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THE EFFECT OF MULTICOMPONENT DIFFUSION ON THE STABILITY OF LAMINAR FLAMES*

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The problem of the stability of the combustion of a three-component gas mixture (fuel, oxidiser and reaction products) is formulated and studied, taking into account the exact mutual diffusion between the components. Boundaries of two regions of thermodiffusive instability TDI-1 and TDI-2 for two mixtures with widely different diffusion properties are constructed, and a comparison is made with the binary theory. The effect of the initial composition of the mixture (a weak, stoichiometric or rich mixture) on the position of the boundaries of the monotonic and oscillatory instability is studied.

The problem of the thermodiffusive instability TDI of a laminar flame in case of a substantial deficiency of fuel was studied in /1-8/. The mechanism of the onset of TDI proposed in /1/ for a Lewis number $L>1$ ($L=D/\varkappa$; D, $\varkappa$ are the effective diffusion coefficient and thermal conductivity) connected with the distortion of the flame front was confirmed quantitatively in /2-5/. The perturbations increase monotonically in this case (region TDI-2, the terminology is that of /3, 4/). Another mechanism responsible for the appearance of TDI was suggested in /6/, and studied within the frame-work of the formulation of the problem in /2/, analytically and numerically in /7/. This instability is oscillatory and is realized when $L<1$ (TDI-1). In /8/**, (**See also, Aldushin A.P. and Kasparyan S.G. Thermodiffusive instability of a stationary combustion wave. Preprint, Chernogolovka, Otd-niye Inst. Khim. Fiziki, Akad. Nauk SSSR, 1978.) where the assumption used in /2-5, 7/ of the quasistationary character of the velocity of perturbed flame front is not used, satisfactory matching of the boundaries of the zones TDI-1 and TDI-2 with those obtained in /3-5, 7/ was achieved (the difference does not exceed 8%).

1. Let an irreversible chemical oxidation reaction take place in a gaseous mixture

$$\nu_1A_1+\nu_2A_2\rightarrow\nu_3A_3 \tag{1.1}$$

where ν_i are the stoichiometric coefficients and A_1, A_2, A_3 are the symbols of the fuel, oxidiser and reaction products respectively. We assume, as in /2-5/, that the chemical reaction front is infinitely thin, which is true in the case of a chemical reaction with a strong (exponential) dependence of the reaction rate on temperature, and a substantially weaker (power) dependence on the concentration of the components in the mixture. When the thickness of the reaction zone is taken into account, we find, according to /8/, (see also the reference in the footnote), the region TDI-2 narrows slightly. In accordance with /2-5/ we regard the chemical reaction front as a surface of a weak discontinuity in temperature and component concentration. In order to simplify the problem we shall neglect the diffusive energy transfer as compared with conductive transfer, which of course can be justified e.g. for a mixture in which the heat capacities of the components are nearly the same. We assume that the binary diffusion coefficient D_{ik} and the thermal conductivity $\varkappa$ are constant /2-5/ and correspond to the temperature of combustion.

The surface of the perturbed reaction zone is given in the form /2/

$$x=\xi(y,t)=\varepsilon\exp(iKy+\Omega t)$$

where ε represents the small amplitude of distortion of the front, K is the real wave number, Ω is the perturbation frequency and $\xi(y,t)$ is a small deviation of the chemical reaction front from its unperturbed position.

We assume that the mass combustion rate m, just as in /2-5/, is known from the theory of the normal combustion of gases /9/

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$$m \sim \exp\left(-\frac{E}{2RT_f}\right) \tag{1.2}$$

Here E is the energy of activation, R is the universal gas constant and T_f is the temperature of the perturbed chemical reaction front.

From (1.2) we see that the problem is studied in the quasistationary formulation and the dependence of the components on concentration is neglected /2-5/. Moreover, we assume that the functional dependence (1.2) of the quantity m on the temperature T_f, which holds for a binary mixture, also holds for a multicomponent gas mixture.

In accordance with what we have said, the process of quasistationary propagation of a combustion wave is described, in a coordinate system attached to perturbed reaction zone, by the following system of equations:

$$\begin{gathered}\frac{dT}{dt} = \varkappa \Delta T, \quad \rho \frac{dC_j}{dt} = -\left(\frac{\partial J_{jx}}{\partial x} + \frac{\partial J_{jy}}{\partial y}\right), \quad j = 1, 2 \\ \sum_{i=1}^{3} C_i = 1 \left(\frac{d}{dt} = \frac{\partial}{\partial t} + u \frac{\partial}{\partial x}, \ \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\end{gathered} \tag{1.3}$$

where C_i is the mass concentration of the i-th component, J_{ix}, J_{iy} are the projections of the diffusion flux of the i-th component on the coordinate axes, ρ is the density of the mixture and u is the steady velocity of the flame, while the indices 1, 2 and 3 corresponds to the fuel, oxidiser and reaction products respectively.

The diffusion fluxes appearing in Eqs.(1.3) are connected with the concentration of the components by the Stefan-Maxwell relations /5/

$$\begin{gathered}\sum_{k=1}^{3} \frac{X_i X_k}{\rho D_{ik}} \left(\frac{J_{kz}}{C_k} - \frac{J_{iz}}{C_i}\right) = \frac{\partial X_i}{\partial z} \\ \sum_{i=1}^{3} X_i = 1, \quad \sum_{i=1}^{3} J_{iz} = 0, \quad z = x, y\end{gathered} \tag{1.4}$$

where X_i is the relative molar concentration of the i-th component.

Using the relation between the molar and mass concentration /9/ we obtain, from (1.4),

$$\begin{gathered}J_{jz} = -\left(a_{j1} \frac{\partial C_1}{\partial z} + a_{j2} \frac{\partial C_2}{\partial z}\right); \quad z = x, y; \quad j = 1, 2 \\ a_{11} = n\,(b_1 b_2 - b_3 b_4), a_{12} = n\,(b_1 b_5 - b_3 b_6), a_{21} = n\,(b_4 b_7 - b_2 b_8) \\ a_{22} = n\,(b_6 b_7 - b_5 b_8), \ n = \rho D_{12}\,(b_1 b_7 - b_3 b_8)^{-1}\end{gathered} \tag{1.5}$$

The coefficients b_i are given by the formulas

$$\begin{gathered}b_1 = \pi_1 + C_1(\pi_2 - \pi_1), \quad b_2 = 1 + C_2(\pi_3 - 1) \\ b_3 = C_1(\pi_4 - \pi_3), \quad b_4 = C_2(1 - \pi_2) \\ b_5 = C_1(1 - \pi_3), \quad b_6 = 1 + C_1(\pi_2 - 1) \\ b_7 = \pi_4 + C_2(\pi_3 - \pi_4), \quad b_8 = C_2(\pi_1 - \pi_2) \\ \pi_1 = \frac{D_{12}}{D_{23}}, \quad \pi_2 = \frac{M_3}{M_1}, \quad \pi_3 = \frac{M_3}{M_2}, \quad \pi_4 = \frac{D_{12}}{D_{13}}\end{gathered}$$

Taking into account Eqs.(1.5), we can write the diffusion equations in the form

$$\rho dC_j/dt = a_{j1} \Delta C_1 + a_{j2} \Delta C_2, \quad j = 1, 2 \tag{1.6}$$

When writing (1.6), we assumed the coefficients a_{ji} to be constant in order to obtain linear equations. This assumption completely retains the main aspect of multicomponent diffusion, namely, the interpenetrating nature of the diffusion of the components of the mixture, and this is essentially analogous to the assumption of /2-5/ that the effective diffusion coefficient is constant.

We shall write the boundary conditions as follows:

$$\begin{gathered}x = -\infty, \quad T = T_0; \quad x = +\infty, \quad T = T_b; \quad x = \pm\infty, \\ \partial T / \partial x = 0 \\ x = -\infty, \ C_j = C_{j0}; \ x = +\infty, \ C_j = C_{jb}; \ x = \pm\infty,\end{gathered} \tag{1.7}$$

$$\partial C_j/\partial x = 0; \quad j = 1, 2 \tag{1.8}$$

where C_{i0} is the initial concentration of the i-th component, T_0 is the initial temperature, T_b is the adiabatic temperature of combustion and C_{ib} is the equilibrium concentration of the i-th component.

The relation connecting the equilibrium concentrations C_{1b} and C_{2b} is

$$C_{20} - C_{2b} = \frac{\nu_2}{\nu_1}\frac{M_2}{M_1}(C_{10} - C_{1b}) \tag{1.9}$$

and is obtained by integrating the one-dimensional non-linear stationary diffusion equations in x from $-\infty$ to $+\infty$, eleminating the source terms with the help of the stoichiometric relation (1.1) and taking into account the boundary conditions (1.8).

Depending on the composition of the mixture in question we must put, in the boundary conditions (1.8) and expression (1.9), $C_{1b} = 0$ for a weak mixture, $C_{2b} = 0$ for a rich mixture and $C_{1b} = C_{2b} = 0$ for a stoichiometric mixture.

We can obtain an expression for the adiabatic combustion temperature by the usual methods

$$T_b = T_0 + q\,(C_{10} - C_{1b})/c_p$$

where q is the thermal effect of the reaction and c_p is the specific heat capacity at constant pressure.

The boundary conditions at the perturbed front of a chemical reaction have the form

$$C_i\,|_{\xi-0} = C_i\,|_{\xi+0} = C_{ib}, \quad T\,|_{\xi-0} = T\,|_{\xi+0} \tag{1.10}$$

$$J_{1x}\,|_{\xi-0} - J_{1x}\,|_{\xi+0} = m\,(C_{10} - C_{1b}) \tag{1.11}$$

$$J_{2x}|_{\xi-0} - J_{2x}|_{\xi+0} = \frac{\nu_2}{\nu_1}\frac{M_2}{M_1} m\,(C_{10} - C_{1b}) \tag{1.12}$$

$$\lambda\left(\frac{\partial T}{\partial x}\bigg|_{\xi-0} - \frac{\partial T}{\partial x}\bigg|_{\xi+0}\right) = qm\,(C_{10} - C_{1b}) \tag{1.13}$$

Conditions (1.11)-(1.13) are obtained by integrating the equations of diffusion and energy with sources in x from $\xi - \delta/2$ to $\xi + \delta/2$, followed by making the thickness of the reaction zone δ tend to zero. The derivative along the normal to perturbed front differs from the derivative with respect to x by the quantity $O\,(\varepsilon^2)$, which is neglected in the linear analysis of the stability.

The system of Eqs.(1.6) has the following solution in the region $x > 0$:

$$C_i\,|_{\xi+0} = C_{ib}, \quad J_{ix}\,|_{\xi+0} = 0, \quad i = 1, 2 \tag{1.14}$$

Taking into account (1.14) we obtain, from (1.11) and (1.12).

$$J_{2x}|_{\xi-0} = \frac{\nu_2}{\nu_1}\frac{M_2}{M_1} J_{1x}|_{\xi-0} \tag{1.15}$$

and this enables us to utilize, in what follows, a single boundary condition

$$J_{1x}\,|_{\xi-0} = m\,(C_{10} - C_{1b}) \tag{1.16}$$

in which $C_{1b} = 0$ for a weak and a stoichiometric mixture, and the value of C_{1b} for the rich mixture is found from (1.9).

We note that relations of the type (1.15) can be obtained for any number of components in the mixture.

2. A stationary solution of the one-dimensional energy equation with boundary conditions (1.7) was obtained in /2/

$$T\,(x) = T_0 + (T_b - T_0)\exp\,(ux/\varkappa) \quad (x < 0)$$

$$T\,(x) = T_b \quad (x > 0)$$

The system of one-dimensional diffusion Eqs.(1.6), for boundary conditions (1.8), has the following stationary solution:

$$C_j\,(x) = C_{j0} + C_{j1}\exp\,(\lambda_1 x) + C_{j2}\exp\,(\lambda_2 x) \quad (x < 0) \tag{2.1}$$

$$C_j\,(x) = C_{jb} \quad (x > 0)$$

$$C_{1j} = (-1)^j\,\lambda_j E_{3-j}, \quad C_{2j} = (-1)^j\,\eta_j E_{3-j}, \quad \eta_j = (m - a_{11}\lambda_j)/a_{12}$$

$$E_j = \frac{(C_{1b} - C_{10})\,(m - a_{11}\lambda_j) - a_{12}\lambda_j\,(C_{2b} - C_{20})}{m\,(\lambda_2 - \lambda_1)}, \quad j = 1,2$$

$$\lambda_{1,2} = \frac{(a_{11}+a_{22}) \pm \sqrt{(a_{11}+a_{22})^2 - 4(a_{11}a_{22} - a_{12}a_{21})}}{2(a_{11}a_{22} - a_{12}a_{21})} m$$

We note that the characteristic equation of the system of one-dimensional stationary diffusion equations, under the conditions

$$\begin{gathered} a_{11}a_{22} - a_{12}a_{21} > 0 \\ (a_{11} + a_{22})^2 - 4(a_{11}a_{22} - a_{12}a_{21}) > 0 \end{gathered} \tag{2.2}$$

will have a double zero root in addition to the positive real roots λ_1 and λ_2. A particular solution of the form $S_1 + S_2x$ corresponds to the former root, where S_i are constants and $S_2 = 0$ by virtue of the boundary conditions (1.8).

Putting $a_{12} = 0$, in (2.1), we obtain the well-known stationary solution /2/.

In accordance with the method of small perturbations, we will seek the solution of the non-stationary energy and diffusion equations, taking into account expressions (1.14), in the form

$$\begin{aligned} & T = T(x) + T_1, \quad C_j = C_j(x) + C_j' \quad (x < 0) \\ & T = T_b + T_2, \quad C_j = C_{jb} \quad (x > 0) \\ & T_j = g_j(x) \exp(iKy + \Omega t), \quad C_j' = A_j(x) \exp(iKy + \Omega t), \\ & j = 1, 2 \end{aligned} \tag{2.3}$$

The functions g_j were found in /2/

$$g_j(x) = D_j(T_b - T_0) \exp(\alpha_j x), \quad \alpha_{1,2} = \frac{u}{2\varkappa} \pm \sqrt{\frac{u^2}{4\varkappa^2} + K^2 + \frac{\Omega}{\varkappa}}$$

where D_j are arbitrary constants.

The characteristic equation of the amplitude boundary value problem for the perturbations in concentration has, under condition (2.2), four real roots

$$\begin{gathered} \beta_{1,3} = \frac{\lambda_1}{2} \pm Q_1, \quad \beta_{2,4} = \frac{\lambda_2}{2} \pm Q_2, \\ Q_i = \sqrt{\frac{\lambda_i^2}{4} + K^2 + \frac{\Omega}{u}\lambda_i}, \quad i = 1, 2 \end{gathered} \tag{2.4}$$

We shall write the final expressions for the amplitudes $A_j(x)$ as follows:

$$\begin{aligned} & A_j(x) = D_3A_{j1} \exp(\beta_1 x) + D_4A_{j2} \exp(\beta_2 x) \\ & A_{1j} = a_{12}(K^2 - \beta_j^2), \quad A_{2j} = a_{12}(\beta_j^2 - K^2) - \rho\Omega - m\beta_j, \\ & j = 1, 2 \end{aligned} \tag{2.5}$$

Substituting the solutions (2.3)-(2.5) into the boundary conditions (1.10), (1.13) and (1.16) and linearizing, we arrive at a system of five linear homogeneous algebraic equations in the unknown constants $\varepsilon, D_1, \ldots, D_4$. The condition that non-trivial solutions of this system should exist, yields the characteristic equation for determining the dimensionless frequency of perturbations

$$\begin{aligned} & (z - a\sqrt{1+\omega})F + zL(1 - a\sqrt{1+\omega})G = 0 \\ & F = B_2\gamma_2\sqrt{1+d_2\omega} - B_1\gamma_1\sqrt{1+d_1\omega} + \gamma_1 - \gamma_2 \\ & \omega = \frac{4\Omega\varkappa}{a^2u^2}, \quad a = \sqrt{1+k^2}, \quad k = \frac{2\varkappa K}{u}, \quad L = \frac{a_{11}}{\rho\varkappa} \\ & z = \frac{E(T_b - T_0)}{2RT_b^2}, \quad G = (\mu_1 - \mu_2)\left(\frac{a_{22}}{a_{11}} - \frac{a_{12}}{a_{11}}\frac{a_{21}}{a_{11}}\right) \\ & B_i = \sqrt{1 + \frac{k^2L^2}{\mu_i^2}}, \quad \mu_i = \frac{\lambda_i a_{11}}{m}, \quad d_i = \frac{a^2L}{\mu_i B_i^2} \\ & \gamma_i = 1 - \mu_i\left(\frac{a_{22}}{a_{11}} - \frac{\nu_2}{\nu_1}\frac{M_2}{M_1}\frac{a_{12}}{a_{11}}\right), \quad i = 1, 2 \end{aligned} \tag{2.6}$$

When $a_{12} = 0$ Eq.(2.6) is identical with the dispersion equation given in /2-5/.

As we know (e.g. /4/), when k is fixed, a 1:1 correspondence exists in the L, z parameter plane between the boundaries of the TDI-1 and TDI-2 zones and the points for which Eq.(2.6) has the root $\omega = i\psi$ $(\psi^2 > 0)$ and $\omega = 0$ respectively.

We shall study these cases separately.

3. Let us consider the case of monotonic instability. When $\omega = 0$ and $k = 0$, Eq.(2.6) is satisfied identically, and this corresponds to the invariance of the plane flame front relative to the displacement of the coordinate system /4, 8/.

In the case of long-wave perturbations $(k \ll 1)$ we have the asymptotic solution

$$L = \frac{z}{z-1} L_*, \quad L_* = \left(1 + \frac{\nu_2}{\nu_1} \frac{M_2}{M_1} \frac{a_{12}}{a_{11}}\right)^{-1} \tag{3.1}$$

which, when $a_{12} = 0$, is identical with the known asymptotic expression /2-5, 8/. The curve described by Eq.(3.1) has a vertical asymptote $z = 1$, which is identical with the results of /2-5, 8/, and a horizontal asymptote $L = L_*$. Unlike in /2-5, 8/, the value of L_* can be greater or smaller than unity, depending on the sign of the coefficient a_{12} (the quantity a_{11} is always positive).

When k is arbitrary, the neutral curve $z = z(L, k)$ will be described by the following equation:

$$z = a \frac{B_2\gamma_2 - B_1\gamma_1 + \gamma_1 - \gamma_2}{L(1-a)G + B_2\gamma_2 - B_1\gamma_1 + \gamma_1 - \gamma_2} \tag{3.2}$$

The curve $L = L(z, k)$ given by Eq.(3.2) has a vertical asymptote

$$z = z_* = \frac{k\sqrt{1+k^2}}{1+k-\sqrt{1+k^2}}$$

which is the same as that in /3-5/, and a horizontal asymptote $L = L_0$ where L_0 is a real positive root of the equation obtained by equating to zero the ratio in (3.2). When $a_{12} = 0$, the root in question $(L_0 = 1)$ will correspond to the case discussed in /3-5/.

In order to be specific, we shall compute the coefficients a_{jk}, using the values of D_{ik} and M_i of the reactions

$$2CO + O_2 = 2CO_2, \quad 2H_2 + O_2 = 2H_2O$$

for which we have, respectively, $\pi_1 = 1.3292$, $\pi_2 = 1.571$, $\pi_3 = 1.375$, $\pi_4 = 1.3555$ and $\pi_1 = 3.6764$, $\pi_2 = 9$, $\pi_3 = 0.5625$, $\pi_4 = 0.8097$. The values of D_{ik} were taken from the monograph /9/.

Fig.1 shows the boundaries of the region of oxidation of CO for $k = 2$. The dot-dash line *1* is the curve obtained in /3-5/. The solid line *2* corresponds to a stoichiometric mixture $(C_{10} = 0.6363, C_{20} = 0.3637)$, and the dotted line *3* represents the boundary of the zone TDI-2 for a rich mixture $(C_{10} = 0.8, C_{20} = 0.2)$. The region TDI-2 is situated above the corresponding neutral curve. Analogous neutral curves for the combustion of hydrogen are shown in Fig.2 (*1* is the curve of /3-5/, *2* corresponds to a stoichiometric mixture $C_{10} = 0.111$, $C_{20} = 0.889$; and *3* corresponds to a rich mixture $C_{10} = 0.8$, $C_{20} = 0.2$). We see that as far as the qualitative aspects are concerned, the behaviour of the boundary of TDI-2 is the same as that of /3-5/ except for the quantity L_0, which was discussed above. The quantitative difference between the first and second reaction is sufficiently large, especially in the case of rich mixtures. Taking into account multicomponent diffusion, in the case of the hydrogen reaction, leads to a noticeable widening of the zone TDI-2 and the appearance of growing perturbations for Lewis numbers $L_0 < L < 1$. In the case of the first mixture, however, taking into account mutual diffusion stabilizes the flame front with respect to its distortions.

The influence of the wave number k on the position of the boundaries of the region TDI-2 in the case of the stoichiometric combustion of CO and H_2 is shown, respectively, in **Figs.**3 and 4 for $k = 0.1, 2, 3$ (curves *1-3* respectively). When k increases, the corresponding neutral curves shift to the right and depart, when $k > 4$, from the domain of real values of z in accordance with the results in /3-5/.

Calculations carried out for weak mixtures have shown that in this case we have satisfactory agreement with the binary theory /3-5/, especially in the case of the combusion of CO. We note that calculating the normal rate of combustion of this mixture taking multicomponent diffusion into account, gave an analogous result /10/.

4. Let us now inspect the oscillatory instability of the flame. Assuming that $\omega = i\psi$ $(\psi^2 > 0)$ in (2.6) and equating to zero the real and imaginary part of this equation, we obtain the parametric representation of the neutral curve in the form

$$f(L, z, k, \psi) = 0, \quad g(L, z, k, \psi) = 0$$

We will omit the functions f and g for brevity. Eliminating the parameter ψ we obtain, for fixed k, the equation of the neutral curve $L = L(z)$.

The results of the computations are shown in **Figs.**1-4 by curves *4-6* corresponding to the same values of the initial parameters as curves *1-3* respectively. The region TDI-1 appears under the corresponding neutral curve.

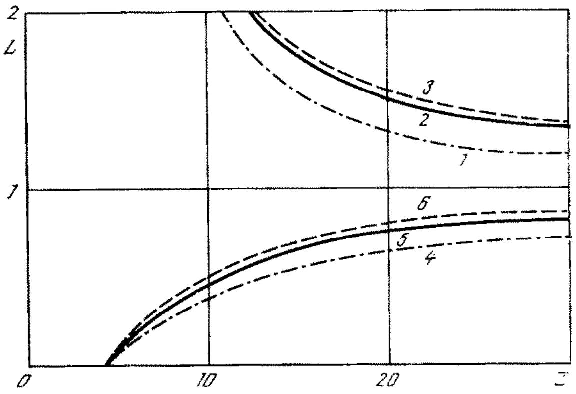

Fig.1

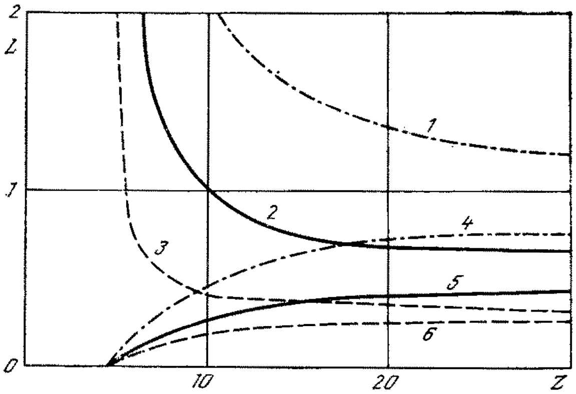

Fig.2

Analysing Fig.2 we find that when mutual intercomponent diffusion is accurately taken into account, we find that in the case of the combustion of H_2 this leads to substantial narrowing of the zone TDI-1.

In the case of the combustion of carbon monoxide we see from Fig.1 that, taking into account multicomponent diffusion causes an insignificant broadening of the region of the appearance of relaxation oscillations in the flame velocity. For both reactions that greatest discrepancy in the position of the boundaries of the region TDI-1 constructed taking into account multicomponent diffusion and in the binary diffusion approximation, is oberved for the case of a rich mixture. For a weak mixtures we have satisfactory agreement with the binary theorem, as well as for the boundaries of the zone TDI-2.

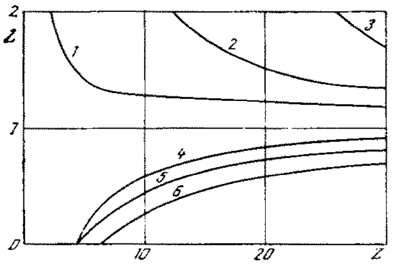

Fig.3

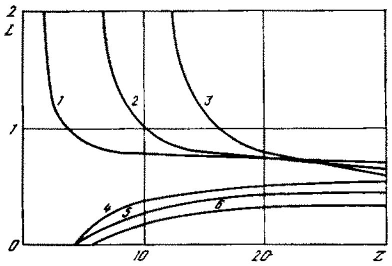

Fig.4

When *k* increases, the neutral curves shift to the right (see Fig.3 and 4) is accordance with the results obtained in /3-5/.

Thus the results of our investigations have shown that in case of mixtures which are almost stoichiometric and of rich mixtures, accurate inclusion of mutual diffusion may lead to an appreciable change in the general pattern of TDI in the case of large differences in the molecular weights of the components. In the case of weak mixtures, the different mobilities of the components are compensated by the deficiency in fuel.

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