VIBRATIONALLY NONEQUILIBRIUM FLOW OF A CO_2–N_2–O_2–H_2O MIXTURE IN A WEDGE NOZZLE

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The combustion of hydrocarbon fuels is used in the production of high temperatures. The composition of the combustion products is dependent on the particular conditions, but as a rule there is usually a fair amount of molecular oxygen accompanying the CO_2 and N_2 (molar fraction $\gamma_{O_2} = 0.2$) along with water vapor ($\gamma_{H_2O} = 0.05$).

The subsequent expansion of the combusion products in a supersonic nozzle is characterized by large deviations from equilibrium. Research on such flows has recently developed considerably on account of the use of gasdynamic lasers, in which wedge and planar or axially symmetrical profiled nozzles are used [1]. The deviation from equilibrium is determined from the weak-signal amplification coefficient for a CO_2 laser probe beam. For simple CO_2–N_2–He mixtures it has been found that there is satisfactory agreement between theory and experiment, so the main problems of energy-exchange kinetics have been solved for such flows [1].

On the other hand, there is an increasing flow of publications on the kinetics of vibrational energy transfer in CO_2–N_2–CO–O_2–H_2–H_2O [2-7] mixtures, and this indicates that simulation of the flow in such mixtures to give agreement with experiment for a wide range of compositions and flow parameters is still an unsolved problem. The complexity in a simulation arises mainly from the uncertainty in the constants, and also from the various simplifications made over the kinetics of vibrational energy transfer.

In each particular case, the theoretical approach may be based on comparison of the calculations with the observed gain, so particular importance attaches to analysis of the measurement conditions. In particular, it is usual to compare the results not with the distribution of the gain along the axis of the flow but with the value obtained in some particular section. Most of the measurements are made as a rule at the end of the nozzle, and then [8] the results should be corrected on account of the real geometry of the jet, whose width may exceed the width of the nozzle by 30%. If experiments are conducted with a shock tube, particular importance attaches to the initiation of the nozzle devices. For example, optical studies have been made of the flow patterns in wedge nozzles [9] and in slots [8], and in some cases it is found that the flow has not attained a steady-state structure throughout the measurements. In order to obtain reliable results, it is best to perform measurements in parallel on several sections within the nozzle provided that there are no perturbations in the flow caused by errors in the profile or detachment of the over-expanded flow from the walls.

Here such a study is performed in order to test theoretical models for vibrational energy transfer. A planar wedge nozzle is considered [9], and in sections at distances $x_1 = 29$ mm, $x_2 = 93$ mm from the inlet part there is an undetached quasistationary flow that is close to one-dimensional [10]. The mixture contained various proportions of CO_2, N_2, O_2, H_2O, and the initial temperatures ranged from 1000 to 2000°K, and the pressures from 2.5 to 7.5 atm.

1. Model for Vibrational Transfer in CO_2–N_2–O_2–H_2O Mixtures. The results of [11-15] indicate that the following are the main relaxation channels for the vibrational energy:

$$CO_2(00^01) + M \rightarrow \begin{cases} CO_2(11^10) + M & W_{312}^M, \\ CO_2(03^10) + M & W_{32}^M, \end{cases}$$

$$CO_2(00^01) + N_2(V = 0) \rightarrow CO_2(00^00) + N_2(V = 1) \quad W_{34},$$

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$$CO_2(00^00) + N_2(V=1) \to \begin{cases} CO_2(11^10) + N_2(V=0) & W_{412}, \\ CO_2(03^10) + N_2(V=0) & W_{42}, \end{cases}$$

$$CO_2(00^00) + O_2(V=1) \to CO_2(10^00) + O_2(V=0) \quad W_{51},$$

$$CO_2(10^00) + M \to CO_2(02^00) + M \quad W_{12}^M,$$

$$CO_2(01^10) + M \to CO_2(00^00) + M \quad W_{20}^M,$$

$$N_2(V=1) + H_2O(000) \to N_2(V=0) + H_2O(010) \quad W_{46},$$

$$O_2(V=1) + H_2O(000) \to O_2(V=0) + H_2O(010) \quad W_{56},$$

$$N_2(V=1) + O_2(V=0) \to O_2(V=1) + N_2(V=0) \quad W_{54},$$

$$N_2(V=1) + M \to N_2(V=0) + M \quad W_{40}^M,$$

$$O_2(V=1) + M \to O_2(V=0) + M \quad W_{50}^M,$$

$$H_2O(010) + M \to H_2O(000) + M \quad W_{60}^M,$$

where M is any unexcited molecule in the mixture, W_{ip} is the probability of vibrational quantum exchange, i = p = 1, 2, 3 correspond to the symmetrical, deformation, and unsymmetrical modes of the CO_2 molecule, i = p = 4, 5, 6 correspond to the lower excited states of the molecules N_2 (V = 1), O_2 (V = 1), and H_2O (010) respectively, p = 0 is the unexcited state of any molecule, and p = 12 is the mixed state of CO_2 (11^10).

The kinetic equations describing the vibrational relaxation were written on the assumption of complete resonance between the symmetrical and deformation vibrations ($\Theta_1 = 2\Theta_2$, where Θ_i are the characteristic vibrational temperatures) and on the basis of equality of the vibrational temperatures ($T_1 = T_2$), by means of a generalized relaxation equation [11]. In that case the probabilities W_{32}^M, W_{312}^M and W_{42}, W_{412} may be combined:

$$W_{3\Sigma}^M = W_{32}^M + W_{312}^M \frac{1-y_2}{1+y_2}, \quad W_{4\Sigma} = W_{42} + \frac{1-y_2}{1+y_2} W_{412},$$

where $y_i = \exp(-\Theta_i/T_i)$; these are the quantities usually measured [11].

Correct choice of the temperature dependence for each of the probabilities of the elementary processes is important in creating the model [7, 16].

The use of various $W_{ip} = f(T)$ relations even within the framework of a single kinetic model can result in about 50% difference in the calculated values for the gain [16]; here the $W_{ip} = f(T)$ relations have been derived from an analysis of the theoretical studies [13-15] and the experimental ones [17-23].

Table 1 gives the approximation relations for the range 300-2000°K.

The gain of the medium at the center of the P(20) line of the 00°1 → 10°0 transition was calculated from

$$\alpha_\nu = \frac{c^2}{4\pi\nu_0 b_D} \sqrt{\frac{\ln 2}{\pi}} A_{mn} \frac{(2j''-1)\rho N_A \gamma_{CO_2} \delta}{\mu Q_{rot} Q_{vib}} H(a, 0)\, DN,$$

$$DN = y_3 B' \exp\left[-\frac{B'j''(j''-1)}{kT}\right] - y_2^2 B'' \exp\left[-\frac{B''j''(j''+1)}{kT}\right],$$

where N_A is Avogadro's number, c is the velocity of light in vacuum, ν_0 is the transition frequency at the line center, b_D is the Doppler width of the line at half height as defined by $b_D = 2\nu_0 \sqrt{\frac{2RT\ln 2}{\mu_{CO_2} c^2}}$, B" and B' are the rotational constants for the lower and upper laser levels respectively, δ is the symmetry factor (δ = 2 for the CO_2 molecules), j" is the rotational quantum number of the lower vibrational state in the transition (j" = 20), ρ and T are the gas density and temperature, k is Boltzmann's constant, R is the universal gas constant, $\mu = \sum_{i=1}^{n} \mu_i \gamma_i$ (γ_i is the molar fraction of component i in the mixture, whose molecular weight is μ_i), $H(a, 0)$ is the Voight function, which was determined in accordance with [1],

TABLE 1

	W_{ip}, cm³/sec	Ref.
$W_{3\Sigma}^{CO_2}$	$\exp(9{,}456-218{,}23T^{-1/3}+1{,}687T^{-2/3}-3909{,}27/T)1{,}36T\cdot10^{-16}$	[20]
$W_{3\Sigma}^{N_2}$	$\exp(15{,}456-424{,}03T^{-1/3}+3852{,}67T^{-2/3}-10672{,}157T^{-1})\times$ $\times1{,}36T\cdot10^{-16}$	[20]
$W_{3\Sigma}^{O_2}$	$W_{3\Sigma}^{N_2}$	[13, 14]
$W_{3\Sigma}^{H_2O}$	$4\cdot10^{-15}T(1-y_{20})^3/(1-y_{30})$	[21, 20]
$W_{20}^{CO_2}$	$T\cdot10^{-21}\exp(-7{,}85+17{,}42T^{-1/3})0{,}136$	[1, 13]
$W_{20}^{N_2}$	$0{,}136\cdot10^{-21}T\exp(-80{,}6+15{,}5T^{-1/3})$	[1, 13]
$W_{20}^{H_2O}$	$0{,}136\cdot10^{-21}T\exp(-5{,}46-21T^{-1/3})$	[1, 17]
$W_{20}^{O_2}$	$W_{20}^{N_2}$	[13, 14]
$W_{40}^{CO_2}$	$\exp(-19{,}414-134{,}727T^{-1/3}+253{,}2T^{-2/3}-2551{,}7T^{-1})$	[13, 14]
$W_{40}^{N_2}$	$\exp(7{,}489-965{,}3T^{-1/3}+6851{,}155T^{-2/3}-17248{,}9T^{-1})$	[13, 14]
$W_{40}^{O_2}$	$W_{40}^{N_2}$	[13, 14]
$W_{40}^{H_2O}$	$2{,}61\cdot10^{-17}\exp(-34{,}88T^{-1/3})$	[13, 14, 23]
$W_{50}^{CO_2}$	$\exp(-146{,}5T^{-1/3})1{,}34\cdot10^{-12}$	[13, 14]
$W_{50}^{N_2}$	$\exp(-131{,}6T^{-1/3})6{,}78\cdot10^{-13}$	[13, 14]
$W_{50}^{O_2}$	$\exp(-157T^{-1/3})1{,}11\cdot10^{-8}$	[13, 14]
$W_{50}^{H_2O}$	$10^2W_{50}^{O_2}$	[13, 14]
$W_{60}^{H_2O}$	$\exp(-11{,}05-222{,}4T^{-1/3}+924T^{-2/3}+82T^{-1})$	[13, 14]
$W_{60}^{O_2}$	$\exp(-32{,}973+2T^{-1/3}+80{,}53T^{-2/3}+68{,}159T^{-1})$	[13, 14]
$W_{60}^{N_2}$	$W_{60}^{O_2}$	[13, 14]
$W_{60}^{CO_2}$	$0{,}5W_{60}^{O_2}$	[13,14]
W_{51}	$1{,}49\cdot10^{-20}T(3T+556)$	[22]
W_{34}	$4{,}16\cdot10^{-14}\sqrt{T}\exp(8{,}84\cdot10^{-7}T^2-T\cdot2{,}07\cdot10^{-3})$	[13, 14]
W_{56}	$1{,}39\cdot10^{-11}\sqrt{T}$	[13]
$W_{4\Sigma}$	$0{,}28W_{3\Sigma}^{N_2}$	[15]
W_{46}	$\exp(-17{,}66-140{,}9T^{-1/3}+347{,}214T^{-2/3}-881{,}9T^{-1})$	[14, 23]

parameter α was determined on the assumption of independence of temperature for the collisional-broadening cross section (the values were taken from the recommendations of [1, 24]), Q_{vib} and Q_{rot} are the vibrational and rotational statistical sums, respectively, and the Einstein coefficient A_{mn} was taken as 0.187 sec^{-1} [24].

The nonequilibrium parameters of the medium were determined by solving the equations of gasdynamics with the relaxation equations; we used the approximation of one-dimensional steady-state flow of an ideal nonconducting gas. Numerical integration of the complete system of equations was by the method of [25] and was based on the given distribution for the density derived for example from the calculation of a flow of a perfect gas in the transonic region. In that case one encounters no difficulties related to passage through the singular point in the critical section of the nozzle, and the integration is carried directly from the subsonic region into the supersonic one. The flow is of nonequilibrium type in the subsonic and transonic parts of the nozzle, and this is utilized in avoiding errors in determining the inversion and the gain of the medium [26], which is particularly important at low values of the dynamic pressure (p_0 < 10 atm). An inexplicit second-order difference scheme [25] reduced the computation time considerably.

2. Methods. The gain measurements were made with a shock tube having a diameter of about 500 mm in the working part. See [27] for a description of the design and the methods of measurement. The planar wedge nozzle with rectilinear generators had an angle of 30° in the supersonic part. The height of the critical section was 2 mm. The subsonic part was rounded to a radius of 4 mm. The mixture was generated in a separate volume with forced circulation and was saturated with water vapor during inlet to the tube. The water vapor

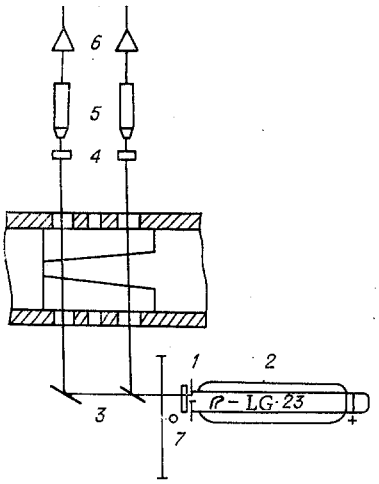

Fig. 1

pressure in the mixture was monitored by sampling by the method of [28]. The samples were taken via a hole in the side wall, so the mixture was first kept in the tube in order to allow the water vapor concentration to equalize over the cross section. Therefore, in most of the experiments the interval between admitting the mixture to the tube and the start of the measurements was not less than 50 min. The error in determining the water vapor concentration was 3%. The pressure before exit through the nozzle was determined with pressure transducers, while the temperature was measured by line reversal [29].

The gain was measured with an LG-23 commercial CO_2 laser. Special measures were taken to isolate the body of the laser from mechanical vibrations and pressure fluctuations in the discharge-tube cooling system. The laser worked almost in the one-mode state with the cavity partially out of adjustment, while within the cavity (Fig. 1) was an iris diaphragm 1, which cut off the transverse modes. Fluctuations in the supply voltage were reduced by means of a smoothing filter, while the discharge current was stabilized with a circuit employing a GU-50 pentode [30]. These measures reduced the instability in the output power to about 3% over 10 min. Figure 1 shows the system used in measuring the gain. The radiation from the laser 2 is split by the two semitransparent plates 3 made of BaF_2 into two beams that pass across the flow and then through the narrow-band filters 4, which isolate the spectral region around 10 μm, and onward to the photoresistors 5 type Svod, then via cathode followers and preamplifiers 6 to the oscilloscope. The intensity was monitored by means of the mechanical chopper 7, which was removed directly before the experiment. The errors in determining the gain were estimated from a sample of six experiments made under similar conditions. We calculated the dispersion ($S^2 = 0.00126$), skewness ($\hat{A} = 1.56$), and excess ($\hat{E} = 0.412$), and we also checked for a normal distribution [31]. The error in determining the gain was 12.6%.

3. Analysis. The flow of the CO_2–N_2–O_2–H_2O mixture was examined over a wide range in the initial parameters, e.g., the temperature of the mixture before inlet to the nozzle varied from 1000 to 2200°K, while the pressure varied from 2.5 to 7.5 atm. The values of the gain measured simultaneously in two sections at the axis of the nozzle were compared with the theoretical values. Figure 2 shows the theoretical results (broken lines) and the measurements (points) for three characteristic complications:

$$\gamma_{CO_2} = 0.078, \quad \gamma_{N_2} = 0.721, \quad \gamma_{O_2} = 0.185, \quad \gamma_{H_2O} = 0.02 \text{ (Fig. 2a);}$$
$$\gamma_{CO_2} = 0.0545, \quad \gamma_{N_2} = 0.784, \quad \gamma_{O_2} = 0.135, \quad \gamma_{H_2O} = 0.025 \text{ (Fig. 2b);}$$
$$\gamma_{CO_2} = 0.081, \quad \gamma_{N_2} = 0.792, \quad \gamma_{O_2} = 0.1, \quad \gamma_{H_2O} = 0.026 \text{ (Fig. 2c).}$$

The flow parameters were correspondingly as follows: $p_0 = 4.3$ atm, $T_0 = 1080$°K; $p_0 = 4.75$ atm, $T_0 = 1630$°K; $p_0 = 5.3$ atm, $T_0 = 2060$°K. The calculations are in satisfactory agreement with experiment only for $T_0 = 1080$°K (Fig. 2a). At $T_0 = 1630$°K (Fig. 2b), the theoretical values of α_ν lie below the observed ones. The discrepancy increases to 30% at $T_0 = 2060$°K (Fig. 2c). This tendency for the discrepancy to increase with temperature may be explained by assuming that the rate of deactivation of the deformation modes of CO_2, and therefore the rate of population of the lower laser level, will be determined in the main by collision with H_2O molecules ($W_{20}^{H_2O}$); the temperature dependence used in the calculations for $W_{20}^{H_2O}$

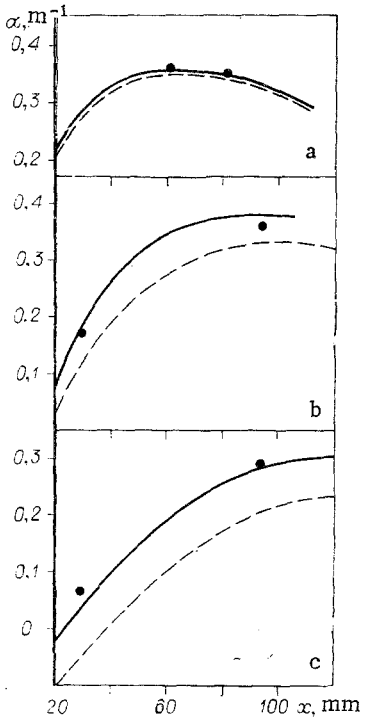

Fig. 2

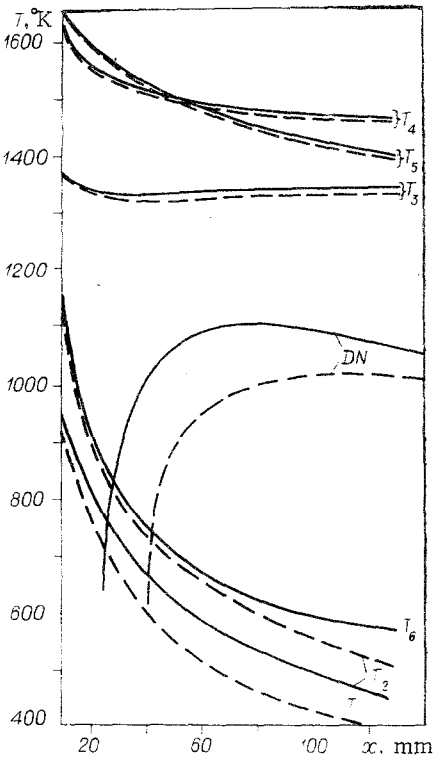

Fig. 3

TABLE 2

No.	p_0, atm	T_0, °K	γ_{CO_2}, %	γ_{N_2}, %	γ_{O_2}, %	γ_{H_2O}, %	Cross section I		Cross section II	
							α_{th}, m^{-1}	α_{exp}, m^{-1}	α_{th}, m^{-1}	α_{exp}, m^{-1}
1	3,2	1500	7,8	70,55	18,5	3,15	—	—	0,335	0,31
2	5,44	1800	5,4	79,7	10,6	4,3	—	—	0,265	0,265
3	6,8	2040	5,4	82,8	10,6	1,8	0,06	0,08	0,305	0,31
4	4,1	1780	8,1	80,1	10	1,7	—	—	0,3	0,26
5	4,67	1930	8,1	79,9	10	1,9	0,07	0,08	0,30	0,275
6	5,4	2000	7,8	71,6	18,5	2,1	0,04	0,065	0,305	0,32
7	6,25	2180	7,8	71,3	18,5	2,4	—	—	0,22	0,23
8	4,31	1080	7,8	72,1	18,5	1,6	0,355	0,36	0,34	0,34
9	4,76	1630	5,45	78,55	13,5	2,5	0,08	0,075	0,38	0,365
10	5,3	2060	8,1	79,3	10	2,6	—	—	0,285	0,29
11	3,17	1180	6,5	78,4	12,3	2,8	0,195	0,155	0,225	0,22
12	4,72	1510	6,5	79,1	12,3	2,1	0,145	0,137	0,305	0,27

is one in which $W_{2_0}^{H_2O}$ decreases [1, 17]. This relationship leads to a slower rate of relaxation of the 10^0 level of the CO_2 molecule as the temperature increases, and thus to freezing of the vibrational temperature T_2 in the supersonic part of the nozzle. A consequence of this is a reduction in the inversion DN, and therefore also in the gain. A difference from [1, 17] is that the $W_{2_0}^{H_2O} = f(T)$ relation derived in [19] was practically constant. In the range in T from 300 to 2000°K, the value of $W_{2_0}^{H_2O}$ may be taken as $W_{2_0}^{H_2O} = 2 \cdot 10^{-11}$ cm³/sec [19]. The temperature curves for $W_{2_0}^{H_2O}$ given in [1, 17, 19] give substantially different results only for temperatures around 1000°K, so the recommendations of [19] should lead to an increased rate of deactivation of the (01^10) and (100) states and to increase in the theoretical values of DN and α_ν at high temperatures ($T_0 \geqslant 1700$°K).

The numerical calculations confirmed this, as Fig. 3 shows for the distributions of the vibrational temperature T_i and translational temperature T, and also the inversion, along the axis of the nozzle derived from the temperature dependence of [1, 17] for $W_{2_0}^{H_2O}$ (broken lines) and of [19] (solid lines) for a mixture with $\gamma_{CO_2} = 0.081$, $\gamma_{N_2} = 0.792$, $\gamma_{O_2} = 0.1$, $\gamma_{H_2O} = 0.026$, $T_0 = 2060$°K, and $p_0 = 5.3$ atm.

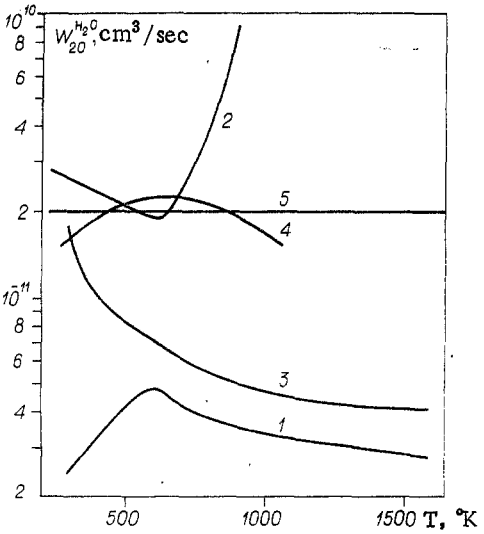

Fig. 4

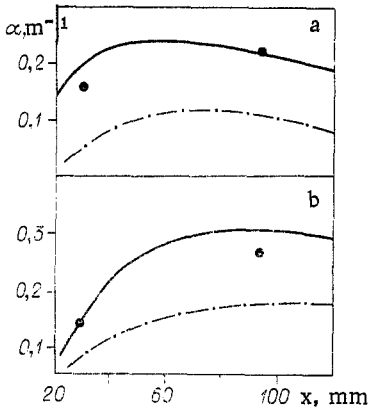

Fig. 5

Parts a-c of Fig. 2 show the distributions of $\alpha_\nu(x)$ calculated from the recommendations of [19] for the above cases (solid lines); use of the value $W_{2_0}^{H_2O} = 2 \cdot 10^{-11}$ cm³/sec constant throughout the temperature range substantially improves the agreement between theory and experiment, particularly at high temperatures. A similar comparison for the gain by the method used here with the $W_{2_0}^{H_2O}$ of [19] and the measured α_ν for other conditions is shown in Table 2, which indicates that the maximum difference between theory and experiment for α_ν is 10%.

A similar correction of $W_{2_0}^{H_2O} = f(T)$ to improve the agreement between theory and experiment has been performed in [2, 6, 7]; for $T > 600°K$ use was made [2, 6] of the temperature dependence derived from [17], while for $T \leqslant 600°K$ it was assumed that $W_{2_0}^{H_2O} = 8.13 \cdot 10^{-11} T$; in [7], allowance was made for the temperature dependence of $W_{2_0}^{H_2O}$, while the absolute values for $450° < T < 850°$ were closer to the value used here. Figure 4 shows curves for $W_{2_0}^{H_2O} = f(T)$ recommended in these studies (the numbers 1-5 correspond to [2, 6], [7], [1, 17], [19] and our results).

Figure 5 compares the observed α_ν with the result from our method (solid lines) and from the method of [10] (dot–dash lines). In [10], no allowance was made for the nonequilibrium flow in the subsonic part of the nozzle, and the model of [1] was used for the vibrational energy transfer in the CO_2–N_2–H_2O mixture, with the oxygen taken as an inert diluent. The temperature dependence $W_{2_0}^{H_2O}(T)$ was taken in accordance with [1, 17] (curve 3 of Fig. 4). The calculations were performed for two characteristic compositions: a) $\gamma_{CO_2} = 0.04$, $\gamma_{N_2} = 0.78$, $\gamma_{O_2} = 0.152$, $\gamma_{H_2O} = 0.028$; b) $\gamma_{CO_2} = 0.04$, $\gamma_{N_2} = 0.797$, $\gamma_{O_2} = 0.152$, $\gamma_{H_2O} = 0.03$. In the first case parameters at the inlet were $T_0 = 1180°K$, $p_0 = 3.17$ atm (Fig. 5a), while in the second (Fig. 5b) they were $T_0 = 1510°K$, $p_0 = 4.72$ atm. The method of [10] reduces the gain by almost a factor two by comparison with experiment or by comparison with the calculation from [19] for our method.

This model for nonequilibrium flow of a CO_2–N_2–O_2–H_2O mixture with $W_{2_0}^{H_2O} = 2 \cdot 10^{-11}$ cm³/sec is thus in satisfactory agreement with experiment throughout the range of parameters and compositions used. The existing discrepancy does not exceed the error of measurement.

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