

Model of Aluminum Agglomerate Evolution in Combustion Products of Solid Rocket Propellant

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A mathematical model was developed for agglomerate evolution in the combustion products of solid rocket propellant. The model is based on the principle of dividing the complex process into relatively simple phenomena with sequential synthesis of their descriptions. The model includes description of many processes observed experimentally. They are 1) metal combustion, 2) chemical interaction between metal and oxide in agglomerates, 3) change in structure of agglomerates, and 4) interaction of agglomerates and carrier flow. Quite high quality of the model was proved by satisfactory agreement between calculation results and experimental data.

Nomenclature

a_k	= oxidizing potential of gas phase (mole fraction of oxidizing components in gas mixture)
c_p	= molar isobaric heat capacity
D	= agglomerate diameter
D_{43}	= mass median diameter of agglomerates
d	= smoke-oxide particle diameter
F	= force
H	= molar enthalpy
J	= mole flux of substance
K_{dep}	= coefficient of deposition of smoke-oxide particles on agglomerate
L	= molar evaporation heat
L_s	= distance from the burning surface of propellant specimen
M	= mass
Nu	= Nusselt number
P	= pressure
Pr	= Prandtl number
Q_{con}	= conductive heat flux
Q_{rad}	= radiant heat flux
Q_{sp}	= intensity of heat consumption on the interface between metal and oxide in agglomerate
R, r	= radius
Re	= Reynolds number
R_0	= 8.314 J/(mole · K), universal gas constant
T	= temperature
t	= time
V	= volume or velocity
Z_M	= share of unburned metal in agglomerates relative to initial metal in propellant
Z_M^a	= share of initial metal in propellant used to form agglomerates
Z_M^{ox}	= share of initial metal in propellant used to form oxide in agglomerates
Z_M^{SOP}	= share of initial metal in propellant used to form smoke-oxide particles
γ	= edge wetting angle

ΔV	= bypass flow velocity
η	= mass share of oxide in agglomerate
λ	= thermal conductivity
π	= 3.14
ρ	= density
σ	= surface tension
χ	= share of metal drop covered by oxide cap in agglomerate
Ω	= area

Subscripts

$A, B, E,$	= boundaries of characteristic zones in agglomerate combustion model
$AE, EB,$	= characteristic zones in agglomerate combustion model
BF, FC	= characteristic zones in agglomerate combustion model
a	= agglomerate
b	= bubble
cap	= oxide cap
g	= gas phase
M	= metal
o	= oxidizer
ox	= oxide
ox.d	= dissociated oxide
p	= smoke-oxide particles

Superscripts

$A, B, E,$	= boundaries of characteristic zones in agglomerate combustion model
F, C	= characteristic zones in agglomerate combustion model
$AE, EB,$	= condensed
BF, FC	= dissociated
c	= gaseous
d	= inflow
g	= inflow
in	= inflow
max	= maximal value
out	= outflow

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Introduction

COMBUSTION of aluminized solid rocket propellant (SRP) results in formation of condensed combustion products (CCP) consisting of two fractions: agglomerates and smoke-oxide particles (SOPs). Agglomerates consist of aluminum and its oxide. Their sizes can reach hundreds and even thousands of micrometers. The SOPs have the size of about 1 μm .

Condensed combustion products have an important influence on rocket motor operation. To provide normal operation of the motor, it is necessary to predict CCP parameters in the motor chamber. A

number of physical and chemical transformations of CCP, that is their evolution, takes place in the combustion chamber. Agglomerates play the most important role in the process of CCP evolution.

In mathematical modeling of physical and chemical transformations of agglomerates in combustion products flow, mainly attention has been paid to the process of metal combustion, and it has been considered acceptable to use descriptions developed for conditions of single particle combustion in various active media. Most studies have used the diffusive combustion model with the main principles of the theory of diffusive vapor-phase burning of drops of hydrocarbon fuels developed by G. A. Varshavskii.¹

For the first time the diffusive vapor-phase model of aluminum particle combustion in a mixture of oxygen and inert gas was formulated by T. Brzustowski and I. Glassman.² The main assumption of the model was that chemical reaction rate is much more than the diffusion rate of reacting components to the thin burning zone located some distance from the particle surface. In the framework of this model, the burning rate is controlled by the process of diffusion of reacting components.

Further the diffusive vapor-phase model has been modified by many authors.^{3–7} Their works have considered combustion of metal particles in various oxidizing media as well as questions associated with 1) influence of dissociation of condensed combustion products on thermal condition of the burning zone, 2) presence of an oxide cap on the surface of a metal drop, and 3) the effect of external flow on the burning process.

A. V. Voronetskii, A. V. Sukhov, and co-authors developed a so-called three-zone model.^{8,9} In the framework of the theory of diffusive vapor-phase combustion, they tried to take into account the influence of pressure on the burning rate. They assumed that beginning from a certain value of concentration of condensed particles in the burning zone these particles have an effect on diffusion of the reacting components. This description does not take into account hydrodynamic effects on motion and accumulation of those particles, that is, on effects, which are important for phenomena under discussion.

The model presented in Refs. 10–12 has a specific place among the models of metal particle combustion. This model was developed to provide determination of both burning rate and sizes of the condensed combustion products. The main assumption of the model is that the characteristic time of diffusion of gaseous components is much less than characteristic time of heterogeneous chemical reaction on surfaces of forming smoke oxide particles. A number of characteristics (geometrical sizes of burning zone, parameters of its temperature field) determined with the help of the model are in contradiction to most available experimental data.

The just-noted model was modified in Refs. 13 and 14 with the aim to describe, first of all, accumulation of oxide on surfaces of burning aluminum particles. In the new version of the model, it was taken into account that before the particle ignition some part of aluminum particle is covered by oxide, and in the process of combustion gaseous oxidizer reacts with aluminum on the free surface (not covered by oxide) with formation of alumina, following partial evaporation of this alumina and aluminum from the surface of burning particle. The model provides determination of the condition for transition from the heterogeneous mode of burning to gas phase.

Reference 15 presents a combustion model of motionless aluminum particles. The model takes into account both kinetics of chemical reaction and diffusion of components. It provides determination not only of burning rate but also fields of interaction potentials in the combustion zone, that is, distribution of temperature and partial pressures of gaseous substances. However formation of final condensed combustion products is not considered in that work.

Reference 16 presents a model, which, for the first time, includes descriptions of agglomerate metal combustion with accounting for 1) its blowing by two-phase flow and 2) real structure of the agglomerate. The model was based on the following principles. An agglomerate consists of two drops: metal and oxide (oxide cap). Oxidizing components of external flow reach the particle surface and take part in heterogeneous reaction with metal. Formed oxide is accumulated on the particle surface as oxide cap. Equilibrium evap-

oration of the mixture Al-Al₂O₃ takes place at the interface between these substances in the agglomerate. Heat transported to the agglomerate controls the kinetics of this process. The evaporation products come to the environment through the oxide cap. Those products condense on nuclei in the environment, which leads to formation of smoke-oxide particles. Condensation process rate is controlled by diffusion of components to the particle surface. The mathematical model consists of description of laminar motion of viscous incompressible liquid, motion of SOPs, condensation process, and heat balance on the agglomerate surface. Potentially, the model provides the possibility to determine burning rate, parameters of agglomerate structure, and sizes of forming SOPs. The authors do not discuss the question of reliability of the model while a number of the model assumptions are in contradiction with experimentally determined facts.

In the works of recent years, Liang and coworkers tried to take into account both real hydrodynamic details of the flow around burning particle and kinetics of chemical reaction and phase transformations.^{17–19} In the origin the model was aimed at describing combustion in cold air, and then it was modified for conditions of solid propellant combustion products. In the framework of the model, the following assumptions were made: 1) the burning particle is spherical and oxide covers some part of its surface, 2) gas flow is viscous and laminar, 3) size of the particle is constant during its burning. The model describes processes of metal evaporation, surface and gas-phase reactions, condensation process, specific details of two-phase flow around particle, and deposition of SOPs from the flow onto its surface. Analysis of the model allowed authors to make the conclusion that characteristic details of combustion (structure of burning zone and dependence of burning rate on different parameters) are close to the details of the diffusive model.

Analysis of available publications allows us to make the following conclusions:

1) Developed models do not describe a number of phenomena taking place in the agglomerate evolution (change in structure, oxide removal from agglomerate^{20,21}), and often they are based on assumptions that are in contradictions with available experimental data.

2) Taking into account the differences in parameters between the two environments, the use of results from modeling single particle combustion in active media to the case of agglomerate evolution in propellant combustion products flow is not correct.

3) The developed models, as a rule, do not provide prediction of parameters of agglomerates in the evolution process in combustion products flow with enough reliability for the practical tasks of designing and perfecting rocket motors.

The circumstances just listed stimulated the present work aimed at development of a mathematical model for agglomerate evolution in the flow of solid propellant combustion products. The model is based on experimental data on evolution of both single agglomerates and assemblies, which were obtained from extensive experimental investigation.^{20–23}

Model

The model of single agglomerate evolution includes a mathematical description of the main phenomena taking place in the process. They are 1) metal burning, 2) chemical interaction between metal and oxide, 3) deposition of SOPs on agglomerate, 4) change in the agglomerate structure, and 5) agglomerate motion.

Combustion of Agglomerate Metal

A great number of experimental data on combustion of aluminum drops in the environment of solid propellant combustion products have been obtained to date. Analysis supports the hypothesis of the diffusive mode of combustion process with formation of a burning area of finite dimensions. The scheme of the model is shown in Fig. 1. The model is based on the following assumptions:

1) The effect of external flow on the burning process is taken into account in the approximation of a "reduced film" (zone AC in Fig. 1). That is, the agglomerate is surrounded by motionless gas situated in zone AC where heat transfer is carried out by conduction, and mass transfer of gaseous substances is carried out by diffusion.

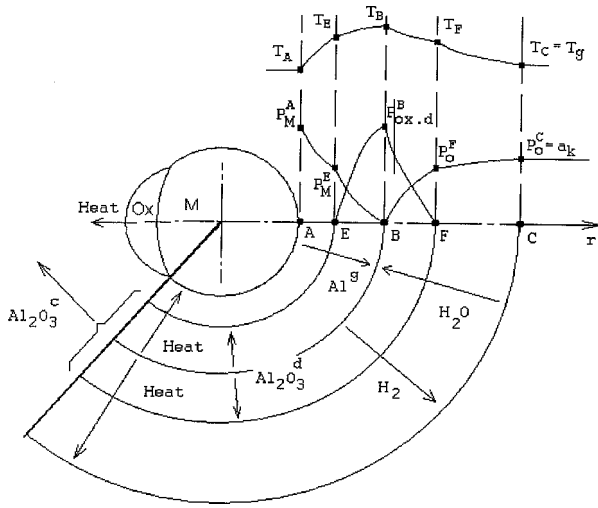


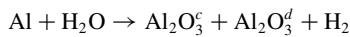
Fig. 1 Schematic of aluminum burning model.

2) Metal evaporation takes place in equilibrium mode from the metal drop surface, which is free of oxide cap.

3) The burning process is quasistationary, isobaric, and axisymmetric.

4) The burning process is controlled by diffusion, that is, the chemical reaction rate is much higher than the rate of component diffusion. The result of this assumption is that all chemical reactions run in equilibrium mode. This important circumstance allowed us to use the following scheme. Interaction of metal vapors and oxidizing components from the environment occurs within a vanishingly narrow area B (Fig. 1). The interaction releases such great energy that a part of the oxide (Al_2O_3) is formed in the gas-phase state and then diffuses into the environment where it condenses to form smoke-oxide particles. Oxide condensation occurs within the area EF (Fig. 1.). Thus reaction between Al vapor and external gaseous oxidizers completes within the area EF with formation of gaseous products and condensed aluminum oxide in the form of SOPs. Thus the area EF can be considered as the burning zone.

5) The oxidizing component from the environment is water vapor. In the combustion products of SRP, water vapor represents the main oxidizing component because the content of the other oxidizing component, carbon dioxide, is comparatively low. According to Ref. 24 the mixture of CO_2 and H_2O can be formally considered as equivalent to a certain amount of pure water vapor. In this case the equation of chemical reaction in area B can be written in the following form (without account for stoichiometric coefficients):



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



7) According to thermodynamic estimate, this reaction runs in the following way:



Approximately three moles of gaseous substances are formed from one mole of Al_2O_3^c .

8) Products of aluminum oxide dissociation ("evaporation") can be considered as a pseudosubstance, that is, pseudovapor (Al_2O_3^d), which has dualistic properties as follows. On one hand, evaporation of one Al_2O_3^c molecule occurs with formation of a few molecules of gaseous substances, that is, the pseudovapor is a mixture of different substances. On the other hand, the products of one molecule of evaporation cannot be separated one from another so that they present an integral unit. That is, the components of the dissociation products are deprived of any individuality and exist only as components of

the pseudosubstance Al_2O_3^d . Pseudovapor is neutral in relation to other components of gas mixture and is capable only of undergoing phase transformation. (It can condense with formation of Al_2O_3^c .) This assumption allows us to describe diffusion and equilibrium condensation of dissociated oxide in the area EF (Fig. 1).

9) Transport coefficients in the characteristic areas are determined by averaged values of temperature and composition parameters.

10) SOPs do not influence diffusion of gaseous substances in zone EF.

11) Binary coefficients of diffusion of all gaseous components are the same in each characteristic area; temperature diffusion does not take place.

12) Lewis number is equal to 1 (Ref. 25).

13) Distributed sources of matter and energy in zone EF are taken into account by means of effective flows.

14) SOPs formed in zone EF are removed into the so-called "trail" under the effect of gas dynamic forces exerted by the environment (Fig. 1). This assumption was made to take into account an obvious feature of the agglomerate burning process, namely formation of "trails" saturated with condensed products of agglomerate metal burning.²³

The mathematical model of agglomerate aluminum burning represents a set of ordinary differential equations describing heat and mass transfer within the "reduced film" AC.

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

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

Here $\bar{P}_i = P_i/P$; P_i is partial pressure of i th component; N is number of components transported through cross section Ω including the i th component. Integration of Eqs. (1) with account for boundary conditions

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

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

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

$$r = R_F : \bar{P}_O = \bar{P}_O^F, \quad \bar{P}_{\text{ox.d}} = \bar{P}_{\text{ox.d}}^F$$

$$r = R_C : \bar{P}_O = \bar{P}_O^C = a_k$$

gives six equations of component diffusion in characteristic areas, namely, 1) two equations for diffusion of metal vapor (in areas AE and EB), 2) two equations for diffusion of water vapor (in areas BF and FC), and 3) two equations for diffusion of dissociated oxide (in areas BE and BF).

Energy transport in the characteristic areas is described by equations of energy conservation written in the following general form:

$$\sum_{i=1}^n J_i^{\text{in}} H_i - \sum_{j=1}^m J_j^{\text{out}} H_j + Q_{\text{sp}} + Q_{\text{con}}^{\text{in}} - Q_{\text{con}}^{\text{out}} + Q_{\text{rad}}^{\text{in}} - Q_{\text{rad}}^{\text{out}} = 0 \quad (2)$$

Integration of Eqs. (2) with the following boundary conditions

$$r = R_A : T = T_A, \quad r = R_E : T = T_E, \quad r = R_B : T = T_B$$

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

gives four equations of heat balance in areas AE, EB, BF, and B.

Assuming equilibrium character of metal evaporation and oxide dissociation, the partial pressures $\bar{P}_M^A, \bar{P}_{\text{ox.d}}^E, \bar{P}_{\text{ox.d}}^F, \bar{P}_{\text{ox.d}}^B$ are connected with temperatures T_A, T_E, T_B , and T_F by the Clausius-Clapeyron equation. For partial pressures of dissociated oxide on the boundaries of area EF, it has been assumed that $\bar{P}_{\text{ox.d}}^F = \bar{P}_{\text{ox.d}}^B = (0.01)P$.

Values of transport coefficients for the gas mixture in the characteristic areas have been calculated on the basis of algorithms and information from Refs. 26 and 27. It was taken into account that transport coefficients depended on the fraction of aluminum burned out, which is characterized by parameter $(1 - Z_M)$. Heat consumption of the interface between metal and oxide in the agglomerate (parameter Q_{sp}) was determined with help of a model of interaction between condensed aluminum and oxide as described next.

The mathematical model for agglomerate metal burning in final form consists of 10 algebraic transcendental equations for heat and mass transfer in the characteristic areas written in general form as follows: $\Phi_i = 0$ ($i = 1, 2, \dots, 10$). The equations are solved using an optimization procedure by means of minimization of the target function of the following form²⁸:

$$\Delta\Phi = \sum_{i=1}^{10} \left(\frac{\Phi_i}{\phi_i} \right)^2 \quad (3)$$

Here ϕ_i is scale factor of function Φ_i . The model provides determination of aluminum burning rate, temperature field, and substances concentrations in zone AC.

Here are some results of parametric analysis of the model:

1) Parameters J_M (mole flux of evaporating metal) and T_A (agglomerate temperature) are quite substantially dependent on transport coefficients in zones BF and FC.

2) Such parameters of the environment as a_k , T_C , and gas flow velocity have the most influence on the agglomerate temperature.

3) Intensity of heat exchange between the metal particle and environment through the oxide cap is rather small, and this process has little influence on thermal state of the particle.

The developed comprehensive model of agglomerate aluminum burning is quite complicated; this fact causes difficulties in its usage. For the aim of calculation of agglomerate aluminum burning rate, a macromodel was developed based on the comprehensive model with a number of additional essential assumptions. The macromodel reproduces principal functional relationships of the comprehensive model and provides determination of agglomerate aluminum burning rate as function of metal drop and oxide cap sizes, pressure, oxidizing potential, and other parameters of the gas flow. It has the following form:

$$J_M = 2\pi R_A \kappa_1 (1 - \chi) Nu \left(\frac{\lambda}{c_p} \right)_{BC} \times \left\{ \frac{2}{3} a_k + \left(\frac{c_p}{\lambda} \right)_{BC} \cdot \left(\frac{\lambda}{c_p} \right)_{AB} \cdot \ln \left[\frac{c_{pM}(T_B - T_A)}{L_M} + 1 \right] \right\} \quad (4)$$

Here $Nu = 2 + 0.6 Re^{0.5} Pr^{0.333}$. Parameter κ_1 is a matching coefficient. Its value as well as values of transport coefficients and other parameters are presented next.

The quality of any model depends, first of all, on its universality and ability to reproduce experimentally fixed regularities. Temperatures of agglomerate T_A and of burning zone T_B , calculated with the help of the present model, are quite close to experimentally measured values.^{29,30} Measurements indicate that the temperature of agglomerates T_A , burning in the gas-phase mode on the propellant surface, is in the interval between 2400 and 2600 K at pressure 0.1 MPa. The model gives this value as 2380 K. Experimentally obtained values of burning zone temperature T_B are 3500–3700 K in the pressure range 2.0–4.0 MPa for the case of agglomerate combustion in SRP combustion products. The model gives this value as 3650 K at 2.0 MPa and 3720 K at 4.0 MPa.

Figure 2 presents dependencies of parameter J_M^Σ on Re and η obtained from the model (curve 6) as well as values of J_M^Σ (lines 1–5) calculated on the basis of relationships obtained experimentally by different researchers.^{31–38} Parameter J_M^Σ is determined as follows: $J_M^\Sigma = J_M + J_{M2}$; here J_M is mole flux of burning metal, and J_{M2} is mole flux of metal taking part in interaction with oxide on the interface between these substances in the agglomerate. (The model

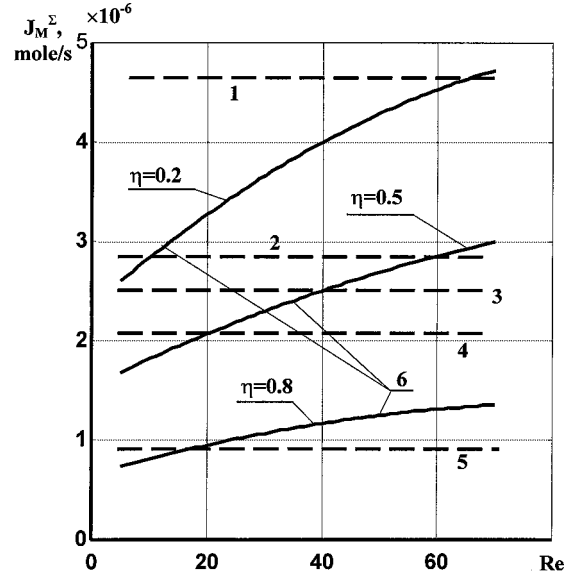
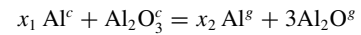


Fig. 2 Mole rate of agglomerate metal burning ($D = 150 \mu\text{m}$, $a_k = 0.2$, and $P = 6.0 \text{ MPa}$).

of interaction presented next provides calculation of J_{M2} .) Comparison (Fig. 2) reveals the most probable reason for great differences in values of burning rates determined by different laws of aluminum particle combustion. Dependencies of J_M^Σ on Re and η and lines 1–5 lie approximately in the same region (Fig. 2). This fact allowed us to conclude that the relationships just noted (Refs. 31–38) give so different results because they do not take into account a number of very important factors, namely, real structure of agglomerates, influence of gas flow, and chemical interaction between substances of the agglomerate.

Chemical Interaction of Condensed Al and Al_2O_3

All experimental data obtained to date and results of thermodynamic estimate shown in Fig. 3 allowed us to consider chemical reaction between aluminum and oxide as the most probable cause of the decrease in the amount of oxide in agglomerates during their evolution. (This phenomenon was named oxide removal.) In Fig. 3 parameter P_i is relative partial pressure of i th component in the gas mixture formed as result of evaporation of aluminum and aluminum oxide. The equation for chemical interaction can be written as follows:



Here:

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

A general layout of the process can be presented as follows (Fig. 4). Chemical reaction runs at the interface of agglomerate substances with formation of bubbles of gaseous products. The bubbles are retained at the interface by surface tension forces. The bubbles grow in size and under certain conditions can reach the outside boundary of the oxide cap. In this case bubbles collapse and release reaction products into the environment. This process causes periodical ballooning of agglomerates and decrease of oxide and metal content in them.^{20,21}

Experimental study has established that the intensity of oxide removal from agglomerates depends on pressure, agglomerate size, and oxide content in the agglomerate.²¹ On the basis of obtained experimental data, a mathematical model of the process of oxide removal from agglomerates has been developed. It is based on the following principal assumptions:

1) Chemical reaction takes place on reaching a certain temperature. It occurs within the adsorption layer formed at the interface of

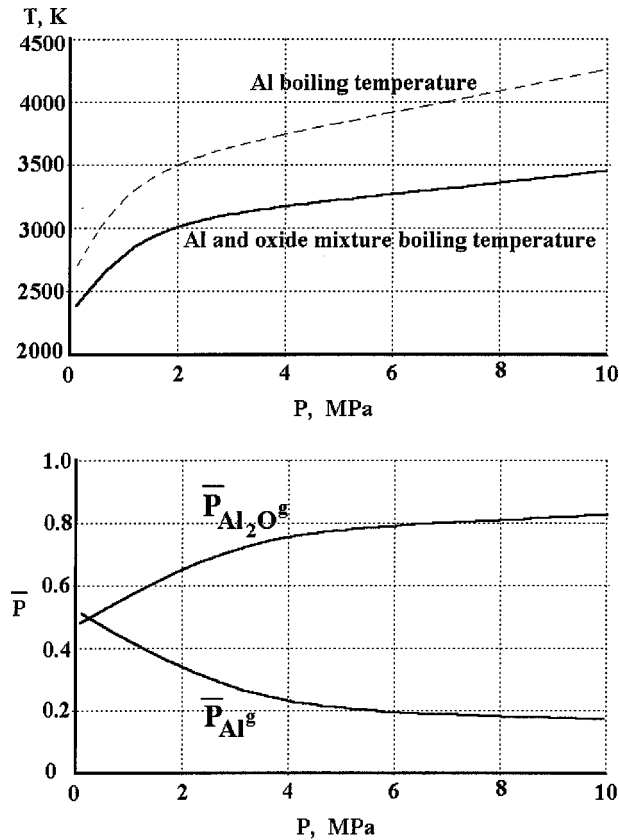


Fig. 3 Results of the thermodynamic estimate for the reaction: $\text{Al}^c + \text{Al}_2\text{O}_3^c \rightarrow \text{Al}_2\text{O}_3^g + \text{AlO}^g + \text{AlO}_2^g + \text{Al}^g + \text{O}^g + \text{O}_2^g$.

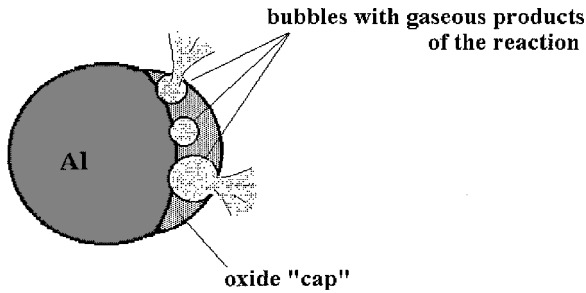


Fig. 4 General layout of oxide removal from agglomerate.

metal and oxide in the agglomerate. Dissolved gases serve as nuclei of gas formation.

2) Primary bubbles of the reaction products grow and merge yielding rather large bubbles going far beyond the boundaries of the adsorption layer.

3) Each large growing bubble has a corresponding area of feeding—an adsorption layer area surrounding that particular bubble and having constant radius R_f . Products of the reaction running within this area are fed into the bubble. It is also assumed that radius R_f does not depend on pressure as well as on other evolution conditions.

4) Initial bubble size does not exceed the thickness of the adsorption layer. Merging of small initial bubbles with large central bubble is the “fast” process.

5) In large bubbles gone beyond the boundaries of the adsorption layer, a reverse process takes place, that is, condensation of gases back to liquid aluminum and aluminum oxide running on the internal surface of the bubble.

6) The condensation process involves only a certain part of bubble internal surface, which does not change in the process of bubble growth.

7) On the whole, the bubble growth is determined by simultaneous running (competing) of processes of evaporation and condensation.

8) If a bubble becomes so large that it reaches the external boundary of the oxide cap, it will collapse and release gases into the environment. Otherwise, only agglomerate ballooning occurs.

9) Oxide cap thickness is assumed to be the same for all large bubbles (equal to some effective value) so that conditions for growth of all large bubbles are the same;

10) Bubbles are spherical.

11) Temperature is the same throughout the agglomerate cross section.

12) Products of the reaction are ideal gases.

These assumptions allowed us to develop the following mathematical model of oxide removal from the agglomerate.

The expression for Al_2O_3 evaporation rate w_1 is in the form of an evaporation law:

$$w_1 = A_1 \exp \left\{ -\frac{E_a}{R_0 T_A} \right\} \frac{1}{\sqrt{T_A}} (P_s - P), \quad \frac{\text{mole of Al}_2\text{O}_3^c}{\text{m}^2 \cdot \text{s}} \quad (5)$$

Here A_1 is a factor depending on properties of evaporating mixture of substances, E_a is an activation energy of the process, P_s is pressure of saturated vapor of the reaction products. The value of P_s is determined by agglomerate temperature T_A .

The equation for the rate of Al_2O_3 formation on the internal surface of a large bubble w_2 has the form:

$$w_2 = A_1 \exp \left\{ -\frac{E_a}{R_0 T_A} \right\} \frac{1}{\sqrt{T_A}} (P - 0), \quad \frac{\text{mole of Al}_2\text{O}_3^c}{\text{m}^2 \cdot \text{s}} \quad (6)$$

The resultant rate of Al_2O_3 consumption in the process of large bubble growth is

$$w_\Sigma = w_1 \Omega_1 - w_2 \Omega_2 \quad (7)$$

Here Ω_1 is replenishment area, and Ω_2 is the area of the bubble surface, where condensation takes place.

The law of single large bubble growth has the form:

$$\frac{dV_b}{dt} = \frac{R_0 T}{P} w_\Sigma (3 + x_2) \quad (8)$$

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

The time of single large bubble growth till its collapse is

$$t = \int_0^{R_b^{\max}} 4\pi R_b \frac{P}{R_0 T_A} \cdot \frac{1}{w_\Sigma (3 + x_2)} dR_b \quad (9)$$

Here R_b^{\max} is maximum radius of the bubble at the moment of its collapse.

The rate of removal of the reaction products from a unit area of interface $\text{Al}^c - \text{Al}_2\text{O}_3^c$ in the agglomerate W :

$$W = \frac{V_b^{\max}}{t\pi R_f^2}, \quad \frac{\text{m}^3 \text{ of the reaction products}}{\text{m}^2 \cdot \text{s}} \quad (10)$$

Here V_b^{\max} is maximum volume of the bubble at the moment of its collapse.

The final expression for the oxide removal J_{ox} is as follows:

$$J_{\text{ox}} = \frac{P}{R_0 T_A} W \frac{1}{3 + x_2} \Omega_{\text{M-ox}}, \quad \frac{\text{mole of Al}_2\text{O}_3^c}{\text{s}} \quad (11)$$

Here $\Omega_{\text{M-ox}}$ is area of the interface $\text{Al}^c - \text{Al}_2\text{O}_3^c$ in the agglomerate.

This model of agglomerate oxide removal takes into account physical principles of the process. It describes dependence of the oxide removal rate on various factors, such as pressure P and parameters of the agglomerate (R_A , η , and T_A). The model contains a number of parameters, which cannot be estimated exactly, including coefficient A_1 , activation energy E_a , radius of replenishment area R_f , etc. These parameters are considered as matching coefficients.

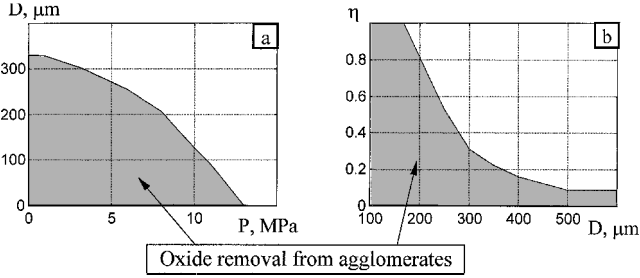


Fig. 5 Results of parametric analysis of the oxide removal model.

Figure 5 presents some calculated results, namely, conditions of realization of oxide removal in the following situation: $a_k = 0.2$, $\Delta V = 2$ m/s, $T_g = 3300$ K. Parameter η is 0.5 in Fig. 5a. Pressure P is 6 MPa in Fig. 5b.

Because of different surface tensions of aluminum and aluminum oxide, large bubbles of the reaction products are almost entirely situated inside the agglomerate oxide cap, therefore causing an increase in the cap volume. This fact is taken into account by using an effective value of oxide cap density ρ_{cap} . On the basis of conducted estimates, it has been determined that ρ_{cap} may be less than the density of liquid oxide ρ_{ox} taken at temperature T_A by approximately 40%. This is why it was assumed that $\rho_{\text{cap}} = 0.6 \cdot \rho_{\text{ox}}$.

Removal of products of the reaction between substances in the agglomerate is a reason of not only the oxide removal from the agglomerate but also removal of metal involved in the reaction. Additional removal of metal because of its chemical interaction with oxide may reach 30% in comparison with metal consumption in the burning process. The mole flux of aluminum consumed in the reaction with oxide and removed from agglomerate J_{M2} is estimated in the following way: $J_{M2} = x_1 \cdot J_{\text{ox}}$.

Deposition of SOPs on Agglomerates

In investigation of the CCP evolution in the solid propellant combustion products flow, we should take into account probable collisions of condensed-phase particles and, first of all, collisions of SOPs with agglomerates. One of the possible results of such collisions may be deposition of SOPs on agglomerates and, as a result, an increase of oxide mass in them. (This occurs in the case where SOPs deposit on the surface of the oxide cap²¹.)

To account for this process, a model of SOPs collisions with agglomerates has been developed. It is based on the following principal assumptions:

- 1) The agglomerate can take different positions in the gas flow relative to direction of the flow (Fig. 6).
- 2) In the case where gas flow blows around the agglomerate from the metal side of drop, conditions for SOPs deposition are worse in comparison with reverse orientation of the agglomerate as a result of metal evaporation from the surface of the drop.
- 3) The gas is viscous and incompressible.
- 4) SOPs have the same size equal to some average value.

The mathematical model of the process is constructed in the following way. At first, the gas velocity field is determined in the region disturbed by the agglomerate, and then trajectories of SOPs motion in this region are calculated.

In the system of coordinates associated with the agglomerate, equations of smoke-oxide particle motion are written in the vector form as follows:

$$\frac{d\mathbf{V}_p}{dt} = \mathbf{F}_p, \quad \frac{d\mathbf{l}_p}{dt} = \mathbf{V}_p \quad (12)$$

Here \mathbf{V}_p is smoke-oxide particle velocity, \mathbf{F}_p is aerodynamic force applied to smoke-oxide particle from the side of gas flow, and \mathbf{l}_p is a spatial coordinate of the particle.

It is assumed that collision of smoke-oxide particles with an agglomerate results in deposition on the agglomerate. Calculations are made for two variants of agglomerate orientation in gas flow. The model provides determination of the rate of deposition of SOPs on

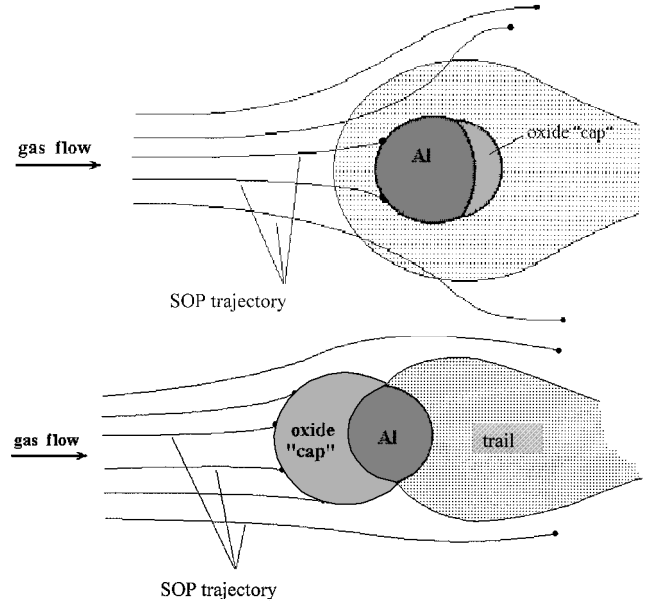


Fig. 6 General layout of SOP deposition on agglomerate.

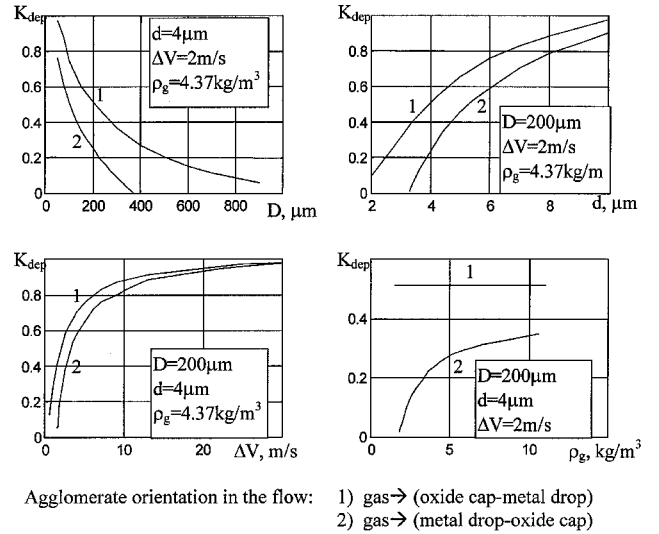


Fig. 7 Dependencies of K_{dep} on D , ΔV , d , ρ_g , and agglomerate orientation.

an agglomerate with dependence on orientation and diameter of agglomerate, gas flow velocity and density, and SOP's size. Figure 7 shows the influence of listed parameters on the coefficient of SOP deposition on the agglomerate K_{dep} , which represents the fraction of SOPs deposited on the agglomerate in relation to SOPs moving in the flow within tube with diameter equal to diameter of the agglomerate.

The method of determination of parameter K_{dep} in the framework of this comprehensive model of SOP deposition is quite complicated and therefore not appropriate for practical tasks, such as calculation of agglomerate population evolution. Therefore a formal macro-model of SOP deposition has been developed on the basis of parametric analysis of the comprehensive model and with use of the "self-adjusting" method.³⁶ The macromodel represents a relationship for calculation of K_{dep} with dependence on D , d , ΔV , ρ_g , and orientation of the agglomerate.

Agglomerate Structure

The agglomerate structure model is based on the assumption that burning agglomerates can be regarded as an equilibrium system, the structure of which is determined by quantities and surface properties of its components.³⁷ Other assumptions are as follows:

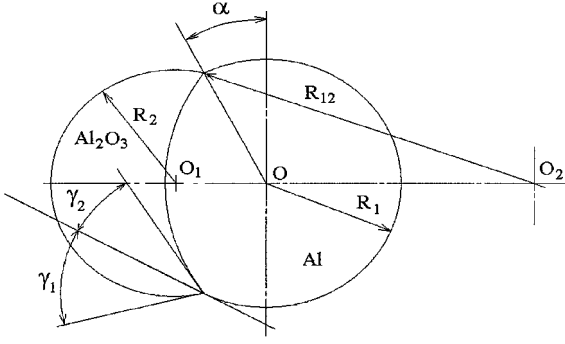


Fig. 8 Schematic of agglomerate structure.

1) Temperature of agglomerate is the same throughout any cross section.

2) Agglomerate diameter corresponds to the biggest of the two: that of metal drop or oxide drop in the agglomerate.

The agglomerate structure model includes equations linking volumes of metal drop and oxide cap with their geometrical characteristics. Figure 8 presents a schematic of agglomerate structure including geometrical parameters, which can be calculated with the help of the model. Input information includes fraction of oxide in the agglomerate η , agglomerate diameter D , densities of aluminum and oxide cap in agglomerate ρ_M and ρ_{cap} , and edge wetting angles γ_1 and γ_2 (Fig. 8). The algorithm of calculation is as follows:

1) Determination of angle α (Fig. 8) as a function of parameter η by solving a set of equations is shown here:

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

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

$$V_M = \frac{\pi}{3} R_1^3 \left\{ [1 + \sin \alpha]^2 [2 - \sin \alpha] + \frac{\cos^3 \alpha}{\cos^3(\alpha + \gamma_2)} \cdot [1 - \sin(\alpha + \gamma_2)]^2 \cdot [2 + \sin(\alpha + \gamma_2)] \right\} \quad (13)$$

Here V_M and V_{cap} are volumes of metal drop and oxide cap in the agglomerate.

2) Calculation of all other parameters of agglomerate structure with determined value of angle α is shown as follows:

$$\chi = 0.5(1 - \sin \alpha) \quad (14)$$

$$\text{If } \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} > 1, \text{ then } \begin{cases} R_2 = 0.5D \\ R_1 = R_2 \frac{\cos(\alpha - \gamma_1)}{\cos \alpha} \end{cases} \quad (15)$$

$$\text{If } \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} \leq 1, \text{ then } \begin{cases} R_1 = 0.5D \\ R_2 = R_1 \frac{\cos \alpha}{\cos(\alpha - \gamma_1)} \end{cases} \quad (16)$$

$$R_{12} = R_1 \frac{\cos \alpha}{\cos(\alpha + \gamma_2)} \quad (17)$$

Volumes V_M and V_{cap} are calculated in accordance with Eqs. (13).

Values of γ_1 and γ_2 were obtained experimentally at two pressure levels.³⁷ Temperature of burning agglomerate T_a at different pressures was estimated using the agglomerate burning model just described. All of this information with addition of data on temperature dependence of oxide surface tension on the interface with

gaseous media σ_2 taken from Ref. 38 was used for determination of temperature dependencies of surface tension on interfaces between 1) aluminum and oxide in agglomerate σ_{12} and 2) agglomerate aluminum and gaseous environment σ_1 . These dependencies were obtained in the following form:

$$\sigma_2(T) = 0.66 - 1.96 \times 10^{-4}(T - 2306), \text{ N/m} \quad (18)$$

$$\sigma_1(T) = 1.267 - 2.6 \times 10^{-4}(T - 933), \text{ N/m} \quad (19)$$

$$\sigma_{12}(T) = 0.687 - 1.6 \times 10^{-4}(T - 933), \text{ N/m} \quad (20)$$

Usage of Eqs. (18–20) is correct for determination of agglomerate structure in the interval of T_a between 2300 and 3500 K. Wetting angles γ_1 and γ_2 should be calculated with use of these equations on the basis of the condition of mechanical equilibrium of contact line at the interface of three media: aluminum, oxide, and surrounding gas.

$$\gamma_1 = \arccos \frac{\sigma_1^2 + \sigma_2^2 - \sigma_{12}^2}{2\sigma_1\sigma_2} \quad (21)$$

$$\gamma_2 = \arccos \frac{\sigma_1^2 + \sigma_{12}^2 - \sigma_2^2}{2\sigma_1\sigma_{12}} \quad (22)$$

For example, at 3000 K and with $\rho_{cap} = 0.6\rho_{ox}$ agglomerates with $D = 200 \mu\text{m}$ and $\eta = 0.3$ have the following parameters of structure: $R_1 = 100 \mu\text{m}$, $R_2 = 96 \mu\text{m}$, $R_{12} = 195 \mu\text{m}$, $\alpha = 0.323 \text{ rad}$, $\chi = 0.341$.

Agglomerate Motion

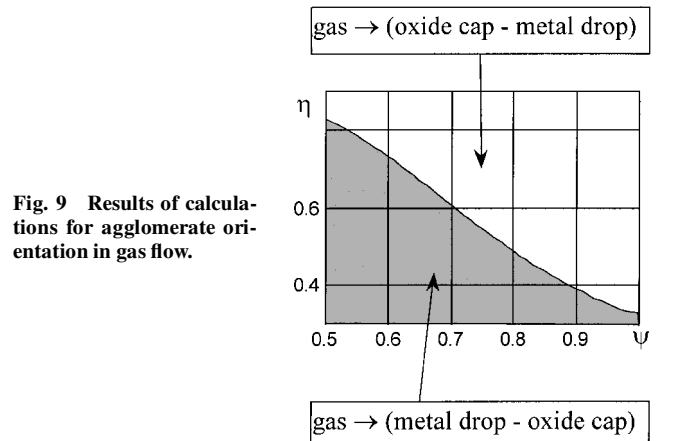
Modeling of the agglomerate motion was carried out in accordance with the approach suggested in Ref. 32. The motion model describes the motion of a single agglomerate in the gas flow under the effect of aerodynamic and inertia forces. Deposition of SOPs on an agglomerate causes additional change of agglomerate impulse. The motion model takes into account this factor; it was assumed that the difference in velocities of the agglomerate and SOPs colliding with it is equal to the difference in velocities of the agglomerate and the gas flow ΔV .

Agglomerate motion equations in vector form are as follows:

$$\frac{dV_a}{dt} = \frac{1}{2} \cdot \frac{C_{drag} S_a \rho_g \Delta V}{M_a} \Delta V + F_m + \frac{K_{dep} \cdot \varphi_{SOP} \cdot S_a \cdot \Delta V}{M_a} \Delta V$$

$$\frac{dI_a}{dt} = V_a \quad (23)$$

Here C_{drag} is agglomerate drag coefficient, S_a is area of agglomerate maximum midsection, F_m is mass force induced by the Earth gravity and rocket acceleration, φ_{SOP} is mass concentration of SOPs in gas flow, and I_a is spatial coordinate of the agglomerate. It was established with calculations that the effect of jet power caused by outflow of metal vapor from the agglomerate metal surface on



agglomerate motion can be neglected. The orientation of the agglomerate in a gas flow was also calculated. It was assumed that the orientation corresponds to the most stable position of the agglomerate as determined by the relative positions of mass and pressure centers of the agglomerate.

Calculation results presented in Fig. 9 show that at the initial stages of evolution gas flow blows around the agglomerate from the side of the metal drop. At the final stages, when a certain magnitude of η is reached the agglomerate can change its orientation so that gas flow will blow around the agglomerate from the side of its oxide cap. The calculations were performed taking into account the phenomena of oxide cap ballooning (in Fig. 9 $\psi = \rho_{\text{cap}} / \rho_{\text{ox}}$). Figure 9 shows that solid (without bubbles) agglomerates, containing more than 30% oxide ($\eta > 0.3$) should be in position **b** (positions **a** and **b** are taken in accordance with Fig. 6) because of higher density of liquid oxide in comparison with liquid aluminum. Ballooning of oxide cap with bubbles causes a decrease in the probability of that position. For example, at $\rho_{\text{cap}} = 0.6\rho_{\text{ox}}$ position **a** is more probable in the case of $\eta < 0.7$. These results are in good correspondence with available experimental data.

Synthesis of Different Models

All of the models just presented together constitute a model of evolution of a single agglomerate in the combustion products of an

model 4 (change in oxide mass in agglomerate for some period of time) and in model 5 (additional force influencing the agglomerate).

4) The model of agglomerate structure (model 4) provides determination of geometrical parameters of the agglomerate, which are used in models 1, 2, 3, and 5.

5) The model of agglomerate motion (model 5) provides determination of agglomerate velocity and spatial coordinates of the agglomerate, which are used in model 1 (a_k , T_g , and ΔV) and in model 3 (ΔV).

Matching coefficients of the model were determined by means of comparison of the model, in describing the evolution of an entire agglomerate population in a combustion flow of SRP, with experimental data. Experimental data were obtained for the case of one-dimensional motion of combustion products flow, taking data at different distances L_S from the burning surface of the propellant specimen.²¹ The mathematical model of evolution of the population of agglomerates in one-dimensional flow was developed on the basis of the model of single agglomerate evolution. Calculations were conducted with use of an optimization procedure. Matching coefficients played the role of variable parameters. The target function Δ^Σ represented disagreement of calculation results and experimental data, namely, values of parameters Z_M and Z_M^{ox} at distance $L_S = 70$ mm. Parameter Δ^Σ was defined on the basis of the total available experimental data as follows:

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

aluminized solid rocket propellant. Figure 10 shows schematically links between the particular models, which provide exchange of the following information:

1) The metal burning model (model 1) provides determination of agglomerate temperature and burning rate, which are used in model 2 (T_A), in model 3 (velocity of flow from metal drop surface), and in model 4 (change in metal mass for some period of time).

2) The model of chemical interaction of Al^c and Al_2O_3^c (model 2) provides determination of the oxide removal rate, which is used in model 1 (heat consumption on the interface $\text{Al}^c - \text{Al}_2\text{O}_3^c$) and in model 4 (change in masses of metal and oxide for some period of time, ρ_{cap}).

3) The model of SOP deposition on agglomerates (model 3) provides determination of flux of deposited SOPs, which is used in

Here N is total number of investigated cases of agglomerate evolution:

Calculated data:

$$[Z_M(70 \text{ mm})]^{\text{calc}}, \quad [Z_M^{\text{ox}}(70 \text{ mm})]^{\text{calc}}$$

Experimental data:

$$[Z_M(70 \text{ mm})]^{\text{exp}}, \quad [Z_M^{\text{ox}}(70 \text{ mm})]^{\text{exp}}$$

At confidence probability 0.997 parameter Δ^Σ is equal to $(5.5 \pm 0.8)\%$, which allowed us to conclude that the quality of both the obtained experimental data and the developed model of agglomerate evolution was relatively good. Figure 11 shows, for example, experimental data and calculation results in the case of one of the propellants used (propellant N4 in accordance with Ref. 21). The obtained values of all of the matching coefficients just

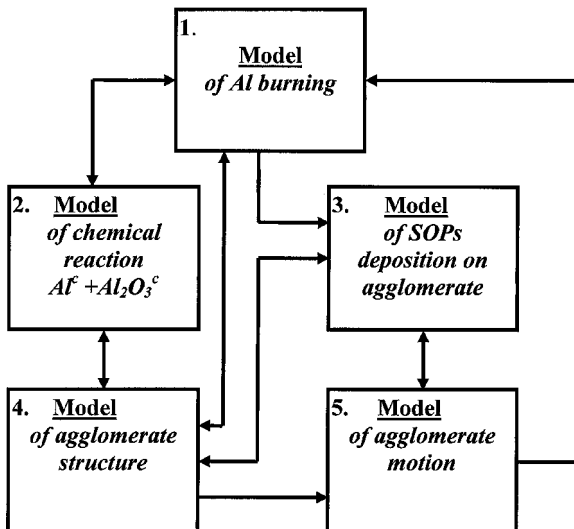


Fig. 10 Block diagram of the model of single agglomerate evolution.

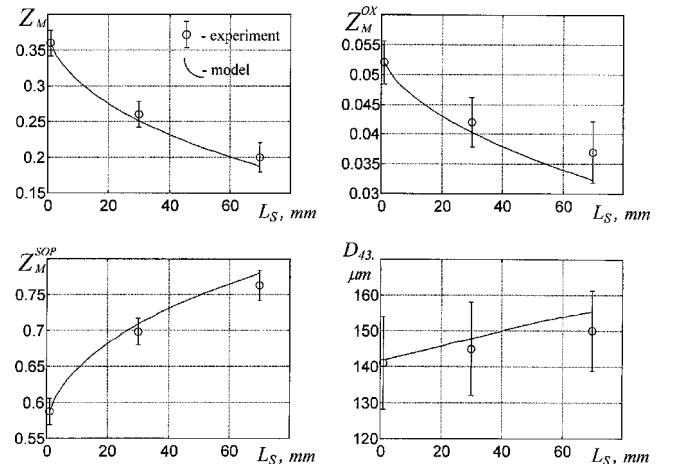


Fig. 11 Comparison of experimental data and calculation results.

mentioned are $k_1 = 0.775$, $A_1 = 1.122 \times 10^{-4}$, $E_a = 74.82$ J/mole, $R_f = 7.516 \times 10^{-5}$ m. Also let us return here to Eq. (4) and present values of some relevant parameters. Transport coefficients were evaluated on the basis of results of thermodynamic calculations performed for all propellants used in the experimental study. The following values were used in calculations of evolution of a population of agglomerates in one-dimensional flow:

$$\lambda_{AB} = 0.20, \text{ J/(m} \cdot \text{s} \cdot \text{K)}, \quad c_{PAB} = c_{PM} = 36.0, \text{ J/(mole} \cdot \text{K)}$$

$$L_M = 2.84 \times 10^5, \text{ J/mole}, \quad Pr = 1$$

$$\lambda_{BC} = 0.2774 + 0.1104 \cdot (1 - Z_M) - 0.0170 \cdot (1 - Z_M)^2, \text{ J/(m} \cdot \text{s} \cdot \text{K)}$$

$$c_{PBC} = 45.08 - 8.210 \cdot (1 - Z_M) - 1.518 \cdot (1 - Z_M)^2, \text{ J/(mole} \cdot \text{K)}$$

It was also assumed in those calculations that temperature T_B was equal to the aluminum oxide boiling point and temperature T_A was equal to the boiling point of the mixture of aluminum and its oxide (Fig. 3).

Conclusions

The main result of this work consists of the development of a mathematical model of evolution of single agglomerates in combustion products of aluminized solid rocket propellant. The model describes a wide range of phenomena that have been observed experimentally. High quality of the model was proved by comparison with experimental data on evolution of both single agglomerates and populations of agglomerates.

The developed model was successfully incorporated into other models of physical and chemical processes taking place in the combustion chamber of solid propellant rocket motor. The model can be used to solve some practical problems of designing and perfecting rocket motors. Future works on mathematical modeling evolution of SRP condensed combustion products are planned to be associated with development of model for SOP evolution.

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