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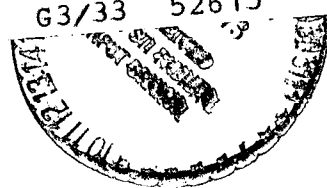
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THEORY OF COMBUSTION OF UNMIXED GASES

Ya. B. Zel'dovich

INTRODUCTION

The author will consider the chemical reaction of two substances (a fuel and oxygen) accompanying the formation of new substances which are the products of combustion and the liberation of heat. /1199*

The author will examine the stationary process with continuous supply of raw materials and removal of products. The special characteristic of the case in question consists in that the fuel and oxygen, or air, are fed separately, i.e., they are not mixed beforehand. Therefore, even in the case when the constant of the reaction rate of oxygen with the fuel is high, the intensity of combustion will not exceed a certain level which depends upon the rate of mixing of the fuel and the oxygen.

Note that the fact of combustion itself significantly changes the distribution of the concentrations relative to the distribution of concentrations when the same gases are mixed without combustion.

It has been known for a very long time, since the time of Faraday, if not earlier, that there is a basic qualitative concept which states that the surface of a flame separates a region in which there is oxygen and no fuel (oxidizing region) from the region in which there is no oxygen but there is fuel (reduction region).

Burke and Schumann [1] calculated the shape of the surface of a flame in a very specific case of combustion in parallel concentric laminar flows of fuel and oxygen or air. They failed to take into account the details of the phenomena that occur in the zone of the flame.

The recently published work by Shvab [2], which was performed in Leningrad back in 1940, is the most complete to date. This paper contains a detailed

*Numbers in the margin indicate pagination in the foreign text.

description of the turbulent tongue of flame, both as a case with feed of pure burning gas and a case of feed of a gas mixture with an insufficient quantity of air, which the author has not taken into account. Shvab finds the relationships between the fields of concentration of gas, oxygen and combustion products, temperature and the velocity field of the gas which the author did not take into account. A number of the results obtained by Shvab (in particular, the constancy of the concentration of combustion products and temperature) at the surface of the flame are given in this article for the sake of completeness.

The detailed discussion of the zone and the kinetics of the chemical reaction are essentially new (§ 5).

In the case of laminar combustion it is possible to go further on the basis of this investigation and to determine the limits of possible intensification of the combustion in view of the fact that at a high rate of feed of fuel and oxygen to the surface of the flame and insufficient rate of the chemical reaction results.

§ 1. General Equations

/1200

One should consider the region in which the gas is moving at a rate the density of the gas is ρ , the weight concentration of the component in which we are interested is a , the coefficient of diffusion is D , the thermal conductivity is λ , the temperature conductivity is the fraction $\frac{\lambda}{\rho a} = \chi$, the temperature is T , with all of the above values variable (depending upon the coordinates). The flow of component a is given by the formula

$$\vec{q}_a = \rho a \vec{w} - \rho D \text{grad } a. \quad (1)$$

The vector \vec{q}_a gives the direction of the flow and its value in grams per centimeter squared per second at a given point. The general equation of diffusion will have the form

$$\text{div } \vec{q}_a = L(a) = -\frac{\partial}{\partial t} (\rho a) + F_a. \quad (2)$$

On the left-hand side is the divisibility of the flow, i.e., the difference between the quantity of the substance brought in by the flow and the amount carried away by it, based on a unit volume; on the right side is the change in

amount of substance a per unit volume $\frac{\partial(\rho a)}{\partial t}$ and the amount of substance a which is formed per unit volume as a result of the chemical reaction.

$L(a)$ is an abbreviated designation for the differential operator

$$L(a) = \operatorname{div}(\rho a \vec{u}) - \operatorname{div}(\rho D \operatorname{grad} a). \quad (3)$$

In a stationary flow

$$\frac{\partial}{\partial t}(\rho a) = 0, \quad \frac{\partial \rho}{\partial t} = -\operatorname{div}(\rho \vec{u}) = 0, \quad (4)$$

$$L(a) = \rho \vec{u} \operatorname{grad} a - \operatorname{div}(\rho D \operatorname{grad} a). \quad (5)$$

In a stationary process

$$L(a) = F_a. \quad (6)$$

The equation of thermal conductivity has an analogous form¹ to that of the heat flux

$$\vec{q}_T = \rho T c \vec{u} - \lambda \operatorname{grad} T = \rho T c \vec{u} - \kappa c \rho \operatorname{grad} T, \quad (7)$$

where c is the specific thermal capacity of a unit mass which the author considers to be constant. From this it follows

$$\operatorname{div} \vec{q}_T = c L(T) = -c \frac{\partial}{\partial t}(\rho T) + Q, \quad (8)$$

where Q is the volume velocity of liberation of heat.

In a stationary process

$$L(T) = \frac{Q}{c}. \quad (9)$$

If the coefficients of diffusion of various substances are originally a , b and the combustion products are g , h and the coefficients of thermal conductivity are all equal to each other

$$D_a = D_b = D_g = D_h = \kappa = D, \quad (10)$$

the operators $L(a)$, $L(b)$, ..., $L(T)$ will also coincide in a formula of the form (6), written for various substances and in formula (9) for the temperature. /1201

In a chemical reaction involving combustion, the amount of raw materials entering into the reaction, the amount of combustion products formed and the

¹Here the author disregards the heat loss through radiation (see below).

amount of heat liberated come within certain strictly constant relationships. Using the F to designate the volume velocity of the reaction, the author uses it to express all of the values

$$F_a = -\frac{F}{\alpha}; \quad F_b = -\frac{F}{\beta}; \quad F_g = \frac{F}{\gamma}; \quad F_h = \frac{F}{\eta}; \quad \frac{Q}{c} = \frac{F}{\tau}, \quad (11)$$

by means of constant and positive coefficients α, \dots, τ . The signs in (11) indicate that a, b are used up and g, h and heat are given off in the reaction. The coefficients α, \dots, τ are placed in the denominator for convenience of further calculation. For example:

$$a = \text{CH}_4; \quad b = \text{O}_2; \quad g = \text{CO}_2; \quad h = \text{H}_2\text{O}; \quad c = 0.5 \left[\frac{\text{cal}}{\text{g} \cdot ^\circ\text{C}} \right];$$

the heat of the reaction is $192,000 \frac{\text{cal}}{\text{mole CH}_4}$.

Expressing F as the consumption of all substances entering into the reaction in $\text{g/cm}^3 \cdot \text{sec}$, one obtains

$$\alpha = 5; \quad \beta = 1.25; \quad \gamma = 1.82; \quad \eta = 2.22; \quad \tau = \frac{1}{4800^\circ} = 2.08 \cdot 10^{-4} (^\circ\text{C})^{-1}.$$

By means of these coefficients, all of the differential equations of diffusion of various substances at equations of thermal conductivity in a chemical reaction will take on a completely identical form, including L and F in all formulas

$$L(\alpha a) = -F; \quad L(\beta b) = -F; \quad L(\gamma g) = F; \quad L(\eta h) = F; \quad L(\tau T) = F. \quad (12)$$

However, it is still not possible to conclude from this that the fields of all of the values a, \dots, T of interest are the same, inasmuch as the field of each value depends not only on the differential equation which this value satisfies but also on boundary conditions.

A combustible gas is fed through a pipe (I); accordingly, within the pipe the following will be valid

$$\text{I: } a = a_0; \quad b = g = h = 0; \quad T = T_0, \quad (13)$$

where a_0 is the concentration of the fuel in the gas reaching the combustion point. This gas may be diluted, for example by nitrogen.

Through another tube (II) air is supplied

$$\text{II. } b = b_0; \quad a = g = h = 0; \quad T = T_0, \quad (14)$$

where b_0 is the concentration of oxygen in the air. During burning of the flame in the open atmosphere condition II refers not to a pipe containing air but to the concentrations in the atmosphere at an infinite distance from the flame.

On the surfaces of the pipes, on the surface of the combustion chamber, etc., the boundary conditions consist in the fact that the flow of any substance through the material surface is equal to zero, so that the component of the corresponding flow vector which is normal to the surface is equal to zero. These conditions are the same for all substances. The boundary conditions for the temperature will be the same as the boundary conditions for a, \dots, h , if one does not remove any heat from the flame, i.e., if only heat-insulated and not heat-radiating surfaces are employed or if the walls in general with temperature T_0 are located only where the gas temperature is equal to T_0 . /1202

One assumes that these conditions are satisfied so that the boundary conditions for all of the substances and the temperatures on the walls are the same.

§ 2. Analysis of Equations. Equation For the Surface of the Flame

Now let us proceed to an analysis of the equations. The principal difficulty in a direct solution of these equations consist in the fact that the rate of the reaction F is highly dependent upon the very values which the author is trying to find, a , b and T .

Calculating the first from the second one has

$$\left[L(\alpha a - \beta b) = L(p) = 0, \text{ where } p = \alpha a - \beta b, \right] \quad (15)$$

with boundary conditions

$$\left[\begin{array}{l} \text{I) } p = \alpha a_0, \quad \text{II) } p = -\beta b_0 \end{array} \right] \quad (16)$$

Hence, the difference between the concentrations of the fuel and oxygen, taken with corresponding stoichiometric coefficients, obeys the equation of diffusion in which the rate of the reaction F is not involved. This is the equation which was discussed by Burke and Schumann to determine the shape of the flame, proceeding on the basis of the fact that the fuel may be viewed as negative oxygen. If we are dealing not with a slow reaction but with combustion,

this means that the function F with simultaneous $a \neq 0$, $b \neq 0$ is very high. Inasmuch as the total amount of substance which is burned per unit time is limited by the amount of fuel which is supplied, the equation $F(a, b, T)$ with a , b and T given means that in reality the width of the reaction zone in which $a \neq 0$ and $b \neq 0$ is decreasing in the flame and the values a and b are decreasing in this zone.

At the limit, with an infinitely rapid reaction, a and b tend toward zero in the reaction zone, so that nowhere (except for an infinitely narrow zone) can a and b differ simultaneously from zero. Thus, in the case of combustion, finding the distribution p in space from the solution of the linear equation (15) with conditions (16), one can readily find the field a and the field b as well

$$\left. \begin{array}{l} \text{with } p > 0, \quad a = \frac{p}{\alpha}, \quad b = 0 \\ \text{with } p < 0, \quad a = 0, \quad b = -\frac{p}{\beta} \end{array} \right\} \quad (17)$$

The condition $p = 0$ also constitutes an equation for the surface of the flame. It is easy to see that the flows of substance a reaching the surface from one side and substance b reaching the same surface from the other side are immediately in a stoichiometric relationship. To prove this, note that on the surface of the flame $p = a = b = 0$, so that the convective parts of the flows $\left[\rho a \vec{u}, \rho b \vec{u}, \rho p \vec{u} \right]$ are also equal to zero. Therefore at the surface

$$\vec{q}_a = \rho D \text{grad } a = \frac{1}{\alpha} \rho D \text{grad } p; \quad \vec{q}_b = \rho D \text{grad } b = -\frac{1}{\beta} \rho D \text{grad } p \quad (17a)$$

and the value $\text{grad } p$ has no special characteristics for the surface of the flame, wherever $p = 0$, so that on this surface F is large when used in the equation for a and b but not in the equation for p .

The surface of the flame ($p = 0$) was found by Burke and Schumann through integration of (15) for the simplest case of concentric flows of fuel and air, moving at the same velocity; their conclusions are in satisfactory agreement with experimental data.

Let us turn now to the equations for the temperature and the combustion products and set up problems for expressing these values through a and b .

Considering the equation for one specific product of the reaction

$$L(\gamma g) = F$$

and comparing it with the equation for a and b

$$L(\alpha a) = -F, \quad L(\beta b) = -F,$$

one can see that it is possible to exclude F from the equation in different ways. However, if one wishes also to obtain the simplest boundary conditions, it will be necessary to select a new variable in a completely determined fashion

$$r = \frac{a}{a_0} + \frac{b}{b_0} + \gamma g \left(\frac{1}{\alpha a_0} + \frac{1}{\beta b_0} \right)^2 \quad (18)$$

it is easy to see that under these conditions

$$L(r) = 0, \quad \text{I) } r = 1, \quad \text{II) } r = 1. \quad (19)$$

§ 3. Distribution of Reaction Products and Temperature

It is clear from equation (18) and conditions (19) that in the entire region r satisfies the equation of diffusion and convective transfer without sources or flows (inasmuch as the first part of the equation is equal to zero). Then r is selected so that $r = 1$ in all flows which enter the region in question both in a flow of gas (I) and in a flow of air or in an atmosphere (II). It is clear that if a certain substance (r) is contained in a certain concentration which is the same in two mixing flows, precisely the same constant concentration of this substance will be found all through the entire mixing area. This is expressed mathematically as follows: $r \equiv 1$ is then the solution of (18) — (19). From this one can find the expression g through a and b

$$g = \frac{\alpha \beta}{\gamma} \frac{a_0 b_0 - a b_0 - a_0 b}{\alpha a_0 + \beta b_0}, \quad (20)$$

and completely analogously for h or T obtained by substituting γ for η or τ .

Thus, the problem which the author set himself has been solved.

²Any combination $z = n(\gamma g + m\alpha a) + (1 - m)\beta b$, where n and m are any constants gives $L = n[F - mF - (1 - m)F] \equiv 0$. However, in order for z to have the same values with $a = a_0, b = 0$ and $a = 0, b = b_0$, it is necessary for $n m \alpha a_0 = n(1 - m)\beta b_0 = 1$, which gives the expression r written above [formula (18)].

In the case of rapid combustion, when (18) is satisfied, one obtains at the front ($p = 0$, $a = b = 0$)

$$g_{00} = \frac{\alpha\beta}{\gamma} \frac{a_0 b_0}{\alpha a_0 + \beta b_0} \quad (21)$$

This result has a very simple and clear physical significance: the values a_0 and b_0 characterize the concentrations of active substances in the burning gases. In order to obtain the stoichiometric mixture in which 1 gram would be subject to reaction, it is necessary to take $\frac{1}{\alpha}$ grams of substance a and $\frac{1}{\beta}$ of substance b; this will give us $\frac{1}{\gamma}$ of substance g. Taking into account the fact that substances a and b in the original gases are dilute, it is necessary to take $\frac{1}{\alpha a_0}$ of one gas and $\frac{1}{\beta b_0}$ of the other. After combustion, the amount $\frac{1}{\gamma}$ of substance g will be contained in the total amount of combustion products which is equal to

$$\frac{1}{\alpha a_0} + \frac{1}{\beta b_0},$$

which gives us a concentration g_{00} which satisfied formula (21).

Hence, in the case of rapid combustion of unmixed gases the reaction zone will contain precisely the same concentration of combustion products as if one had mixed burning gases in a stoichiometric ratio and had performed a chemical reaction involving combustion without any diffusion volume.

In precisely the same way, in the absence of heat loss through radiation and cooling of the surfaces in the flame, and with equality of the coefficient of temperature conductivity and diffusion, it can be shown that the temperature in the combustion zone of the diffusion flame $T_{00} = \frac{\alpha\beta}{\gamma} \frac{a_0 b_0}{\alpha a_0 + \beta b_0}$ is also equal to the temperature of combustion at a constant pressure of the stoichiometric mixture of the gases in question.

§ 4. Comparison of the Temperature of the Flame With Experiment

The conclusion which was reached earlier, namely that the temperature of the unmixed flame is equal to the temperature of combustion of a stoichiometric mixture is contradictory with respect to experience: it is well known from daily laboratory experience that in the combustion of a given amount of illuminating gas in a Bunsen burner with the openings used for air intake at the

bottom closed, the flame temperature is lower than when the same gas is burning with the openings open, so that a prepared mixture of gas and air reaches the flame.³

However, this contradiction with respect to experience is explained not by an error in the calculations but by the fact that the condition for usability of the calculation was not fulfilled earlier; in reality, in the thermal balance of a laboratory burner it is impossible to disregard the amount of heat which is given off by the form of radiation.

With an equal amount of burning gas, i.e., with equal liberation of heat, without feeding air, the size of the flame is much larger than when air is fed, so that the radiant surface is larger and the liberation of chemical energy per unit surface is less. In addition, the brightness of the flame when air is not supplied is greater due to the appearance of tiny particles of carbon in it which come from the decomposition of the hydrocarbons in the fuel; when air is supplied, the carbon (lamp black) disappears. The presence of the soot in the flame of unmixed gas is also quite natural; let us examine the point A in the reducing area (Figure 1), i.e., within the surface of the flame. Let us say that this point which is located near the surface of the flame; in this instance, the temperature at point A will be high, the gas will already have been diluted strongly by the products of combustion and nitrogen, but there will be no oxygen in it. Combustion in the absence of oxygen will also lead to the formation of soot.

In the flame of a mixture of a gas with air, the gas is also heated ahead of the flame front, but this heating takes place in the presence of oxygen, and those accumulations of hydrocarbon molecules which could become starting points for the formation of soot particles, are immediately oxidized. As a result, the thermal radiation of the flame of the mixture is much less, while the temperature is much higher than in a flame of an unmixed gas, although the

³Here and below, it is assumed that the stoichiometric amount of air is drawn in. It is possible to consider (although one shall not do so here) the case of an insufficient quantity, when two "cones" of flame are formed, the inner (compressing mixture) and outer (burning in the surrounding air).

initial "ideal" theoretical value of the temperature of combustion in both cases is the same. As has been already pointed out, this theoretical value constitutes the temperature which must be reached in combustion in the absence of losses through radiation and side reactions, but with complete consideration of the conductive and convective heat exchange of the flame with the gas and air.

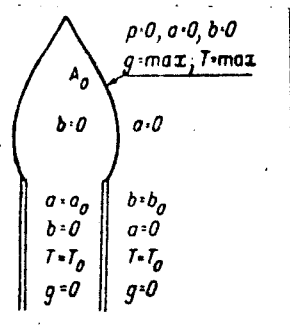


Figure 1.

Consideration of the conductive and convective heat exchange is theoretically necessary, inasmuch as this heat exchange is unavoidably bound up with the processes which feed the fuel and oxygen to the flame. Regardless of the ratio between the present and supplied amounts of air and gas, in the case when they are fed separately the flame is always located in the same

position, such that the fuel and oxygen reach the surface in a stoichiometric ratio. When the coefficients of diffusion and temperature conductivity are equal (especially in a turbulent motion, which ensures such equality), the concentration of combustion products and temperature in the flame correspond precisely to the combustion of a stoichiometric mixture (with equal losses to radiation), as the result of this calculation.

§ 5. Combustion Limit of Unmixed Gases

The methods described above makes it possible to calculate the position of a surface of a flame when any quantity of gas and air is fed at a caloric content of the gas which can be as small as desired; this calculation is based on the assumption of a high rate of chemical reaction at the surface of the flame (and at combustion temperature), which leads to a narrow thickness of the zone in which the chemical reaction takes place and to the possibility of viewing the flame as a geometric surface.

It is obvious that when the rate of the chemical reaction is insufficient one can expect deviations from this picture. By analogy with other phenomena of combustion and explosion, one can expect that a decrease in the rate of

reaction with all other conditions equal will cause first of all a slight quantitative change -- expansion of the reaction zone and then after achieving a certain critical value, damping of the flame; combustion becomes impossible and ceases, and instead of combustion there will be mixing of cold gas and air without any reaction. In the following the author will attempt to discuss the critical conditions for damping in a simple schematic case.

In 1940 the author discussed [3] the conditions for the possibility of combustion (propagation of a flame) in a prepared mixture of gases. In this case, the limits depended upon a drop in temperature of combustion due to heat loss by the side walls of the tube and heat loss through radiation. The decrease in the temperature of combustion in turn led to a drop in the rate of flame propagation, i.e., to a decrease in the heat emission per second. With a decrease in the speed of the flame the relative thermal losses increase, etc. Therefore, the critical condition for the possibility of combustion of a prepared mixture can be written so that the decrease in the combustion temperature from the action of thermal losses must not exceed a certain low limit (RT_c^2/A where A is the heat of activation of the combustion reaction and T_c is the combustion temperature). /1206

However, this theory of the effect of external experimental losses cannot be applied to combustion of unmixed gases. As a matter of fact, the decrease in the combustion temperature in this case does not lead to a change in the quantity of gas which is burned per unit surface of the flame, inasmuch as the rate of combustion is governed here exclusively by the rate of diffusion feed of oxygen and fuel to the surface of the flame, and not by the rate of propagation (which depends upon the rate of the reaction), as was the case for the prepared mixture.

The combustion limit of unmixed gases is set by the decrease in temperature which depends upon the finite velocity of the chemical reaction.

Let us examine the distribution of the concentrations and the temperature in the zone of the reaction. If the rate of the reaction were infinite, the distribution would be given by Figure 2. The dashed line shows the position of the zone in which $a = 0$, $b = 0$. Using it as the origin for calculating the

coordinate x , one directs the x -axis perpendicular to the surface of the flame. If the total amount of material which reacts per unit surface in a unit time is represented by M , the diffusion flows a and b will be equal (respectively)⁴

$$-\rho D \frac{\partial a}{\partial x} = \frac{1}{\alpha} \frac{M}{1 + \frac{1}{\beta}} = \frac{\beta}{\alpha + \beta} M; \quad \rho D \frac{\partial b}{\partial x} = \frac{\alpha}{\alpha + \beta} M, \quad (22)$$

so that the distribution of the reacting components near the zone is given by formulas

$$\left. \begin{aligned} x < 0, \quad a &= \frac{M}{\rho D} \frac{\beta}{\alpha + \beta} (-x), \quad b = 0 \\ x > 0, \quad a &= 0, \quad b = \frac{M}{\rho D} \frac{\alpha}{\alpha + \beta} x \end{aligned} \right\} \quad (23)$$

By means of formula (20) and (21), rewritten for T , one obtains the corresponding distribution of temperature. In the general case

$$T = T_{\infty} \left(1 - \frac{a}{a_0} - \frac{b}{b_0} \right), \text{ where } T_{\infty} = \frac{\alpha\beta}{\tau} \frac{a_0 b_0}{\alpha a_0 + \beta b_0}. \quad (24)^5$$

Substituting expression a and b in (23), one has

$$\left. \begin{aligned} T &= T_{\infty} \left(1 - \frac{1}{a_0} \frac{M}{\rho D} \frac{\beta}{\alpha + \beta} (-x) \right), \quad x < 0 \\ T &= T_{\infty} \left(1 - \frac{1}{b_0} \frac{M}{\rho D} \frac{\alpha}{\alpha + \beta} x \right), \quad x > 0 \end{aligned} \right\} \quad (25)$$

The corresponding curve is also shown in Figure 2.

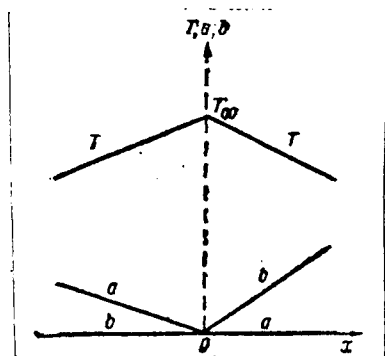


Figure 2.

How do the curves change when the reaction is not instantaneous?

It is obvious that with an equal amount of substance burning per unit surface M , the concentration gradient and the entire distribution of a and b does not change far from the zone. However, now curves a and b cannot undergo sharp curvature (corresponding to an

⁴Inasmuch as the author is interested in the vicinity of the zone of the reaction in these formulas and below ρ and D will be taken at a temperature in the reaction zone which is rounded off to T_{00} .

⁵More exactly, $T_{00} = \frac{\alpha\beta}{\tau} \frac{a_0 b_0}{\alpha a_0 + \beta b_0} + T_0$. In the following, one shall consider the initial temperature T_0 to be small in comparison with the combustion temperature and will disregard T_0 everywhere.

instantaneous reaction) at the origin of the coordinates, as in Figure 2. They will cross, as shown in Figure 3, asymptotically approaching zero in the region occupied by the second component. The dashed line in this curve shows the distribution for an instantaneous reaction.

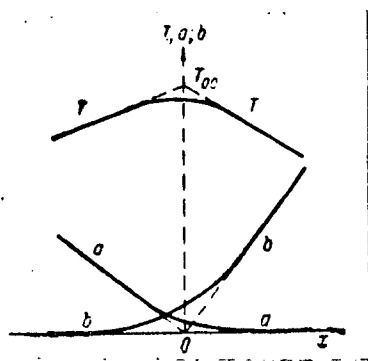


Figure 3.

In order to be able to determine precisely the curves for the distribution of concentration in Figure 3, it would be necessary to integrate the equations for the diffusion of the form (12), substituting a specific expression for the rate of the chemical reaction, for example

$$F = abKe^{-\frac{E}{RT}}. \quad (26)$$

Inasmuch as F depends on at least 3 values a , b and T , it is necessary to view the system of 3 equations of the second order. However, thanks to the concepts developed above, one can find first of all p , which links a and b , and then express T through a and b and thereby reduce the problem to a single equation of the second order, for example for a , in which F will be expressed by a and a known function $p(x)$.

However, for our purposes such an approach is too complicated, and the conclusions in which the author is interested in will be obtained (admittedly with an accuracy up to numerical factors which the author does not know) by methods of analysis of dimensionality and the theory of similarity.

Let us introduce the effective values -- the width of zone x_1 , the temperature in the zone of the reaction T_1 , concentrations a_1 and b_1 . The total amount of substance which reacts in the entire zone is expressed by these effective values

$$M = a_1 b_1 K x_1 e^{-\frac{E}{RT_1}}, \quad (27)$$

Let us express all of the values on the right-hand side by x_1 . In order to link them, the author notes that one is given a relationship a and b upon x at distances from the zone which are large in comparison with the wide zone of

the chemical reaction x_1 , and this relationship is linear, i.e., it is characterized by certain values set externally, by gradients $\frac{\partial a}{\partial x}$, $\frac{\partial b}{\partial x}$ or relationships $\frac{a}{x}$ or $\frac{b}{x}$ [see formulas (23)⁶]. It is clear from the dimensionality that relationship a_1 , b_1 and x_1 must be given by similar formulas

$$a_1 = \frac{M}{\rho D} \frac{\beta}{\alpha + \beta} x_1, \quad b_1 = \frac{M}{\rho D} \frac{\alpha}{\alpha + \beta} x_1. \quad (28)$$

The relationship of T_1 with a_1 and b_1 as given by formula (24) from which /1208 by substituting (28) we can have

$$T_1 = T_{00}(1 - \varphi M x_1), \text{ where } \varphi = \frac{1}{\rho D} \frac{\frac{\beta}{a_0} + \frac{\alpha}{b_0}}{\alpha + \beta} = \frac{\alpha \beta}{\alpha + \beta} \frac{1}{\rho D T_{00}}. \quad (29)$$

Substituting in (27), one finally obtains the equation which links M and x_1

$$M = \frac{\alpha \beta}{(\alpha + \beta)^2} \frac{M^2}{(\rho D)^2} K x_1^3 \exp\left(-\frac{E}{RT_{00}(1 - \varphi M x_1)}\right) = \\ = \frac{\alpha \beta}{(\alpha + \beta)^2} \frac{M^2}{(\rho D)^2} K e^{-\frac{E}{RT_{00}}} x_1^3 e^{-\frac{\varphi M x_1 E}{RT_{00}}}. \quad (30)$$

In the second line of (30) the author expanded the value in the exponent in a series using the Frank-Kamenetskiy method [4]. The value on the right-hand side, depending upon x_1 , has the form $x^3 e^{-mx}$ and consequently passes through a maximum at a certain critical value $x_1 = x_{cr}$.

$$x_{kp.} = \frac{3RT_{00}}{\varphi M E}, \quad (31)$$

$$M_{kp.}^2 = \frac{\alpha \beta}{(\alpha + \beta)^2} \left(\frac{RT_{00}}{E}\right)^3 \rho D \left(\frac{\alpha + \beta}{\frac{\beta}{a_0} + \frac{\alpha}{b_0}}\right)^3 K e^{-\frac{E}{RT_{00}}}. \quad (32)$$

The significance of the maximum M as a function of x_1 consists in the fact that when x_1 is small the covered areas a and b are small and the reaction zone is narrow so that the concentrations of reacting substances in it are small.

⁶The ratios $\frac{a}{x}$ and $\frac{b}{x}$ change slightly only at distances which are greater than x_1 , but less than the size of the flame. At distances which are comparable to the dimensions of the flame it is not permissible to disregard the convective terms in the diffusion equation.

When x_1 is small and the given temperature is T_{00} , the temperature T will differ slightly from T_{00} , $M \sim x_1^3$, $a_1 \sim b_1 \sim x_1 \sim \sqrt[3]{M}$, in accordance with the known results for flames which show good diffusion in a high vacuum [5]. When x_1 is large, the concentrations of reacting substances are high, so that the temperature decreases leading to a drop in the total amount of reacting substance.

By intensifying the combustion, increasing the feed of reacting substances a and b to the zone, one simultaneously intensify the cooling of the reaction zone. Until the rate of the reaction is sufficiently high, x_1 is less than maximum, the temperature practically does not decrease. However, at a certain critical value M_{cr} [formula (32)] a decrease occurs in the temperature of the zone which leads to a further decrease in the rate of the reaction and a further drop in the temperature; combustion is interrupted, and instead of combustion there is a mixing of cold gases. The maximum possible temperature drop prior to separation is the fraction $\frac{3RT_{00}^2}{E}$. Note the curious similarity of expression M_{cr} (32) with the expression for the amount of substance burnt per unit time in the flame propagating according to a previously prepared stoichiometric mixture of combustible gas and air, whose combustion in unmixed forms was discussed above.

For the propagation of a flame in a stoichiometric mixture, according to the work of Frank-Kamenetskiy and the author [6], one obtains in the designations employed here

$$M_{cr} = \frac{(\alpha\beta)^2}{\left(\frac{\beta}{a_0} + \frac{\alpha}{b_0}\right)^3} \left(\frac{RT_{00}}{E}\right)^3 6\rho DK e^{-\frac{E}{RT_{00}}}, \quad (33)$$

which differs from (32) only in the factor $\frac{6\alpha\beta}{\alpha + \beta}$ which does not have a numerator inasmuch as when (32) is derived the author discarded the numerical factors. This coincidence is very interesting from the theoretical standpoint inasmuch as it shows that the maximum intensity of combustion of the mixture even of unmixed gases, if the mixing is sufficiently intensified, is of the same order.

In the theory of combustion of an explosive mixture, the author has shown that the chemical reaction proceeds in a zone in which the concentration of the reacting gas (that which is inadequate in the mixture or of both in the case of a stoichiometric mixture) is very small -- on the order of $\frac{RT_{00}}{E}$ of the initial

concentration. As the calculations above indicate, in the combustion of unmixed gases the concentrations of both reacting substances (fuel and oxygen) in the reaction zone is very small. These concentrations depend upon the intensity of combustion: in contrast to an explosive mixture, in which there is a characteristic value for the intensity of combustion (speed of the flame), the intensity of combustion of a flame of unmixed gases M depends upon the external conditions. However, at the maximum possible M , at the point of separation of the flame, the concentrations in the reaction zone do not exceed the concentrations in the stoichiometric mixture in terms of the order of magnitude of the amount $\frac{RT_{00}}{E}$.

The limit found above to the intensity of combustion of unmixed gases explains at least qualitatively the fact that when a rapid stream emerges from a tube the flame breaks down completely some distance from the outlet cross-section of the tube, so that at the outlet, where mixing of the reacting components is most intensive, the flame separates. Previously the author did not take into account the thermal losses due to radiation. When they are taken into account, for example, in formulas (29-32) instead of the theoretical value T_{00} which was calculated on the basis of thermal capacity, one must use the maximum possible temperature for an instantaneous reaction, but with consideration of radiation -- the temperature T_{00}' . This T_{00}' is less than T_{00} even in the infinitely thin zone where $x_1 \rightarrow 0$, due to the radiation of heated gases on the left and right of the reaction zone. The temperature T_{00}' decreases as M decreases, so that at small M less heat is given out and the radiant zone is wider. When the radiation is taken into account, when M decreases due to the decrease in T_{00}' M_{cr} will also decrease according to (32) and with very small M a second, lower limit M will arise, with separation of the flame at too low an intensity of combustion. Finally, with low caloric content of the gas, the upper and lower limits of M may coincide, and combustion will become completely impossible. Qualitatively speaking, the picture is similar to the simpler case of exothermal reaction in a flow with consideration of the heat loss considered by Zysin and the author [7].

The author will not take up in this paper the practically significant but more complex problem of the limit of intensification of turbulent combustion

of unmixed gases: the complexity of this problem has to do with the fact that in the presence of turbulence we cannot correctly link the average velocity of the reaction with the average temperature. Apparently it will be necessary to determine experimentally the limits of possible combustion conditions.

Finally, it should be pointed out that by means of equations (12) with a concrete form of function F [for example (26)] it is also possible to solve the very interesting problem of the diffusion flashback of the fuel through the zone of the flame; as was shown in Figure 3, the concentration of the mutually penetrating reacting substances during the transition through the reaction zone decreases sharply but does not return to zero. Inasmuch as the temperature and the rate of the reaction also drop on both sides of the reaction zone, the concentration of the fuel which has already reached a certain distance from the flame in the oxidizing zone will not change further. However our approximate method will not do for solving this problem; it is necessary to solve equations in the manner described in the text after formula (26). /1210

Conclusions

The distribution of the concentration products and temperature is discussed in the combustion of mixed gases. It is shown that in the simplest conditions these concentrations and temperature at the surface of a flame are the same as in the combustion of previously prepared stoichiometric mixtures of the same gases.

A possible limit was found to the intensification of combustion of unmixed gases, depending upon the limiting rate of the chemical reaction. In order of magnitude, this limit is close to the rate of combustion of stoichiometric mixtures.

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