MUTUAL EFFECT OF ATOMIC RECOMBINATION AND VIBRATIONAL RELAXATION OF Br_2 UNDER CONDITIONS OF SUPERSONIC EXPANSION IN A NOZZLE

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The mutual effect of processes of vibrational relaxation, dissociation, and recombination in a diatomic gas has been widely studied in recent years [1-13]. This interaction between vibrational and chemical kinetics mainly determines the picture of the phenomena taking place with the high-temperature dissociation of a molecular gas in shock waves and with atomic recombination in expanding flows. The principal part of the investigations in this region relates to the study of the interaction between dissociation and vibrational relaxation [1-11]. Considerably less attention has been paid to study of the mutual effect of chemical and vibrational kinetics in processes where recombination predominates [12, 13]. In [12] an attempt was made to examine the conditions under which vibrational—recombination interaction is possible in an expanding gas stream. An investigation was made of the recombination of atoms of oxygen strongly diluted with argon. This made it possible to exclude vibrational—vibrational exchange of energy from the discussion. Assuming that in the process of recombination there is established a quasi-steady-state distribution of the forming molecules with respect to vibrational states, the authors of [12] predicted the possibility of an excess population of the upper vibrational levels and a corresponding slowing-down of atomic recombination.

Article [13] discussed the effect of recombination in an expanding flow on the rate of vibrational relaxation of the forming molecules and the conditions under which a considerable deviation from an equilibrium distribution with respect to vibrational levels takes place in the flow before the recombination started to "freeze." Modelling a molecule by a cut-off harmonic oscillator, the author showed that such conditions are realized with $T_V > (1/16)D/R$, where T_V is the vibrational temperature of the gas at the moment when the equilibrium distribution breaks down; D is the dissociation energy. It is worthy of note that under these circumstances there is the possibility of a considerable increase in the "effective" time of vibrational relaxation.

The present article is devoted to a numerical investigation of the flow of partially dissociated Br_2 through a nozzle, taking account of the mutual effect of vibrational and chemical kinetics, carried out on the basis of a physical model which is more real than in preceding work.

Partially dissociated bromine is a convenient subject for such investigations. The relatively low dissociation energy of Br_2 makes it possible to expect that with stagnation temperatures around 3000°K and with the satisfaction of determined conditions for expansion in a nozzle, the mutual effect will be found to be considerable.

In the work an attempt was made to find criteria making it possible to judge when it is necessary to take account of vibrational—recombination with the flow of a gas with vibrational and chemical disequilibrium through a nozzle.

To solve the problem posed, we consider the steady-state one-dimensional flow of an ideal gas mixture through a nozzle, without taking account of the viscosity, the thermal conductivity, or diffusion. It is described by the usual equations of gasdynamics

$$\rho u S = \text{const};\quad \rho u\,\frac{du}{dx} = -\frac{dp}{dx};\quad \frac{d}{dx}\left(H + \frac{u^2}{2}\right) = 0$$

$$(p = \rho RT/\mu;\ \mu = 1/\alpha;\ \alpha = \alpha_1 + \alpha_2;\ H = H_1\alpha_1 + H_2\alpha_2),$$

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where ρ, u are the density of the gas mixture and the velocity of the flow; p, T are the pressure and the temperature of the gas mixture; S(x) is the area of the surface of the velocities, equal to the velocity at the axis of the nozzle at the point x (in the case of small aperture angles of the nozzle, S coincides with the area of its transverse cross section); H is the enthalpy of unit mass of the gas mixture; μ is the molecular weight of the mixture; H_i is the enthalpy of one mole of the i-th component; and α_i is the number of moles of it in unit mass of the mixture (i takes on the values 1 and 2, which correspond to Br_2 and Br).

In the given case, the process of vibrational relaxation in a diatomic gas in the presence of dissociation and recombination is described by the equation [4, 5]

$$\frac{u}{(1-\gamma)^2}\frac{d\gamma}{dx}=\frac{1}{\tau}\frac{(\beta-\gamma)}{(1-\beta)(1-\gamma)}+\frac{(D/R\Theta-\varepsilon)}{\alpha_1}u\frac{d\alpha_1}{dx} \tag{1}$$

$$(\Theta=\hbar\omega_0/k;\quad \gamma=\exp(-\Theta/T_v);\quad \beta=\exp(-\Theta/T);\quad \varepsilon=\gamma/(1-\gamma)),$$

where Θ is the characteristic vibrational temperature; ω_0 is the frequency of the vibrations; $\hbar$ is the Planck constant; ε is the number of quanta of vibrational energy per one molecule; D is the dissociation energy; and k is the Boltzmann constant.

The equation has two terms in its right-hand side, the first of which describes relaxation with an unchanged number of molecules, while the second takes account of the change in the vibrational energy due to a change in the number of molecules with dissociation and combination.

The equations of chemical kinetics describing the reaction

$$Br_2+M\underset{k_r}{\overset{k_d}{\rightleftarrows}}Br+Br+M, \text{ where } M=Br_2,\quad Br,$$

have the form

$$u\frac{d\alpha_1}{dx}=-k_d\alpha_1+k_r\rho\alpha_2^2,\quad u\frac{d\alpha_2}{dx}=-2\left(-k_d\alpha_1+k_r\rho\alpha_2^2\right), \tag{2}$$

where $k_r = (k_{r1}\alpha_1 + k_{r2}\alpha_2)\rho$; k_{r1} and k_{r2} are the constants of the atomic recombination of bromine, when the third partner with respect to the collision is Br_2 or Br; k_d is the nonequilibrium rate constant of the dissociation.

The proposed macroscopic description of relaxation and chemical reaction under nonequilibrium conditions is possible if among the relaxation processes there is a process which assures a rapid exchange of energies between the lower states of the reacting molecule. Such exchange leads to a relatively rapid establishment of a quasi-steady-state distribution with respect to these states, characterized by the effective temperature T_v. The lack of equilibrium of the chemical reaction in this case is reflected in the fact that the rate constant k_d depends on the parameters T and T_v.

With $T_v = T$, $k_d(T)k_r = K_e$ is the equilibrium constant. With $T_v \neq T$, $k_d(T, T_v)$ retains the sense of the true rate constant and can be determined with satisfactory accuracy using a model proposed in [8].

In this model, a molecule is regarded as an anharmonic oscillator. The concentration of molecules is assumed as sufficient so that, due to the rapid exchange of vibrational quanta, a quasi-steady-state Treanor distribution [14] is established in the lower vibrational levels. Above some limiting level n*, the rate of translational—vibrational exchange becomes greater than the rate of vibrational—vibrational exchange, and a Boltzmann distribution is established with the temperature of the translational motion.

The rate constant of the dissociation is proportional to the population of the vibrational levels near the threshold of the dissociation; therefore [8],

$$k_d(T,T_v)=k_d(T)\frac{Z(T)}{Z(T_v)}\exp\left[\frac{n^*\hbar\omega_0}{k}\left(\frac{1}{T}-\frac{1}{T_v}\right)+2\varkappa n^*\left(2\frac{\varepsilon(T_v)}{\varepsilon(T)}-1\right)\right],$$

where Z is the vibrational statistical sum; $\varkappa$ is the constant of the anharmonicity. The limiting level n* is found from the condition of the equality of the rates of vibrational—vibrational and translational—translational exchanges

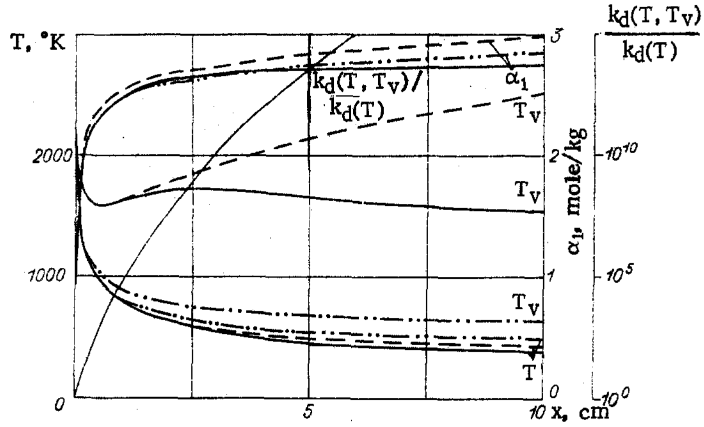

Fig. 1

$$\sum_j X_j Q^{j,j+1}_{n+1,n} = P^{(1)}_{n+1,n} + P^{(2)}_{n+1,n}\alpha_2/\alpha_1. \tag{3}$$

For calculating n*, the probability SSH-theory [15] is used.

The probability of a transition from the (n + 1)-th vibrational level to the n-th level, with a collision with the i-th particle, has the form

$$P^{(i)}_{n+1,n} = \frac{8}{z_0}(n+1)\Phi^2\Lambda^2\chi^{1/2}_{n,i}\exp\left(-3\chi_{n,i}\right),$$

$$\chi_{n,i} = \left(\frac{\pi^2\mu_i\omega_n^2}{2\alpha^2 kT}\right)^{1/3}, \quad \Phi^2 = \frac{\hbar\lambda^2 a^2}{2m\omega_0}, \tag{4}$$

$$\Lambda_i = \frac{2\pi\mu_i\omega_n}{a^2\hbar}, \quad \lambda^2 = \frac{1}{2}\frac{m_1^2+m_2^2}{(m_1+m_2)^2},$$

where $z_0 = 3$ is an orientation factor; m, μ_i are the reduced masses of the oscillator and the colliding particles; i takes on the values 1 and 2 for Br_2 and Br, respectively; and m_1 and m_2 are the masses of the oscillator. For Br_2, $m_1 = m_2$ and $\lambda^2 = 1/4$; $1/a \approx 2 \cdot 10^{-9}$ cm is the effective radius of the action of volumetric forces.

The probability of the transfer of a vibrational quantum by an oscillator in the (n + 1)-th state to an oscillator in the state j has the form

$$Q^{j,j+1}_{n+1,n} = \frac{8\mu_1 kT\Phi^4}{a^2\hbar}(n+1)(j+1)\operatorname{sech}(A\omega_{nj}),$$
$$A = (2/a)\sqrt{\mu_1/3kT}, \quad \omega_{nj} = \omega_n - \omega_j. \tag{5}$$

Formula (5) was obtained by approximate averaging with respect to the velocities of the colliding particles and is valid with the condition $A|\omega_{n,j}| < 1.5$. Calculations carried out in the present work have shown that the solutions of the equation for determining n* lie in the region of values of $\omega_{n,j}$ such that this condition is satisfied.

Substituting (4), (5) into (3), we obtain

$$\frac{3}{4}\frac{kT}{\hbar\omega_0}\sum_j (j+1)X_j\operatorname{sech}(A\omega_{nj}) = \Lambda_1^2\chi^{1/2}_{1,n}\exp\left(-3\chi_{1,n}\right) + \frac{\alpha_2}{\alpha_1}\Lambda_2^2\chi^{1/2}_{2,n}\exp\left(-3\chi_{2,n}\right). \tag{6}$$

Taking into consideration that the main contribution to the sum (6) is that of the region of small values of j, following [8], we set

$$X_j = \exp\left(-j\hbar\omega_0/kT_V\right)/Z(T_V), \quad \operatorname{sech}(A\omega_{nj}) = 4\exp\left(-2A|\omega_{nj}|\right).$$

Using these relationships, condition (6) can be transformed to the form

$$\frac{3kT}{\hbar\omega_0}\frac{\exp(-2A\omega_{0n})(1-\gamma)}{(1-\gamma)^2} = \Lambda_1^2\chi^{1/2}_{1,n}\exp\left(-3\chi_{1,n}\right) + \frac{\alpha_2}{\alpha_1}\Lambda_2^2\chi^{1/2}_{2,n}\exp\left(-3\chi_{2,n}\right). \tag{7}$$

From the solution of (7) we find n = n*.

With a numerical solution of the problem, the equations of gasdynamics were integrated by the Euler method with a recalculation [16] and the kinetic equations, by the Newton method [17]. This made it possible to start the calculation from the subsonic equilibrium region and to overcome the difficulties usually arising in problems of this kind, connected

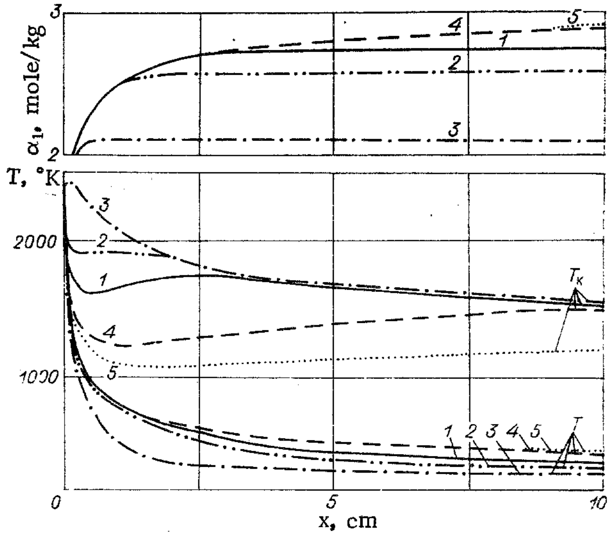

Fig. 2

with integration of the kinetic equations near equilibrium. The singular point, appearing near the critical cross section, was found by iterational matching of the mass flow rate and the joining of the branches of the solution at this point. In each step of the integration, the transcendental equation (7) was solved and $k_d(T, T_v)$ was calculated. All the calculations were made on a BÉSM-6 digital computer.

A study was made of the flow of partially dissociated bromium through a flat-wedge-shaped nozzle with total aperture angles of 15, 30, and 60°. The critical cross sections of these nozzles had the form of a slit with a height of 1.6 or 0.3 mm. The width of the nozzles was 76 mm in all cases. The contour of the subsonic section was part of a circle with a radius of 1 mm. The conditions at the inlet to the nozzle $T_0 = 3000$°K and $p_0 = 15$ atm were determined by the expediency of studying kinetic flows far from quasiequilibrium, as well as from quasifrozen conditions. In addition, the nonequilibrium flow must contain a sufficient concentration of molecules to assure a rapid exchange by vibrational quanta.

Data, summarized in the review [18], were used for the value of the time of the vibrational relaxation of Br_2. These values are approximated by the temperature dependence $p\tau_1 = 2.6 \cdot 10^{-9} \exp(40.6 \cdot T^{-1/3})$; $p\tau_1$, atm·sec.

Atomic bromine was assumed to be twice as effective as molecular. The relaxation time of the mixture was determined from the usual relationship

$$\frac{1}{\tau} = \frac{1}{\tau_1}\frac{\alpha_1}{\alpha_1+\alpha_2} + \frac{1}{\tau_2}\frac{\alpha_2}{\alpha_1+\alpha_2}.$$

The rate constants of the recombination k_{r1} and k_{r2} were borrowed from [19]. They are approximated by the temperature dependence

$$\log k_{r1}\,(\mathrm{cm}^6/(\mathrm{mole}^2\cdot\mathrm{sec})) = (16.89 \pm 0.85) + (3.01 \pm 0.28)\log(300/T),$$
$$\log k_{r2}\,(\mathrm{cm}^6/(\mathrm{mole}^2\cdot\mathrm{sec})) = (18.22 \pm 2.00) + (4.30 \pm 0.62)\log(300/T).$$

Calculations of the flow in nozzles having a height of the critical cross section of 1.6 mm showed that the flow is chemically nonequilibrium, but equilibrium with respect to the vibrational degrees of freedom is retained (the calculation was made up to 15 cm downstream from the critical cross section). This means that, under the given conditions, the processes of relaxation and recombination differ in their times and do not interact.

Another picture is observed with the consideration of the results of a calculation for a nozzle with a height of the critical cross section of 0.3 mm and an aperture angle of 60°. Figure 1 shows the distributions of the vibrational and translational temperatures, the concentrations of Br_2, and the ratios $k_d(T, T_v)/k_d(T)$ with the flow of partially dissociated bromine in such a nozzle. The solid lines denote a solution taking account of the mutual effect of chemical and vibrational kinetics and the dashed lines, a solution in which account was taken of the effect of the chemical kinetics on vibrational relaxation, but neglecting the inverse effect; the dashed-dot lines relate to a solution for the case where

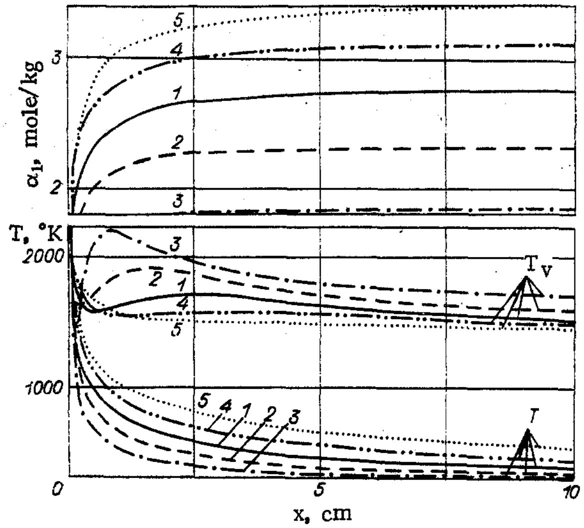

Fig. 3

the chemical and vibrational kinetics are not connected [in the relaxation equation (1) no account is taken of the change in the vibrational energy due to the reaction, and, in Eqs. (2), the rate constant of the dissociation $k_d(T)$ depends only on the translational temperature].

The qualitative explanation of the picture obtained consists in the following. Taking account of the mutual effect of the chemical and vibrational kinetics of the competition between the decrease in the vibrational energy due to deactivation and its rise due to recombination leads to a situation in which, even at a distance of 0.5 cm from the critical cross section, the decrease in the vibrational energy of the molecules ceases. Under these circumstances, there is a sharp increase in the ratio $k_d(T, T_v)/k_d(T)$ and a decrease in the rate of formation of Br_2. The value of n^* varies approximately from 14, at the moment of the start of the breakaway of T_v from T, to 43 at the end of the calculation. When the rate of recombination becomes equal to the rate of nonequilibrium dissociation, the chemical composition in the flow becomes almost constant. Under these circumstances, the influx of energy to the vibrational degree of freedom ceases, and T_v, attaining a maximum, starts to drop; its decrease is determined only by the process of vibrational deactivation. It must be emphasized that the constancy of the composition in the flow does not mean "freezing" of the chemical process, but is a consequence of an increase of the rate of dissociation under nonequilibrium conditions. This can be seen clearly from the solution in which no account was taken of the effect of the high vibrational temperature on the rate of decomposition (dashes). In this case, the kinetic region of the flow extends along the nozzle, and the influx of energy to the vibrational degree of freedom does not cease along its entire extension.

When the vibrational and chemical kinetics are not connected (dashed-dot line), there is no additional source of energy in the flow. The rate of its decrease is determined only by the process of vibrational relaxation, and the corresponding vibrational temperature almost does not differ from the temperature of the translational motion.

To clarify how the results obtained are affected by the rate constants of the recombination and the time of vibrational relaxation, calculations were made in which these quantities were varied.

Figure 2 shows the distribution of T, T_v, and α_1 along the nozzle, obtained taking account of vibrational–chemical interaction. The numbers 1-5 denote distributions where the time of vibrational relaxation was taken equal to τ, $\tau/0.66$, $\tau/0.3$, $\tau/2$, $\tau/3$, respectively. A slow run-off of the vibrational energy (large times of the vibrational relaxation) leads to a large breakaway of T_v from T and to a decrease in the rate of formation of Br_2 in the initial part of the flow.

An analogous connection between the processes of chemical and vibrational kinetics came out in calculations where the rate constant of the recombination was varied (Fig. 3, where the numbers 1-5 denote distributions of T, T_v, and α_1 along the nozzle, obtained with k_r, $k_r/3$, $k_r/10$, $k_r/0.33$, $k_r/0.1$, respectively). Large rate constants of the recombination lead

TABLE 1

m	n	τ, sec	τ_g, sec
1	1	$2,2 \cdot 10^{-5}$	$1,5 \cdot 10^{-5}$
1/0,66	1	$4,3 \cdot 10^{-6}$	$4 \cdot 10^{-6}$
1/0,3	1	$2,4 \cdot 10^{-7}$	$7,6 \cdot 10^{-7}$
1/2	1	$1,1 \cdot 10^{-4}$	$6,7 \cdot 10^{-5}$
1	1/3	$9,2 \cdot 10^{-6}$	$9,6 \cdot 10^{-6}$
1	1/10	$5,3 \cdot 10^{-6}$	$9,7 \cdot 10^{-6}$
1	1/0,33	$2,0 \cdot 10^{-5}$	$2,0 \cdot 10^{-5}$

to smaller breakaways of T_v from T and to a corresponding increase in the rate of formation of Br_2. This is connected with the fact that the parameters T and ρ depend to a considerable degree on the value of the recombination constant.

To clarify the conditions with which it is necessary to take account of the vibrational—recombination interaction in a supersonic flow, a comparison was made of the characteristic times of the flow of the gas τ_g ($\tau_g = [ud(\ln\rho)/dx]^{-1}$) and the times of vibrational relaxation τ. Table 1 gives these values for the point where the vibrational temperature passes through a local maximum, obtained in calculations with variation of τ and k_r. The values of m and n given in the table are factors of the variation with τ and k_r, respectively. As can be seen, in the region of a strong vibrational—recombination interaction, τ is close in value to τ_g. In fact, this condition, along with the requirement $\tau_r < \tau_g$, where by τ_r there is understood the characteristic time of the chemical reaction [20] ($\tau_r = [k_{r1}\rho^2/(\alpha_1 + \alpha_2)\mu_2^2\mu_1]^{-1}$), is a criterion of the need to take account of the virational—recombination interaction in an expanding flow.

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RESONANCE CO_2 ABSORPTION (10.6 μ) BEHIND A SHOCK FRONT

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Carbon dioxide is widely used in various molecular laser systems, and therefore there is interest in measurements of the coefficient of absorption of CO_2 at the frequency of the usually used laser transition (100, J) → (001, J + 1) at the wavelength of 10.6 μ in a wide range of temperatures and densities of the gas. For this purpose it is helpful to use the method of compression and heating of the gas being studied in shock waves [1, 2]. The dependence of the coefficient of absorption α of CO_2 at the 10.6-μ wavelength on the pressure of the gas at room temperature has been studied in sufficient detail at present [3, 4], although even in this region of variation of the parameters attempts at comparing the calculated values of α with experiment (at $p \geq 1$ atm) have revealed considerable discrepancies, connected primarily with the imperfection of the approximation of the shape of the lines of the vibrational–rotational spectrum of the CO_2 molecule by a Lorentzian contour.

It is interesting to trace the behavior of the coefficient of absorption at high gas temperatures, where the values of α can be especially large. The temperature dependence of α at $p \simeq 1$ atm in the temperature range of 300-420°K was determined in [5]. It was discovered that the measured values of the coefficient of absorption are considerably higher than the calculated values, and therefore to explain the dependence $\alpha(T)$ it was assumed in [5] that the cross section for collisional broadening of the line contour of CO_2 molecules varies as a function of the temperature in the form $\sigma_c \sim T^{-1/2}$. In [6] similar measurements were made up to temperatures of ~615°K. On the basis of the calculations of [7] the authors explained the high values of α which they obtained by the contribution of transitions of "mixed" states ($11^10 \to 01^11$, $12^00 \to 02^01$) without resorting to the temperature dependence of the broadening cross section. Measurements of α up to $T \sim 1600$°K were made in [8] using the shock-tube technique. In the calculations of the coefficient of absorption the authors of this report allowed for the contribution of "mixed" transitions and they also assumed a dependence $\sigma_c \sim T^{-1/2}$.

In the present work the temperature dependence was determined both for CO_2 and for a mixture of CO_2 and nitrogen in a wide range of temperatures (500-2100°K). Measurements of α were also made as a function of the pressure at high gas temperatures (1100 and 1600°K) behind both the incident and the reflected shock waves. The purpose of the tests described

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