

## HEAT AND MASS TRANSFER PROCESSES IN CHEMICALLY REACTING MEDIA

### TOWARD GENERALIZATION OF CALCULATIONS OF THE STATIONARY VELOCITY OF DIFFUSION FLAME PROPAGATION OVER THE SURFACE OF A FUEL

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*The results of numerical investigations of the laws governing the spreading of a two-dimensional diffusion flame over thermally thin layers of a cellulose material using the algorithm of calculation of stationary velocity of flame propagation, which is based on the principle of minimum entropy production, are presented. The obtained dependences of the velocity of flame propagation on thermophysical parameters (thickness of the fuel, oxygen concentration and pressure of the ambient, velocity of the blowing flow) agree quantitatively with the well-known experimental data.*

In [1] an algorithm for calculating the stationary velocity of flame propagation was suggested which was based on the principle of minimum entropy production. Further calculations of the velocity of propagation of a one-dimensional laminar flame through a mixed gas mixture [2, 3] have shown that the use of this algorithm allows one to adequately describe the well-known physical laws (there is a certain quantitative deviation from a physically exact solution based on the integral balance of mass in the course of chemical conversion in a combustion wave, which is reasonably expected considering the substantially nonlinear "process characteristics–temperature" dependence which brings this solution beyond the framework of the classical linear nonequilibrium thermodynamics). The thermodynamic analysis of this classical problem on the propagation of flame was carried out in a number of works in which the parameters of entropy production in a flame [4] are investigated, and the principle of minimum production of entropy is used to obtain an analytical solution [5] and, which is closest to the subject of the present work, the extreme properties of entropy production are considered as a functional with a view to formulate the variational principle for determining the velocity of flame propagation [6].

It should be noted that solution of the problem of *one-dimensional* propagation of a flame can be obtained within the framework of the mathematical statement which is based on phenomenological conservation laws of continuum mechanics (the first law of thermodynamics) and which does not call for the use of additional relations following from the second law of thermodynamics, one of the possible formulations of which is the principle of minimum entropy production. Various algorithms for calculating a stationary velocity of flame propagation over a mixture of stirred gases are considered and verified (e.g., [7]), the results of the application of which correspond to experimental data with an accuracy comparable with the accuracy of measurements, and their further development is directed mainly at refinement of the kinetic schemes of combustion reaction. On the other hand, an analysis of the problem of *two-dimensional* wave propagation over the surface of a fuel [8, 9] has shown that on its formulation in steady-state equations written in a coordinate system moving with the flame front one fails to obtain a unique solution with respect to the sought-for value of the stationary velocity of flame propagation. Here, unlike the one-dimensional flame propagation, transition to a stationary formulation of the problem by the coordinate transformation of the form  $x' = x + v_{fl}t$

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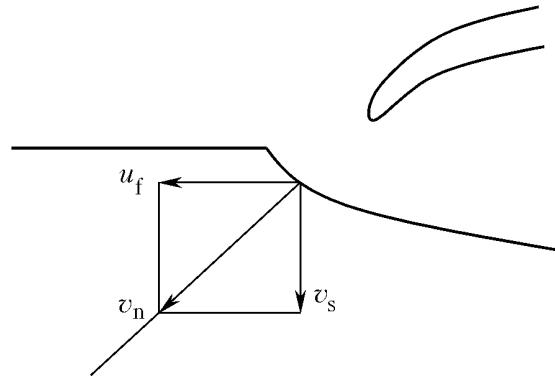


Fig. 1. Configuration of flame propagation.

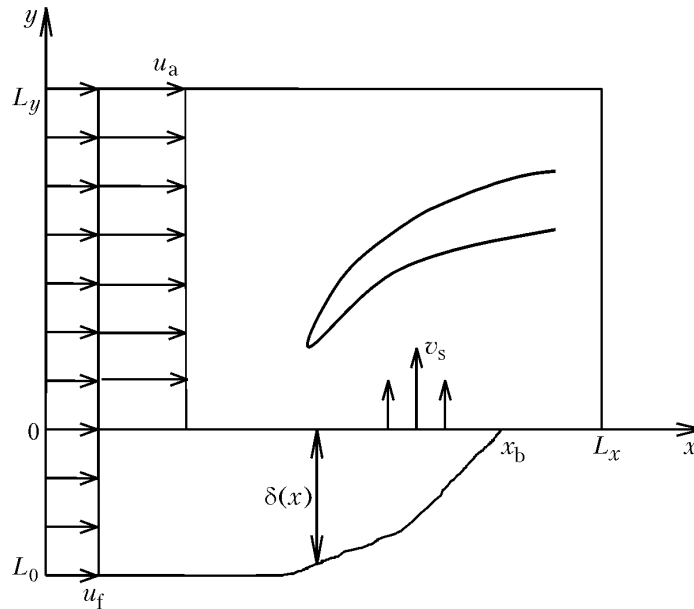


Fig. 2. Physical model of the problem.

does not lead to a single-valued formulation, since in expansion of the true vector of the flame propagation velocity  $v_n$  in a Cartesian coordinate system (Fig. 1) there are two significant components: the linear rate of thermal decomposition of material  $v_s$  and the velocity of flame propagation along its surface  $u_f$ . Their determination requires the application of a relation that represents the condition of mass conservation in the course of the chemical reaction of thermal decomposition of material. The use of a single equation for determining two values leads to a situation where, in the general case, one fails to obtain a closed mathematical statement of the problem of calculating a stationary flame propagation velocity. In [8] it is shown that the earlier approaches were based on a priori relations that redefine a conjugate mathematical model of the problem. The following additional conditions were suggested that make it possible to close the problem and calculate the flame propagation velocity: in classical work [10] a fixed value of temperature is prescribed on the surface of a fuel, in [11] a fixed value of the coordinate of the burning out of a fuel  $x_b$  (Fig. 2), and in [12] a certain fixed value of temperature is prescribed at a certain point on the fuel surface. As is shown in the analysis given below, the above-mentioned conditions are a priori ones and they do not correspond to the statement of a conjugate problem, since the values of the parameters considered are determined as a result of solving the conservation equations.

The aim of the present work is the parametric estimation of the algorithm for calculating a stationary velocity of flame propagation which would be based on the principle of minimum entropy production using the results of investigation into the laws governing the propagation of a diffusion flame over the surface of a fuel. The basic criterion

for the comparison of the obtained results with experimental data is the value of the stationary velocity of flame propagation; it is a microscopic value which reflects the resulting effect of the entire process of combustion as an ensemble of various physicochemical phenomena.

**Formulation of the Problem.** In the formulation of a model of steady propagation of a two-dimensional diffusion flame over the surface of a fuel the following basic assumptions were made: in the fuel there is a single macroscopic reaction of thermal decomposition of material; in the gas phase between the gaseous products of pyrolysis and an oxidizing reagent of the surrounding medium there occurs one gross reaction of combustion; the thermophysical characteristics of all the components of the gas mixture are assumed to be identical; the gas motion is laminar; radiative heat transfer is not considered.

In a coordinate system moving with the flame front (Fig. 2) the steady-state equations of transfer in a gas phase, which describe laminar motion of a viscous chemically reacting gas with account for natural convection, have the form [9]

$$\rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial y} = \frac{\partial}{\partial x} \frac{\text{Pr} \lambda}{C} \frac{\partial u}{\partial x} + \frac{\partial}{\partial y} \frac{\text{Pr} \lambda}{C} \frac{\partial u}{\partial y} - \frac{\partial p}{\partial x} + (\rho_a - \rho) g \sin \theta, \quad (1)$$

$$\rho u \frac{\partial v}{\partial x} + \rho v \frac{\partial v}{\partial y} = \frac{\partial}{\partial x} \frac{\text{Pr} \lambda}{C} \frac{\partial v}{\partial x} + \frac{\partial}{\partial y} \frac{\text{Pr} \lambda}{C} \frac{\partial v}{\partial y} - \frac{\partial p}{\partial y} + (\rho_a - \rho) g \cos \theta, \quad (2)$$

$$\frac{\partial \rho u}{\partial x} + \frac{\partial \rho v}{\partial y} = 0, \quad (3)$$

$$C \rho u \frac{\partial T}{\partial x} + C \rho v \frac{\partial T}{\partial y} = \frac{\partial}{\partial x} \lambda \frac{\partial T}{\partial x} + \frac{\partial}{\partial y} \lambda \frac{\partial T}{\partial y} + Q \rho W, \quad (4)$$

$$\rho u \frac{\partial Y_o}{\partial x} + \rho v \frac{\partial Y_o}{\partial y} = \frac{\partial}{\partial x} \frac{\text{Le} \lambda}{C} \frac{\partial Y_o}{\partial x} + \frac{\partial}{\partial y} \frac{\text{Le} \lambda}{C} \frac{\partial Y_o}{\partial y} - v_o \rho W, \quad (5)$$

$$\rho u \frac{\partial Y_{fu}}{\partial x} + \rho v \frac{\partial Y_{fu}}{\partial y} = \frac{\partial}{\partial x} \frac{\text{Le} \lambda}{C} \frac{\partial Y_{fu}}{\partial x} + \frac{\partial}{\partial y} \frac{\text{Le} \lambda}{C} \frac{\partial Y_{fu}}{\partial y} - v_{fu} \rho W, \quad (6)$$

$$\rho = p/RT. \quad (7)$$

In the fuel, the equations of the conservation of energy and concentration of the initial reagent of the reaction of pyrolysis are considered:

$$C_s \rho_s u_f \frac{\partial T_s}{\partial x} = \frac{\partial}{\partial x} \lambda_s \frac{\partial T_s}{\partial x} + \frac{\partial}{\partial y} \lambda_s \frac{\partial T_s}{\partial y} + Q_s \rho_s W_s, \quad (8)$$

$$u_f \frac{dY_s}{dx} = -Y_s W_s. \quad (9)$$

The rates of the reactions of combustion in the gas phase and of the reaction of thermal decomposition of the fuel are defined by the Arrhenius law:

$$W = k Y_o Y_{fu} \exp(-E/R_0 T), \quad (10)$$

$$W_s = k_s \exp(-E_s/R_0 T_s). \quad (11)$$

According to the scheme of the computational domain presented in Fig. 2, the boundary conditions have the form

$$x = 0: u = u_f + u_a, \quad v = 0, \quad T = T_a, \quad Y_{fu} = 0, \quad Y_o = Y_{o,a}, \quad (12)$$

$$y = 0 \quad (0 < x < x_b): u = u_f, \quad \rho v = \rho_s v_s, \quad (13)$$

$$T = T_s, \quad (14)$$

$$-\lambda \frac{\partial T}{\partial y} + \rho v C T = -\lambda_s \frac{\partial T_s}{\partial y} + \rho_s v_s C_s T_s, \quad (15)$$

$$-\frac{Le \lambda}{C} \frac{\partial Y_o}{\partial y} + \rho v Y_o = 0, \quad (16)$$

$$-\frac{Le \lambda}{C} \frac{\partial Y_{fu}}{\partial y} + \rho v Y_{fu} = \rho_s v_s, \quad (17)$$

$$y = 0 \quad (x > x_b): \rho v = 0, \quad \frac{\partial}{\partial y} = 0 \quad \text{for } u, T, Y_o, Y_{fu}, \quad (18)$$

$$y = -\delta(x): \frac{\partial T_s}{\partial n} = 0, \quad (19)$$

$$x = L_x: \frac{\partial}{\partial x} = 0 \quad \text{for } u, v, T, Y_o, Y_{fu}, \quad (20)$$

$$y = L_y: u = u_f + u_a, \quad \frac{\partial v}{\partial y} = 0, \quad T = T_a, \quad Y_{fu} = 0, \quad Y_o = Y_{o,a}. \quad (21)$$

**Algorithm of Calculation of the Flame Propagation Velocity.** The problem considered is represented by Eqs. (1)–(9) for nine sought-after variables:  $u$ ,  $v$ ,  $p$ ,  $T$ ,  $Y_o$ ,  $Y_{fu}$ ,  $\rho$ ,  $T_s$ , and  $Y_s$ . In addition to them, the conservation equations and boundary conditions contain additional variables: the linear rate of pyrolysis of a fuel,  $v_s$ , the shape of the boundary of the fuel burning out  $\delta(x)$ , and, as a parameter of coordinate system transformation, the velocity of stationary propagation of flame  $u_f$ . These quantities are characteristic parameters of the problem and, by definition, must be expressed in terms of the remaining parameters obtained as a result of solving the boundary-value problem, Eqs. (1)–(9), with corresponding boundary conditions. In order to obtain the needed relations we will consider the process of thermal decomposition of a fuel in the following model [9]: in the fuel there occurs a volume reaction of pyrolysis which leads to gasification of the solid at the burning-out boundary  $\delta(x)$ . Correspondingly,  $Y_s = 1$  in the solid ( $-\delta(x) < y < 0$ ) and  $Y_s = 0$  after the gasification of the fuel ( $-L_0 < y < -\delta(x)$ ). Integration of Eq. (9) over the coordinate  $y$  yields the equation

$$u_f \frac{d\delta(x)}{dx} = - \int_{-\delta(x)}^0 W_s dy, \quad (22)$$

the right-hand side of which represents the well-known equation for the linear rate of combustion of a solid fuel [13]:

$$v_s(x) = \int_{-\delta(x)}^0 W_s dy. \quad (23)$$

Integration of Eq. (22) over the coordinate  $x$  yields an expression for the shape of the fuel surface  $\delta(x)$ :

$$\delta(x) = L_0 - \frac{1}{u_f} \int_0^x v_s dx. \quad (24)$$

Thus, integration of Eq. (9), which accounts for the mass balance in the solid in the course of chemical reaction of thermal decomposition, leads to the needed relations (23) and (24) for determining the linear rate of pyrolysis  $v_s$  and the boundary of the burning-out of the fuel  $\delta(x)$ , respectively. In actual fact, the transformation of differential equation (9) into two integral ones allows one to replace the two-dimensional variable  $Y_s(x, y)$  by two one-dimensional —  $v_s(x)$  and  $\delta(x)$ . It is evident that the velocity of flame propagation  $u_f$  remains the unknown parameter, and for this velocity to be determined, it is necessary to enlist some additional relations the derivation of which in the present work is based on application of the principle of minimum entropy production. In accordance with the adopted model of heat transfer in a fuel two irreversible processes participate in entropy production: heat conduction and chemical reaction. We will represent the total entropy production in the given thermodynamic system as [14]

$$P = \int_0^{x_b} \int_{-\delta}^0 \left( J_x X_x + J_y X_y + J_W X_W \right) dy dx. \quad (25)$$

The thermodynamic fluxes and generalizing forces for the heat conduction process [14] are

$$J_x = -\lambda_s \frac{\partial T_s}{\partial x}, \quad J_y = -\lambda_s \frac{\partial T_s}{\partial y}, \quad (26)$$

$$X_x = -\frac{1}{T_s^2} \frac{\partial T_s}{\partial x}, \quad X_y = -\frac{1}{T_s^2} \frac{\partial T_s}{\partial y}. \quad (27)$$

The thermodynamic flux corresponding to the chemical reaction is proportional to its rate:

$$J_W = \rho_s W_s. \quad (28)$$

For the generalized force corresponding to the chemical reaction the following relation [9] was obtained:

$$X_W = Q_s \left( \frac{1}{T_s} - \frac{1}{T_s^*} \right) - R \ln \left( 1 - \frac{\delta}{L_0} \right), \quad (29)$$

where  $T_s^*$  is the maximum temperature in the fuel adopted here as a certain characteristic temperature closest to an equilibrium one with respect to the considered reaction of pyrolysis.

The algorithm of determining the velocity of flame propagation is as follows. The solution of the problem, Eqs. (1)–(8), is found at different values of  $u_f$ . Using the temperature distributions found, we calculate the values of integral (25) in which the thermodynamic fluxes and generalized forces are determined from Eqs. (26)–(29). Among the values of entropy production  $P(u_f)$  obtained, the minimum value is determined, which is assumed to correspond to the steady state of the considered thermodynamic system, and the value  $u_f$  corresponding to it is assumed to correspond to the sought-for value of the stationary velocity of flame propagation.

TABLE 1. Initial Data for the Base Variant [17]

Parameters	Gas phase	Material	Parameters	Gas phase
$C$	1005.6	1257	$\nu_F$	1.0
$\lambda$	0.0254	0.1257	$\nu_0$	1.185
$\rho$	1.0	650	Le	1
$Q$	$1.68 \cdot 10^7$	$-7.54 \cdot 10^5$	Pr	0.7
$k$	$10^9$	$10^{10}$	$p$	$10^5$
$E$	62 850	125 700	$Y_{o,a}$	0.21
$L_o$	—	0.00013	$\theta$	$\pi/2$
$T_a$	300	300	$u_a$	0

**Results of Calculations.** The procedure of numerical solution of the conjugate problem of heat and mass transfer in a reacting heterogeneous system "gas–solid body" is based on combined application of the control volume method [15] for a gas phase and finite element method [16] for a solid fuel. This approach [9] allows one to use the advantages of both methods consisting, on the one hand, in the efficient algorithm of flow field calculation and, on the other, in the higher accuracy of calculation of the integral characteristics in a fuel and approximation of the variable boundary of burning-out of the fuel which is formed in accordance with the obtained distribution of the rate of thermal decomposition of material, according to Eq. (24). For a more accurate approximation of the nonlinear source term which describes heat release in the course of the combustion reaction, as well as of the ambient boundary conditions, the computational domain in the gas phase was composed of two subdomains: the inner region which includes the flame zone, with a constant step of a difference grid (for the base variant  $60 \times 20$  nodes along the coordinates  $x$  and  $y$ , respectively) and the outer region with the grid step increasing toward the boundaries of the computational domain (additionally 15 and 10 nodes along the coordinates  $x$  and  $y$ , respectively). On increase in the concentration of oxygen in the ambient entailing a growth of the physical dimensions of the flame, the number of computational nodes in the inner subdomain increased up to  $120 \times 40$ , with preservation of the characteristic dimension of the grid step in the flame zone.

The calculations were made for a flame propagating over both surfaces of a thermally thin (entirely burning) layer of cellulose material (paper) down a vertical surface opposite to the blowing flow caused by natural and forced convection. For the given conditions an appreciable number of consistent experimental data on the flame propagation velocity were found that were obtained by various authors. As the basic conditions we accepted "normal" outer conditions (atmospheric pressure, oxygen concentration in air, absence of forced convection). The values of the thermophysical and kinetic parameters used are presented in Table 1.

The dependences of the flame propagation velocity on different initial conditions are presented in Fig. 3. On the whole, one can note a satisfactory quantitative agreement between the calculated and experimental data. The results of investigation of the dependence of the flame propagation velocity on the material layer thickness (Fig. 3a) were used to calibrate the kinetic parameters of the combustion reaction rate in a gas phase by the value of the pre-exponential factor, since a great quantity of experimental values are available for this case. The good agreement between the calculated and experimental results on the influence of oxygen concentration in the ambient (Fig. 3b) is due to the fact that here the initial data [17] were used which, in turn, had been verified by the results of our own experiment [18]. The results of calculations (Fig. 3c) demonstrate a qualitatively correct description of the dependence of the flame propagation velocity on ambient pressure; however, the quantitative correspondence with measurements was obtained only for atmospheric conditions. This is due to the fact that the pressure exerts a substantial effect on the kinetics of the reactions taking place in the flame, which needs a separate analysis.

Figure 4 presents the dependences of the distributions of entropy production on the flame propagation velocity at different velocities of the blowing flow. As the latter velocity increases, the point of minimum on the corresponding curve becomes less distinct and at a certain value of it ( $u_a > 0.7$  m/sec) the local minimum disappears, and the distribution of entropy production acquires a monotonic character. The general form of the dependence of the flame propagation velocity on the blowing flow velocity (Fig. 5) shows that the latter has a critical value at which there exists the

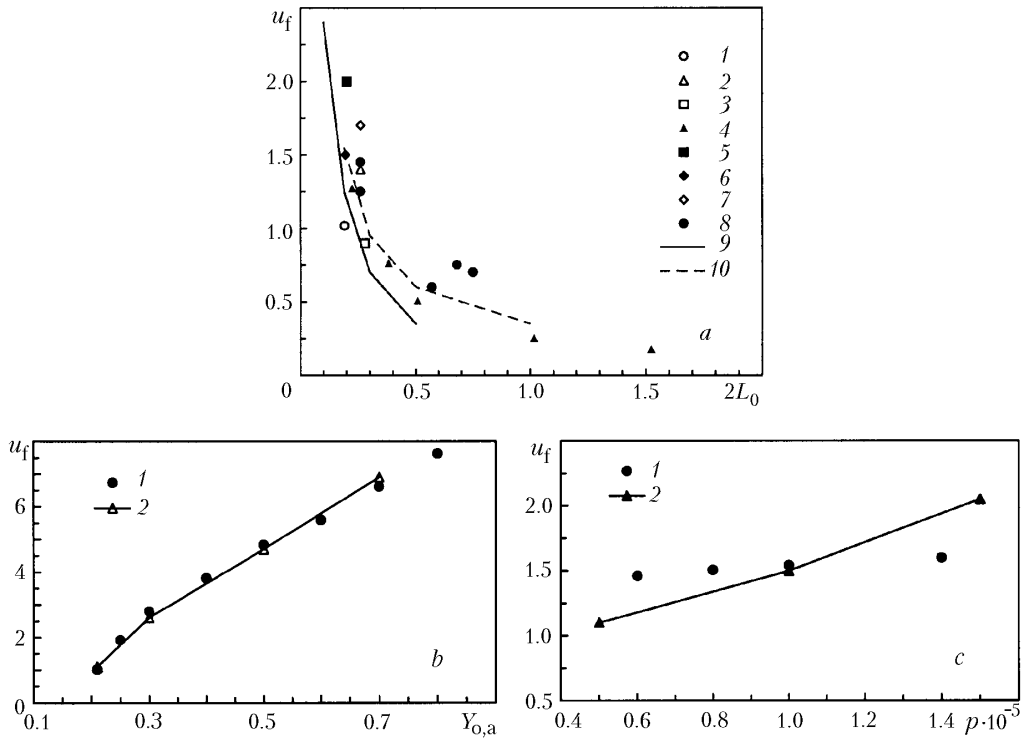


Fig. 3. Flame propagation velocity vs. the process parameters: a) fuel layer thickness, experiment: 1) [18]; 2) [19]; 3) [20]; 4) [21]; 5) [22]; 6) [23]; 7) [24]; 8) [25]; calculation: 9)  $k = 8 \cdot 10^8$  1/sec, 10)  $k = 1 \cdot 10^9$  1/sec; b) oxygen concentration in the ambient: 1) experiment [18]; 2) calculation; c) ambient pressure: 1) experiment [23]; 2) calculation.  $u_f$ , mm/sec;  $L_0$ , mm;  $p$ , Pa.

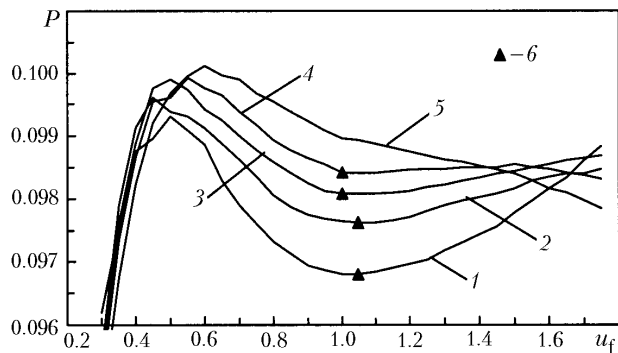


Fig. 4. Distribution of entropy production vs. flame propagation velocity at different blowing flow velocities: 1)  $u_a = 0$ ; 2) 0.15; 3) 0.3; 4) 0.7; 5) 1.0 m/sec; 6) points of a minimum.  $u_f$ , mm/sec;  $P$ , W/(m·K).

thermodynamic system considered, which corresponds to the minimum entropy production. At higher values of  $u_a$  the minimum value of entropy production corresponds to the point of a global minimum  $u_f = 0$ , i.e., to the absence of flame propagation. This feature reflects the fundamental difference between a *stationary* propagation of flame and its propagation at a *constant velocity*. Experimental investigations [19] have demonstrated the existence of two regimes of flame propagation opposite to the blowing flow. Until the critical blowing velocity is attained there is a stable regime; thereafter the flame propagation becomes *locally unstable* — the propagation velocity is determined as a time-averaged value. The unstable regime is a substantially nonstationary process, and its characteristics cannot be calculated on the basis of the proposed algorithm, since at its basis is the determination of the steady-state thermodynamic state of the

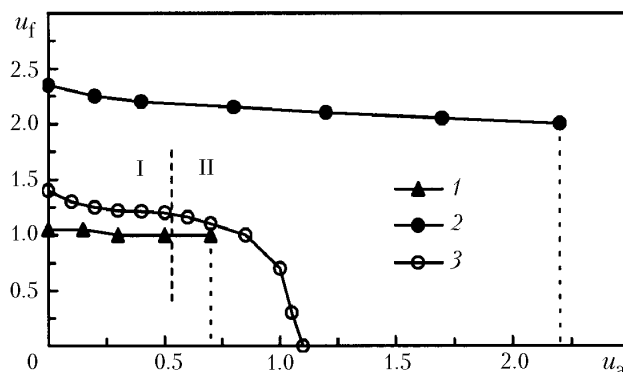


Fig. 5. Flame propagation velocity vs. the blowing flow velocity: 1, 3)  $Y_{o,a} = 0.21$ ; 2) 0.3 (1, 2) calculation; 3) experiment [19]); I, stable regime; II, unstable regime.  $u_f$ , mm/sec;  $u_a$ , m/sec.

system. In considering a stable regime of flame propagation the results of calculation of its velocity (Fig. 5) correspond well to experimental data.

Thus, the results obtained in the present work confirm the fundamental suitability of the principle of minimum entropy production for describing the base laws governing flame propagation (at least, for the above-investigated conditions). It should be emphasized that the problem considered can be solved in the statement which employs the unsteady-state equation of transfer [26]. In such a case, no uncertainties arise which are typical of the stationary formulation of the problem considered here; however, it is also evident that solution of a system of nonlinear unsteady-state equations involves a good deal of expenditures of computational resources. In conclusion we note that the necessity of applying relations based on the concepts of the thermodynamics of irreversible processes (which, despite their dynamic development [27], are still far from being generally recognized) is due to the reduction of the problem's dimensionality in the transition to steady-state equations.

## NOTATION

$C$ , heat capacity, J/(kg·K);  $E$ , activation energy, J/mole;  $g$ , free fall acceleration, m/sec<sup>2</sup>;  $k$ , pre-exponential factor, 1/sec;  $L_0$ , initial thickness of a layer of a fuel, m;  $L_x$ , length of the computational domain along the coordinate  $x$ , m;  $L_y$ , length of the computational domain along the coordinate  $y$ , m;  $Le$ , Lewis number;  $n$ , coordinate normal to the material surface;  $P$ , entropy production, W/(m·K);  $Pr$ , Prandtl number;  $p$ , pressure, Pa;  $Q$ , thermal effect of reaction, J/kg;  $R$ , specific gas constant, J/(kg·K);  $R_0$ , universal gas constant, J/(mole·K);  $T$ , temperature, K;  $T_s^*$ , characteristic temperature of a fuel, K;  $t$ , time, sec;  $u$ , velocity along the coordinate  $x$ , m/sec;  $v$ , velocity along the coordinate  $y$ , m/sec;  $u_f$ , stationary flame propagation velocity, m/sec;  $v_n$ , normal rate of combustion, m/sec;  $v_s$ , linear rate of pyrolysis, m/sec;  $W$ , chemical reaction rate, 1/sec;  $x$ , coordinate along the fuel surface, m;  $x_b$ , coordinate of the burning-out point of a fuel, m;  $X$ , generalized force;  $Y$ , mass concentration;  $y$ , coordinate normal to the surface of a fuel, m;  $\delta$ , variable thickness of a layer of a fuel, m;  $\theta$ , angle between the coordinate  $y$  and direction of action of the buoyancy force, rad;  $\lambda$ , thermal conductivity, W/(m·K);  $\nu$ , stoichiometric coefficient;  $\rho$ , density, kg/m<sup>3</sup>. Subscripts: 0, initial; a, ambient; b, burning-out point; fu, fuel; f, flame; n, normal; o, oxidant; s, solid fuel. Superscript: \*, characteristic.

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