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**Propagation Speed of Stretched Premixed Flames
with Multi-step Reaction Mechanisms**

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Ausbreitungsgeschwindigkeit gestreckter Vormischflammen mit mehrstufigem Reaktionsmechanismus

Zusammenfassung

Es wird die Ausbreitungsgeschwindigkeit einer dünnen Vormischflamme betrachtet, die durch ein großskaliges Strömungsfeld gestört wird. Im Gegensatz zu vorausgegangenen Veröffentlichungen wird die hier dargestellte asymptotische Analyse für einen realistischeren mehrstufigen Reaktionsmechanismus ausgeführt. Eine Eigenschaft des vorgestellten Ansatzes ist die relativ einfache asymptotische Methode, die auf einem verallgemeinerten krummlinigen Koordinatensystem, das fest mit der Flamme verknüpft ist, beruht. Die erste Reaktion besitzt eine hohe Aktivierungsenergie und erzeugt Radikale, aber ihre Wärmefreisetzung ist vernachlässigbar klein, während die anderen Reaktionen den Verbrennungsprozeß beenden und dabei Wärme freisetzen. Die hier betrachtete Flamme besitzt zwei ausgezeichnete Zonen: Die Aktivierungszone und die Wärmefreisetzungszone. Es wird gezeigt, dass die Ausbreitungsgeschwindigkeit der Flamme von der in diesem Bericht eingeführten effektiven Zeldovich Zahl Z_f und der Flammstreckung abhängt. Im Gegensatz zur konventionellen Zeldovich-Zahl ist die effektive Zeldovich-Zahl nicht unmittelbar mit den Aktivierungsenergien der einzelnen Reaktionen verknüpft. Die exponentielle Abhängigkeit der Flammgeschwindigkeit von der Flammstreckung, die bei Einschrittmeechanismen auftritt, wird durch eine lineare Abhängigkeit abgelöst. Im Falle einer zweistufigen Reaktion, der einfachste Fall, der eine Unterscheidung einer Aktivierungs- und einer Wärmefreisetzungszone erlaubt, kann die effektive Zeldovich-Zahl analytisch ermittelt werden. Im Falle realistischer Reaktionen wird die effektive Zeldovich-Zahl aus Experimenten ermittelt.

Abstract

The propagation speed of a thin premixed flame disturbed by a fluid flow of a larger scale is considered. Unlike in preceding publications, the presented asymptotic analysis is performed for a more realistic multi-step reaction mechanism. Another feature of the present approach is represented by the relatively simple asymptotic technique based on the adaptive generalised curvilinear system of coordinates attached to the flame. The first reaction with high activation energy generates radicals but its heat effect is negligible while the other reactions complete the combustion process and release heat. The flame considered here has two distinct reaction zones: the activation zone and the heat release zone. The flame propagation speed is shown to depend on the effective Zeldovich number Z_f , which is introduced in the paper, and the flame stretch. Unlike the conventional Zeldovich number, the effective Zeldovich number is not directly linked to the activation energies of the reactions. The exponential dependence of the flame speed on the flame stretch, known for a single step reaction, appears to be replaced by a linear dependence. For a two-step reaction, which is the most simple reaction mechanism allowing for the distinct activation and heat release zones, the effective Zeldovich number can be determined analytically. For realistic reactions, the effective Zeldovich number is determined from experiments.

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1 Introduction

In premixed combustion, a mixture of fuel and oxidizer reacts in a flame to form the burned products. The self-propagation mechanism of premixed flames is determined by a sophisticated balance of the chemical reaction, mass diffusion and convective transport of heat and species. In non-uniform time-dependent flows, strong interactions of the flow with this balance result in a non-constant local flame-propagation speed. Since the flame is, typically, thin in comparison with the characteristic scales of the fluid flow [1, 2], the most simple approach to premixed combustion is based on viewing the flame as a surface of discontinuity which separates the dense cold mixture from the light hot products. The flame behaves as a gas-dynamic discontinuity. A first model of this kind was proposed by Darrieus [3] and independently by Landau [4]. In the Darrieus-Landau model, the flame is described as two incompressible fluids of different densities which are separated by the flame front. The model is completed by assuming that the flame propagates relative to the fluid at a fixed speed - the adiabatic flame speed. A major problem with the Darrieus-Landau model is its implication with respect to the stability of planar flames. It can easily be shown [5] that planar flames are unstable with respect to perturbations of any wave length. This result is in contradiction with the properties of planar flames which are known to exist in laboratory experiments. In order to overcome this deficiency of the Darrieus-Landau model, Markstein [6] proposed a simple model, where the flame speed is assumed to be proportional to the flame curvature. The constant of proportionality is called the Markstein number. It should be noted that this model is empirical: the dependence of the flame speed on the flame curvature is postulated but not derived from the first principles. In the Markstein model, the short-wave perturbations are stabilized but the Markstein model fails to explain that the flame speed of flat flames in flows involving velocity gradients is not identical to the adiabatic flame speed in a quiescent mixture.

Zeldovich [7] introduced the concept of a more realistic flame by considering the flame as a one-step irreversible reaction between fuel and oxidizer. This reaction is deemed to have a high activation energy and, therefore, the reaction rate strongly depends on the temperature so that the reaction zone is restricted to a thin zone near the highest temperature. Mathematically, this common property of the flames is expressed by a large value of the Zeldovich number Z . The thickness of the reaction zone can be assessed as $\sim l_f/Z$ where l_f is the flame thickness. Behind the reaction zone, there is no reaction since all the available fuel is consumed in the reaction zone while, ahead of the reaction zone, the reaction rate is negligibly small due to the low temperature. The preheat zone is located ahead of the reaction zone. Within the preheat zone, the incoming mixture is preheated by heat conduction until reaction sets in at high temperatures. Also, within the preheat zone fuel diffuses towards the reaction zone where it is consumed. Due to the low heat conductivity and mass diffusivities of gas mixtures, the preheat zone is thin under typical conditions encountered in combustion.

The interactions of the premixed flames formed by a one-step irreversible reaction with fluid flows are considered in many publications [8–10] and reviewed by Clavin [11]. For these asymptotic models, Zeldovich's solution represents a zero-order approximation. At the next order of approximation, the corrections induced by the flame stretch are accounted for. The flame stretch κ [12] is defined as time derivative of logarithm of the area of a flame segment and involves two terms: a term which is proportional to velocity gradients and another term which is proportional to the flame curvature [13, 14]. The major asymptotic analyses of the influence of the flame stretch on the flame speed are performed by Sivashinsky [8] and Matalon and Matkowsky [9]. They consider a thin flame formed by a one-step irreversible reaction with a high activation energy disturbed by an arbitrary external flow and assume that the flame curvature is weak as compared to the flame thickness. The flame characteristics depend on the Lewis number L which is defined as the ratio of heat diffusivity, to mass diffusivity. Sivashinsky [8] assumed that the Lewis number significantly deviates from unity $\Delta L \equiv (L - 1) \sim 1$ while Matalon and Matkowsky [9] considered the case when the Lewis number is close to 1 so that $\Delta L \sim 1/Z$. These formulations are different characteristic limits and result in different equations for the flame propagation speed. The analysis of the flames for the case of a near-stoichiometric mixture and a one-step reaction was performed only recently [10].

Realistic chemical mechanisms are, of course, much more complicated than a one-step irreversible reaction. The combustion process is usually activated by a reaction generating free radicals. Typically, this reaction has a very high activation energy and the corresponding Zeldovich number can be as high as 50. The heat

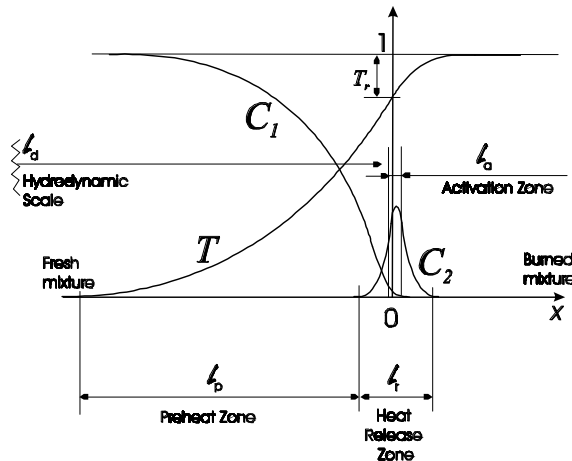


Figure 1: Asymptotic zones in a premixed flame with the two-step reaction mechanism.

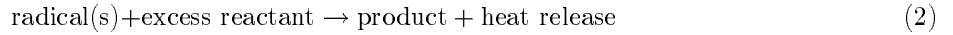
effect of the initializing reaction is minimal. Subsequently, the radicals are consumed in a series of reactions which release heat and the products. While the thickness of the activation zone can be as small as $\sim 1/50$ of the flame thickness, the other reactions take place within a wider zone. About 90% of the heat release occurs in the reaction zone which can be characterized by a different characteristic scale: about $1/5$ of the flame thickness. The question of how the equations obtained for the flames with one-step reaction mechanism can be applied to realistic flames is by no means simple. Abdel-Gayed et al. [15,16] suggested that the activation energy of a realistic flame can be determined by plotting logarithm of the flame speed versus the inverse adiabatic temperature of the products. The flame is formally treated as being generated by a one-step reaction. The Zeldovich numbers determined by this method appear to be ~ 10 [15,16]. These values of the Zeldovich number do not necessarily correspond to the activation energy of the initializing reaction. Thus, analyses of the interactions of fluid flows and flames with more realistic chemical mechanisms seem to be imperative.

While numerical calculations performed for undisturbed, planar premixed flames using realistic chemical kinetics are quite common [1], evaluations of the realistic flames disturbed by the flow are more problematic. This is mostly related to the conventional asymptotic technique [8–10] which makes the mathematics of the asymptotic analysis quite tedious even for a one-step reaction. Recently, Klimenko and Class [?,17] suggested an alternative technique based on the adaptive curvilinear system of coordinates and generalized the flame speed equation [9] for arbitrary values of the diffusion coefficient. The advantage of this new technique is its relative simplicity which should allow for more complicated physical formulations of the problem.

In the present work, we investigate how the propagation speed of realistic flames is affected by flame wrinkling in an arbitrary fluid flow. We assume that the combustion process is initialized in a very thin activation zone by a primary reaction which has a very high activation energy but its heat effect is minimal. This reaction converts a reactant into radicals and is followed by a series of secondary reactions which generate the final combustion products and release heat. If the secondary reactions are very fast then the combustion process would be controlled by the activating reaction and heat would be released in the activation zone. As it is discussed above, in a realistic flame, the heat release zone is normally wider than the activation zone. Hence, at least some of the secondary reactions must be slower. The two-level (activation/heat release) structure of the reaction zone can not be modelled by a one-step reaction mechanism since, in this case, the activation and the heat release zones are the same. We begin our consideration from a two-step reaction mechanism – the most simple mechanism which still can have the expected structure of the reaction zone. This simplified mechanism allows to obtain a complete analytical solution of the problem. The results are then generalized for reaction mechanisms involving a series of reactions releasing heat.

2 The Reaction Zone with the Two-Step Mechanism

We consider a flame whose characteristic width l_f is much smaller than the characteristic scale of hydrodynamic fluctuations l_d . The chemical kinetic mechanism is represented by two reactions



The reactions are controlled by equations

$$\rho \frac{\partial C_1}{\partial t} + \rho \mathbf{v} \cdot \nabla C_1 - \nabla \cdot \left(\frac{D}{L_1} \nabla C_1 \right) = -W_1 \quad (3)$$

$$\rho \frac{\partial C_2}{\partial t} + \rho \mathbf{v} \cdot \nabla C_2 - \nabla \cdot \left(\frac{D}{L_2} \nabla C_2 \right) = W_1 - W_2 \quad (4)$$

$$\rho \frac{\partial T}{\partial t} + \rho \mathbf{v} \cdot \nabla T - \nabla \cdot (D \nabla T) = W_2 \quad (5)$$

where C_1 is the mass concentration of the deficient reactant, C_2 is mass concentration of the radicals and T is the temperature. The concentration C_1 is normalized so that $C_1 = 1$ in the fresh mixture and $C_1 = 0$ in the burned gas. Normalizations of C_2 and T_2 are consistent with normalization of C_1 , as it is determined by (4) and (5), so that $T = 0$ in the fresh mixture and $T = 1$ is the adiabatic temperature (that of undisturbed flame) in the products. The density ρ and the "dynamic" diffusion coefficient $D = \rho \mathcal{D}$ are, generally, functions of the temperature: $\rho = \rho(T)$ and $D = D(T)$.

As shown in Fig.1, we distinguish the following zones: the preheat zone $\sim l_p$; the activation zone whose characteristic length scale is l_a and the heat release zone whose characteristic length scale is l_r . Typically, both of the two reaction zones are thinner than the preheat zone. The rate of the first reaction (1) is essential only within the activation zone. If the reaction (2) is very fast, then the radicals are immediately converted into products, C_2 is small and $W_1 \approx W_2$ so that equation (4) may be discarded. This case is equivalent to the conventional one-step mechanism [8, 9]. In the present work we are interested in the case when the rate of (2) is sufficiently slow to provide essential concentration of C_2 and, at the same time, sufficiently fast to ensure that $l_r \ll l_p$. This can be summarized by

$$l_a \ll l_r \ll l_p = l_f \ll l_d \quad (6)$$

The flame thickness l_f is determined by the thickness of its widest zone l_p . The relation between the characteristic scales in (6) and the other parameters involved is discussed later when the specific zones are considered. The inequality $l_a \ll l_r$ ensures that the case under consideration is essentially different from the conventional one-step mechanism.

The first reaction specified by (1) has a high activation energy. This reactions is confined to a very thin activation zone. The radicals produced in the first reaction diffuse upstream and downstream from the activation zone into the heat release zone where the radicals are converted into products by the second reaction (2). The second reaction is assumed to be moderately fast so that the heat release zone is relatively thin. The temperature variation is relatively small within the reaction zone $\Delta T \sim l_r/l_p$ and so is the variation of the concentration of the excess reactant $\Delta C_0 \sim l_r/l_p$. Thus, within the reaction zone, we neglect terms $O(l_r/l_p)$ and represent the parameters involved in form of the expansions

$$D = D_b + \dots, \quad \rho = \rho_b + \dots \quad (7)$$

$$W_1 = A_1^\circ C_1 \exp\left(-\hat{T}_{a1}/\hat{T}\right) = A_1 C_1 \exp(Z_1 \Delta T) + \dots \quad (8)$$

$$W_2 = A_2^\circ C_2 \exp\left(-\hat{T}_{a2}/\hat{T}\right) = A_2 C_2 \exp(Z_2 \Delta T) + \dots \quad (9)$$

where the subscript "b" is used to denote values in the burned mixture, $\hat{T} = \hat{T}_u + T(\hat{T}_{b*} - \hat{T}_u)$ is the absolute temperature and ΔT is defined as $\Delta T \equiv T - 1$. Particular forms of the expressions for A_1° and A_2° depend on the specific features of the reactions. For the leading-order analysis, these values are treated as constants. The Zeldovich numbers Z_1 and Z_2 are related to corresponding activation energies of the reactions \hat{T}_{a1} and \hat{T}_{a2} . The equations for W_1 and W_2 are obtained by expanding the conventional Arrhenius reaction rates into a series in regions where the reaction rates are significant. The parameter Z_1 is assumed to be large - the activation of the reactions always needs a sufficiently high temperature. The second reaction is expected to be uniformly fast within the reaction zone (otherwise the radicals would diffuse upstream and initiate reactions earlier, ahead of the reaction zone). The simplest way to satisfy this requirement is to assume that Z_2 is either zero or sufficiently small and can be neglected within the reaction zone. Although the dependence of W_2 on the temperature is neglected within the reaction zone and the coefficient A_2 does not depend on T , large variations of the adiabatic temperature may affect A_2 since $A_2 \sim \exp(\hat{T}_{a2}/\hat{T}_{b*})$.

2.1 The Activation Zone

Within the activation zone, the temperature can be represented by the expansion $T = 1 + b_0 + b_1 x + O(l_a/l_r)$ where x is local normal coordinate coefficients b_0 and b_1 are not known and to be determined by matching with the heat release zone. The activating reaction is started when the temperature becomes sufficiently high and terminated when $C_1 \rightarrow 0$. The leading order terms of equation (3) are given by

$$-\frac{D_b}{L_1} \frac{d^2 C_1}{dx^2} = -A_1 C_1 \exp(Z_1 b_0 + Z_1 b_1 x) \quad (10)$$

Here we neglect the convective terms ($\sim l_a/l_p$) and the terms related to the flame stretch ($\sim l_a/l_d$). The solution of (10), which satisfies $C_1 \rightarrow 0$ as $x \rightarrow \infty$, is given by

$$C_1(x) = M \frac{2L_1}{Z_1 b_1 D_b} \text{BesselK}_0 \left(\frac{2}{Z_1 b_1} \left(\frac{A_1 L_1}{D_b} \right)^{1/2} \exp \left(\frac{Z_1 b_0}{2} + \frac{Z_1 b_1}{2} x \right) \right) \quad (11)$$

The main characteristics of this solution are given by the integrals

$$M \equiv \int_{-\infty}^{+\infty} W_1 dx = -\frac{D_b}{L_1} \lim_{x \rightarrow -\infty} \left(\frac{dC_1}{dx} \right) \quad (12)$$

$$x_0 \equiv \frac{1}{M} \int_{-\infty}^{+\infty} x W_1 dx = \frac{D_b}{M L_1} \lim_{x \rightarrow -\infty} \left(C_1 - x \frac{dC_1}{dx} \right) = \frac{1}{Z_1 b_1} \left(\ln \left(\frac{Z_1^2 b_1^2 D_b}{A_1 L_1} \right) - \gamma_0 \right) - \frac{b_0}{b_1} \quad (13)$$

where $\gamma_0 \equiv 2\gamma$ and $\gamma \approx 0.577$ is the Euler constant.

2.2 The Heat Release Zone

Within the heat release zone, the radicals which are produced in the activation zone diffuse upstream and downstream while they are converted into products and heat. The source of radicals is located somewhere near $x = 0$ and can be treated as the delta-function since $l_a \ll l_r$. A more precise location of the radicals source, which increases accuracy of matching, is x_0 determined by (13). The leading order equations order are given by

$$-\frac{D_b}{L_2} \frac{d^2 C_2}{dx^2} = M \delta(x - x_0) - A_2 C_2 \quad (14)$$

$$-D_b \frac{d^2 T}{dx^2} = A_2 C_2 \quad (15)$$

As in the previous section, we neglect the convective terms ($\sim l_r/l_p$) and the terms related to the flame stretch ($\sim l_r/l_d$). The solutions of (14) and (15), which satisfy $C_2 \rightarrow 0$ as $x \rightarrow \pm\infty$ and $T \rightarrow T_b$ as $x \rightarrow \infty$ are given by

$$C_2 = \frac{M}{2} \left(\frac{L_2}{A_2 D_b} \right)^{1/2} \exp \left(- \left(\frac{A_2 L_2}{D_b} \right)^{1/2} |x - x_0| \right) \quad (16)$$

$$T = T_b - \frac{M}{2} \left(\frac{1}{A_2 D_b L_2} \right)^{1/2} \exp \left(- \left(\frac{A_2 L_2}{D_b} \right)^{1/2} |x - x_0| \right) + \frac{M}{D_b} (x - x_0) H(x_0 - x) \quad (17)$$

where H denotes the Heaviside function. Note that in a disturbed flame the temperature excess $\Delta T_b \equiv T_b - 1$ may be non-zero.

2.3 The Released Heat

The coefficients b_0 and b_1 are determined by expanding T given by (17) near $x \approx x_0$

$$T \rightarrow 1 + b_0 + b_1 x, \quad b_0 = \Delta T_b - T_r - b_1 x_0, \quad b_1 = \frac{M}{2D_b}, \quad T_r \equiv \frac{M}{2} \left(\frac{1}{A_2 D_b L_2} \right)^{1/2} \quad (18)$$

The matching conditions are given by

$$Z_1 (b_0 + b_1 x_0) = Z_1 (\Delta T_b - T_r) = \ln \left(\frac{Z_1^2 b_1^2 D_b}{A_1 L_1} \right) - \gamma_0 \quad (19)$$

The middle term of this equality is found from (18) while the right-hand side is determined from (13). The equation determining the heat release takes the form

$$M = \frac{\gamma_1}{Z_1} (D_b A_1 L_1)^{1/2} \exp \left(\frac{Z_1}{2} \left(\Delta T_b - \frac{M}{2} \left(\frac{1}{A_2 D_b L_2} \right)^{1/2} \right) \right) \quad (20)$$

where $\gamma_1 = 2 \exp(\gamma_0/2) \approx 3.56$. This equation determines how M depends on ΔT_b . When compared with the conventional equation

$$M = \frac{1}{Z_1} (2D_b A_1 L_1)^{1/2} \exp \left(\frac{Z_1}{2} \Delta T_b \right) \quad (21)$$

which is obtained for the one-step reaction mechanism, equation (20) looks quite similar with new additional correcting term in the exponent and a new factor γ_1 instead of $2^{1/2}$. In fact, the relationship $M(\Delta T_b)$ generated by equation (20) is quite different from the conventional relationship $M(\Delta T_b)$ generated by (21). This point needs a more detailed analysis.

First we define exact values of the characteristic scales and note that the average rate of production and recombination of the radicals are the same.

$$l_a = \frac{1}{b_1 Z_1} = \frac{2D_b}{M Z_1}, \quad l_r = \left(\frac{D_b}{A_2 L_2} \right)^{1/2}, \quad M = \int_{-\infty}^{+\infty} W_2 dx = \int_{-\infty}^{+\infty} W_1 dx \quad (22)$$

Using $\zeta = l_r/l_a$ we can rewrite (20) in the form $Z_1 \Delta T_b = \zeta + 2 \ln(\zeta) + 2 \ln(l_1/l_r) - \gamma_0$ where $l_1 \equiv (D_b/(A_1 L_1))^{1/2}$. We use the subscript "*" to denote values in the undisturbed flame. That is ζ_* is determined

by

$$0 = \zeta_* + 2 \ln(\zeta_*) + 2 \ln(l_1/l_r) - \gamma_0 = \zeta_* + 2 \ln(\zeta_*) + \ln\left(\frac{A_2 L_2}{A_1 L_1}\right) - \gamma_0 \quad (23)$$

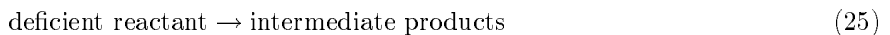
For $\Delta\zeta \equiv \zeta - \zeta_*$ we have $Z_1 \Delta T_b = \Delta\zeta + 2 \ln(1 + \Delta\zeta/\zeta_*)$. Since the conditions in (6) require that $\zeta_* \gg 1$, the order of the logarithmic term is smaller than the order of $\Delta\zeta$ for any order of $\Delta\zeta$. Thus, we have $Z_1 \Delta T_b = \Delta\zeta + O(\Delta\zeta/\zeta_*)$ and the excess heat release $\Delta M \equiv M - M_*$ is determined by the equation

$$\frac{\Delta M}{M_*} = \frac{\Delta\zeta}{\zeta_*} = \frac{\Delta T_b}{T_{r*}} = \frac{Z_f}{2} \Delta T_b, \quad Z_f \equiv \frac{2}{T_{r*}} \equiv 2 \frac{Z_1}{\zeta_*} = 4 \frac{(A_2 D_b L_2)^{1/2}}{M_*} \quad (24)$$

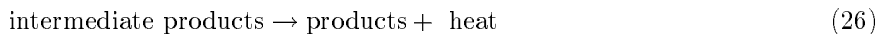
where M_* is the value of M in the undisturbed flame, $T_{r*} = 1 - T(x_0)$ is the temperature change in the reaction zone of the undisturbed flame and Z_f can be called as the "effective Zeldovich number" of the flame. Equation (24) determines the excess heat release as a function of excess temperature of the products. It appears that equation (24) does not depend on D_b since both values ζ_* and T_{r*} do not depend on D_b as determined by equation (23).

3 Reaction zone in the case of multi-step reaction mechanisms

Here, we generalize the results obtained in previous section for the more realistic reaction mechanisms. The combustion process is assumed to be initialized by the activating reaction



which consumes the deficient reactant C_1 as specified by equations (3) and (8). The activation energy of this reaction, \hat{T}_{a1} must be very high since the reaction rate in the unburned mixture is extremely slow. The activating reaction is followed by many secondary reactions



which generate final products and release heat. Of course, the analysis can not be performed without making assumptions constraining the reaction mechanism. First, we assume that the combustion process is initialized by a single reaction while there could be several competing initializing reactions. Second, we assume that the heat is released in a zone whose characteristic thickness is wider than the thickness of the zone where the activating reaction takes place although the overall thickness of the reaction zone remains small as determined by (6). Third, we assume that the activation energy of the activating reaction is so high that its reaction rate depends on the temperature much stronger than the reaction rates of any of the secondary reactions. Forth, we assume that the heat effect of the activating reaction is negligible. As it is discussed in Introduction, these assumptions generally correspond to physics of the realistic flames. We also assume that C_1 is the deficient reactant. Hence the activating reaction, which is treated as irreversible, is terminated when $C_1 \rightarrow 0$. If C_1 is the excess reactant, its consumption is terminated when remaining concentration C_1 is in equilibrium with the products. This terminating condition requires to treat the activating reaction as reversible. In a simplified model, one can assume that only stoichiometric portion of the excess reactant is subject to chemical reactions.

Since the heat release zone is much wider than the activation zone and the activating reaction does not release heat by itself, we can still expand the temperature $T = 1 + b_0 + b_1 x + O(l_a/l_r)$ within the activation zone. Hence, the previous analysis of the activation zone, which is based on this expansion, remains valid for the multistep mechanism considered in this section. This analysis results in

$$b_0 + b_1 x_0 = \frac{1}{Z_1} \ln\left(\frac{Z_1^2 b_1^2 D_b}{A_1 L_1}\right) - \gamma_0 = \Delta T_b - T_r \quad (27)$$

where $T_r = T(x_0) - \Delta T_b$. This equation is similar to equation (19) but, unlike in the two-step analysis, b_1 and T_r are not determined by the corresponding formulae in (18). Let M_* and T_{r*} be the values of M and

T_r in the undisturbed flame where $\Delta T_b = 0$ by definition. When the flame is disturbed, the reaction zone is affected by excess temperature $\Delta T_b \neq 0$. The excess temperature affects primarily the activating reaction since this reaction is most sensitive to temperature changes. As the result the rate of radical generation by the activating reaction (M defined by (12)) appears to be different from its undisturbed value M_* and $\Delta M \equiv M - M_* \neq 0$. This affects the rates of the secondary reactions resulting in a different value of T_r and $\Delta T_r \equiv T_r - T_{r*}$ is a function of ΔM . If ΔM is relatively small, we can write

$$\Delta T_r = \left(\frac{\partial T_r}{\partial M} \right)_* \Delta M \quad \text{or} \quad \frac{\Delta M}{M_*} = \frac{Z_f}{2} \Delta T_b, \quad Z_f \equiv \frac{2}{M_*} \left(\frac{\partial T_r}{\partial M} \right)_*^{-1} \quad (28)$$

The dependence of T_r on M is determined by specific properties of the secondary reactions. In the previous section where the simplified two-step mechanism is considered, the concentration of the radicals C_2 increased lineally with the increase of M – the radical supply rate by the activating reaction. The larger values of C_2 correspond to larger variations of the temperature in the heat release zone T_r . This tendency is likely to be also valid for more complicated multi-step mechanisms when M is not very large. However, a significant further grows of M would increase the overall reactions rates but not the radical concentration due to various radical destruction mechanisms. Thus, T_r would be a weak function of M . This corresponds to small values of the derivative $\partial T_r / \partial M$ and large values of the effective Zeldovich number Z_f .

4 Estimation of the effective Zeldovich number

In this section we estimate the Zeldovich number using experimental data for propagation speed. Abdel-Gayed et al. [15, 16] suggested a method of determining the activation energy and the Zeldovich number from experimental data for the flame propagation speed. They considered a model one-step reaction whose propagation speed S can be estimated

$$\ln(S) \sim \frac{1}{2} \left(a - \frac{\hat{T}_{a1}}{\hat{T}_b} \right) \quad (29)$$

where a is the logarithm of pre-exponential factor whose dependence on temperature is logarithmically weak and can be neglected. The activation energy is found from experiments by plotting $\ln(S)$ vs $1/\hat{T}_b$ and determining the slope of the curve which corresponds to \hat{T}_a . The Zeldovich number (Z_1 in our notations) is given by

$$Z_1 = \frac{\hat{T}_{a1}}{\hat{T}_b^2} (\hat{T}_b - \hat{T}_u) \approx \frac{\hat{T}_{a1}}{\hat{T}_b} \sim 2\hat{T}_b \frac{d \ln(S)}{d \hat{T}_b} = 2 \frac{d \ln(S)}{d \ln(\hat{T}_b)} \quad (30)$$

The estimation $Z_1 \approx \hat{T}_{a1}/\hat{T}_b$ is valid when $\hat{T}_b \gg \hat{T}_u$ which is usually the case realistic flames.

In the present work the burning velocity which is determined by the rate of consumption of C_1 : $S_* = M_*/\rho_u$, can be estimated as

$$\ln(S) \sim \frac{1}{2} \left(a - \frac{\hat{T}_{a1}}{\hat{T}_b - \hat{T}_r} \right) \quad (31)$$

where $\hat{T}_b - \hat{T}_r$ is the absolute temperature in the activation zone at $x = x_0$

Let us assume that, initially, $S = S_*$ and $\hat{T}_b = \hat{T}_{b*}$. Due to some changes in the flame the temperature of the burned products is changed $\Delta \hat{T}_b = \hat{T}_b - \hat{T}_{b*} = (\hat{T}_{b*} - \hat{T}_{u*}) \Delta T_b$. The propagation speed is also changes $\Delta S = S - S_*$. Substitution of $\Delta \hat{T}_r = (\hat{T}_{b*} - \hat{T}_{u*}) \Delta T_r$ and $\Delta T_r \approx (\partial T_r / \partial M)_* \Delta M \approx 2Z_f^{-1} \Delta M / M_* \approx 2Z_f^{-1} \Delta S / S_*$ (as determined by (28)) yields

$$2 \frac{\Delta S}{S_*} \sim \frac{\hat{T}_{a1}}{(\hat{T}_b - \hat{T}_r)^2} \left(\Delta \hat{T}_b - (\hat{T}_{b*} - \hat{T}_{u*}) \frac{2}{Z_f} \frac{\Delta S}{S_*} \right) \sim Z_1 \left(\frac{\Delta \hat{T}_b}{\hat{T}_b} - \frac{2}{Z_f} \frac{\Delta S}{S_*} \right) \quad (32)$$

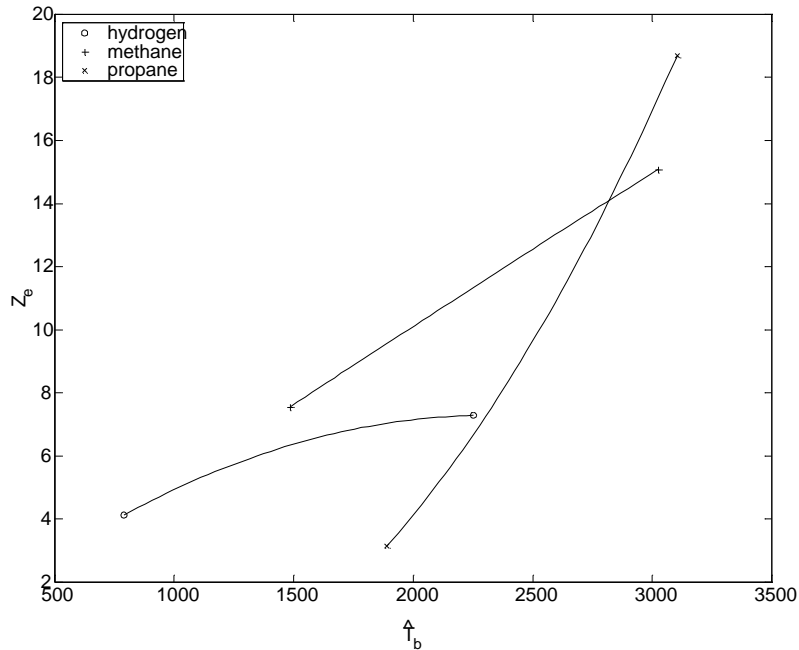


Figure 2: Dependence of the Zeldovich number defined as $Z_e = 2d \ln(S)/d \ln(\hat{T}_b)$ on the absolute temperature of the products \hat{T}_b . The data are taken from [15,16].

where \hat{T}_b is assumed to be much larger than \hat{T}_r and Z_1 is substituted for \hat{T}_{a1}/\hat{T}_b . Equation (32) can be rewritten as

$$Z_e \equiv \frac{1}{\frac{1}{Z_f} + \frac{1}{Z_1}} \sim 2 \frac{\hat{T}_{b*}}{S_*} \frac{\Delta S}{\Delta \hat{T}_b} = 2 \frac{d \ln(S)}{d \ln(\hat{T}_b)} \quad (33)$$

Hence, the method of Abdel-Gayed et al. [15,16] effectively gives the estimate of either Z_1 or Z_f whichever is smallest. This corresponds to the physics of the problem: if Z_1 is not sufficiently large then the activation zone is no different from the heat release zone and the effective Zeldovich number becomes irrelevant.

The value Z_e defined by equation (33) and determined from the curves presented in [15,16] is plotted in Figure 2. The experimental points have quite a significant scattering so that Figure 2 gives only rough indication how Z_e depends on \hat{T}_b . However, the tendency of Z_e to increase for larger \hat{T}_b can not be ignored. The value of $Z_1 \approx \hat{T}_{a1}/\hat{T}_b$ should decrease for increasing \hat{T}_b while the value of Z_f which is defined by (28) is a complicated function of the reaction mechanism and may behave differently from Z_1 .

5 The Preheat Zone

The preheat zone is the widest zone of the flame front and, as such, this zone is primarily affected by the fluid flow. Here, we follow our previous work [?,17] and select the moving generalized curvilinear system of coordinates x^1, x^2, x^3 so that $x^1 = 0$ specifies the location of the reaction zones. The coordinate x^1 can be selected to be normal to the flame surface. The covariant and contravariant metric tensors for this system of coordinates are given by g_{ij} and g^{ij} where $i, j = 1, 2, 3$. We can assume that locally $g^{11} = g_{11} = 1$ and the coordinate x^1 coincides with physical normal coordinate x used in previous sections: $x^1 = x$. The proof that this can be done without loss of generality and other details of this approach were presented earlier [?]. We introduce also the flame surface segment area as the square root of $G_f \equiv \det(g_{\alpha\beta})$ where $G = g/g_{11} = g$, $g \equiv \det(g_{ij})$ and $\alpha, \beta = 2, 3$ while $i, j = 1, 2, 3$. The subscript "f" indicates that the indexed value is taken

at the flame surface, related to whole flame structure or remain constant within the flame.

Considering that $g_{11} = 1$ and that the reaction zone is relatively thin, we can write the system of governing equations in the form

$$\frac{\partial G_f^{1/2} \rho}{\partial t} + \frac{\partial G_f^{1/2} m^i}{\partial x^i} = 0 \quad (34)$$

$$\rho \frac{\partial C_1}{\partial t} + m^i \frac{\partial C_1}{\partial x^i} - \frac{1}{G_f^{1/2}} \frac{\partial}{\partial x^j} \left(g^{ij} G_f^{1/2} \frac{D}{L_1} \frac{\partial C_1}{\partial x^i} \right) = -M \delta(x^1) \quad (35)$$

$$\rho \frac{\partial T}{\partial t} + m^i \frac{\partial T}{\partial x^i} - \frac{1}{G_f^{1/2}} \frac{\partial}{\partial x^j} \left(g^{ij} G_f^{1/2} D \frac{\partial T}{\partial x^i} \right) = M \delta(x^1) \quad (36)$$

where $m^i \equiv \rho U^i$ and U^i is the fluid velocity relative to the moving system of coordinates $U^i \equiv v^i + dx^i/dt$. Without loss of generality the system of coordinates can be selected to nullify U_α , $\alpha = 2, 3$ [?, 17]. The chemical source terms are approximated by the delta functions. For simplicity, we shall also denote $m \equiv m^1$ and $x \equiv x^1$. Note that G_f is determined by outer flow and depends on stretched coordinates $X^i = x^i/\varepsilon$ and $\tau = t/\varepsilon$ where the small parameter ε represents the ratio $\varepsilon \equiv l_p/l_d$.

The values involved are represented by the asymptotic expansions

$$\begin{aligned} m &= m_0 + \varepsilon m_1 + \dots, \quad c \equiv 1 - C_1 = c_0 + \varepsilon c_1 + \dots, \quad T = T_0 + \varepsilon T_1 + \dots, \\ \rho &= \rho_0 + \dots, \quad D = D_0 + \varepsilon D_1 + \dots, \quad M = M_0 + \varepsilon M_1 + \dots \end{aligned} \quad (37)$$

It is convenient to determine c first and then find C_1 (by calculating $1 - c$) and T (by formally putting $L_1 = 1$). For $x \leq 0$, the leading order solution is, obviously, given by

$$m_0 = M_0, \quad c_0 = \exp(L_1 B), \quad B(x) \equiv M_0 \int_0^x H(-x) \frac{dx}{D_0}. \quad (38)$$

We take into account here that $c = 1$ in the burned mixture since the deficient reactant is fully consumed. If we select D_b as the characteristic value of the diffusion coefficient, the characteristic scale of the preheat zone is given by

$$l_p = \frac{D_b}{M_0} = \frac{Z_1}{2} l_a = \frac{Z_f}{4} l_r \quad (39)$$

The next order terms are given by

$$m_1(x) = \int_x^0 \Psi_m dx + m_{f1} \quad (40)$$

$$c_1(x) = \exp(L_1 B) \left(\frac{M_1}{M_0} + \frac{1}{M_0} \int_x^0 \frac{\Psi_C}{\exp(L_1 B)} dx \right) + \frac{1}{M_0} \int_{-\infty}^x \Psi_C dx \quad (41)$$

$$\Psi_C \equiv -\frac{\partial m_1 c_0}{\partial x} - K_f c_0 \rho_0 - \frac{\partial c_0 \rho_0}{\partial \tau} + \frac{\partial}{\partial x} \left(\frac{D_1}{L_1} \frac{\partial c_0}{\partial x} \right) \quad (42)$$

where m_{f1} is constant, $K_f \equiv \partial \ln(G_f^{1/2})/\partial \tau$ is the rescaled flame stretch and $\Gamma_f \equiv \left(\partial \ln(G_f^{1/2})/\partial X \right)_f$ is the rescaled flame curvature which are evaluated at $X = 0$. It should be noted that $\Psi_C = 0$ for $x > 0$. We find the next order temperature term T_1 by putting $L_1 = 1$ in equations (38), (41) and (42). Then we take into

account that $c_1 = 0$ in the burned mixture while T_{1b} may be non-zero. Thus, in the burned mixture we have

$$M_1 = M_C, \quad T_b = 1 + \varepsilon \frac{M_C - M_T}{M_0} \quad (43)$$

where M_C and M_T represent the integrals

$$M_I \equiv m_{f1} + \left(K_f \phi_I + \frac{d\phi_I}{d\tau} \right) - M_0 \left(\frac{D_1}{D_0} \right)_b \quad (44)$$

$$\phi_C \equiv \int_{-\infty}^0 \rho_0 c_0 dx, \quad \phi_T \equiv \int_{-\infty}^0 \rho_0 T_0 dx \quad (45)$$

the index "I" in equation (44) is either "T" or "C" and $(D_1/D_0)_b$ denotes D_1/D_0 at $x = 0$.

6 Propagation Speed

6.1 Non-Equidiffusion Flames

The case considered here has some similarities with the limit introduced by Sivashinsky [8] for the one-step reaction mechanism. We assume that $\varepsilon Z_f = z_f \sim 1$ that is $l_d/l_p \sim l_p/l_r$. For this limit, the value of M_0 is, according to (24) and (43), given by

$$M_0 = M_* \left(1 + \frac{Z_f}{2} \Delta T_b \right) = M_* \left(1 - \frac{z_f}{2} \frac{K_f \phi + d\phi/d\tau}{M_0} \right) \quad (46)$$

where $\phi \equiv \phi_T - \phi_C$. Using the burning velocity $S = M_0/\rho_u$, the physical flame stretch $\kappa_f \equiv \partial \ln(G_f^{1/2})/\partial t = \varepsilon K_f$ and replacing integration over x by integration over T_0 in

$$\phi \equiv \int_{-\infty}^0 \rho_0 (T_0 - C_0) dx = \frac{1}{M_0} \int_{-\infty}^0 \rho_0 D_0 (1 - T_0^{\Delta L_1}) dT_0 \quad (47)$$

we obtain

$$S = S_* \left(1 - \Phi \frac{Z_f}{2} \frac{\mathcal{D}_u}{S^2} \left(\kappa_f - \frac{d \ln(S)}{dt} \right) \right), \quad \Phi \equiv \int_0^1 \hat{\rho} \hat{D} (1 - T^{\Delta L_1}) dT \quad (48)$$

where $D \equiv D/\rho$ is the "kinematic" diffusion coefficient and $\Delta L_1 \equiv L_1 - 1$ and the symbol " $\hat{\cdot}$ " denotes the parameters normalized by their unburned values: $\hat{D} \equiv D/D_u$, $\hat{\rho} \equiv \rho/\rho_u$. Note that the flame speed determined by (48) may have significant speed variations $(S - S_*)/S_* \sim 1$. However, the case of $S \rightarrow 0$ may not be consistent with the condition $\zeta \gg 1$ and it is not covered by the present asymptotic theory.

6.2 Near-Equidiffusion Flames

Here we consider the case when $\lambda \equiv Z_f \Delta L_1 \sim 1$ which has some similarities with the limit introduced by Matalon and Matkowsky [9] for one-step reactions. For this case the integral ϕ can be assessed as $\phi \rightarrow \Delta L_1 \phi^\circ$ where

$$\phi^\circ \equiv \int_{-\infty}^0 \rho_0 T_0 x dx \quad (49)$$

Equation (24) determines the terms in the expansion $M = M_0 + \varepsilon M_1 + \dots$

$$M_0 = M_*, \quad M_1 = M_* \frac{Z_f}{2} T_{1b} = -\frac{\lambda}{2} (K_f \phi^\circ + d\phi^\circ/d\tau) \quad (50)$$

This gives the following estimation for the excess temperature $\Delta T_b \sim \varepsilon T_{1b} \sim \varepsilon \Delta L_1$. The constant m_{f1} , which determines m_1 at $x = 0$, can be found from (44)

$$m_{f1} \equiv M_1 - \left(K_f \phi_C + \frac{d\phi_C}{d\tau} \right) - \Omega \left(K_f \phi^\circ + \frac{d\phi^\circ}{d\tau} \right) \Delta L_1 \quad (51)$$

where $\Omega \equiv (d \ln(D)/dT)_{T=1}$ is evaluated in the reaction zone of the undisturbed flame. Since $T_{1b} \sim \Delta L_1 \sim 1/\Delta Z_f$ is small the last term in (51) can be neglected. The unburned mass flow rate $m_u = m_{u0} + \varepsilon m_{u1} + \dots$ is linked to m_{f1} by the following equation [?,17]

$$m_{u0} = M_0, \quad m_{u1} = m_{f1} - \left(K_f \phi_\rho + \frac{d\phi_\rho}{d\tau} \right), \quad \phi_\rho \equiv \int_{-\infty}^0 (\rho_u - \rho_0) dx \quad (52)$$

Since the flame speed changes are small $S = S_* + \varepsilon S_1 + \dots$, the order of the time derivatives is small and $d/d\tau$ terms can be neglected. Finally, we introduce the physical flame stretch $\kappa_f \equiv \partial \ln(G_f^{1/2})/\partial t$, note that $\phi_C \rightarrow \phi_T$ as $\Delta L_1 \rightarrow 0$, substitute the integration over dT_0 for the integration over dx and combine (50), (51), (52) and $S = m_u/\rho_u$ into

$$S = S_* - \kappa_f \frac{\mathcal{D}_u}{S_*} \left(\frac{\lambda}{2} \Phi^\circ + \Phi_T + \Phi_\rho \right) \quad (53)$$

$$\Phi^\circ = - \int_0^1 \hat{\rho} \hat{D} \ln(T) dT, \quad \Phi_T \equiv \int_0^1 \hat{\rho} \hat{D} dT, \quad \Phi_\rho \equiv \int_0^1 (1 - \hat{\rho}) \hat{D} \frac{dT}{T}$$

We note that, for practical purposes, equation (53) and the steady version of equation (48) can be combined into one equation which is more likely to be valid for a wider range of Lewis numbers

$$S = S_* \left(1 - \kappa_f \frac{\mathcal{D}_u}{S_*^2} \left(\frac{Z_f}{2} \Phi + \Phi_C + \Phi_\rho \right) \right), \quad \Phi_C \equiv \int_0^1 \hat{\rho} \hat{D} T^{\Delta L_1} dT \quad (54)$$

7 Conclusions

The flame speed of a thin premixed flame disturbed by an arbitrary fluid flow of a larger scale is considered. The asymptotic theory is suggested for the reaction mechanism which is more realistic than the one-step reactions considered previously in asymptotic theories. The mechanism is formed by two reactions: the activating reaction and the heat release reaction. The first reaction has a very high activation energy and triggers the combustion process by generating radicals. The second reaction converts the radicals into heat and products.

If the reaction rate of the second reaction is extremely fast, this problem is essentially equivalent to the conventional case of a one-step reaction with high activation energy. However, slower rates of the second reaction correspond to a distinct problem with three asymptotic zones within the flame: the widest zone is the preheat zone, the heat release zone is somewhat thinner and the activation zone is the thinnest. The propagation speed of this flame is different from the conventional propagation speed calculated for the one-step reaction mechanism.

The results obtained for two-step reactions are generalized for multi-step reaction mechanism represented by a single activation reaction followed by several secondary reactions releasing heat. The most important features of the flames with distinct activation and heat release zones are summarized now.

1) The variations of the speed of the disturbed flames is determined by the effective Zeldovich number introduced in the paper rather than by the conventional Zeldovich number linked to the activation reaction energy.

2) The effective Zeldovich number, Z_f , which a) is not directly linked to the activation energies of both reactions, b) is generally smaller than the conventional Zeldovich number of the first, initializing reaction and c) is proportional to the ratio the flame thickness to that of the heat release zone. It is shown that the approximate method [15,16] gives an estimation of this effective Zeldovich number rather than the Zeldovich number of the initializing reaction.

3) In the case of near-equidiffusion flames, the equation for the flame speed is similar to the conventional equation [9]. There is, however, a significant difference: the effective Zeldovich number should be substituted for the conventional Zeldovich number which is based on the activation energy of the initializing reaction.

4) In the case of $\Delta L_1 \sim 1$, the resultant equation for the flame speed is, unlike the conventional exponential equation [8], linear with respect to the flame stretch. It is likely that the near-equidiffusion equation (which is also linear) has a wider range of applicability for realistic flames than for the flames with the one-step reaction mechanism.

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