$

$$r = R_B : \quad T = T_B,$$

$$r = R_F : \quad T = T_F,$$

$$r = R_C : \quad T = T_C.$$

makes it possible to write down the equations of heat balance for areas AE, EB, BF, and B.

Assuming the equilibrium character of metal evaporation and oxide dissociation, the partial pressures P_M^A , $P_{\text{ox.d}}^E$, $P_{\text{ox.d}}^F$, $P_{\text{ox.d}}^B$ are connected with temperatures T_A , T_E , T_B , and T_F by Clausius-Clapeyron equation.

For partial pressures of dissociated oxide on the boundaries of combustion area EF it has been assumed: $P_{\text{ox.d}}^E = P_{\text{ox.d}}^F = 0,01 \cdot P$

The values of transfer coefficients for gas mixture in the characteristic areas have been calculated on the basis of algorithms and information support [56, 57]. In so doing the dependence of transfer coefficients on the degree of metal fuel burning out which is characterized by parameter $(1 - Z_m)$ has been accounted for.

In the final form the basis of the mathematical model for agglomerate metal combustion process is a set of ten algebraic transcendental equations for heat and mass transfer in the characteristic areas written in the following general form: $F_i = 0$ ($i = 1, 10$). The solving of this set of equations is accomplished with help of optimization procedure [58] by means of minimization of the target function of the following form:

$$\Delta_F = \sum_{i=1}^{10} \left(\frac{F_i}{f_i} \right)^2 \quad (1.3)$$

where f_i - scale factor of function F_i .

Solution of the equation set allows one to determine the burn rate, as well as distribution of temperatures and substances concentrations in the «reduced film».

Complexity of the process description results in difficulties arising in application of comprehensive model of combustion. To determine the rate of aluminum combustion in calculations of agglomerates evolution in the solid propellant combustion products flow a macromodel has been developed. It has been derived from the comprehensive

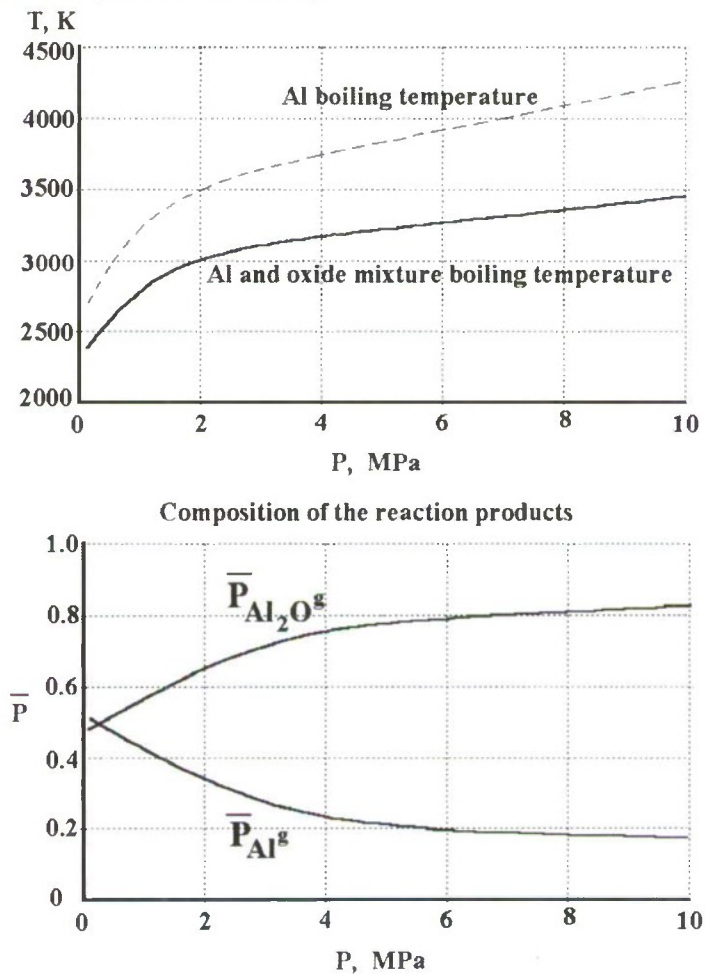
model of combustion, with a number of additional essential assumptions simplifying the process description being introduced. It reproduces very well the principal functional relationships of the comprehensive model and makes it possible to determine the aluminum burn rate as functions of metal drop and oxide "cap" sizes, pressure and oxidizing potential of environments, and velocity of blowing out the agglomerate by the external gas flow. The given macromodel has the following form:

$$J_M = 2\pi R_A \kappa_1 (1 - \chi) \text{Nu} \left(\frac{\lambda}{c_p} \right)_{BC} \left\{ \frac{2}{3} a_k + \left(\frac{c_p}{\lambda} \right)_{BC} \left(\frac{\lambda}{c_p} \right)_{AB} \ln \left(\frac{c_{PM} (T_B - T_A)}{L_M} + 1 \right) \right\} \quad (1.4)$$

where κ_1 is agreement coefficient.

B. Agglomerate oxide removal.

The totality of known experimental data and the results of thermodynamic estimate shown in Figure 1.29 make it possible to see chemical reaction between aluminum and oxide as the most probable cause of a decrease in quality of oxide in agglomerates in the course of their evolution (oxide removal).

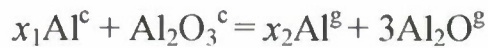


\bar{P}_i - relative partial pressure of i -th component in gas mixture.

Figure 1.29. Results of the thermodynamic estimate for the reaction:



Equation for the chemical interaction may be written as follows:



$$\text{where } x_1 = \frac{\overline{P_{\text{Al}_2\text{O}}} + 3}{\overline{P_{\text{Al}_2\text{O}}}}; \text{ and } x_2 = \frac{3 \cdot (1 - \overline{P_{\text{Al}_2\text{O}}})}{\overline{P_{\text{Al}_2\text{O}}}}$$

A general layout of the process can be presented as follows (Figure 1.30).

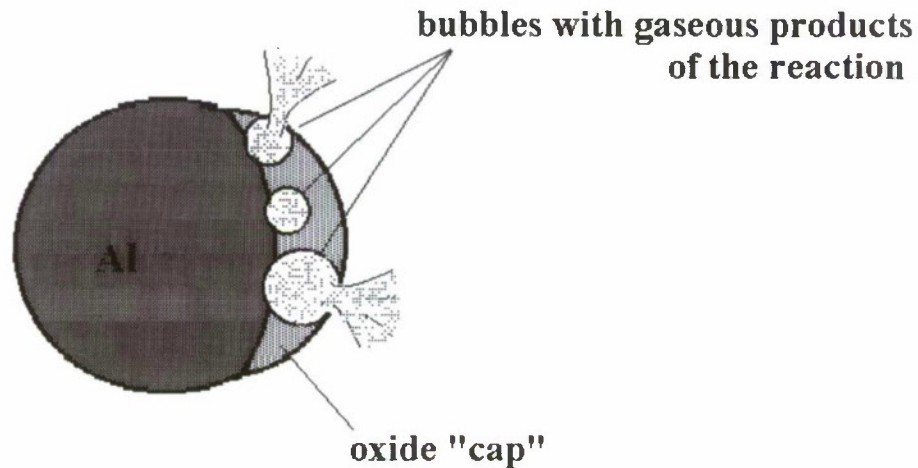


Figure 1.30. General layout of oxide removal from agglomerate.

Chemical reaction runs at the interface of agglomerate substances with formation of gaseous products which form bubbles. The bubbles are retained on the surface of interface by the surface tension forces. The bubbles grow in size and under certain condition can reach the outside boundary of an oxide "cap". In that case bubbles are broken and reaction products are released into environment. The given process causes periodical blowing out of agglomerates, and reduction of oxide and metal content in them.

The experimental study has established that the intensity of oxide removal from agglomerates depends on pressure, agglomerate size, and oxide content in them. On the basis of these data a mathematical model of the process of oxide removal from agglomerates has been developed. It is based on the following principal assumptions:

- Chemical reaction takes place on reaching a certain temperature, it occurs within the adsorption layer which is formed at the interface of metal and oxide in the agglomerate. Bubbles of dissolved gases serve as nuclei of gas formation.
- Primary bubbles of reaction products grow and fuse. It yields rather big bubbles going far beyond the boundaries of the adsorption layer.
- Each large growing bubble has a corresponding area of feeding - an adsorption layer area surrounding that particular bubble and having constant radius R_f . The products of reaction running within this area are fed into the bubble. Let's also assume that value R_f doesn't depend on pressure and evolution conditions.
- Initial bubbles size doesn't exceed the thickness of adsorption layer. Their fusion with large central bubble is a «fast» process.

- In a large bubble gone beyond the boundaries of adsorption layer a reverse process takes place - the process of condensation of the gaseous reaction products accompanied by formation of condensed aluminum and its oxide on the internal surface of bubble.
- The condensation process involves only some part of bubble internal surface. The portion of that surface doesn't change in the process of bubble growth.
- On the whole, the bubble growth process is determined by simultaneous running (competing) of gas formation and condensation processes.
- In case when the bubble reaches the external boundary of oxide "cap" the former breaks down and releases gaseous products into environments. Otherwise, only the agglomerate blowing out occurs.
- Oxide «cap» thickness is the same for all the bubbles and equal to the effective value, conditions of bubbles growth are the same;
- Bubbles shape is spherical;
- The temperature is the same throughout the agglomerate cross-section;
- Products of the reaction are ideal gas.

The accepted assumptions allow one to give the following mathematical description of the process.

The expression for estimating Al_2O_3^c evaporation rate (w_1) is formulated in form of evaporation law:

$$w_1 = A_1 \exp\left\{-\frac{E_a}{R_0 T_A}\right\} \frac{1}{\sqrt{T_A}} (P_S - P) , \quad \frac{\text{mole of Al}_2\text{O}_3^c}{\text{m}^2 \cdot \text{sec}} \quad (1.5)$$

where A_1 - factor depending on properties of evaporating mixture of substances,

E_a - energy of the process activation,

P_s - pressure of saturated vapor of interaction products.

The value of P_s is determined by agglomerate temperature T_A .

The equation for estimating the rate of condensed oxide formation on the internal surface of the large-size growing bubble (w_2) has the form:

$$w_2 = A_1 \exp\left\{-\frac{E_a}{R_0 T_A}\right\} \frac{1}{\sqrt{T_A}} (P - 0) , \quad \frac{\text{mole of Al}_2\text{O}_3^c}{\text{m}^2 \cdot \text{sec}} \quad (1.6)$$

The resultant rate of Al_2O_3^c consumption in the process of bubble growth:

$$w_\Sigma = w_1 \Omega_1 - w_2 \Omega_2 \quad (1.7)$$

where Ω_1 - dimensions of replenishment area,

Ω_2 - area of internal surface of the bubble whereupon the condensation of reaction products takes place.

The law of single large-size bubble growth has the form:

$$\frac{dV_b}{dt} = \frac{R_0 T}{P} w_\Sigma (3 + x_2) \quad (1.8)$$

where $(3 + x_2)$ - mole number of gaseous products which are formed as a result of reaction with involvement of 1 mole of Al_2O_3^c .

The time of single large-size bubble growth till the moment of its breaking:

$$t = \int_0^{R_b^{\max}} 4\pi R_b \frac{P}{R_0 T_A} \cdot \frac{1}{w_{\Sigma}(3+x_2)} dR_b \quad (1.9)$$

where R_b^{\max} - maximum radius of the bubble at the moment of its breaking.

The rate of reaction products removal from a unit of surface area of $Al^c - Al_2O_3^c$ contact in agglomerate (W):

$$W = \frac{V_p^{\max}}{t\pi R_f^2}, \frac{m^3 \text{ of the reaction products}}{m^2 \cdot \text{sec}} \quad (1.10)$$

The final expression for estimating the rate of $Al_2O_3^c$ removal (J_{ox}) is derived in the following form:

$$J_{ox} = \frac{P}{R_0 T_A} W \frac{1}{3+x_2} \Omega_{M-ox}, \frac{\text{mole of } Al_2O_3^c}{\text{sec}} \quad (1.11)$$

where Ω_{M-ox} - area of $Al^c - Al_2O_3^c$ contact surface in agglomerate.

The given model accounts for physical principles of agglomerate oxide removal process and allows one to take into account how the oxide removal rate is affected by various factors, such as pressure P and parameters of agglomerate (R_A , η , and T_A).

The model contains a number of parameters the values of which can't be estimated exactly. They include, for example, such constituents of the above equations as coefficient A_1 , activation energy E_a , radius of replenishment area R_f , et al. The given parameters serve as agreement coefficients.

The results of modeling can be seen in Figure 1.31 which shows the conditions for realization of oxide removal under the following conditions: oxidizing potential of environment $a_k=0.2$, velocity of agglomerate blowing out $\Delta V=2$ m/s, surrounding gas temperature $T_g=3300K$, $\eta=0.5$ (for picture *a*) and $P=6MPa$ (for picture *b*).

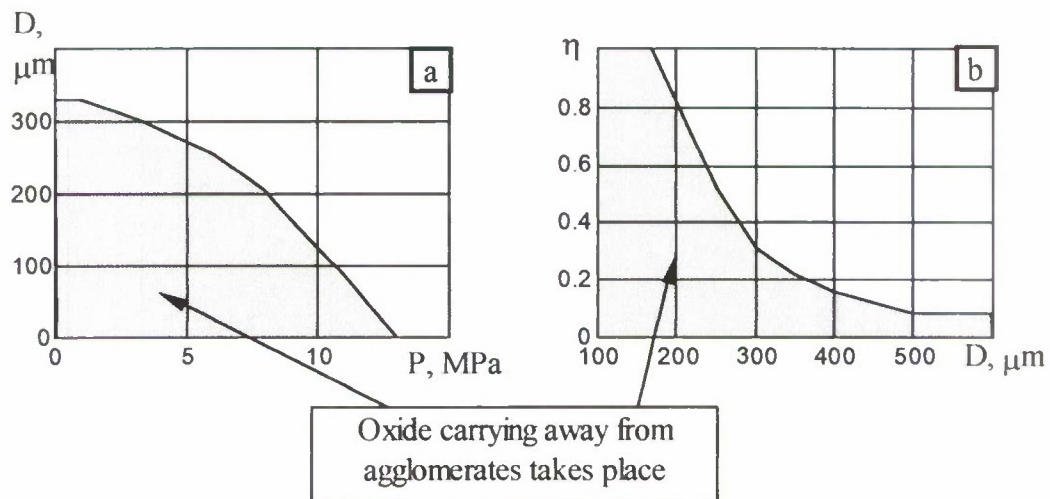


Figure 1.31. Results of modeling the oxide removal process.

The bubbles of gaseous reaction products are almost all found in agglomerate oxide «cap» therefore causing its increase in volume. That factor is accounted for by using the effective value of «cap» density (ρ_{cap}). On the basis of the conducted estimates it has been assumed that the effective density of burning agglomerate «cap» is less by approximately 40% in comparison with the density of liquid oxide (ρ_{ox}) taken at temperature T_A .

$$\rho_{\text{cap}} = 0.6 \rho_{\text{ox}}$$

The removal of chemical interaction products from agglomerate is a reason for both the oxide removal and additional removal of metal involved in that interaction. Additional evaporation of metal due to chemical interaction with oxide may come up to 30% as compared with metal combustion. The mole flow of additionally evaporating metal (J_{M+}) is estimated in the following way:

$$J_{M+} = x_1 J_{\text{ox}}$$

C. Deposition of HDO particles on agglomerates.

While investigating the CCP evolution in the solid propellant combustion products flow we can't but account for probable collisions of condensed phase particles and, first of all, HDO particles with agglomerates. One of the probable results of such collisions may be the deposition of HDO particles on agglomerates and, as a result of it, an increase of oxide mass in them.

To account for the given process a model of HDO particles collisions with agglomerates has been developed. It is based on the following principal assumptions:

- Agglomerate can be in various positions in the flow relatively to direction of its blowing out (Fig.1.32). Orientation of agglomerate is determined by the mutual positions of its mass and pressure centers. The calculation results presented in Fig.1.33 allow us to suppose that at the initial stages of evolution the agglomerate blowing out occurs from the direction of metal drop. But at the final stages, when a certain magnitude of η is reached, blowing out comes from the direction of oxide «cap». In carrying out these calculations the blowing up of oxide «cap» with bubbles containing gaseous products of chemical reaction between metal and its oxide has been accounted for. Figure 1.33 shows that the agglomerate blowing up, characterized by the value of «cap» mean density and oxide density ratio (parameter ψ in Fig.1.33), affects the agglomerate orientation. On the basis of carried out estimates it has been accepted in this model: $\psi=0,6$.
- In blowing out of agglomerate by external gas flow from the direction of metal drop the conditions for its collision with HDO particles deteriorate due to blowing in of metal vapors from the surface of agglomerate metal drop.
- The gas flowing around the agglomerate is viscous and incompressible.
- HDO particles size is the same and equal to typical size.

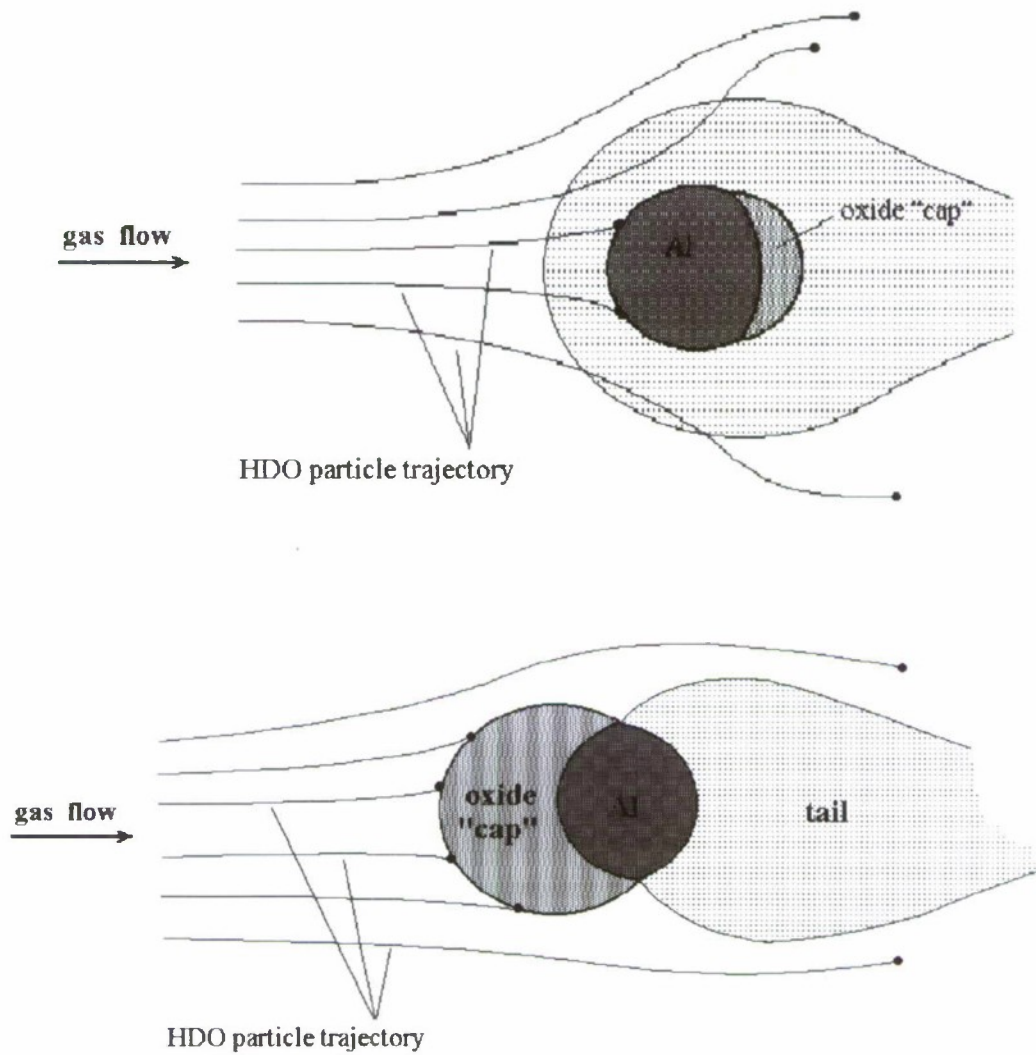


Figure 1.32. General layout of HDO particles deposition on agglomerate.

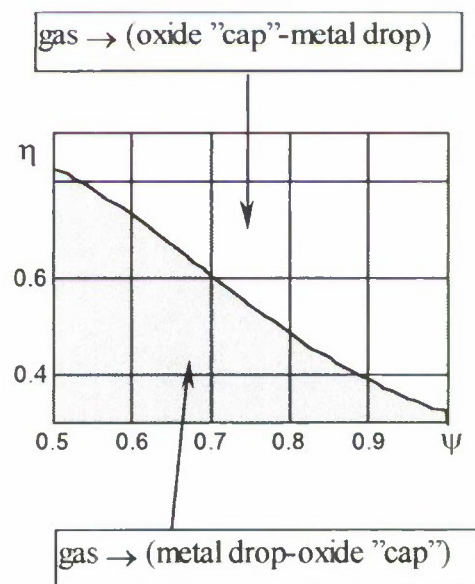


Figure 1.33. Results of calculations for agglomerate orientation in the external flow at various conditions.

Mathematical description of the process is constructed in the following way. First, we determine the field of gas phase velocities in the region disturbed by the agglomerate, then, calculate the trajectories of HDO particles movement in this region.

In the system of coordinates connected with agglomerate the equations of HDO particle motion written in the vector form are as follows:

$$\begin{aligned} \frac{d\vec{V}_p}{dt} &= \vec{F}_p \\ \frac{d\vec{l}_p}{dt} &= \vec{V}_p \end{aligned} \quad (1.12)$$

where \vec{V}_p - HDO particle velocity;

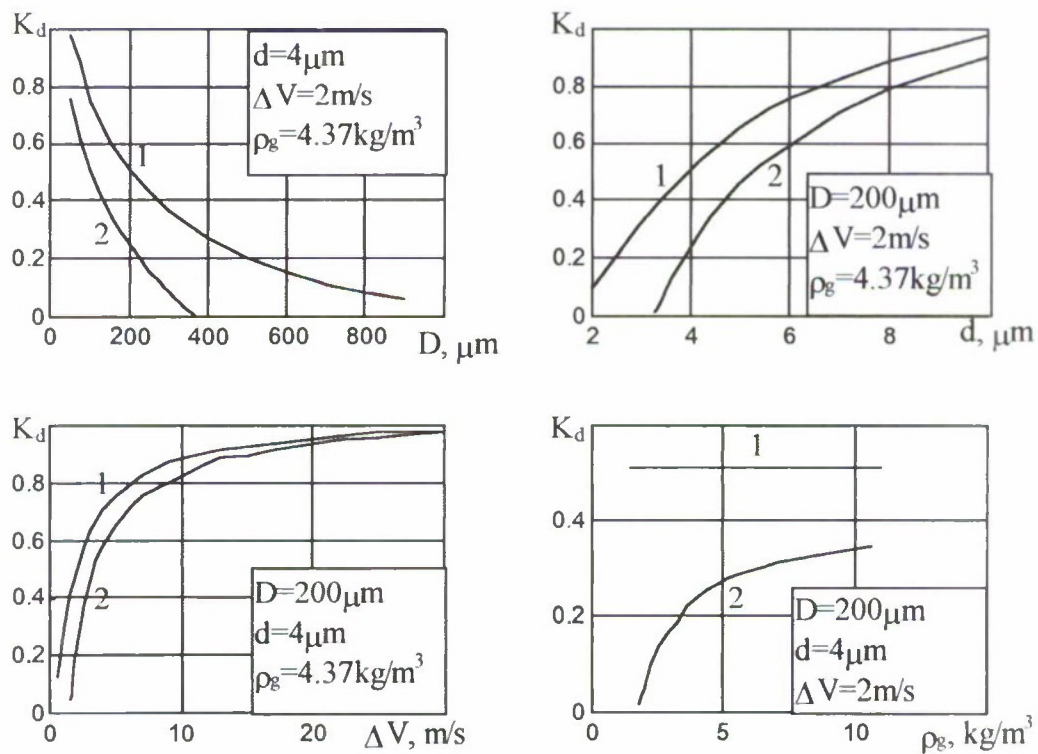
\vec{F}_p - aerodynamic force applied to HDO particle from the side of gas flow,

\vec{l}_p - movement of HDO particle.

It is assumed that HDO particle collision results in its deposition on the agglomerate. The calculations are made for two variants of agglomerate orientation in the gas flow.

On the basis of the given calculations we find the rate of capturing HDO particles by the agglomerate depending on orientation and diameter (D) of agglomerate, its blowing out velocity (ΔV), HDO particle size (d), and gas density (ρ_g).

Fig.1.34 shows influence the above parameters on coefficient of HDO particles capturing by agglomerate (K_d).



Agglomerate orientation in the flow: 1 - gas->(oxide «cap»-metal drop)
2 - gas->(metal drop-oxide «cap»)

Figure 1.34. Dependencies of K_d on D , ΔV , d , ρ_g , and agglomerate orientation.

The presented comprehensive deposition model is quite bulky and therefore it is not convenient for calculating the agglomerate population evolution. For that purpose a formal macromodel has been developed with help of self-adjusting method [32] on the results of parametric analysis of a comprehensive model for HDO particles deposition on agglomerates.

D. Agglomerate structure.

The agglomerate structure model is based on the previously formulated statement that the agglomerate residing in the flow of solid propellant combustion products can be regarded as an equilibrium dispersible system. In this case its internal structure is determined by the surface properties and quantities of substances entering into the composition of agglomerate: metal, oxide, and gaseous products of their interaction.

Other assumptions are as follows.

- The temperature of agglomerate is the same at its any point.
- The density of oxide «cap» is equal to the effective value $\rho_{\text{cap}} = 0.6 \rho_{\text{ox}}$. (The given assumption allows for the presence of gas inclusions).
- Agglomerate diameter corresponds to the biggest of the two: that of metal drop or oxide drop in agglomerate.

The diagram of agglomerate structure is shown in Figure 1.35.

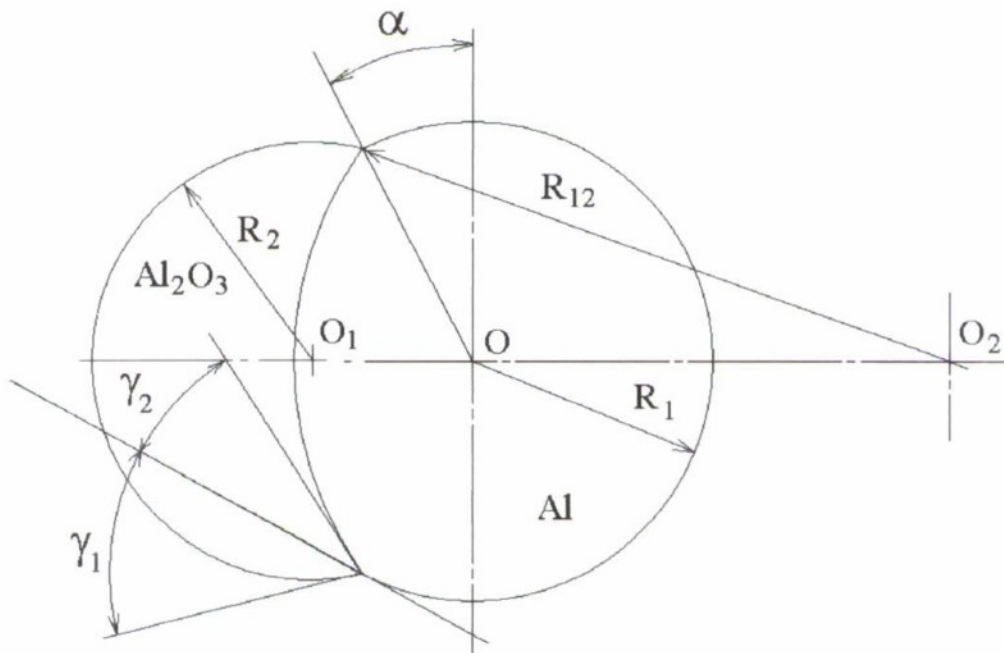


Figure 1.35. Scheme of agglomerate structure.

To calculate structure parameters the following initial information is used: oxide share in agglomerate η , agglomerate diameter D , and edge wetting angles γ_1 and γ_2 [30].

The algorithm for calculating the agglomerate structure parameters consists in the following:

1) to find the value of angle α (Fig. 1.35) as a function of parameter η value by resolving a set of equations:

$$\frac{V_{\text{cap}}}{V_M} = \frac{\rho_M}{\rho_{\text{cap}}} \cdot \frac{\eta}{1 - \eta}$$

$$V_{\text{cap}} = \frac{\pi}{3} R_1^3 \left\{ \frac{\cos^3 \alpha}{\cos^3(\alpha - \gamma_1)} \cdot [1 - \sin(\alpha - \gamma_1)]^2 \cdot [2 + \sin(\alpha - \gamma_1)] - \frac{\cos^3 \alpha}{\cos^3(\alpha + \gamma_2)} \cdot [1 - \sin(\alpha + \gamma_2)]^2 \cdot [2 + \sin(\alpha + \gamma_2)] \right\} \quad (1.13)$$

$$V_M = \frac{\pi}{3} R_1^3 \left\{ [1 + \sin \alpha]^2 [2 - \sin \alpha] + \frac{\cos^3 \alpha}{\cos^3(\alpha + \gamma_2)} \cdot [1 - \sin(\alpha + \gamma_2)]^2 \cdot [2 + \sin(\alpha + \gamma_2)] \right\}$$

where V_M and V_{cap} - volumes of metal drop and oxide «cap» in agglomerate.

2) to find all the other parameters of agglomerate structure by the known value of angle α .

$$\chi = 0.5 (1 - \sin \alpha)$$

$$\text{If } \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} > 1, \text{ then } \begin{cases} R_2 = 0,5D \\ R_1 = R_2 \frac{\cos(\alpha - \gamma_1)}{\cos \alpha} \end{cases}$$

$$\text{If } \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} \leq 1, \text{ then } \begin{cases} R_1 = 0,5D \\ R_2 = R_1 \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} \end{cases} \quad (1.14)$$

$$R_{12} = R_1 \frac{\cos \alpha}{\cos(\alpha + \gamma_2)}$$

The volumes V_M and V_{cap} are found in accordance with the above-written expressions (1.13).

E. Agglomerate movement.

The modeling of the given process is carried out in accordance with the approach suggested in [31]. The movement model describes the movement of a single agglomerate in the gas flow under effect of aerodynamic and mass forces. Deposition of HDO particles out of the external flow on agglomerate is a reason of additional change of agglomerate impulse. The movement model takes into account this factor. In so doing it is assumed that the difference in agglomerate velocity and that of HDO particles colliding with it is equal to the difference in velocity of agglomerate and that of external flow (ΔV).

Motion equations in the vector form are as follows:

$$\frac{d\bar{V}_a}{dt} = \frac{1}{2} \cdot \frac{C_a S_a \rho_g \Delta V}{M_a} \Delta \bar{V} + \bar{F}_m + \frac{K_d \cdot \gamma_{HDO} \cdot S_a \cdot \Delta V}{M_a} \Delta \bar{V} \quad (1.15)$$

$$\frac{d\bar{l}_a}{dt} = \bar{V}_a$$

where C_a - coefficient of agglomerate aerodynamic drag,

S_a - area of agglomerate maximum midsection,

F_m - mass force induced by the Earth gravity and rocket acceleration,

γ_{HDO} - mass concentration of HDO particles in the gas flow,

l_a - movement of agglomerate.

As shown by calculations, the effect of jet power, caused by outflow of metal vapors from the agglomerate surface, on agglomerate movement is unessential and it can be neglected.

Synthesis of models describing metal combustion, oxide removal, HDO particles deposition, agglomerate structure formation, and agglomerate movement provides for development of single agglomerate evolution model. The use of this model for agglomerate population if a description of carrier gas phase is available allows to carry out modeling evolution for the entire agglomerates population.

Evolution model for population of agglomerates.

The use of the model for a single agglomerate evolution makes it possible to describe the evolution process for the population of agglomerates in a random three-dimensional flow of SRP combustion products. Modeling the evolution of agglomerate population presumes that it is necessary to describe their interaction with the carrier gas phase, namely, exchange of energy, matter, and momentum. As a result of such an interaction the properties of agglomerates and carrier gas phase change.

The totality of phenomena taking place in the agglomerates evolution leads to the following changes in the properties of the carrier environment: rise in temperature, a drop in oxidizing potential (content of oxidizing components), an increase in HDO particles concentration. The temperature and oxidizing potential of the gas phase are determined by the degree of metal burning out and amount of oxide incorporated in agglomerates in form of «caps».

Within the framework of the given investigation the agglomerates evolution has been studied under conditions of linear flow of combustion products. To compare with the experimental results there has been accomplished the modeling of agglomerate population evolution under mentioned conditions. The developed model for agglomerate population evolution in the linear flow of SRP combustion products quite comprehensively accounts for phase interaction and allows one to calculate characteristics of the flow at any cross-section. The given model is based on the following main principles:

- The flow of combustion products is quasi-steady and adiabatic.
- Agglomerates are presented by set of fractions. There is no interaction between agglomerates in the flow.
- HDO particles form monofraction of particles which have size equal to d_{43} .

- Composition and temperature of gas phase at each cross-section of the flow correspond to thermodynamically equilibrium state and are determined by propellant properties and content of active metal at the given cross-section of the flow (parameter Z_m) [30].
- HDO particles are in complete thermal and velocity equilibrium with gas. The concentration of HDO particles is determined by parameter Z_m^{HDO} .

The use of the given model allows one to carry out a comparison of modeling results with experimental ones, to find the values of agreement coefficients, and to assess the quality level of the process modeling.

More precise definition of the evolution model.

The carried out experimental investigation of CCP evolution in the flow of SRP combustion products has allowed to obtain quite big volume of experimental data. Those data complement previously obtained information which has served as a base for development of the agglomerates evolution model. Newly obtained information can be used for more precise definition of the model. More precise definition regards the problems of HDO particles deposition on agglomerates and estimating the model agreement coefficients.

A. Improvement of the model for deposition of HDO particles on agglomerates.

To improve the given model some special experiments have been carried out to study the evolution under conditions of high-velocity flow of combustion products which have been created with help of narrowing tubes (items 1.1.3, 1.1.4, 1.1.5). The analysis of those experiments results and other experimental information carried out in the mentioned items has permitted to establish the following. HDO particles can deposit on agglomerates. But under conditions of the carried out experiments the given phenomenon doesn't result in accumulation of oxide incorporated in agglomerates. Most probably it can be explained by evaporation of HDO particles deposited on agglomerates due to their chemical interaction with agglomerate metal. Taking into account those conclusions at the present stage of CCP evolution process investigation, it can be concluded that under conditions, when HDO particles deposit onto the metal of agglomerates, the oxide accumulation in agglomerates doesn't take place. So, under mentioned conditions the given phenomenon and the corresponding model should be neglected in the agglomerates evolution modeling.

B. More precise definition of model agreement coefficients.

The evolution model contains a number of parameters which are determined by means of agreement with experimental data. The values of those parameters (agreement coefficients) have been estimated on the basis of quite a big volume of experimental information obtained before. That information has been essentially complemented by the results of experiments conducted within the framework of the present study. The use of the total accumulated to date experimental data, including previously and recently obtained information, allows to define more precisely the agreement coefficients of the model.

The technique for defining the values of agreement coefficients consists in the following. Determination of agreement coefficients values is done with help of optimization procedure. In so doing, the agreement coefficients play role of variable parameters. The target function is parameter Δ^Σ representing relative disagreement of experimental and calculated data characterizing the evolution process. As those data there are used the values of parameters Z_m and Z_m^{ox} corresponding to sampling cross-section of 70mm. Parameter Δ^Σ is defined on the basis of the total available experimental data as follows.

$$\Delta^\Sigma = \sum_{i=1}^N \left\{ \frac{[Z_m(70\text{mm})]_i^{\text{model}} - [Z_m(70\text{mm})]_i^{\text{exp}}}{[Z_m(70\text{mm})]_i^{\text{exp}}} \right\}^2 + \sum_{i=1}^N \left\{ \frac{[Z_m^{\text{ox}}(70\text{mm})]_i^{\text{model}} - [Z_m^{\text{ox}}(70\text{mm})]_i^{\text{exp}}}{[Z_m^{\text{ox}}(70\text{mm})]_i^{\text{exp}}} \right\}^2$$

where N - total number of investigated cases of agglomerate evolution;

$[Z_m(70\text{mm})]_i^{\text{model}}$, $[Z_m^{\text{ox}}(70\text{mm})]_i^{\text{model}}$ - calculated data estimated with help of the evolution model;

$[Z_m(70\text{mm})]_i^{\text{exp}}$, $[Z_m^{\text{ox}}(70\text{mm})]_i^{\text{exp}}$ - experimental data.

According to the indicated technique the more precise determination of the agreement coefficients has been carried out. It can be noted that the values of the agreement coefficients have changed unessentially (not more than by 10%). Newly obtained agreement coefficients values ensure more accurate agreement of modeling results and the experimental data obtained in the framework of the present study (item 1.1.4). Thus, the mean error in predicting metal combustion has decreased from 7% down to 5% and the mean error in predicting oxide removal has decreased from 8.5% down to 7% as a result of more precise defining of the agreement coefficients.

1.2. EVOLUTION OF CCP IN COLD OXIDIZING ENVIRONMENTS.

To describe the process of MF combustion as a component of SRP there are used various experimental data obtained in setups of different types including those where cold gases are employed as oxidizing environment. It seems of a considerable interest to assess how the properties of this environment influence various aspects of combustion process, and consequently a possibility to apply the obtained results in conditions of a rocket engine.

1.2.1. ANALYSIS OF LITERATURE.

FORMULATION OF THE INVESTIGATION TARGET.

Appearance of metallized SRP has given a stimulus to carrying out intensive investigations of Al particle combustion in various environments [1 et al.] All the elaborated techniques for experimental investigation can be conventionally divided into two groups:

- techniques ensuring determination of characteristics of metal combustion as a component of SRP;
- techniques based on creation of special conditions for metal combustion which consist, as a rule, in using comparatively cold (in comparison with conditions of propellant) active environments.

The second group of techniques ensures a possibility of carrying out more detailed studies of combustion process and rather simple ways of obtaining various experimental information (temperature of particles at ignition and combustion, qualitative and quantitative characteristics regarding the condition of surface, time of fusion and combustion of particles) [15, 43, 44, 45].

When interpreting the obtained results the main idea, as a rule, is the assertion that combustion is realized in the mode close to diffusion one which results in a weak dependence of combustion characteristics on environment temperature. For example, it has been registered that the time of particle combustion in the environment temperature range of 2,000 - 3,000K changes insignificantly [1]. (Data on this dependence in a wider temperature range (at essentially lower temperatures) are not available.)

At the same time the results of several works give grounds to suppose that metal particle combustion can be also influenced by kinetic factors. The data of work [43] allows one to consider that probably the combustion of Al particles in air environment takes place both on their surface and in the gas phase, i.e., the combustion area is spatial.

The results of several works [46-48, 50] give evidence of a possibility of an essential accumulation of oxide on the surface of particles in the process of their combustion. (It should be noted that taking into account the kinetics of oxide formation within the framework of the model developed by the authors of work [54] doesn't allow one to explain the scale of oxide accumulation in combustion of metal particles in air and water vapor environment, the scale which has been registered by the same authors in work [46].) The obvious manifestation of the kinetic factors influence on metal particle combustion in the air environment has apparently been a reason for development of new models of combustion [50-52].

The available experimental information on realization of oxide accumulation phenomenon has not been confirmed by the results of investigating the evolution of CCP incorporated in the two-phase flow of propellant combustion products which have been obtained within the framework of both the given investigation and other investigations we have carried out.

The carried out analysis gives grounds to suppose that the environment parameters might essentially influence the mechanism of Al particle combustion. But this supposition lacks direct experimental confirmation. (In comparison of the results obtained in various works one or another methodical factor may possibly prove to be most decisive). At the same time the given problem seems to be rather important for both interpretation of the results obtained in the present investigation of CCP evolution in the gas phase and development of methodology for investigating MF as a component of SRP.

In connection with that the target of the present investigation has been formulated in the following way.

For the population of agglomerates, for which the characteristics of evolution process in propellant combustion products have been obtained, to find the corresponding

characteristics in comparatively cold oxidizing environments by using the analogous methodical procedures.

Below there are given the data on parameters of agglomerates which have been used in the investigation. The propellants in that case have played the role of agglomerates «generators».

Two different propellants (W1 and W2) were used in the experiments. Near by the propellant burning surface agglomerates are characterized by the following parameters:

Propellant W1: $D_{43} = 180 \pm 20 \mu\text{m}$, $\eta = 0.19 \pm 0.02$;

Propellant W2: $D_{43} = 340 \pm 35 \mu\text{m}$, $\eta = 0.35 \pm 0.03$

Agglomerate size distribution for the propellants is presented in Figure 1.36.

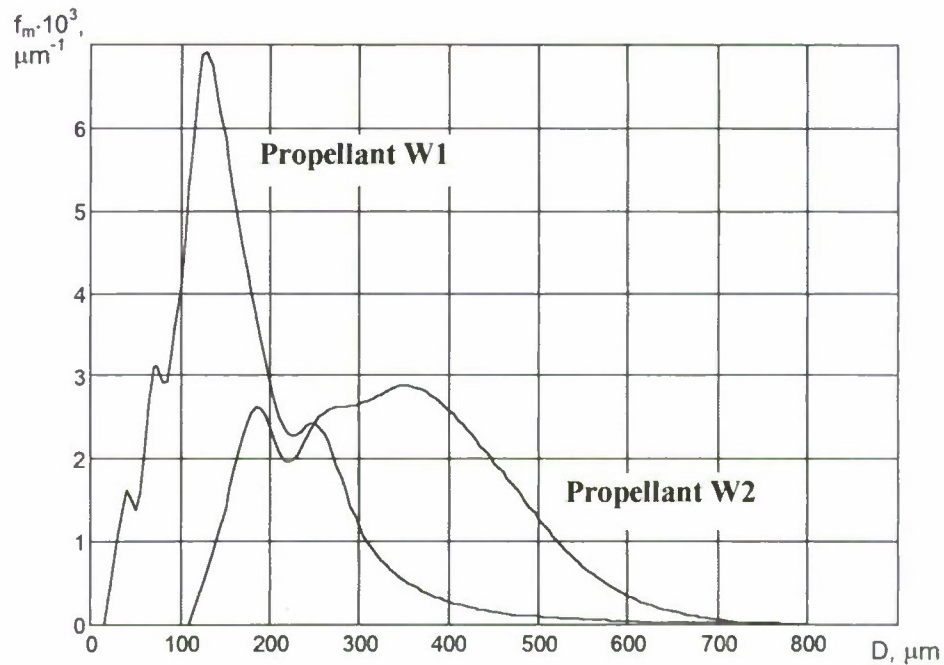


Figure 1.36. Agglomerate size distribution for propellants W1 and W2.

1.2.2. EXPERIMENTAL TECHNIQUE.

Experimental technique is illustrated by Figure 1.37. The experimental method is as follows. Combustion of agglomerates was studied in conditions of low-temperature gaseous mixture of air and nitrogen in CVB. Specimen of SRP is source («generator») of burning Al-Al₂O₃ particles. Oxygen concentration in CVB is varied by means of changing proportion of the mixture components. Rather small samples (5x5x10mm) were used to minimize influence of combustion process on gaseous media in CVB. Thus, the burning agglomerates quickly leave stream of a sample combustion products and get into turbulent boundary layer between the combustion product jet and surrounding motionless gas. During further moving of the agglomerates in the CVB, environmental parameters are approximating to initial values which were before the specimen ignition (that is, gas temperature is reducing and oxygen concentration is increasing). Thus, burning of agglomerates occurs in conditions of variable temperature and oxygen concentration.

Experiments were conducted for pressure 1 MPa and for different oxygen concentration in CVB (14%, 10.5%, 7%, 3.5%).

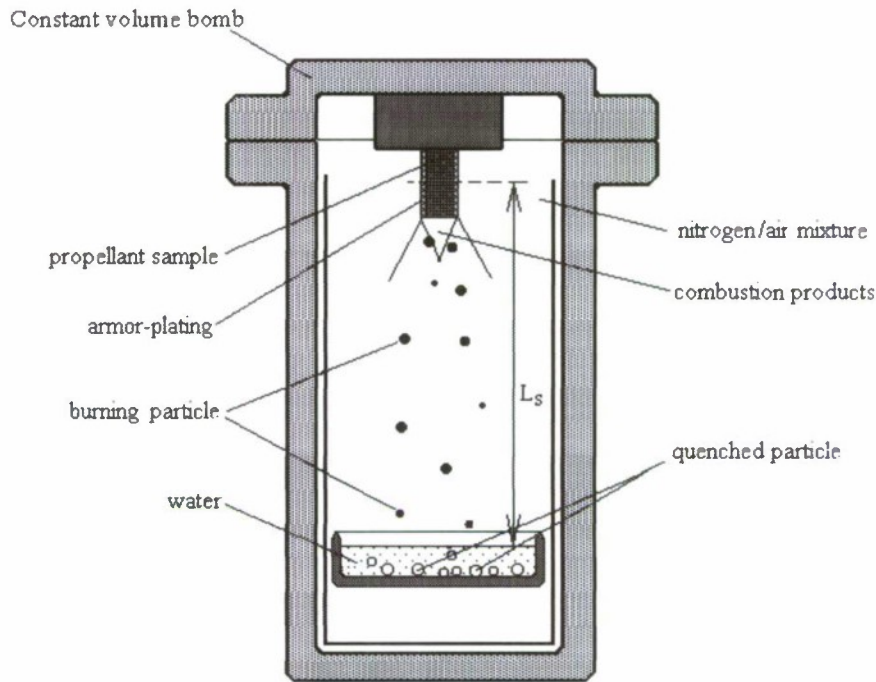


Figure 1.37. Scheme of experiments.

Burning process of agglomerates was interrupted at different stages of their evolution in CVB by means of quenching in liquid (water). Water was used as freezing liquid because it does not burn in oxygen-containing medium. But water is an active medium for burning agglomerates. Hence, agglomerates are able to interact with water in the process of their freezing. Special experiments showed that chemical interaction between agglomerates and water might actually take place. However, for conditions of the experiments made (pressure 1MPa), the results of the interaction were not considerable. Thus, it was obtained that parameter Z_m^{OX} increases by 5% in the process of agglomerates freezing in the water. This fact allowed us to conclude that discussed freezing method may be used to evaluate agglomerates evolution in oxygen-containing cold medium.

1.2.3. RESULTS AND DISCUSSION

The given investigation has been directed to studying the quantitative behavior of agglomerate oxide (changes in its quantity) when agglomerates evolve in low-temperature oxygen-containing gas environment. The experimental results are presented in Tables 1.1, 1.2, 1.3 and in Figure 1.38. They show that metal combustion (a decrease of parameter Z_m) is accompanied by an essential increase in amount of oxide in agglomerates, that is the accumulation of oxide in agglomerates takes place.

Table 1.1. (Propellant W1)

CCP parameters at burning propellant surface.

L_s , mm	Z_m	Z_m^{OX}
1	0,315	0,038

CCP parameters after burning in CVB:

in the case when mole share of oxygen (a_k^{CVB}) is equal to 0,14.

L_s , mm	Z_m	Z_m^{OX}
70	0,091	0,072

in the case when mole share of oxygen (a_k^{CVB}) is equal to 0,105

L_s , mm	Z_m	Z_m^{OX}
350	0,042	0,105

in the case when mole share of oxygen (a_k^{CVB}) is equal to 0,07

L_s , mm	Z_m	Z_m^{OX}
350	0,113	0,099

Table 1.2 (Propellant W2)

CCP parameters at burning propellant surface.

L_s , mm	Z_m	Z_m^{OX}
1	0,490	0,140

CCP parameters after burning in CVB:

in the case when $a_k^{CVB} = 0,14$.

L_s , mm	Z_m	Z_m^{OX}
350	0,176	0,277

in the case when $a_k^{CVB} = 0,105$

L_s , mm	Z_m	Z_m^{OX}
350	0,25	0,252

Table 1.3.

a_k^{CVB}	$-\frac{\Delta Z_m^{OX}}{\Delta Z_m} \cdot 100\%$	
	Propellant W1	Propellant W2
0,07	30 %	-
0,105	25 %	47 %
0,140	15 %	43 %

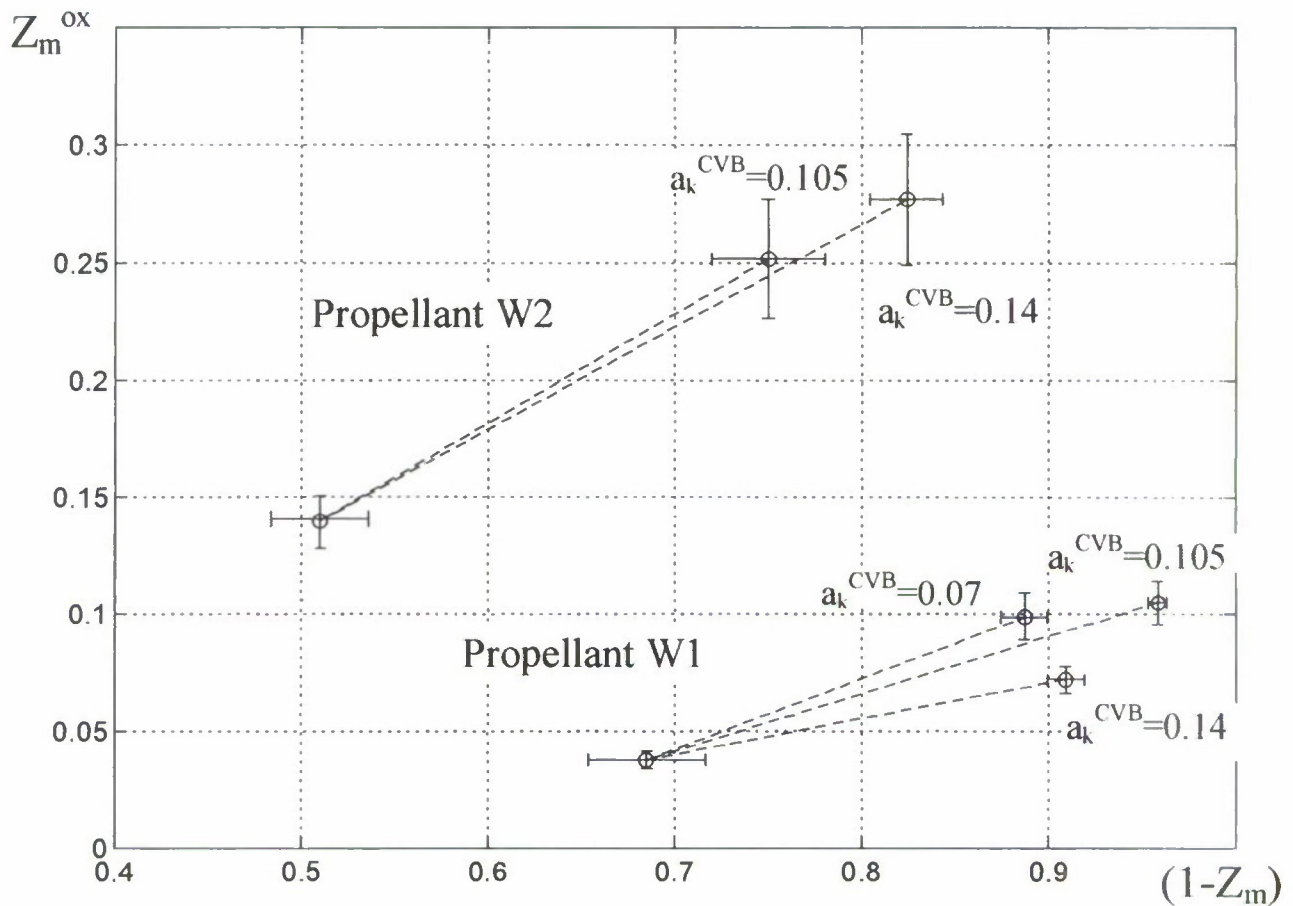


Figure 1.38. Experimental dependence of Z_m^{ox} on $(1-Z_m)$.

The experimental data allow to conclude that rate of oxide accumulation grows with agglomerate size increasing and oxygen concentration reducing in gas media. In the case of propellant W2 and $a_k^{CVB} = 0.105$, approximately 47% (almost a half) of oxide, which is formed in agglomerate metal burning, accumulates in the «caps» of the agglomerates. These data are in good correspondence with Prof. A. Zenin's data obtained in experiments with aluminum particle combustion in similar conditions.

Reduced concentration of oxygen in CVB down to 3.5% is found out to make burning process impossible: the agglomerate extinction occurs at very little distances from propellant burning surface.

The main result of the study is as follows. It was experimentally found that burning process of Al-Al₂O₃ particles in low-temperature oxygen-containing gaseous media is accompanied by significant accumulation of alumina in the particles. Intensity of the oxide accumulation depends on the particle size and oxygen concentration.

Experimental data concerning evolution of the agglomerates in the flow of propellant combustion products are shown in Tables 1.4, 1.5 and in Figure 1.39. The experiments were done in accordance with experimental technique described in item 1.1.3. In the experiments, agglomerates were frozen with the help of water.

Table 1.4. (Propellant W1)

L_s , mm	Z_m	Z_m^{ox}
1	0,315	0,038
70	0,200	0,030

Table 1.5. (Propellant W2)

L_s , mm	Z_m	Z_m^{ox}
1	0,49	0,14
70	0,42	0,135

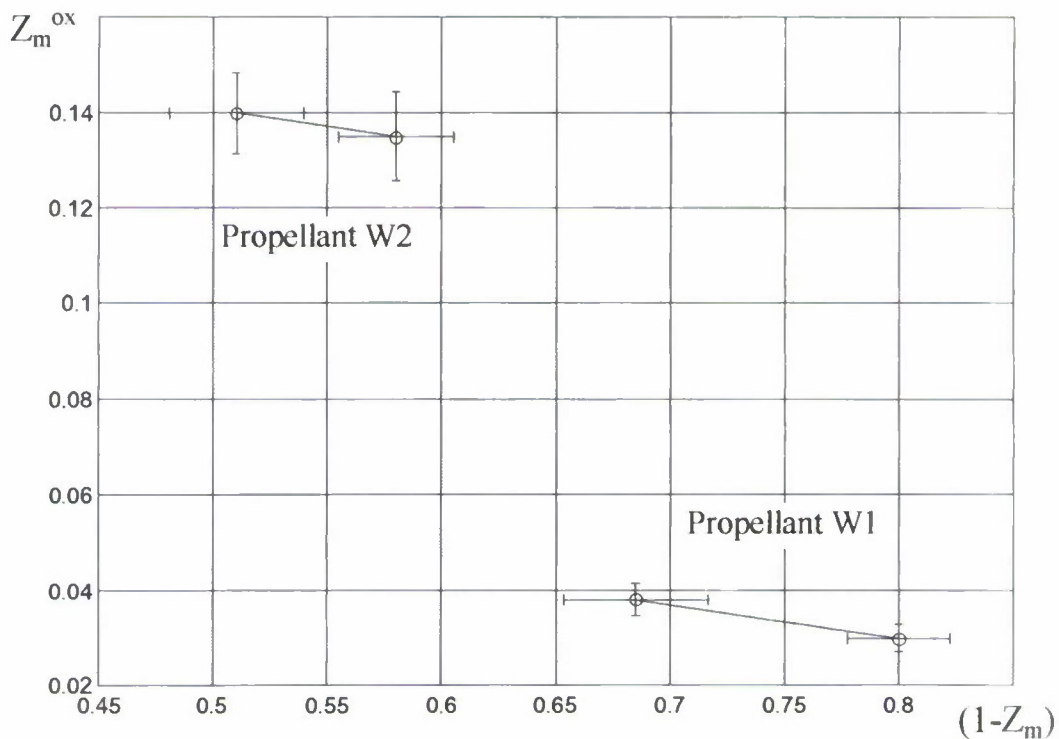


Figure 1.39. Evolution of agglomerates in combustion products flow.
(pressure 1MPa).

The results of the carried out investigation give grounds for thinking that the presence of cold oxidizing environment leads to the transformation of metal particle combustion mechanism as compared with high-temperature environments. The following concepts of metal particle combustion mechanism can be formulated.

The agglomerates involved in the studied process of evolution are of conventional structure. They consist of metal drops and «cap» oxide. (In the general case the formation of «cap» oxide is a natural result of the first stage of combustion after ignition - heterogeneous combustion).

Due to essential heat losses the area of agglomerate metal combustion taking place in the evolution is of finite dimension (spatial area). It is located both in gas phase and on metal surface. Metal combustion in the gas phase results in formation of HDO particles; that on the metal surface - formation of oxide remaining incorporated in agglomerate. The ratio of metal involved in gas-phase reaction and that in heterogeneous

one is the factor determining the share of oxide accumulating in agglomerate as a component of the latter.

A necessary condition for realization of combustion in heterogeneous mode is a removal of formed oxide from the metal surface to ensure that there is no blocking the contact between metal and oxidizer. Due to comparatively low rates of chemical interaction taking place on the surface of metal particles the surface forces are capable of transferring this oxide into «cap» one.

Following a decrease in heat losses of a burning particle (its temperature rises), first the share of metal evaporating from its surface increases, then metal combustion completely shifts into gas phase. The given hypothesis is indirectly confirmed by the data obtained in work [47] which show that the amount of oxide accumulated on the particle surface decreases with a drop in pressure. (A pressure drop leads to an increase in thermodynamic probability of gasification of the condensed Al and Al_2O_3 mixture.) Figure 1.40 shows a diagram illustrating combustion of Al particles in cold environment.

It is quite natural that for development of a detailed description, including a mathematical one as well, of metal particle combustion in cold oxidizing environments it is necessary to carry out more extensive experimental investigations comprising determination of: combustion rate as a function of various factors, parameters of burning particle structure, and dispersity of combustion products being formed. But the carried out investigation makes it possible to establish the fact of influence exerted by the oxidizing environment temperature on metal particle combustion mechanism, and that it is necessary to take this factor into account when using the results obtained in different model setups for description of MF combustion with MF being a component of SRP.

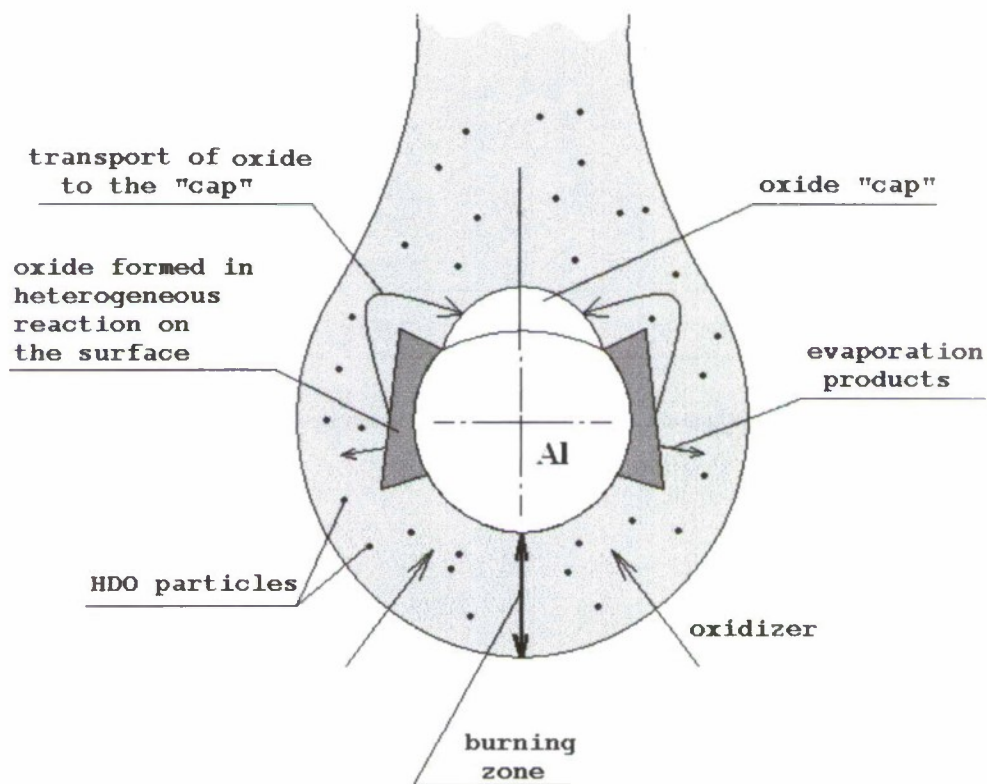


Figure 1.40. A diagram for illustrating possible nature of Al particle combustion in cold oxidizing medium.

2. STUDYING THE CCP FORMATION IN VICINITY OF PROPELLANT BURNING SURFACE WITH THE USE OF A MODIFIED METAL FUEL.

As soon as agglomeration phenomenon was discovered, the work started on development of techniques (approaches) ensuring lowering the negative effects related to that phenomenon. (As a rule, it concerned increasing the dispersity of agglomerates). As the agglomerate formation occurs due to fusion of initial particles and products of their transformation, then one of the obvious techniques to affect the agglomeration process consists in coating of MF initial particles with films of particular materials (modification of MF).

The subject of the present study is to experimentally find out the nature in which different coatings affect the CCP formation with the purpose to find how to most effectively use MF as a component of propellant and define more accurately the mechanism of MF behavior in the propellant surface layer.

2.1. ANALYSIS OF LITERATURE RELATED TO THE EFFECT OF COATING ON MF COMBUSTION.

By now a number of works have been carried out where combustion of propellants containing Al coated with different types of films [33, 34, 35] has been examined. As materials for coating there have been used substances with higher melting temperatures as compared to Al. They are Ni, Fe, Cu. It was expected that the use of high-melting coatings should have resulted in agglomerate size reduction.

In works [33, 34] the experiments on agglomerate dispersity assessment were conducted by combustion of binary composites on the basis of Al and AP or cesium nitrate. The obtained results allow one to conclude that encapsulation of metal particles with high-melting coatings makes it possible to reduce agglomerate size. It has been established that there exists an optimum content of coating substance which corresponds to maximum agglomerate dispersity. Coating efficiency decreases when environment pressure rises and excess factor of composite oxidizer decreases. The most essential reduction (by 6-7 times) of agglomerate size is observed when Cu coating is used (Cu content is about 0.5-0.7%) in stoichiometric composites of AP-Al at atmospheric pressure. The composites combustion rate grows in monotone mode with an increase in coating material content. (Maximum content was 5%).

In work [35] the study was carried out on model propellants on the basis of AP and plexiglas (polymethylmethacrylate) at pressures of up to 6.0MPa. Efficiency of coating use influence on agglomerate parameters was assessed by indirect data: change in propellant combustion rate, maximum temperature of combustion products in gas phase, and dispersity of final products of MF combustion. It was thought that an increase in both combustion rate and temperature, and dispersity of these products was a sign of agglomerate size reduction. Besides, qualitative data on intensity of agglomeration process running were also obtained with help of

filming the burning surface. The obtained results have allowed the work's authors to draw a conclusion that encapsulation of Al particles with films of high-melting metals (Ni, Cu, Fe) decreases intensity of agglomeration. It was assumed that a similar effect could be reached by using organic substances as coating materials.

It should be noted that the authors of the reviewed works proceeded from assumptions that particles fusion occurs before the former are ignited after which they should leave the burning propellant surface.

An interesting study has been completed by the authors of works [36, 37] who assessed how encapsulation of Al particles affects flame propagation rate in aerosuspensions (W) of same particles and their induction times. As coating materials they used Ni and fluorine-containing organic substances. The given study is distinguished by methodical correctness, and the obtained results - by clearly expressed physical nature, and they point to the fact that presence of coatings, due to an increase in particle reaction capacity, causes a decrease in their ignition temperature and results in W value growth. However, the conditions of MF combustion in aerosuspensions and in propellants are essentially different, and, as a result, the data obtained for combustion of dispersed Al in active media don't allow one to describe MF behavior as a propellant component.

On the whole, the results of the carried out analysis of literature show the lack of systematic studies regarding the effect of MF modification of CCP characteristics. The available information is not complete and limited which doesn't allow one to define the principles of using coatings which will ensure development of more effective propellants.

Formulation of Study Objectives.

Taking into account the results of analysis of the literature, the objectives of studying the processes of CCP formation in vicinity of burning propellant surface with modification of MF accomplished can be formulated as follows:

1. To obtain comprehensive experimental information of quantitative character on the properties of entire CCP in vicinity of burning propellant surface with different methods of MF modification.
2. On the basis of available experimental material to date to formulate main principles of MF modification influence on MF combustion as a propellant component.

It is expedient to use as coatings high-melting metals and organic substances ensuring to a considerable degree protection of metal against reaction with environment until the moment of ignition and which are essentially different by their physico-chemical properties.

For the purpose of carrying out the study the authors of works [36, 37] were kind enough to make available to us the powders of modified Al with parameters given in Table 2.1.

Table 2.1
Modified Al powders.

Powder symbol	A1	A2*	A3	A4**	A5**
$d_{M_{43}}$, μm	22,5	21,9	10,4	10,5	10,2
Coating	-	Ni	-	$\text{Cl}_2\text{Si}[\text{OCH}_2(\text{CF}_2\text{-CF}_2)_2\text{H}_2]_2$ (APPRET)	polymethylfluoroacrylate (PMFA)

Notes:

* - This modified aluminum powder was made by coating of the powder A1 with Ni film;

** - These modified aluminum powders were made by coating of the powder A3 with fluorine-containing organic films;

$d_{M_{43}}$ - mass-medium diameter of modified MF particles determined within the present study.

Mass share of coating in these powders doesn't exceed 3.2%. The principles of manufacturing these powders are described in [36]. They were used in development of propellants similar in properties to those used in the first stage activities [30]. These propellants have the following components with given mass shares:

- AP (64%);
- binder (isoprene rubber and oil) (11.4%);
- modified MF (24%);
- additives (0.6%).

AP consists of two fractions: large-size (160-315 μm) and small-size (<50 μm), their proportion in propellant is 40/60.

Propellants differ by type of modified MF (Table 2.2).

Table 2.2.
Propellant compositions.

Propellant symbol	M1	M2	M3	M4	M5
MF type	A1	A2	A3	A4	A5

2.2. METHODS OF STUDY AND ITS RESULTS.

The study included sampling with subsequent analysis of CCP in above-the-surface area of burning propellant. The technique on the whole is similar to one used previously in carrying out the first stage activities [30].

The changes are mainly related to the method of dispersion analysis of sampled agglomerates. To assess dispersity parameters we have used more advanced apparatus «Malvern Mastersizer, Ver.2.12» instead of diffraction microanalyzer «Analyzette 22».

The experiments were being conducted at two pressure levels: low (1.5MPa) and high (6.0MPa). The experimental results are given in Tables 2.3 - 2.4 and shown in Fig. 2.1 - 2.14. In the tables the parameters' mean values and their confidence intervals with confidence probability of 0.997 are indicated.

Table 2.3.
P=6.0 MPa

Propellant	Coating	Z_m	Z_m^{ox}	Z_m^a	η	D_{43} , μm	d_{43} , $m\mu m$	U, mm/sec
M1	-	0.155 ± 0.016	0.080 ± 0.007	0.235 ± 0.023	0.494 ± 0.050	213 ± 20	-	9.2 ± 0.7
M2	Ni	0.197 ± 0.020	0.058 ± 0.005	0.255 ± 0.025	0.359 ± 0.035	104 ± 10	-	7.9 ± 0.6
M3	-	0.133 ± 0.013	0.060 ± 0.006	0.193 ± 0.019	0.460 ± 0.050	126 ± 14	-	8.1 ± 0.6
M4	APPRET	0.126 ± 0.011	0.033 ± 0.004	0.159 ± 0.015	0.334 ± 0.035	138 ± 15	-	6.9 ± 0.6
M5	PMFA	0.110 ± 0.010	0.041 ± 0.004	0.151 ± 0.014	0.414 ± 0.040	133 ± 12	-	6.0 ± 0.5

Table 2.4.
P=1.5 MPa

Propellant	Coating	Z_m	Z_m^{ox}	Z_m^a	η	D_{43} , μm	d_{43} , $m\mu m$	U, mm/sec
M1	-	0.259 ± 0.026	0.065 ± 0.006	0.324 ± 0.032	0.320 ± 0.030	223 ± 20	570 ± 100	7.3 ± 0.6
M2	Ni	0.307 ± 0.030	0.090 ± 0.008	0.397 ± 0.038	0.356 ± 0.035	138 ± 13	840 ± 120	5.7 ± 0.5
M3	-	0.333 ± 0.035	0.114 ± 0.013	0.447 ± 0.048	0.393 ± 0.038	210 ± 20	370 ± 80	7.05 ± 0.6
M4	APPRET	0.321 ± 0.032	0.059 ± 0.005	0.380 ± 0.037	0.255 ± 0.025	185 ± 20	730 ± 110	5.7 ± 0.5
M5	PMFA	0.356 ± 0.034	0.073 ± 0.009	0.429 ± 0.043	0.279 ± 0.027	150 ± 15	-	5.18 ± 0.4

Let's analyze the results of studying agglomeration as regards low and high pressure levels.

A. Low pressure.

The use of nickel coating leads, first, to an increase in dispersity of agglomerates (mass-medium diameter decreases from approximately 220 μm down to approximately 140 μm) and, second, to disappearance of large-size mode in mass function of agglomerate size distribution density (f_m) (Fig. 2.1, 2.3). Substitution of A1 powder with A2 also results in some increase in relative quantity of agglomerates and oxide content in them. (Values Z_m^a and η go up approximately by 20% and 10% respectively).

Fluorine-containing organic films exert similar influence on agglomerate dispersity (mass-medium diameter decreases from approximately 210 μm down to approximately 180 μm for propellant M4 and to approximately 150 μm for

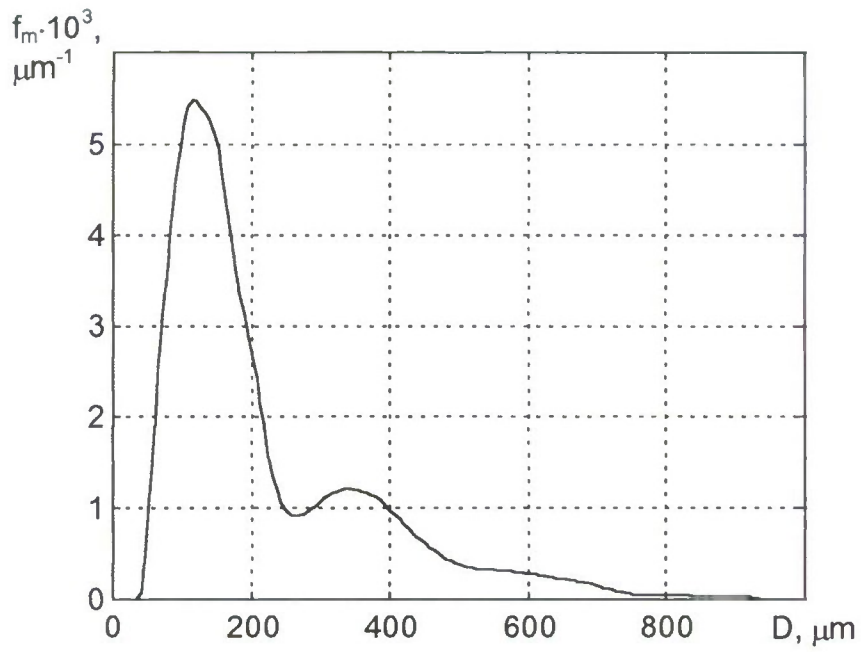


Figure 2.1. Agglomerate size distribution for propellant M1 at pressure 1.5MPa.

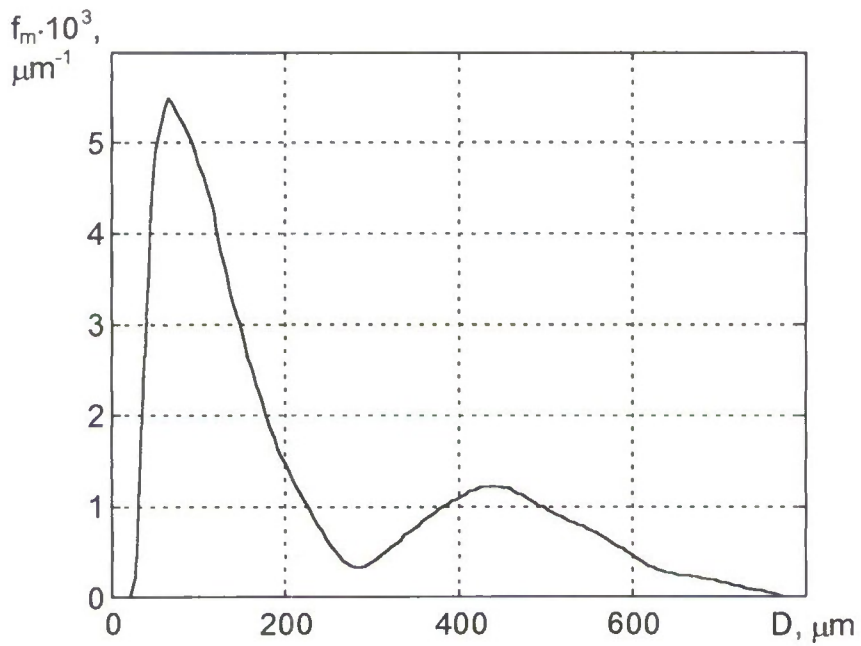


Figure 2.2. Agglomerate size distribution for propellant M1 at pressure 6.0MPa.

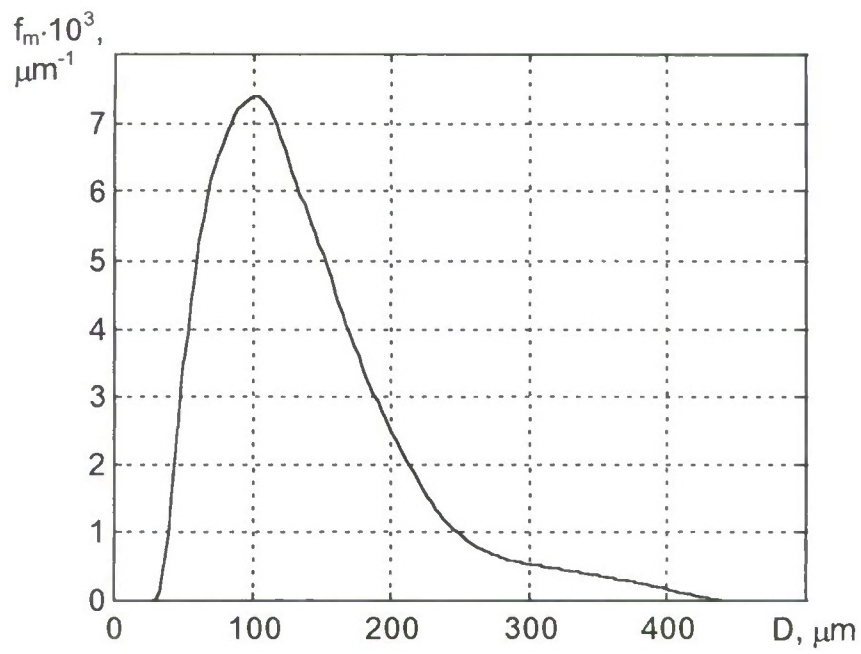


Figure 2.3. Agglomerate size distribution for propellant M2 at pressure 1.5MPa.

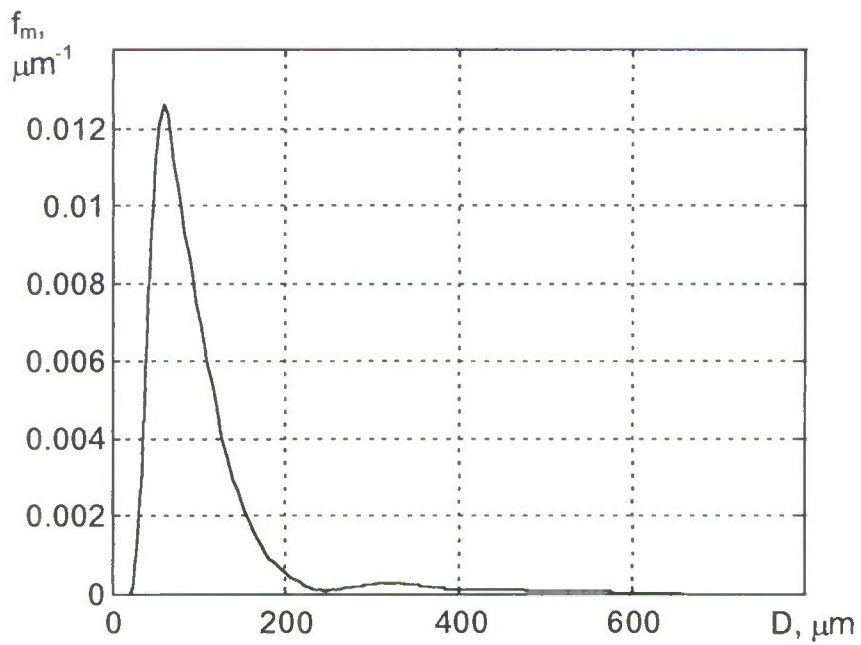


Figure 2.4. Agglomerate size distribution for propellant M2 at pressure 6.0MPa.

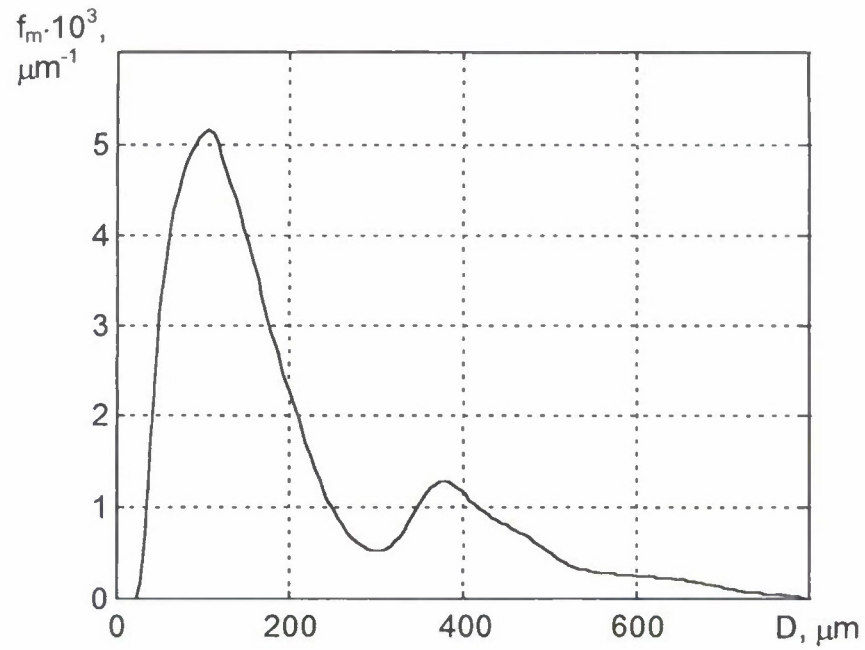


Figure 2.5. Agglomerate size distribution for propellant M3 at pressure 1.5MPa.

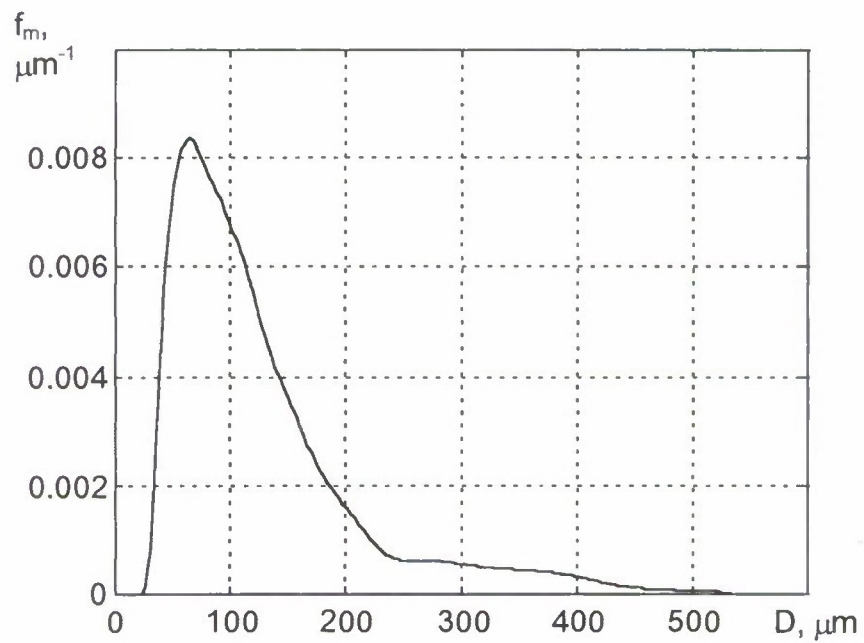


Figure 2.6. Agglomerate size distribution for propellant M3 at pressure 6.0MPa.

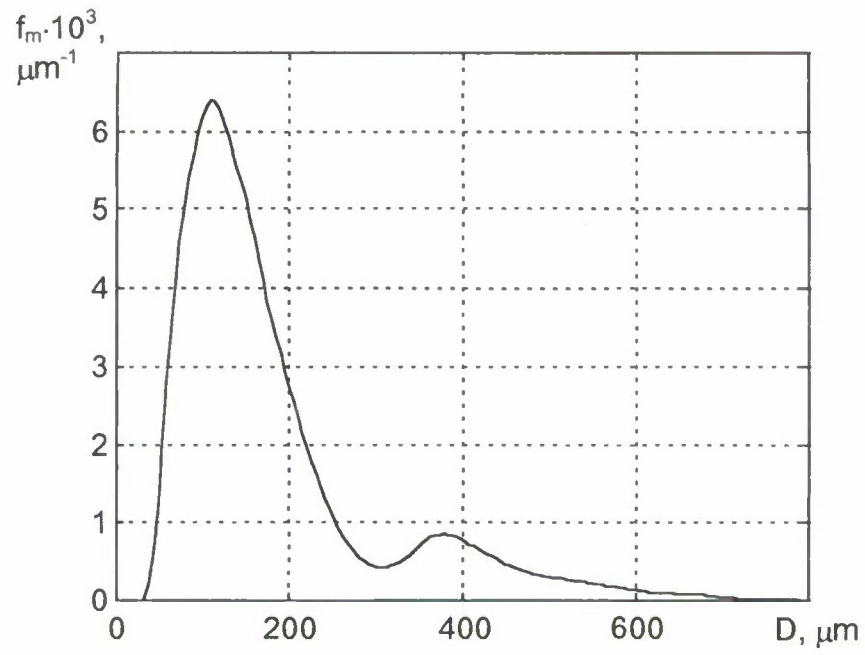


Figure 2.7. Agglomerate size distribution for propellant M4 at pressure 1.5MPa.

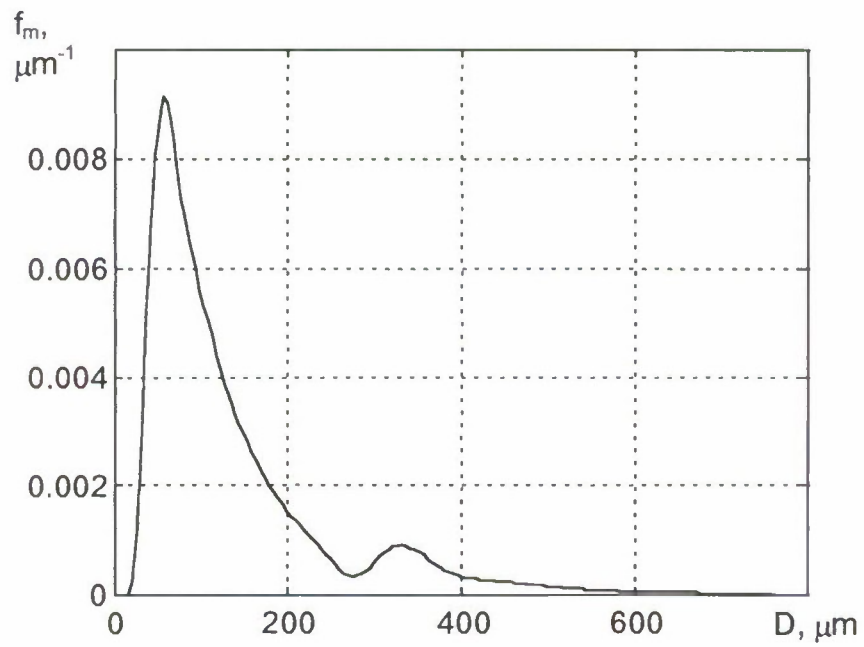


Figure 2.8. Agglomerate size distribution for propellant M4 at pressure 6.0MPa.

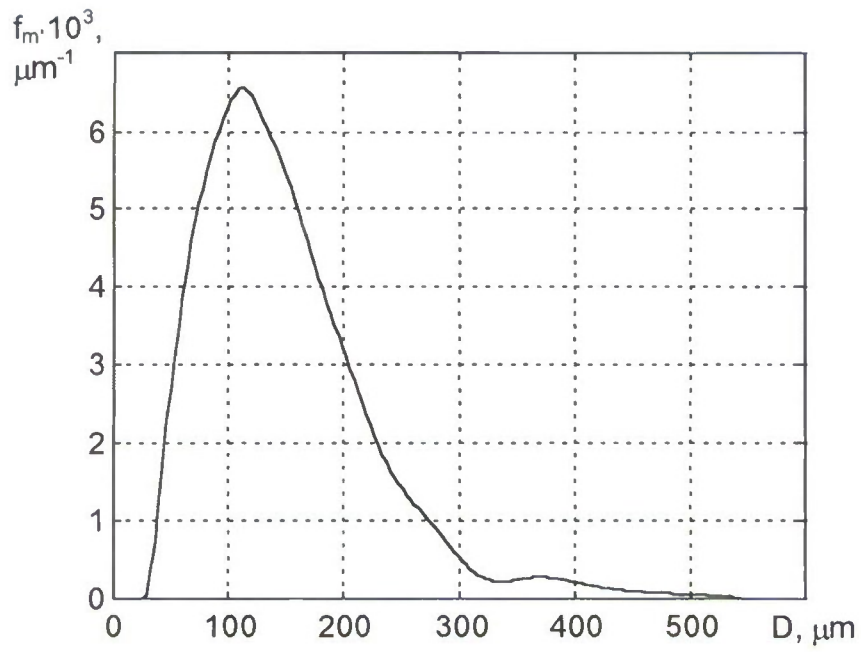


Figure 2.9. Agglomerate size distribution for propellant M5 at pressure 1.5MPa.

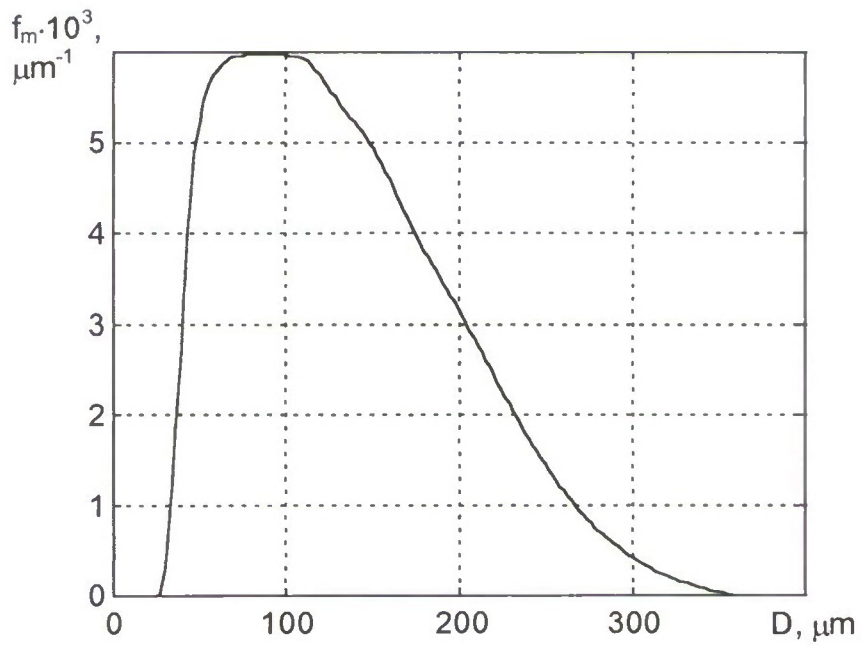


Figure 2.10. Agglomerate size distribution for propellant M5 at pressure 6.0MPa.

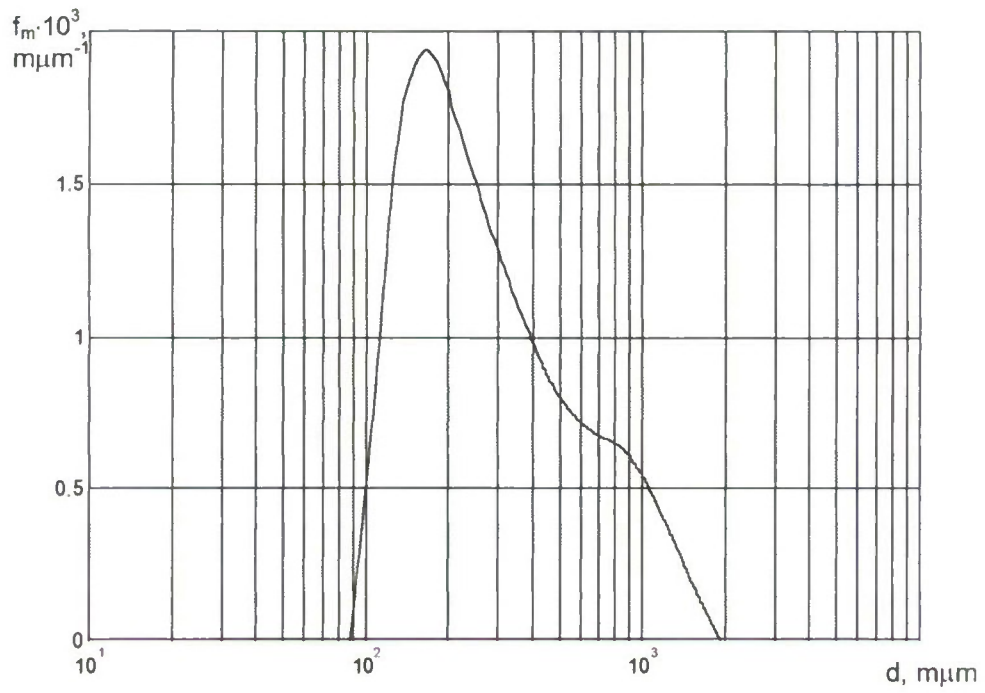


Figure 2.11. HDO particle size distribution for Propellant M1 at pressure 1.5MPa.

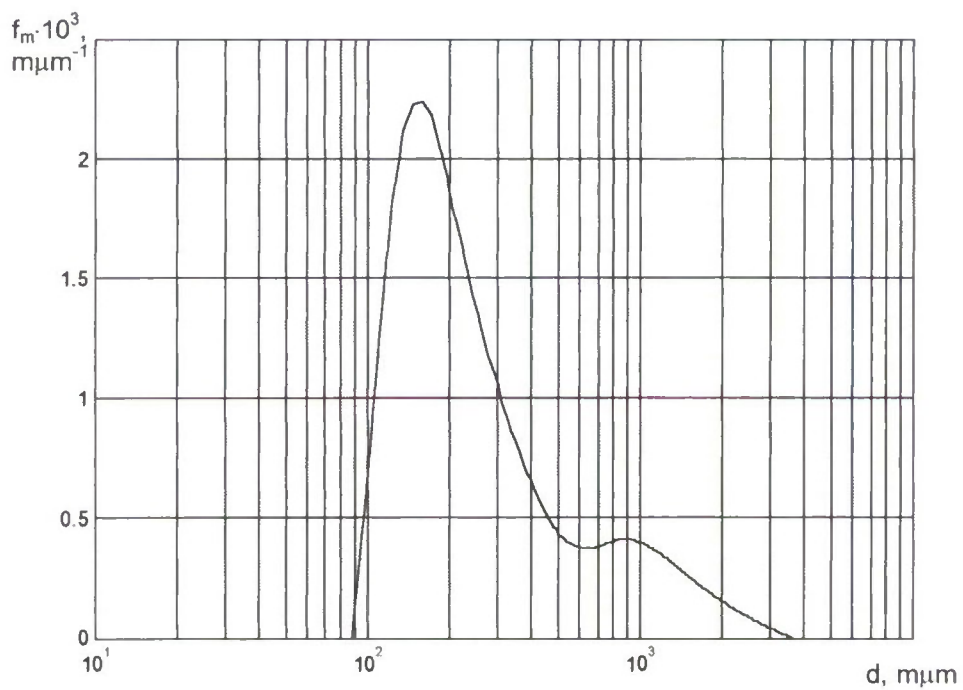


Figure 2.12. HDO particle size distribution for Propellant M2 at pressure 1.5MPa.

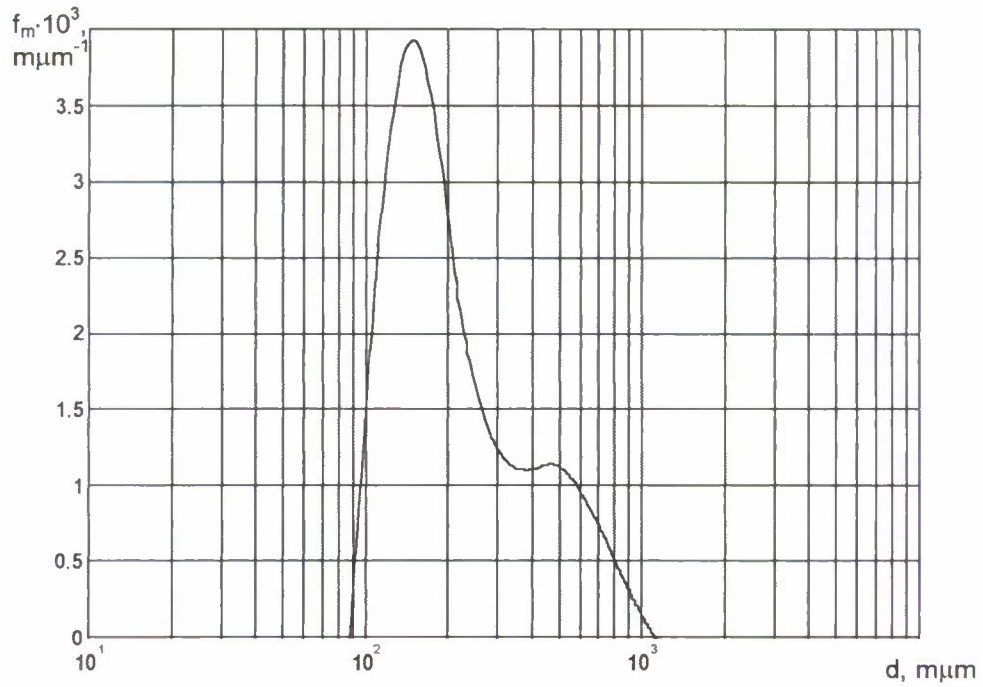


Figure 2.13. HDO particle size distribution for Propellant M3 at pressure 1.5MPa.

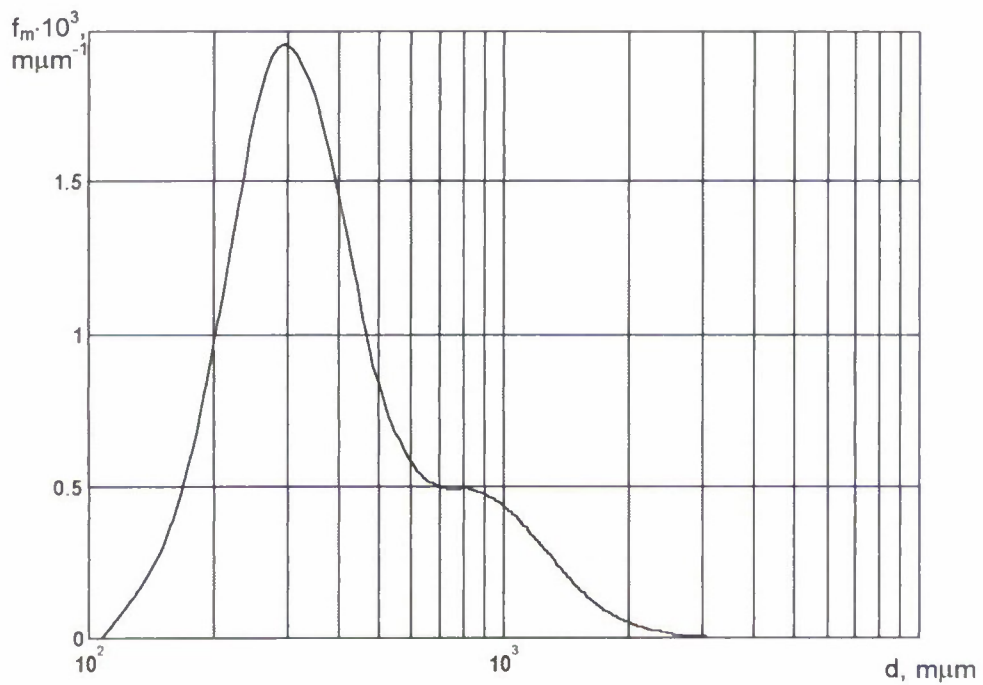


Figure 2.14. HDO particle size distribution for Propellant M4 at pressure 1.5MPa.

propellant M5). The mass functions of agglomerate size distribution density (f_m) in combustion of propellants M3, M4, and M5 are bimodal and the use of those coatings leads to significant reduction in the number of particles which are responsible for formation of the mode in the large-size region (Fig. 2.5, 2.7, 2.9). Besides, the use of those coatings decreases the share of oxide in agglomerates (parameter η) by approximately 1.5 times and the total mass of agglomerates to some extent (parameter Z_m^a). As a result, the oxide content in agglomerates considerably decreases (the decrease of parameter Z_m^{OX} can reach nearly times two).

B. High pressure.

The use of Ni coating leads to an essential increase in agglomerate dispersity, with function f_m becoming practically unimodal (Fig. 2.2, 2.4). Besides, the use of A2 powder causes some decrease of oxide content in agglomerates (by about 1.4 times) and an increase in metal content (parameter Z_m increases by about 25%).

The presence of fluorine-containing coating leads, like in the case of low pressure, to a decrease of oxide content in agglomerates. However, that effect is less pronounced than in the case of low pressure. Agglomerate dispersity changes insignificantly when similar coatings are used. (Probably it can be stated that the available data on dispersity parameters are within their natural range of scattering.) Function f_m is close to unimodal one in its character (Fig. 2.6, 2.8, 2.10).

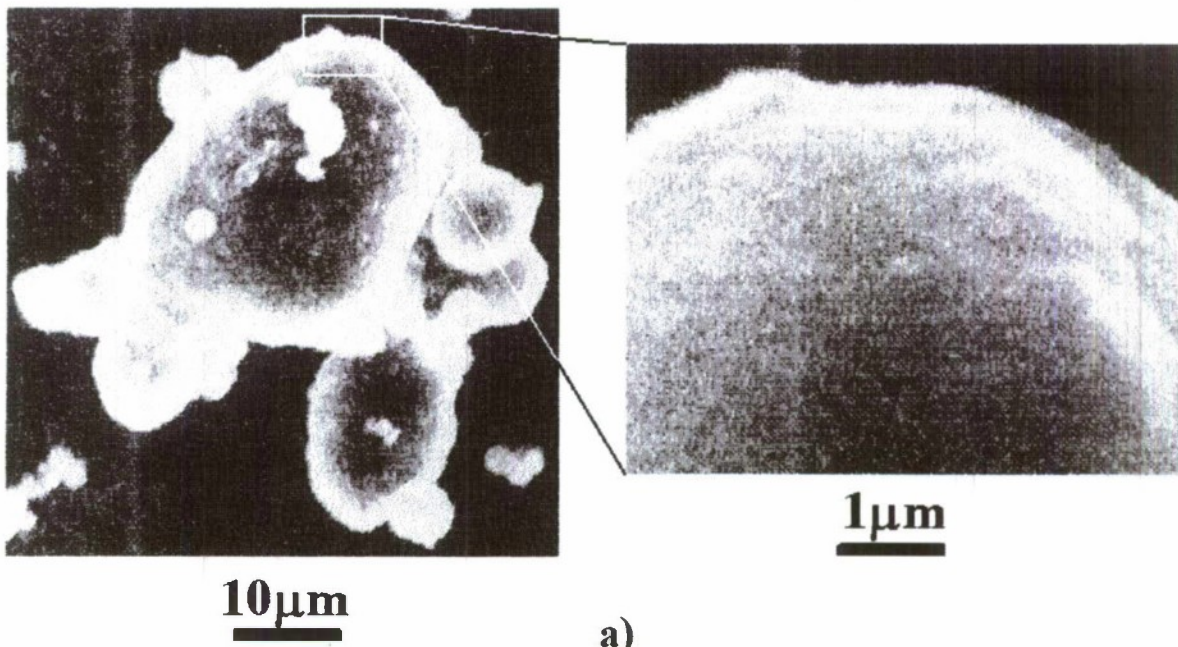
HDO parameters have been investigated at low pressure level. The results (Table 2.4 and Figures 2.11 - 2.14) of this investigation has allowed to establish the following:

The use of higher-dispersity MF leads to an increase in HDO dispersity. Thus, if in combustion of propellant M1 the value of d_{43} is 570nm, then for propellant M3 d_{43} is about 370nm.

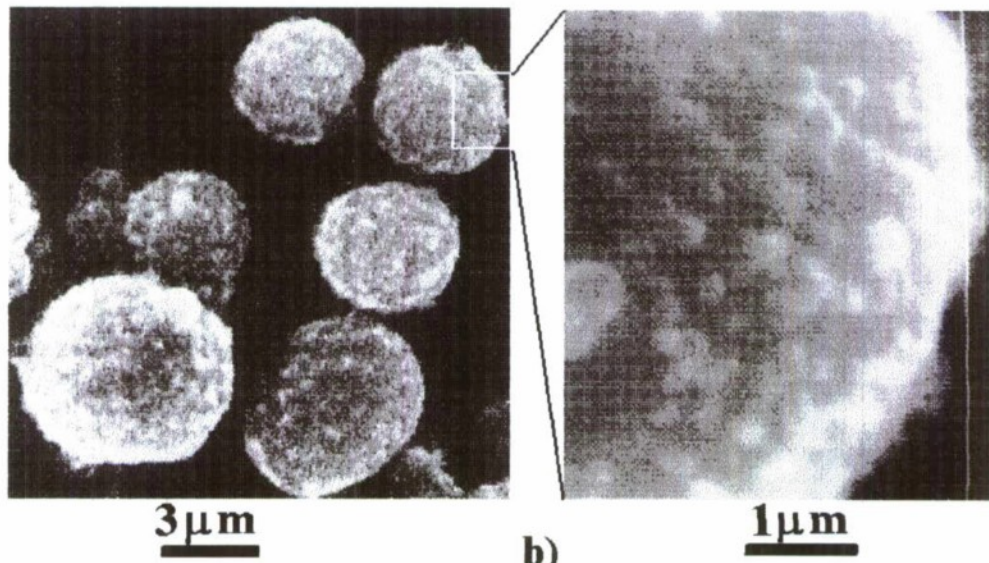
The presence of coatings causes a decrease in HDO dispersity, and in so doing this effect, as applied to coatings of fluorine-containing organic substances, is essentially more pronounced.

In addition to CCP investigation the initial MF powder particles have been studied. This study has included more precise determination of dispersion characteristics and analysis of surface of the given particles by means of their visualization with help of electronic microscope.

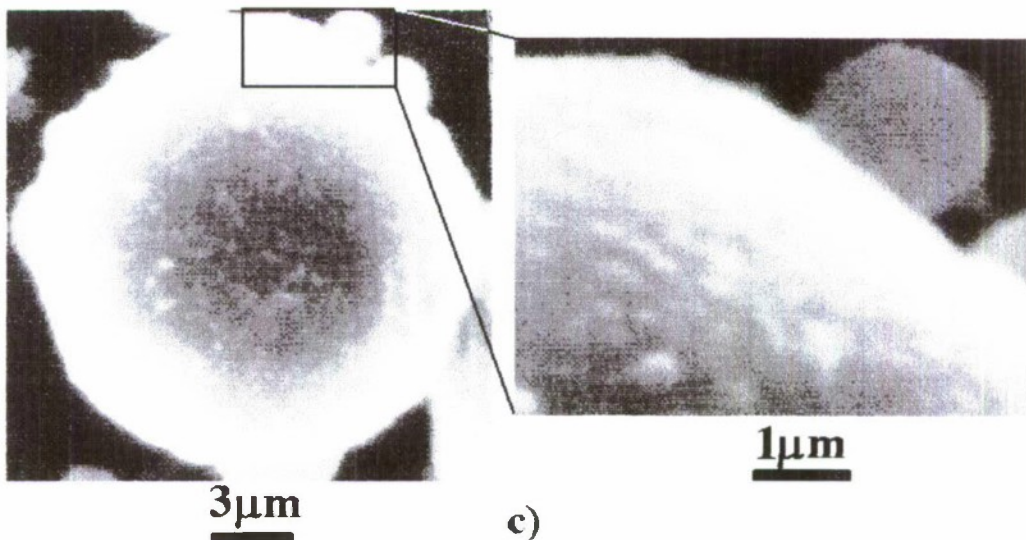
The results of dispersion characteristics determination are given in Table 2.1, they give grounds to assume that deposition of coatings insignificantly influences the particle size. The visualization of particle surface makes it possible to draw a conclusion that no coating defects are available (continuity of coating is not broken), and the particles have more regular spherical form when fluorine-containing organic substances are used (Fig. 2.15 - 2.16).



a)

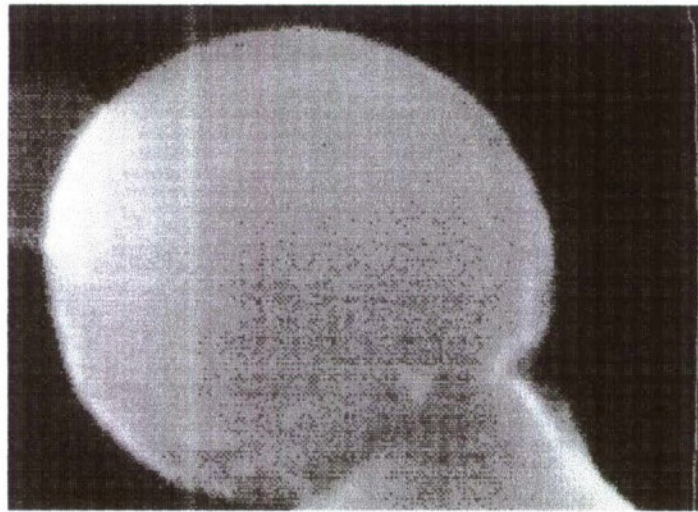


b)



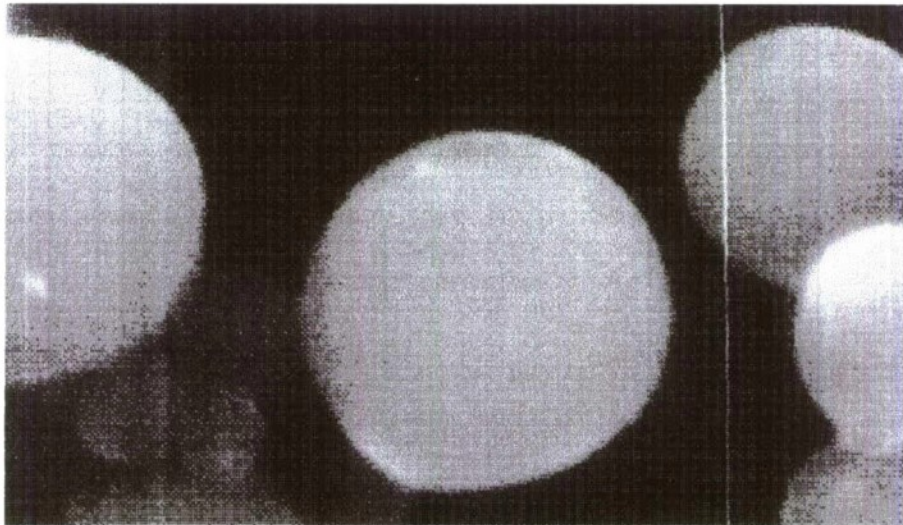
c)

Figure 2.15. Particles of MF (a-A1, b-A2, c-A3).



1 μ m

a)



1 μ m

b)

Figure 2.16. Particles of MF (a-A4, b-A5).

2.3. PHYSICAL CONCEPTS OF MF MODIFICATION INFLUENCE ON CCP FORMATION IN VICINITY OF BURNING PROPELLANT SURFACE.

According to the present understanding [38, 39 and al.], the MF combustion within the propellant surface layer is an integral part of agglomeration process. The final formation of agglomerates takes place only after MF ignition.

Absolute quantity of agglomerates being formed is determined by the share of MF involved in agglomeration, (this value is essentially determined by propellant structure

[30, 38, 39]), and the extent of MF burning out in gas-phase mode within the surface layer (SL).

The agglomerate size depends on residence time of agglomerating particles in SL which, in turn, depends on the level of adhesion (F_{adh}) and aerodynamic (F_{acr}) forces, discontinuity of SL and combustion process dynamism. Depending on various combinations of the above factors the agglomerate formation might be realized by the following mechanisms: «pre-pocket», «pocket», and «inter-pocket» [38, 40].

The presence of coating on MF particles might have the following obvious consequences for realization of agglomeration:

- changing MF ignition temperature (T_{ign}) and, as a result, changing the properties of the frame layer (FL) (SL upper portion) where the agglomerate formation takes place [38, 10];
- changing the surface properties of agglomerate 'substance' which inevitably influences the value of F_{adh} .

On the whole, the FL properties might be determined by the ratio between T_{ign} and the degradation temperature of carbonaceous elements (carbon frame) (T_{degr}). The given ratio might serve as a characteristic for placing propellants into one or another class [10]. If condition $T_{ign} > T_{degr}$ is fulfilled, then MF combustion is going on within the FL limits in presence of high coherence of metal particles with FL elements. As a result, the first stage of combustion which is going on in heterogeneous mode is distinguished by a comparatively high share of burning out metal and consequently a significant amount of oxide in FL composition and in agglomerates being formed on its surface. Otherwise ($T_{ign} < T_{degr}$) the ignition of MF particles takes place on the FL upper surface. In so doing the combustion in heterogeneous mode takes place under conditions of lesser heat losses, and this fact is reflected in a decrease of metal share burning out in this mode, and consequently in decrease of amount of oxide in agglomerates.

In combustion of propellants M1 and M3 at low pressure level the comparatively large amounts of oxide in agglomerates being formed give grounds to think that in this case the condition $T_{ign} < T_{degr}$ is fulfilled. As a result, the FL is saturated with liquid Al and Al_2O_3 which leads to a growth of adhesion forces supporting agglomerating particles on the FL surface. The given circumstance causes intensification of agglomerate formation by «inter-pocket» mechanism according to which the agglomerating particles being formed while the «pocket» burns out do not leave the burning propellant surface but are captured by another «pocket». The present mechanism is responsible for mode formation in the large-size region on functions f_m . (The first mode (the mode in the small-size region) is determined by agglomerates which are mainly formed by «pocket» mechanism.)

The use of Ni-containing coatings apparently doesn't change the value of T_{ign} , or insignificantly decreases it. It should be noted that in combustion of metal particles as a component of propellants the melting temperature of protective films (for example, Al_2O_3 or Ni) in principle doesn't matter for ignition. The decisive factor in the given process is the high rate of heating resulting in intensive cracking of protective film in the presence of temperature stress [41]. Just this circumstance

causes comparatively low values of T_{ign} (about 1300K) registered in the experiment [42].

It can be supposed that MF combustion in presence of Ni leads to changes in Al_2O_3 surface properties and decreases adhesion forces. (Delay of growing particle on the FL surface is caused by this particle 'cap' bonds with elements of this layer.) As a result, there take place a suppression of «inter-pocket» fusion of particles and a transformation of agglomerate size distribution function resulting in nearly complete disappearance of the mode in the large-size region on the function f_m .

Organic coatings unlike a Ni or Al_2O_3 have fundamentally different physical properties. It can be supposed that ignition of A4 and A5 powder particles occurs only after gasification of coating substance, delaying ignition. It results in an increase of T_{ign} . A change in relation between T_{ign} and T_{degr} leads to a transformation of FL properties which is reflected in lowering the level of adhesion forces. A drop in F_{adh} contributes to decreasing the intensity of «inter-pocket» fusion and increasing the agglomerate dispersity. Hence, in spite of the different mechanism of influence, the use of fluorine-containing organic coatings leads to the effect similar to one taking place in case of Ni-containing coating applications, that is, an essential reduction in the number of agglomerates being formed by the «inter-pocket» mechanism.

In case of high pressure the registered values of Z_m , Z_m^{ox} , Z_m^a and characteristics of function f_m give grounds to draw the following conclusions on the mechanism of combustion for propellants with unmodified MF (propellants M1, M3):

- values of temperature $T_{\text{ign}} \sim T_{\text{degr}}$ or T_{ign} slightly exceed T_{degr} , that is, we can say that with pressure rise the relation between T_{ign} and T_{degr} changes;
- some amount of MF incorporated in agglomerates burns in above-the surface zone of gas phase due to realization of non-steady effects resulting in fragmentation of particles (importance of this phenomenon is considerably less in comparison with previously studied compositions [30]);
- a decrease in initial MF dispersity leads to changing the level of adhesion forces resulting in keeping the intensive formation of agglomerates by the «inter-pocket» mechanism in case of raising pressure for propellant M3.

Thus, the studied propellants display characteristic peculiarities in combustion inherent in propellants of different classes (the classification in works [38,10] defines them as classes A and B) and the dispersity of initial MF influences the realization of various mechanisms of agglomeration.

The effect of the use of Ni-containing coating at high pressure seems to be similar to the respective effect at low pressure. With an insignificant change in the value of T_{ign} a decrease in the value of F_{adh} takes place, and, as a result, a reduction in the amount of agglomerates being formed by the «inter-pocket» mechanism occurs.

The use of organic coatings leads to some growth in T_{ign} at high pressure. But this factor doesn't essentially influence FL properties, and consequently the dispersity of agglomerates being formed.

The diagram illustrating the character of MF modification influence on the agglomeration process is given in Fig. 2.17.

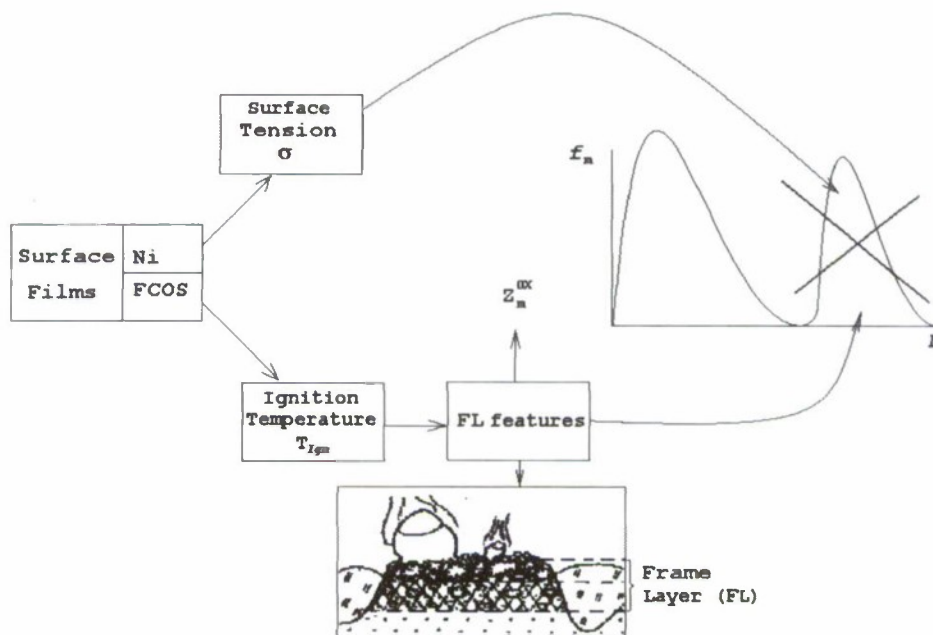


Figure 2.17. Influence of MF modification on the MF agglomeration.

The obtained results allow us to formulate some conclusions regarding desirability of using coatings.

The use of Ni-containing coating is a quite effective means for decreasing in the two-phase flow the content of the largest agglomerates being formed by the «inter-pocket» mechanism, the agglomerates primarily responsible for realization of slag formation process in the engine chamber. It can be supposed that under certain conditions the given coatings will be able to ensure transition from the «pocket» agglomeration mechanism to the «pre-pocket» one.

It seems reasonable to use the fluorine-containing organic coatings in case when we have active MF combustion within FL. These coatings are capable of not only increasing agglomerate dispersity due to decreasing the amount of the largest ones, but of reducing oxide content in agglomerates as well. The latter factor is quite important as the given oxide is a source of formation for large-size fraction of the final CCP.

It is quite natural that for resolving the practical problems of modified MF applications it is necessary to carry out additional studies targeted at identification of various aspects of modification influence on the combustion process (influence on the propellant combustion rate law, optimum content of coating substance, etc.).

However, on the whole, it is possible to state that the use of MF with coatings made of different materials can essentially change characteristics of agglomeration process.

At present there is no sufficiently complete picture of the physical process of HDO formation in vicinity of the burning propellant surface. It is clear that these

particles are mainly formed in combustion of MF not involved in agglomeration. There are some reasons to suppose [30] that an important role in formation of the given particles is played by the processes of MF combustion in heterogeneous mode and transition from this mode to gas-phase one. The results obtained within the framework of the present study confirm this hypothesis. These results give evidence that the HDO particle size tends to decrease following an increase in burning particle dispersity and the temperature of environment.

Thus, a substitution of powder A1 with A3 in the propellant leads to an essential increase in HDO dispersity. The use of Ni-containing coating doesn't lead to a considerable change in MF combustion within SL, and as a result, there are no essential differences in HDO dispersity in combustion of M1 and M2 propellants. The fluorine-containing organic coatings, on the contrary, contribute to decreasing the share of MF burning within SL, and their use results in a drop of HDO dispersity (propellants M4).

CONCLUSION.

Within the framework of the present work various aspects of MF evolution in combustion as a component of SRP have been studied which has allowed to solve two problems:

- description of evolution process of CCP as a component of two-phase flow;
- defining the methods of influencing agglomeration process with the purpose to reduce the size of agglomerates.

The research has yielded the following principal scientific results:

1. A technique of experimental investigation ensuring a possibility to determine various characteristics of CCP particles (HDO and agglomerates) at different stages of evolution during their movement as components of two-phase flow has been worked out.
2. A big volume of quantitative information on changes in CCP chemical composition, structure, and dispersity depending on evolution process time has been obtained.
3. A general physical picture of CCP evolution process has been formulated. The following principal phenomena of the given process have been determined:
 - agglomerate metal combustion with formation of HDO particles which are supplied into carrier gas phase;
 - chemical interaction of condensed Al and Al_2O_3 with formation of gaseous products;
 - changes in agglomerate structure;
 - deposition of HDO particles of the carrier gas phase on agglomerates;
 - coagulation of HDO particles of the carrier gas phase between themselves.
4. Improvement of the previously developed mathematical model for evolution of agglomerate population has been carried out. A possibility to use the given model for predicting agglomerate characteristics in evolution has been shown.
5. A possibility of thermal conditions in which the agglomerate evolution takes place to influence the given process has been established. With an essential decrease in temperature of environment the mechanism of agglomerate metal combustion changes, and, as a result, the laws of evolution process change.
6. It has been established that the use of Ni and organofluoric coatings for initial Al powder particles contributes to elimination of agglomerate formation by «inter-pocket» mechanism which results in disappearance of the largest-size agglomerate fraction and, on the whole, to growth of their dispersity. Besides, organofluoric coatings contribute to reduction of oxide content in agglomerates. Physical concepts regarding the mechanism of how the coatings influence agglomeration have been formulated.

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