

## DETERMINATION OF THE CATALYTIC ACTIVITY OF MATERIALS BY SOLVING THE EQUATIONS OF A NONEQUILIBRIUM MULTICOMPONENT BOUNDARY LAYER ON A FLAT PLATE

S. V. Peigin and V. Yu. Kazakov

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*A method is presented for determining the dependence of the probability of heterogeneous recombination  $\gamma_w$  from results of measurements of the heat flux  $Q_w$  to the surface of a catalytic sensor exposed to a pulsed supersonic flow of gas dissociated by an incident shock wave propagating in a shock tube. It is shown that the accuracy of the determination of  $\gamma_w$  depends not only on the accuracy of the measurements in the experiment, but also on the results of mathematical modeling of the flow of the dissociated gas over the surface of the body. Results from an analysis of an experiment are presented.*

**Introduction.** The catalytic properties of materials in relation to heterogeneous recombination in supersonic flows of dissociated gases have a significant effect on the level of thermal loads on a body in such a flow [1]. In connection with the development of the "Shuttle" and "Buran" reusable spacecraft, the 1980s marked the beginning of extensive experimental and theoretical research to determine the basic regularities of the processes occurring on a surface in heterogeneous catalytic reactions and the quantitative characteristics of these reactions for different materials [2–6]. The effect of the state of the gas interacting with the surface on the probability of heterogeneous recombination  $\gamma_w$  is still unknown. Of the known experimental methods, the method that most closely approaches conditions that arise when bodies enter the dense atmospheric layers involves determination of  $\gamma_w$  by exposing a model to a pulsed supersonic flow of a gas that has been dissociated by an incident shock wave propagating in a shock tube [7]. The only measurement in these experiments is the one of the heat flux  $Q_w$  to the surface of the test material, which depends on the catalytic heterogeneous reactions that occur and on the processes that take place in the gas layer over the surface. Thus, the accuracy of the determination of  $\gamma_w$  depends not only on the accuracy of measurement of heat flux  $Q_w$  but also on the results of mathematical modeling of the flow of dissociated gas over the surface of the body.

In this paper, we present results of determining the probability of heterogeneous recombination of oxygen atoms from the experimental data of [8, 9]. We use laminar boundary layer theory as the mathematical model and allow for the nonequilibrium of the chemical reactions and multi-component diffusion [1]. The initial equations are solved by a highly efficient numerical algorithm characterized by a high order of accuracy [10]. The algorithm does not require preliminary solution of the Stefan–Maxwell relations for diffusion flows. The calculations were performed for different sets of conditions in the incoming flow, and the efficiency and accuracy of the algorithm were evaluated. Approximation solutions of the given problem were obtained in [11, 12] with simplifying assumptions on the character of the gas flow in the boundary layer.

**Formulation of the Problem.** We are examining a hypersonic nonequilibrium flow of a mixture of  $O_2$ , O, and Ar gases over a flat plate consisting of two parts ( $M_1$  and  $M_2$ ) coated with substances having different catalytic properties. The surface  $M_1$  is noncatalytic at  $x < L$ , and the surface  $M_2$  is coated with the material whose catalytic properties are being studied.

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The initial mathematical model was the theory of a laminar boundary layer (LBL). The equation of a laminar boundary layer is the limiting form of the Navier–Stokes equations for  $Re \rightarrow \infty$ . This model is asymptotically exact in describing the flow structure near the surface of bodies at large Reynolds numbers and is even quite accurate at  $Re_\infty > 10^{3.5}$ . Since  $Re_\infty \sim 10^4$  for conditions in the incoming flow in the experiment, LBL theory can be used to solve the problem formulated here.

The initial system of equations written in the coordinate system  $(x, y)$  with allowance for the nonequilibrium chemical reactions and the multicomponent diffusion process and ignoring barometric diffusion, thermal diffusion, and the diffusional heat effect has dimensional form

$$\frac{\partial}{\partial x}(\rho u) + \frac{\partial}{\partial y}(\rho v) = 0; \quad (1)$$

$$\rho \left( u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} \right) = \frac{\partial}{\partial y} \left( \mu \frac{\partial u}{\partial y} \right); \quad (2)$$

$$\rho c_p \left( u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} \right) = \frac{\partial}{\partial y} \left( \frac{\mu c_p}{Pr} \frac{\partial T}{\partial y} \right) + \mu \left( \frac{\partial u}{\partial y} \right)^2 - \frac{\partial T}{\partial y} \sum_{k=1}^N c_{pk} I_k - \sum_{k=1}^N h_k \dot{W}_k; \quad (3)$$

$$\rho \left( u \frac{\partial c_k}{\partial x} + v \frac{\partial c_k}{\partial y} \right) + \frac{\partial I_k}{\partial y} = \dot{W}_k, \quad k = 1, 2, \dots, N-1; \quad (4)$$

$$\frac{\partial P}{\partial y} = 0; \quad (5)$$

$$P = \rho R_A T \sum_{k=1}^N \frac{c_k}{m_k}; \quad (6)$$

$$\mu \frac{\partial x_i}{\partial y} = \sum_{j=1}^N \frac{m^2}{m_j m_i} S_{ij} (c_i I_j - c_j I_i), \quad i = 1, 2, \dots, N-1; \quad (7)$$

$$\sum_{j=1}^N I_j = 0, \quad \sum_{j=1}^N c_j = 1, \quad x_i = c_i \frac{m}{m_i}. \quad (8)$$

Here the  $y$  coordinate is directed along the normal to the surface of the plate, the  $x$  coordinate is directed along the surface,  $u$  and  $v$  are physical components of the velocity vector along  $x$  and  $y$ , respectively,  $P$ ,  $\rho$ , and  $T$  are the pressure, density, and temperature of the gas mixture,  $\mu$ ,  $c_p$ ,  $Pr = \mu c_p / \lambda$ , and  $m$  are the viscosity, specific heat, the Prandtl number, and the molecular weight of the gas mixture,  $c_i$ ,  $m_i$ ,  $h_i$ ,  $c_{pi}$ ,  $I_i$ , and  $\dot{W}_i$  are the mass concentration, molecular weight, specific enthalpy, and specific heat of the  $i$ th component, the normal component of the vector of its diffusion flow, and the rate of its formation, and  $S_{ij}$  is the binary Schmidt number.

We solve system (1)–(8) subject to the following boundary conditions:

— on the external boundary of the boundary layer

$$y \rightarrow \infty: \quad u = u_e(x), \quad T = T_e(x), \quad C_i = C_{ie}(x);$$

— on the surface of the plate

$$y = 0: \quad u = v = 0, \quad T_w = 300 \text{ K}, \quad x < L: \quad I_O = I_{O_2} = I_{Ar} = 0,$$

$$x \geq L: \quad I_O = -\gamma_w \sqrt{\frac{RT_w}{2\pi m_O}} \rho_w C_O, \quad I_{O_2} = -I_O, \quad I_{Ar} = 0.$$

The following dissociation-recombination reactions of oxygen proceed in the flow:

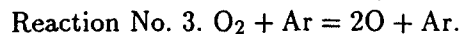
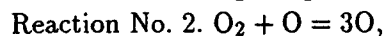
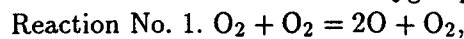


TABLE 1

Reaction No.	A	a	$E_a/R$	B	b
1	$2.3 \cdot 10^{19}$	-1.0	59,400	$1.9 \cdot 10^{16}$	-0.5
2	$8.5 \cdot 10^{19}$	-1.0	59,400	$7.1 \cdot 10^{16}$	-0.5
3	$3.0 \cdot 10^{18}$	-1.0	59,400	$2.5 \cdot 10^{15}$	-0.5

TABLE 2

Variant No.	Material of coating $M_2$	$V_\infty$ , m/sec	$T_\infty$ , K	$P_\infty$ , mm Hg	$x_{O_\infty}$	$x_{O_2\infty}$	$x_{Ar\infty}$	$Q_w$ , cal/(cm <sup>2</sup> ·sec)
1	Platinum	2760	3180	79.0	0.03	0.29	0.68	40.0
2	➤	2740	3180	90.0	0.03	0.29	0.68	50.2
3	➤	2830	3300	87.0	0.02	0.30	0.68	42.0
4	➤	2590	3020	78.0	0.05	0.25	0.70	37.1
5	Aluminum	2800	3240	84.0	0.02	0.30	0.68	33.0
6	Silicon monoxide	2790	3240	89.0	0.02	0.30	0.68	34.3
7	➤	2820	3280	85.0	0.02	0.30	0.68	35.6
8	➤	2820	3290	96.0	0.02	0.30	0.68	37.1
9	➤	2800	3240	84.0	0.02	0.30	0.68	35.0
10	➤	2830	3310	92.0	0.02	0.30	0.68	36.8
11	➤	2760	3180	82.0	0.02	0.30	0.68	31.5

The coefficients of the forward reactions  $D_j$  [cm<sup>3</sup>/(mole·sec)] and reverse reactions  $R_j$  [cm<sup>6</sup>/(mole<sup>2</sup>·sec)] have the form

$$D_j = AT^a \exp\left(-\frac{E_a}{RT}\right), \quad R_j = BT^b.$$

Table 1 gives the values of  $A$ ,  $a$ ,  $E_a/R$ ,  $B$ , and  $b$  for reaction Nos. 1-3.

The thermodynamic parameters and the mixture transport coefficients are calculated using formulas [13-15]

$$\mu = \sum_{i=1}^N \frac{\mu_i c_i / m_i}{c_i / m_i + \sum_{j=1, j \neq i}^N G_{ij} c_j / m_j}, \quad \lambda = \sum_{i=1}^N \frac{\lambda_i c_i / m_i}{c_i / m_i + 1.065 \sum_{j=1, j \neq i}^N G_{ij} c_j / m_j},$$

$$\mu_i = 2.6693 \cdot 10^{-5} \sqrt{m_i T} / (\sigma_i^2 \Omega_i^{(2,2)*}), \quad \lambda_i = \frac{15}{4} \mu_i \frac{R_A}{m_i} (\text{O, Ar}),$$

$$\lambda_i = \frac{15}{4} \mu_i \frac{R_A}{m_i} \left(0.115 + \frac{0.354 c_{pi}}{R_A}\right) (\text{O}_2), \quad D_{ij} = 0.00268 T^{3/2} \sqrt{\frac{m_j + m_i}{2m_i m_j}} \left(P \sigma_{ij}^2 \Omega_{ij}^{(1,1)*}\right)^{-1},$$

$$G_{ij} = \frac{1}{\sqrt{8}} \sqrt{\frac{m_j}{m_i + m_j}} \left[1 + \left(\frac{m_j}{m_i}\right)^{0.25} \left(\frac{\mu_i}{\mu_j}\right)^{0.5}\right]^2,$$

$$\Omega_{ij}^{(1,1)*} = 1.074 \left(\frac{T k}{\sqrt{\varepsilon_i \varepsilon_j}}\right)^{-0.1604}, \quad \Omega_i^{(2,2)*} = 1.157 \left(\frac{T k}{\varepsilon_i}\right)^{-0.1472}, \quad \sigma_{ij} = 0.5(\sigma_i + \sigma_j),$$

where  $i$  is the component number. The values of  $\varepsilon_i/k$  and  $\sigma_i$  are given below:

$$\varepsilon_1/k = 106.7, \quad \sigma_1 = 3.467, \quad \varepsilon_2/k = 106.7, \quad \sigma_2 = 3.050, \quad \varepsilon_3/k = 93.3, \quad \sigma_3 = 3.542.$$

TABLE 3

$\eta$	$U/U_e$	$T$	$C_O$	$U/U_e$	$T$	$C_O$
	$N = 12$			$N = 120$		
0	0	300.0	0.14027	0	300.0	0.1405
0.2	—	—	—	0.0498	611.6	0.1405
0.4	—	—	—	0.1105	962.3	0.406
0.5	0.14403	1143.7	0.14051	0.1441	1144.8	0.1407
0.6	—	—	—	0.1796	1328.5	0.1407
0.8	—	—	—	0.2554	1689.7	0.1408
1.0	0.3356	2027.3	0.1409	0.3357	2028.4	0.1410
2.0	0.7275	3055.5	0.1414	0.7277	3055.7	0.1415
3.0	0.9436	3201.7	0.1417	0.9436	3201.6	0.1417
4.0	0.9949	3187.3	0.1417	0.9949	3187.3	0.1417
5.0	0.9998	3181.8	0.1417	0.9998	3181.8	0.1417
6.0	1.0000	3180.4	0.1417	1.0000	3180.4	0.1417

TABLE 4

$\eta$	$U/U_e$	$T$	$C_O$	$U/U_e$	$T$	$C_O$
	$N = 3$			$N = 6$		
0	0	300.0	0.1380	0	300.0	0.1396
2.0	0.71754	3031.2	0.1411	0.7266	3053.9	0.1413
3.0	—	—	—	0.9434	3202.3	0.1417
4.0	0.99713	3179.4	0.1417	0.9949	3187.0	0.1417
5.0	—	—	—	0.9998	3181.8	0.1417
6.0	1.00000	3180.4	0.1417	1.0000	3180.4	0.1417

We examined 11 variants of relations among the velocity, pressure, temperature, and molar concentrations of the individual components on the external boundary of the boundary layer for the three materials studied: platinum, aluminum, and silicon monoxide. Table 2 shows the experimental conditions and the experimentally determined heat flux  $Q_w$  at the point  $x = L = 18$  mm.

We calculated the probability of heterogeneous recombination  $\gamma_w$  for different materials using experimental data on heat flux to the surface of the body at the point  $x = L$ .

**Method of Numerical Solution.** We write the initial system of equations in Dorodnitsin variables, thus resolving the singularity problems at the point  $x = 0$ :

$$\xi = \int_0^x \rho_e \mu_e u_e dx, \quad \eta = \frac{u_e}{\sqrt{2\xi}} \int_0^y \rho dy, \quad f'_\eta = \frac{u}{u_e}, \quad \theta = \frac{T}{T_e},$$

$$X_i = \frac{\sqrt{2\xi}}{\xi'_x} I_i, \quad l = \frac{\mu\rho}{\mu_e\rho_e}, \quad X_q = -\frac{lT_e c_p}{Pr} \theta'_\eta + \sum_{k=1}^N h_k X_k.$$

The Stefan–Maxwell relations are used in the form

$$X_i = \alpha_i c'_{i\eta} + \beta_i c_i, \quad i = 1, 2, \dots, N-1, \quad (9)$$

where

$$\alpha_i = -\frac{lb_{(ii)}}{S_{iN} a_{(ii)}}, \quad \beta_i = a_{ii}^{-1} \sum_{j=1, j \neq i}^{N-1} \left( a_{ij}^* X_j + c'_{j\eta} \frac{lb_j^*}{S_{iN}} \right), \quad a_{ij} = -a_{ij}^* c_i, \quad b_{ij} = -b_j^* c_i \quad (i \neq j),$$

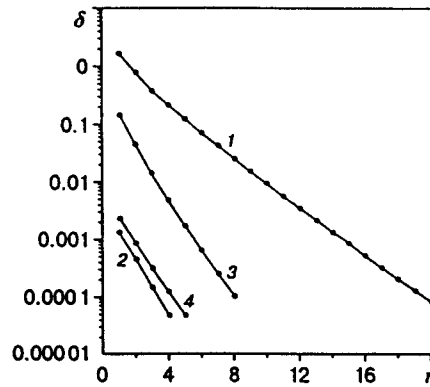


Fig. 1. Dependence of the convergence parameter  $\delta$  on the number of iterations  $n$  with a route coordinate  $x = 0, 18, 18.01, \text{ and } 30 \text{ mm}$  (curves 1-4).

$$a_{ij}^* = \frac{m_N S_{ij}}{m_j S_{iN}} - \frac{S_{iN}}{S_{iN}}, \quad b_j^* = \frac{m_N}{m_j} - 1, \quad a_{ii} = \frac{S_{iN}}{S_{iN}} + \sum_{j=1, j \neq i}^{N-1} a_{ij}^* c_j, \quad b_{ii} = 1 + \sum_{j=1, j \neq i}^{N-1} b_j^* c_j.$$

The problem is solved on the basis of a finite-difference scheme that is implicit with respect to  $y$  and has the order of approximation  $O(\Delta y)^4$  [10]. Each of the initial equations is reduced to a system of first-order equations which are then solved in the following sequence: the equations of motion, the energy equations, and the equations for the concentrations of  $O_2$  and  $O$ .

The equations for the concentrations are solved by the approach used in [16], which includes the following steps:

(a) the concentration equations are regarded as a Cauchy problem (with boundary conditions on the surface of the body) for first-order equations in diffusion fluxes  $X_i$ , with the concentrations and their derivatives taken from the preceding iteration;

(b) the coefficients  $\alpha_j$  and  $\beta_j$  are calculated after the determination of  $X_i$ ;

(c) the Stefan-Maxwell relations (9) with the known coefficients  $\alpha_j$  and  $\beta_j$  are substituted into the concentration equations and a solution is found to the resulting boundary-value problem for second-order equations in  $c_i$ ; the iteration is then repeated beginning with step (a).

The present computational procedure does not require solution of the Stefan-Maxwell equations for diffusion fluxes.

The following linearization is used in the problem:

— in the energy equation, linearization of the rate of formation of the  $k$ th component

$$\left(\frac{\dot{W}_k}{\rho}\right)^i = \left(\frac{\dot{W}_k}{\rho}\right)^{i-1} + \left(\frac{\partial \dot{W}_k \rho^{-1}}{\partial \theta}\right)^{i-1} (\theta^i - \theta^{i-1}),$$

where  $i$  is the current iteration number;

— in the concentration equations [17],  $(\dot{W}_k/\rho)^i = W_{0k}^{i-1} - W_{1k}^{i-1} c_k^i$ .

**Results of the Calculations.** An analysis of the numerical results shows that the proposed algorithm is stable and efficient. Tables 3 and 4 show the effect of the grid step on the accuracy of the calculations at the point  $x = 10 \text{ mm}$  for the first variant.

The following iterative process is used to determine the probability of heterogeneous recombination  $\gamma_w$ . In the first step after the point of discontinuity of the catalytic properties, we calculate the heat flux  $Q_w$  for the given  $\gamma_w$ . The resulting value is compared with experimental data on the heat flux. The value of  $\gamma_w$  is refined, and the boundary-layer equations are calculated again. Six or seven iterations are needed for convergence.

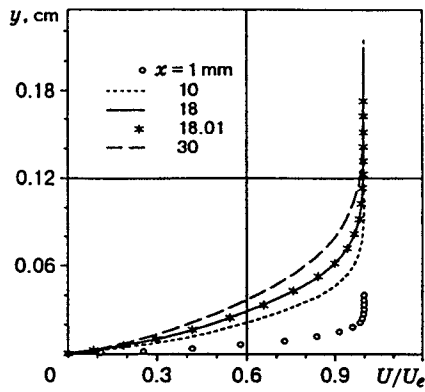


Fig. 2

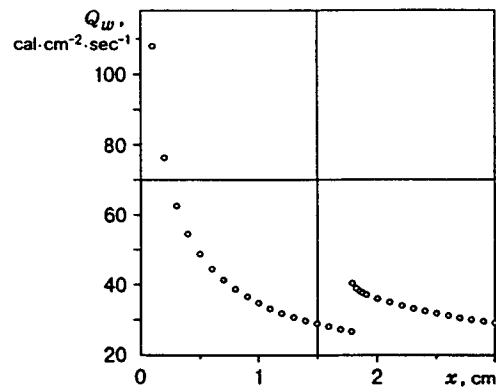


Fig. 3

Fig. 2. Velocity profiles along the surface of the plate (calculation for variant No. 9 of conditions).

Fig. 3. Distribution of heat flux along the surface of the plate (calculation for variant No. 9 of conditions).

TABLE 5

Variant No.	Coating material $M_2$	$\gamma_w$	$\gamma_w(\dot{w}_k = 0)$	$\gamma_w [11]$
1	Platinum	$1.2572 \cdot 10^{-2}$	$1.3604 \cdot 10^{-2}$	$1.0 \cdot 10^{-2}$
2	»	$1.8442 \cdot 10^{-2}$	$1.9723 \cdot 10^{-2}$	$2.5 \cdot 10^{-2}$
3	»	$1.0162 \cdot 10^{-2}$	$1.1347 \cdot 10^{-2}$	$1.0 \cdot 10^{-2}$
4	»	$1.5298 \cdot 10^{-2}$	$1.6156 \cdot 10^{-2}$	$1.5 \cdot 10^{-2}$
5	Aluminum	$3.8468 \cdot 10^{-3}$	$4.9752 \cdot 10^{-3}$	$2.5 \cdot 10^{-3}$
6	Silicon monoxide	$4.0001 \cdot 10^{-3}$	$5.2323 \cdot 10^{-3}$	$2.0 \cdot 10^{-3}$
7	»	$5.4632 \cdot 10^{-3}$	$6.5990 \cdot 10^{-3}$	$3.0 \cdot 10^{-3}$
8	»	$4.3169 \cdot 10^{-3}$	$5.6620 \cdot 10^{-3}$	$2.0 \cdot 10^{-3}$
9	»	$5.5321 \cdot 10^{-3}$	$6.6644 \cdot 10^{-3}$	$3.5 \cdot 10^{-3}$
10	»	$4.7177 \cdot 10^{-3}$	$5.9781 \cdot 10^{-3}$	$2.8 \cdot 10^{-3}$
11	»	$3.6286 \cdot 10^{-3}$	$4.7475 \cdot 10^{-3}$	$1.8 \cdot 10^{-3}$

In addition, we use local iterations with respect to nonlinearity to obtain a solution of the problem on the current ray ( $x = \text{const}$ ). The iterative process is considered convergent if the parameter  $\delta$  (the maximum difference of all sought functions at all points of the difference grid in two neighboring iterations) does not exceed  $10^{-4}$ . Calculations show that the numerical method described above is very economical and has a high rate of convergence in nonlinearity. Figure 1 gives an example of the dependence of the parameter  $\delta$  on the number of iterations  $n$  that characterizes the rate of convergence in nonlinearity for variant No. 9 of conditions on the external boundary of the boundary layer and different values of the route coordinate  $x$ . It is evident that except for the first calculation point, 4–6 iterations are needed to obtain a solution with an accuracy of  $10^{-4}$ .

Figures 2 and 3 show typical examples of calculation of the velocity profile across the boundary layer and the distribution of heat flux along the surface of the plate according to the numerical solution of the problem for variant No. 9 of conditions on the external boundary. It is apparent that the solution of the boundary-layer equations (including the distribution of the parameters across the boundary layer) strongly depends on the route coordinate. Thus, the widely used approximation of local similarity can result in a substantial error in the given case.

Table 5 shows the probabilities of heterogeneous recombination  $\gamma_w$  calculated on the basis of experimental data on heat flux to the surface at the point  $x = 18$  mm. The data in the third column are obtained by solution of the problem in the complete formulation, and the data in the fourth column are obtained on the assumption that the homogeneous chemical reactions were frozen inside the boundary layer. The results in [11] were obtained on the assumption of local similarity of the flow and the absence of chemical reactions in the gas phase. Obviously, the accuracy of the mathematical model of flow used here has a significant effect on the values of the probability of heterogeneous recombination (the difference can reach 70–100%).

It can be concluded from the above results that within the framework of an asymptotically correct hydrodynamic model of flow and with allowance for the complex character of the nonequilibrium physico-chemical processes that take place in the flow, the approach proposed here and the numerical algorithm developed on the basis of it make it possible to determine the probability of heterogeneous recombination of oxygen atoms from experimental measurements of heat flux to the surface coated with the materials that we studied. The numerical method can be recommended for use in analyzing experimental data for other coating materials and gas mixtures.

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