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THE STUDY OF CARBON MATERIAL'S AEROTHERMOCHEMICAL DESTRUCTION IN COMBUSTION PRODUCTS OF LIQUID-PROPELLANT ROCKET ENGINES

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Abstract

The process of hypersonic vehicles' movement in the dense layers of the atmosphere is accompanied by the considerable combustion of heat shield, which effects on the aerodynamic, mass-inertial and centering characteristics of the product.

For correct calculation of model's movement parameters is necessary to:

- Using the theoretical and computation methods for determining ablative characteristics of thermal-protective materials;
- Taking into account all the basic physical and chemical processes, involved in their ablation, using the above mentioned methods;
- Testing these techniques on the wide range of experimental data;
- Consideration the full range of issues related on the movements and ablation the thermal protective materials in the mutual conjugate formulation of the problem.

At the present time, carbon materials are widely used as the thermal protection for high-temperature heating elements of hypersonic air vehicles. The subject of this paper is one of the aspects of determining the ablation characteristics of carbon materials (CM).

1 Physico-mathematical statement of the problem

There is voluminous bibliography to issues related to CM ablative properties calculations. It can be reference to papers [1-4] and this issue's review in the monograph [5].

The main physico-chemical transformations of carbon during the ablation are:

- Kinetic oxidation on the surface in air stream (for which we shall use the term 'wall');
- Its sublimation, which runs on the wall;
- Its thermomechanical 'erosion' (mechanical ablation), which emerges on adjacent to a wall layer of material.

Fig. 1 shows typical dependence G_w mass velocity of thermochemical CM destruction from the wall T_w temperature.

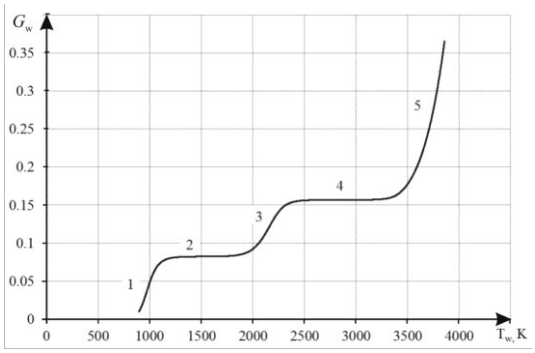

Fig. 1. Mass velocity of thermochemical CM destruction dependence from the wall temperature.

Herein:

1, 2 - 'Kinetic branch' and 'diffusion shelf' in conditions when the carbon dioxide is the only stable chemical compound of carbon;

3 - Kinetic branch in conditions when carbon monoxide is added to stable chemical compounds of carbon;

4 - Diffusion shelf in conditions, when the carbon monoxide is the only stable chemical compound;

5 – 'Sublimate branch' in conditions, when pure carbon and cyanogen are added to stable chemical compounds of carbon;

G_w is measured in parts of the heat transfer coefficient $A_{h,n}$ on impenetrable wall; T_w, K.

The process of oxidation, used in practice by sublimation mode ablation of carbon, is considered in the diffusion approximation as a part of the basic methods of calculation [1-4]. Moreover, the products of the sublimation of carbon are not involved in chemical reactions occurring on the wall.

The full model of thermochemical carbon's ablation (in which the above limitation is deleted) was represented in the papers [6, 7]. This was achieved due to the fact that in the whole range of definition of the temperature of the wall, the rate of oxidation of carbon is considered in the kinetic formulation.

However the specification of estimated theoretical model of carbon ablation (at using of methodology from the papers [6, 7]) is jointed with the necessity of determining the carbon corrosion kinetic constants by atomic oxygen (mainly oxygen is in atomic condition on sublimation branch).

Significant impact on the CM ablation depends on the methods of carbon erosion calculation and the calculation of increasing heat exchange through roughness of the wall [4, 8-11].

Thus, the number of unknowns, occurring in theoretical and computational methodology for calculating the ablation of carbon material, is very large. It provides high requirements for testing this technique on the experimental data obtained under different operating conditions of heat-shield materials.

Among the most important elements of ground tests of thermal protection materials, it is testing them in the jets of the combustion products of liquid-propellant rocket engines (LPRE).

It is the type of tests, which are realized levels of gas pressure on the wall, greater than 10 MPa and the turbulent gas flow in the boundary layer over almost the entire surface of the model. Therefore, the analysis of experimental data of this type can provide important information on the mechanism of erosion of CM. Example of solutions of this problem is addressed in this work.

Using the results of tests of thermal protection in LPRE combustion products streams for determination of its physical properties is possible at presence of enough strict physico-mathematical description of occurring processes only. Proposed methodology of carbon erosion mechanism determining is based on complex solution of the following tasks:

- Thermodynamic computation of LPRE combustion products and its properties estimations (estimation program [12] is used);
- Computation of LPRE combustion products stream (which is flew down to the flooded space) parameters (computational program [13]);
- Computation of perfect gas stream in shock layer over the model surface;
- Computation of convection heat transfer on impenetrable model surface (for effective length method [14] combined with mass-average values method [15] is used);
- Computation of carbon ablation characteristics (full thermo-chemical model of its mass loss [6], analogy between processes of heat and mass transfer in a boundary layer [5] and described below carbon erosion mechanism are used);
- Computation of two-dimension non-stationary heating model made of CM with mobile outer boundary (method [16] is used);
- Computation of the wall roughness influence on location of transition area on the model's surface and heat transfer intensity

strengthening are carried out under PANT methodology [8 - 10].

- Computation of model ablation form change for one step of integration in the time coordinate (implicit iteration method [17] is used).

The main feature of this test is essential change of model's shape during the experiment due to the ablation of thermal protection. Thereby all abovementioned tasks (except the first two ones) can be solved at each step of the integration in the time coordinate in complex mutual coupling formulation. So the integration step is chosen automatically on condition that the value of ablation of the thermal protection material unexceeds of the maximum allowed value and numerical stability is saved.

Having rather high pressure level in boundary layer, all known CM compositions have particular mass loss (in condensate phase), which is one of the less studied elements of CM destruction. We will use the term 'full aerothermochemical model' for the model where full thermochemical model of CM destruction is added by taking into account CM erosion.

So, this physico-mathematical model of CM aerothermochemical destruction is based on following:

- Arrhenius equations [5] to calculate carbon kinetic oxidation;
- Langmuir-Knudsen formula to calculate the velocity of non-equilibrium carbon's sublimation written the same way as in the paper [2];
- Carbon erosion law written as unique dependence of this process velocity from gas pressure on the wall ;
- Mass balance of chemical elements on the wall,
- Equations of equilibrium execution of chemical reactions on the wall;
- Chemical composition of gas in a boundary layer limited by the following set of substances

$$O, O_2, N, N_2, NO, C, C_2, C_3, CO, CO_2, CN, H, H_2, OH, H_2O, Ar, \quad (1)$$

formed by chemical elements O, N, C, H, Ar .

- Parallels between processes of heat and mass transfer in the boundary layer.

Considering the abovementioned this statement of the problem can be described by the following system of nonlinear algebraic equations (2)-(7).

The following form of Arrhenius equation is used to calculate carbon heterogeneous oxidation [5].

$$G_{Ox} = \frac{\rho_w}{A_{h,n}}\left[2Z_{O2}K_{w,O2}\exp\left(-\frac{E_{w,O2}}{R_{un}T_w}\right) + Z_O K_{w,O}\exp\left(-\frac{E_{w,O}}{R_{un}T_w}\right)\right]M_C \quad (2)$$

Herein:

G_{Ox} – mass velocity of carbon oxidation in parts of heat transfer coefficient on impenetrable wall;

ρ_w – density of gas mixture on the wall, kg/m^3;

$A_{h,n}$ – heat transfer coefficient on impenetrable the wall, kg/(m^{2}sec);

Z_i – the ratio between mass concentration of i substance in gas mixture on the wall and its molar mass, kmol/kg;

$K_{w,i}$ – pre-exponential factor in Arrhenius equation for i oxidating substance, m/sec;

$E_{w,i}$ – energy of activation in Arrhenius equation for i oxidating substance, J/kmol;

R_{un} – absolute gas constant, J/(kmol$\cdot$K);

T_w – wall temperature, K;

M_i – molar mass of i substance, kg/kmol.

Then, based on research results published in the paper [7], the same value for energy activation by atomic oxygen as for molecular oxygen is used in present research, $E_{w,O} = E_{w,O2} = 1,72\cdot 10^8$ J/kmol.

Pre-exponential factors from Arrhenius equation written in form (2) are considered below as one of the main parameter of coordination of the calculated and experimental data for thermal protection ablation. Moreover, considering the fact that simultaneous participation of atomic and molecular oxygen in process of heterogeneous carbon oxidation is

scarcely (confirmed by test results) there are no differences between these two factors below so it is taken as the following:

$K_{w,O} = K_{w,O2} = K_w$.

Considering the admissions made, equation (2) is modified into

$$G_{Ox} = \frac{\rho_w}{A_{h,n}}\left(2Z_{O2} + Z_O\right)K_w \exp\left(-\frac{1{,}72 \cdot 10^8}{R_{un}T_w}\right)M_C. \quad (3)$$

While researching the carbon sublimation we will use non-equilibrium sublimation model [2], based on Langmuir-Knudsen formula, using as indicated below:

$$G_{Sub} = \sum_{k=1}^{3} \frac{\zeta_k}{A_{h,n}} \cdot \frac{p_{Ck,\oplus}\left(T_w\right) - p_{Ck,w}}{\sqrt{2\pi R_{un} T_w / M_{Ck}}} \quad (4)$$

Herein:

G_{Sub} - mass velocity of sublimation, measured in heat transfer coefficient parts on impenetrable wall;

ζ_k - wall accommodation coefficient in relation to molecules of C_k type (for their values are used data from paper [2]: $\zeta_C = 0.3$, $\zeta_{C2} = 0.5$, $\zeta_{C3} = 0.1$);

$p_{Ck,w}$ - partial pressure k substance on the wall, Pa;

$p_{Ck,\oplus}$ - saturated stream pressure k substance.

Almost all utilized CMs are composite materials. Modern CM is a bar structure full of pyrocarbon appeared as a result of multiple pyrolysis of organic filler called ‘carbon loam’. Carbon composite has the same structure after component organic resin decompounding. It takes place under the low temperature influence in the inner layers of material.

Presence of some condensed components in the material inevitably leads to the difference in velocities of its thermo-chemical destruction resulted from heterogeneous carbon oxidation and its sublimation. The result of this circumstance is change of the wall roughness.

Hypothesis of connection between local pressure in a boundary layer, wall roughness and carbon erosion is rather logical. Indeed from the position of heat stability by pressure rising in a boundary layer the altitude of roughness (which can withstand force impact from the side of flowed around gas stream and then increase the velocity of the carbon erosion) should be decreased.

So, we can suppose that both carbon erosion velocity and wall roughness are pressure functions in a boundary layer. Herewith carbon erosion velocity should increase and the altitude of wall roughness should decrease due to growth of the noted pressure.

It makes sense that the carbon erosion appears only at rather high pressure in a boundary layer for the right composition of heat protective material.

Under this methodology, we can accept the following:

- Intensity of carbon erosion’s process depends only on the pressure on the wall;
- p_{Lim} is the limit value of this pressure is set up (carbon erosion is neglected up to this value);.

Considering the abovementioned, the following functional dependence of carbon mass loss erosion intensity from pressure on the wall is proposed:

$$G_{Er} = \frac{K_{Er}}{A_{h,n}}\left\{\exp\left[\max^{n_{Er}}\left(1, \frac{p_w}{P_{Lim}}\right) - 1\right] - 1\right\} \quad (5)$$

Herein:

G_{Er} – carbon erosion mass loss velocity, measured in heat transfer coefficient parts on impenetrable wall;

K_{Er} – pre-exponential factor, kg/(m^{2}sec);

n_{Er} – index.

This task is closed by equations of chemical elements mass balance, by equations of equilibrium execution of chemical reactions on the wall and Dalton equations at given value of the wall temperature T_w.

In addition, mass balances of oxygen, hydrogen, nitrogen and carbon are written as the following:

$$Z_O + 2Z_{O2} + Z_{OH} + Z_{H2O} + Z_{NO} + Z_{CO} + 2Z_{CO2} = \frac{A_{h,n}\Theta_{O,e}}{\left(A_{h,n} + G_{Gas}\right)M_O};$$

$$Z_H + 2Z_{H2} + Z_{OH} + 2Z_{H2O} = \frac{A_{h,n}\Theta_{H,e}}{\left(A_{h,n} + G_{Gas}\right)M_N};$$

$$Z_N + 2Z_{N2} + Z_{NO} + Z_{CN} = \frac{A_{h,n}\Theta_{N,e}}{\left(A_{h,n} + G_{Gas}\right)M_N}; \qquad (6)$$

$$Z_C + 2Z_{C2} + 3Z_{C3} + Z_{CO} + Z_{CO2} + Z_{CN} = \frac{A_{h,n}\Theta_{C,e} + G_{Gas}}{\left(A_{h,n} + G_{Gas}\right)M_C}.$$

Herein:

$\Theta_{j,e}$ – concentration of j chemical element on the outer boundary of the boundary layer;

$G_{Gas} = G_{Ox} + G_{Sub}$ – mass velocity of material gasification in heat transfer coefficient parts on impenetrable wall.

Conditions of equilibrium execution of chemical reaction on the wall are written as the following:

$$\begin{aligned}
&p_{O,w}^2 = k_{eq,O2}(T_w)\,p_{O2,w};\\
&p_{H,w}^2 = k_{eq,H2}(T_w)\,p_{H2,w};\\
&p_{O,w}p_{H,w} = k_{eq,OH}(T_w)\,p_{OH,w};\\
&p_{O,w}p_{H,w}^2 = k_{eq,H2O}(T_w)\,p_{H2O,w};\\
&p_{N,w}^2 = k_{eq,N2}(T_w)\,p_{N2,w};\\
&p_{N,w}p_{O,w} = k_{eq,NO}(T_w)\,p_{NO,w}; \qquad (7)\\
&p_{C,w}^2 = k_{eq,C2}(T_w)\,p_{C2,w};\\
&p_{C,w}^3 = k_{eq,C3}(T_w)\,p_{C3,w};\\
&p_{C,w}p_{O,w} = k_{eq,CO}(T_w)\,p_{CO,w};\\
&p_{C,w}p_{O,w}^2 = k_{eq,CO2}(T_w)\,p_{CO2,w};\\
&p_{C,w}p_{N,w} = k_{eq,CN}(T_w)\,p_{CN,w}.
\end{aligned}$$

Herein $k_{eq,i}$ – constant of chemical reaction equilibrium of formation i molecular substance from atoms.

According to Dalton's equation

$$\sum_i C_{i,w} = 1; \qquad \sum_i p_{i,w} = p_e$$

It follows that

$$\sum_i Z_i M_i = 1; \qquad M_w = \left(\sum_i Z_i\right)^{-1}.$$

Herein:

p_e – pressure on the outer border of the boundary layer;

M_w – molar mass of gas mixture on the wall.

All 16 substances from the list (1) participate at sum operation of i index hereinafter and density of gas mixture on the wall from equation (2) is estimated by Mendeleev-Clapeyron equation.

Herewith, q_w specific heat flux streaming to warming of thermal protection and using as boundary condition on the wall for Fourier equation can be calculated by the following formulae:

$$q_w = A_{h,n}\left[A_h\left(h_r - h_w\right) + G_{Gas}h_{C,w}^{(s)} - \left(A_h + G_{Gas}\right)h_w\right] - \varepsilon_w\sigma T_w^4 \qquad (8)$$

$$h_w = \sum_i C_{i,w}h_{i,w}.$$

Herein:

h_r - enthalpy recovery in the incoming gas stream, J/kg;

h_w - gas mixture enthalpy on the wall;

$h_{C,w}^{(s)}$ - enthalpy of condensed carbon on the wall;

$h_{i,w}$ - enthalpy i gas mixture on the wall;

ε_w - wall emissivity factor;

σ - Stefan-Boltzmann factor, W/(m^2 K^4).

2 Methodic bases of thermal shield ablation experiment conducting in LPRE combustion products jet

Industrial propulsion systems are usually used in gas-dynamic stands designed to conduct thermal shield ablation experiment in LPRE combustion products jet. The following processes take place there to increase the stream stagnation pressure:

- Supersonic part size of nozzle section is being decreased (final propulsion system nozzle is usually almost the same as sonic nozzle);
- Tested model made by heat protection material under study should be mounted on minimum distance from the nozzle edge where stable mode of engine combustion products exhaust in flooded space is preserved.

Fig. 2 shows the principal scheme of conducting of the ablation experiment in LPRE combustion products jet (results are given below).

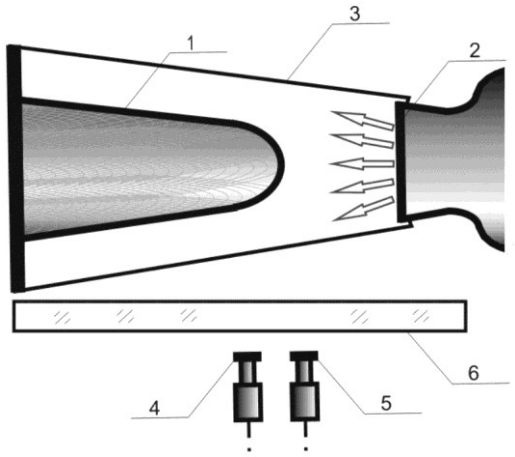

Fig. 2. Principal scheme of conducting of the ablation experiment in LPRE combustion products jet: 1 – model made by heat protection material under study; 2 – nozzle edge of the engine; 3 – water-cooling holder; 4 – high speed video camera; 5 – thermal vision camera; 6 – quartz optical window.

Hydrogen gas is used on this stand as a fuel and liquid oxygen – as an oxidizer. These fuel components are delivered through the blowpipe to the combustion chamber where chemical interaction with water vapor formation happens. Fuel used to freeze nozzle walls only is delivered through peripheral blowpipes and k_{Jet} ratio of fuel component (oxidizer to fuel) in jet core should be no more than its stoichiometric value (which is equal to 8) to prevent the nozzle walls from the burnout.

All initial data on motion of combustion materials in engine nozzle (Figs. 3, 4) are appeared during experiment in combustion materials products jet of oxygen-hydrogen liquid engine. Results are given below:

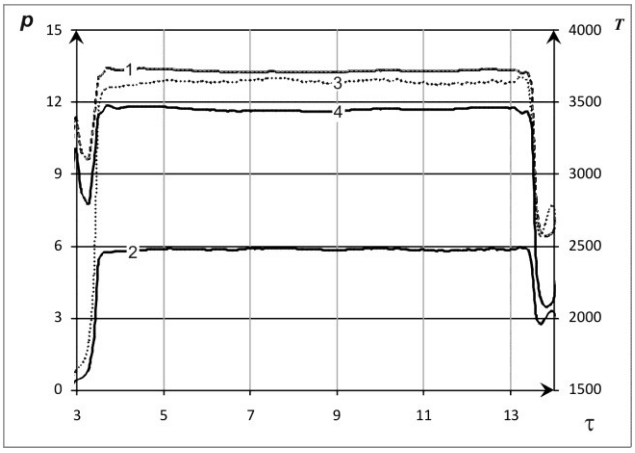

Fig. 3. Dependence of gas parameters in nozzle channel from time coordinate during experiment: 1 - p_k ; 2 - p_c ; 3 - T_k ;4 - T_c ; p , MPa; T , K; τ , sec

Herein:

p_k - gas pressure in combustion chamber, MPa;

p_c - gas pressure on the nozzle edge, MPa;

T_k - gas temperature in combustion chamber, K;

T_c - gas temperature on the nozzle edge, K;

τ - time, sec.

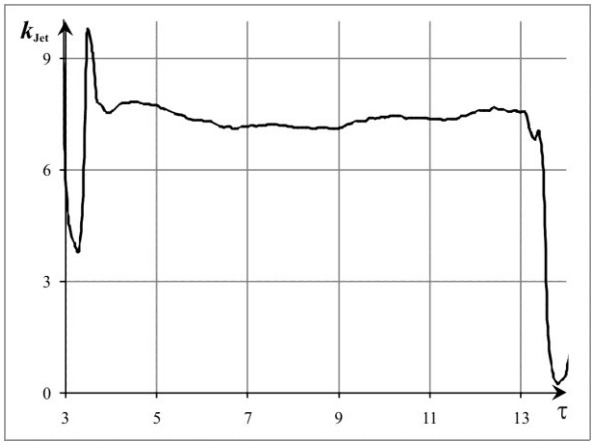

Fig. 4. Dependence of fuel component ratio in jet core from time coordinate during experiment; τ , sec

Essential change of all gas flow parameters on nozzle channel is observed at initial stage of experiment. This is definitely is the obstacle to conduct the correct analyzing of experimental data. Meanwhile gas flow mode in nozzle channel on the main time line slightly differs from stationary. It allows to change this type of test into a tool for quality determination of material physical properties by using widespread way (in aero-dynamical experiment) of preserving the model from influence of nonstationary gas jet. It is currently based on using secure caps made of easily ablative material (for example fluorine).

Unfortunately, these measures weren't taken during experiments analyzed below. It definitely effects on quality of conducted estimated theoretical analysis of experimental data.

Strictly nonstationary nature of change in time of all gas-dynamic characteristics in combustion chamber of engine at initial phase of ablation experiment conducting causes serious problems at comparison of computational and experimental data on development of model ablation shape in

progress. That is why we made noted comparison in fixed point of time only which corresponded the duration of model flowing by gas jet at stationary mode of gas generator work during 8 seconds.

Analysis of ablation experiments conducted for CM in high-pressure oxidizer jets of LPRE combustion products is matched with necessity of determining the following: kinetic constants of condensed carbon oxidizing by gas jet oxidizing components; the main rules of carbon erosion process execution; effective rigidities of the wall to location of transition area on the model's surface and to strengthening of convective heat transfer.

It is common knowledge (paper [11] for example) – the roughness of wall has a high influence on convective heat transfer intensity on the surface of the blunt body at high Reynolds numbers. Some effective sizes of roughness ($b_{Rou,\,Tr}$, $b_{Rou,\,L}$ and $b_{Rou,\,T}$) are introduced to calculate this influence. They are used to determine the following:

- 'Transition area' location;
- Heat transfer gain rate at laminar flow of gas jet at the boundary layer;
- Heat transfer gain rate at turbulent flow of gas jet at the boundary layer.

3 Results of investigation

The object of theoretical and calculated analysis (results are presented below) is a model made of modern CM in the shape of spherically blunted cone.

At the first stage, there was the research to determine the influence on changing the shape of the model by the main elements of thermal protection mass loss mechanism.

The elements of CM mass loss mechanism, which were analyzed, are the following:

- Kinetic constants of carbon heterogeneous oxidizing by the components of gas jet incoming on the model;
- Carbon erosion velocity-force impact on material from the side of gas jet incoming on the model dependence;
- The size of the wall roughness to the location of the transition area on the model surface and to the strengthening of convective heat transfer intensity.

As the result of the theoretical and calculated analysis of experimental data was found the following:

- Satisfactory fit of calculated and experimental data on ablation shape of tested model at fixed values of the wall roughness can be obtained by single combination of carbon erosion law and kinetic of its heterogeneous corrosion only;
- Roughness of wall to the location of transition area almost doesn't influence on the model ablation's shape;
- Value of the wall roughness influences only on kinetics of the carbon heterogeneous oxidizing and does not influence on the law of carbon erosion.

Therefore, we unambiguously specified the law of the carbon erosion under which satisfactory matching of estimated and experimental data on burn form of model made of CM is provided.

Fig. 5 shows the quality of this calculation.

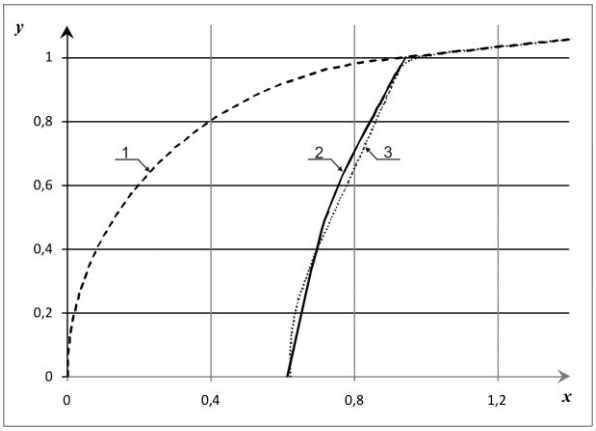

Fig. 5. Comparison of calculated and experimental data on model's ablation shape: 1 - initial form; 2 - experimental data; 3 - calculated data.

Calculated data (Fig. 5) corresponds to the following initial data: $p_{Lim} = 5$ MPa; $K_{Er} = 8$ kg/(m^{2}sec); $n_{Er} = 0.4$.

Herewith:

- Calculation of model warming and ablation is being conducted in nonstationary statement;
- Oncoming flow of gas jet were accepted as plane parallel and homogeneous.

Conclusion

As the results of performed comparison of calculated theoretical data and the result of ablation experiment of thermal protection in the jet of LPRE combustion products for CM are the following:

1. New approach of processing of ablation experiments in the stream of LPRE combustion products is enunciated. The using of this approach allows to turn this experiments for CM from comparative tests to tests for determination ablation properties of thermal protection. Moreover, using the results of these stand tests at full-scale conditions of thermal protection functioning.

2. The law of carbon erosion was established by the first time.

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