Influence of hydrodynamics and diffusion upon the stability limits of laminar premixed flames

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(Received 26 October 1981)

An analytical theory is developed for the stability properties of planar fronts of premixed laminar flames freely propagating downwards in a uniform reacting mixture. The coupling between the hydrodynamics and the diffusion process is described for an arbitrary expansion of the gas across the flame. Viscous effects are included with an arbitrary Prandtl number. The flame structure is described for a large value of the reduced activation energy and for a Lewis number close to unity. The flame thickness is assumed to be small compared with the wavelength of the wrinkles of the front, this wavelength being also the characteristic lengthscale of the perturbations of the flow field outside the flame. A two-scale method is then used to solve the problem. The results show that the acceleration of gravity associated with the diffusion mechanisms inside the front can counterbalance the hydrodynamical instability when the laminar-flame velocity is low enough. The theory provides predictions concerning the instability threshold. In particular, the dimensions of the cells are predicted to be large compared with the flame thickness, and thus the basic assumption of the theory is verified. Furthermore, the quantitative predictions are in good agreement with the existing experimental data.

The bifurcation is shown to be of a different nature than predicted by the purely diffusive-thermal model.

The viscous diffusivities are supposed to be independent of the temperature, and then the viscosity is proved to have no effect at all on the dynamical properties of the flame front.

1. Introduction

Owing to heat release Q, the temperature T increases inside a premixed flame of thickness d from $T_{-\infty}$ to $T_{\rm b}$. According to a practically isobaric condition, the corresponding density decrease from $\rho_{-\infty}$ to $\rho_{\rm b}$ can be expressed in terms of a parameter γ , defined by $\gamma = (\rho_{-\infty} - \rho_{\rm f})/\rho_{-\infty} = 1 - (T_{-\infty}/T_{\rm f})$, $0 < \gamma < 1$, describing the gas expansion through the flame. The subscripts $-\infty$ and b refer to conditions in the fresh mixture and in the local burned gases respectively. Subscript f refers to the burned gases in the case of planar and adiabatic flames, i.e. $T_{\rm f} = T_{-\infty} + Q/C$, where C is the specific heat of the mixture. When the activation energy E of the chemical reaction is large, $\beta = (E/RT_{\rm f}) (T_{\rm f} - T_{-\infty}/T_{\rm f}) > 1$, the reaction takes place only in a thin zone of vanishing thickness d/β located close to the maximum of temperature $T_{\rm b}$, while the combustion rate is a strongly increasing function of the temperature of combustion $T_{\rm b}$. In the planar case, the gradients of the temperature and of species concentration are oriented only in the direction perpendicular to the front, and the temperature of combustion is equal to $T_{\rm f}$. The thickness d and the velocity $u_{\rm L}$ of the

front are determined by a simple balance of the corresponding fluxes of heat and mass with the chemical production characterized by the reaction time τ_r :

$$\tau_{\mathbf{r}}(T) \sim e^{E/kT}, \quad u_{\mathbf{L}}^2 \sim \frac{\rho D_{\mathrm{th}}}{\rho_{\mathrm{f}} \tau_{\mathrm{r}}(T_{\mathrm{f}})}, \quad d = \frac{\rho D_{\mathrm{th}}}{\rho_{-\infty} u_{\mathrm{L}}}, \tag{1}$$

where $D_{\rm th}$ is the thermal diffusivity of the gas mixture. For a wrinkled front, the transverse gradients modify the local balance of energy and affect the temperature of combustion so that $T_{\rm b} \neq T_{\rm f}$, and the local velocity u of the front is also modified, so that $u \neq u_{\rm L}$. Thus the dynamic behaviour of a wrinkled front must depend on the diffusive and convective transport of energy and mass, which control $T_{\rm b}$.

However, apart from these transport mechanisms occurring inside the flame, the motion of a wrinkled front depends also on hydrodynamical effects that are developed on a scale larger than d (see figure 1). In fact, the combustion rate controls only the front velocity u relative to the local upstream flow velocity $u_{-\infty}$, which produces a convective displacement of the front. However, owing to hydrodynamics, the upstream flow field $u_{-\infty}$ is itself modified by the wrinkling of the front on a distance of the order of magnitude of the wavelength Λ of the wrinkles. Thus a feedback mechanism is developed in the motion of the wrinkled fronts where hydrodynamic effects are coupled to the transport processes taking place inside the flame. The origin of this feedback lies in the fact that the gas expands through the flame thickness $\gamma \neq 0$, and so the conservation of mass and momentum causes a deflection of the streamlines of the gas flow at the tilted front. This local deflection induces a modification of the flow, which, as a consequence of negligibly small Mach number, can be considered as instantaneous and essentially incompressible. To point out the typical difficulty involved in the study of the dynamic properties of a wrinkled front, let us recall that this coupling between hydrodynamics and transport processes is not involved in the calculation of the steady and unidimensional basic solution where the planar front, perpendicular to a uniform flow (with a velocity equal to $u_{\rm L}$), is not affected by any deflection of the streamlines. The solution for this freely propagating planar flame, which is characterized by (1), has been obtained a long time ago by Zel'dovich & Frank-Kamenetzki (1938). However, for non-steady wrinkled fronts the above-mentioned coupling causes great difficulties in analytical studies, and, to the best of our knowledge, a complete and satisfactory analytical solution of the stability of the planar flame has not yet been obtained.

It is the purpose of the present work to provide such an analysis on the basis of recent results of Clavin & Williams (1982) (referred to hereinafter as I). Attention is focused on a realistic situation where γ is not small. In this case, according to the pioneering studies of Darrieus (1938) and Landau (1944), the hydrodynamic effects are known to be of particular importance because they produce a strong mechanism of instability of the front, which leads one to question the physical relevance of the basic planar solution. The method employed in the present study is based on the difference of scale between the flame thickness d and the relevant wavelengths Λ ; $\epsilon = d/\Lambda < 1$. A two-scale method is used that identifies the scale d for the longitudinal variation in the temperature and the scale Λ for the transverse variation. The flow is then separated into two parts: a far flow field, which varies on scale Λ , and the flow inside the flame thickness, which varies on scale d. This method allows us to obtain local relations between the flame velocity and the characteristic properties of the far flow field at the front. Thus it is shown, as was anticipated in the pioneering studies quoted above, that the problem is reduced to the solution of an incompressibleflow problem outside the flame thickness with boundary conditions specified on both

sides of the front. The final result is sensitive to these boundary conditions, which can be specified only by a detailed analysis of the structure of the wrinkled flame. Such an analysis is completely absent not only in the first studies mentioned above but also in the more recent phenomenological description reported in Markstein (1964) and in Zel'dovich et al. (1980). The phenomenological boundary conditions used there limit the validity of the final result. Even in the simplifying approximation $\gamma = 0$ it is only recently that the structure of the wrinkled front has been completely described by Sivashinsky (1977a) and by Joulin & Clavin (1979), who have used an asymptotic expansion in $\beta \rightarrow \infty$ for solving the purely thermal diffusive model introduced and first studied by Barenblatt, Zel'dovich & Istratov (1962). When the gas expansion is retained, $\gamma \neq 0$, this local structure is not only of a diffusive nature but also involves transverse convective transport, which evolves within the flame as a consequence of the expansion. In addition to the hydrodynamic instability described by Darrieus and Landau, which is associated with a longitudinal convection, the modification of the flame structure by the transverse convective transports is a supplementary effect of the gas expansion. This particular aspect has been partially solved in I, where it is shown how the flame structure can be determined for small values of $\epsilon = d/\Lambda$ for any arbitrary expansion, $0 < \gamma < 1$. In this case the front is only weakly wrinkled and the deflection of the streamlines produces only weak effects on the flame structure even for strong expansion ($\gamma \sim 1$). But the corresponding modifications are found to be of the same order of magnitude as the diffusive effects. The study of Clavin & Williams (I) was concerned primarily with the establishment of the local equation for the evolution of the front expressed in terms of the value of the upstream far-flow velocity at the front, which is considered as a given quantity. In the present paper we are concerned with the calculation, in the laminar case, of this flow field produced by the wrinkling of the front. In contrast to I, where the determination of the local temperature of combustion $T_{\rm b}$ is sufficient, the flow calculation requires further computations of the jump conditions across the flame for flow velocity and pressure. These relations are derived in §3, and associated with the evolution equation of I they constitute the boundary conditions necessary to solve the fluid-mechanical problem involved in the study of the front stability developed in §4. As reported in the review monograph of Markstein (1964), the size of the cellular structures appearing at the instability threshold of premixed flames are observed to be large (~ 1 cm) compared with the flame thickness d (~ 10^{-2} cm) for most of the reactive mixtures he studied. Thus the perturbation analysis in small values of $\epsilon = d/\Lambda$ used herein is expected to be good for the study of the limit of stability of planar flames.

As suggested by Landau (1944) for the combustion of liquids and by Einbinder (1953) in gases, the hydrodynamical instability can be damped out for all wavelengths when the acceleration due to gravity g is added to the transport mechanisms. The corresponding criterion is that the Froude number $F = u_{\rm L}^2/(1-\gamma) gd$, must be smaller than some critical value $F_{\rm c}$. This corresponds to the experimental observations (see Lewis & von Elbe 1961, p. 390; Markstein & Somers 1953), which show that it is only for slow-burning flames that planar freely propagating fronts can be stabilized, in a uniform laminar flow, provided that they propagate in the downward direction. The expression for $F_{\rm c}$ is derived in this paper in terms of the thermal expansion γ and of the diffusive properties of the reactive mixture. The corresponding instability threshold is predicted by this expression for $F_{\rm c}$ to be experimentally observable in usual hydrocarbon-oxygen mixtures diluted by nitrogen when the flame velocity $u_{\rm L}$ of the planar front is between 5 cm/s and 17 cm/s. This threshold is also predicted

to concern only the wavelengths close to $\Lambda_c = \pi u_L^2/(1-\gamma)g$, and to correspond to a fuel-rich composition for slow-burning flames and to a lean composition for fast flames. This is in qualitative and quantitative agreement with the experimental results reported by Markstein (1951, 1964) concerning slow-burning freely propagating flames. The predicted bifurcation is of a different nature than those described previously, either by the phenomenological theory of Markstein (1970) or by the purely thermal-diffusive model studied by Sivashinsky (1977*a*) and by Joplin & Clavin (1979), even when the modifications proposed by Markowsky & Sivashinsky (1979) are introduced to take into account the acceleration due to gravity in the presence of a negligible expansion.

In addition to the considerations concerning the acceleration due to gravity, the results obtained here provide the two first orders in the development in powers of ϵ of the exact dispersion relation associated with the instability of the basic planar solution,[†] the first term corresponding to the analyses of Darrieus and Landau.

The viscous effects cannot be neglected inside the flame, since the Prandtl number is of order unity. Nevertheless, it turns out that for temperature-independent diffusivities the viscous effects balance themselves, and do not play any role in the dynamical properties of the flame front. A similar result has been independently obtained by Frankel & Sivashinsky (1982). This clarifies the controversy concerning the stabilizing or destabilizing influence of viscosity reported, for example by Markstein (1964) and by Zel'dovich *et al.* (1980).

The mathematical formulation is given in §2. The analysis is presented in §3, which can be omitted in a first reading. In §4 the results are discussed and compared with the previous studies. A quantitative discussion and the concluding remarks are presented in §5.

2. Formulation

The flame model is identical with that of I, and the formulation is similar. The reader is referred to I for more details. Slight modifications are introduced here to take into account the acceleration due to gravity. Furthermore, the laminar character of the upstream flow and the linearization appropriate to the stability analysis introduce further simplifications.

Attention is restricted to an overall exothermic reaction controlled by one limiting reactant. An Arrhenius rate is adopted with a large activation energy $\beta \ge 1$; d and $d/u_{\rm L}$ are the units of length and time respectively, $x = \alpha(y, t)$ defines the location of the reactive zone and the moving coordinates; $\xi = x - \alpha$, $\eta = y, \tau = t$ are introduced, $r = \rho/\rho_{-\infty}$ denotes the density ratio, U and v are the non-dimensional longitudinal (x-direction) and transverse (y-direction) components of the flow velocity. The subscript \mathbf{L} refers to the one-dimensional steady-state solution, and the nonsubscripted variables refer to the perturbation. The linearized versions of the conservation equations are written in the moving system of coordinates; we introduce

$$s_{\rm L} = r_{\rm L} U_{\rm L}, \quad s = r_{\rm L} \left(U - \frac{\partial \alpha}{\partial t} \right) + r U_{\rm L},$$
 (2)

[†] According to Zel'dovich (1981) there may exist steady but non-planar flame fronts propagating in tubes, which could be intrinsically stable with respect to hydrodynamical effects. In such cases, which are beyond the scope of the present paper, the stability analysis is expected to be of a different nature. Such a situation cannot be excluded also for planar fronts in 'eurved flows' such as a stagnation-point flow.

‡ In order to simplify the notation, only one transverse coordinate is specified.

where $r_{\rm L} = 1$, $U_{\rm L} = 1$, r = 0, U = 0 when $\xi \to -\infty$. Here s identifies the nondimensional longitudinal mass flux in the moving coordinate system. The mass conservation gives $s_{\rm L} = 1$ for the steady-state solution and

$$\frac{\partial r}{\partial \tau} + \frac{\partial s}{\partial \xi} + \frac{\partial (r_{\rm L} v)}{\partial \eta} = 0$$
(3)

for the perturbation.

Energy conservation is written with the non-dimensional temperature

$$\theta = (T - T_{-\infty})/(T_{\rm f} - T_{-\infty}).$$

When the Mach number $M = [\rho_{-\infty} u_{\rm L}^2/p_{-\infty}]^{\frac{1}{2}}$ is negligibly small, the conservation of energy outside the reaction zone gives

$$\theta_{\rm L} = \begin{cases} e^{\xi} & (\xi < 0, \\ 1 & (\xi > 0) \end{cases}$$

for the steady-state solution and

$$r_{\rm L}\frac{\partial\theta}{\partial\tau} + \frac{\partial\theta}{\partial\xi} + \left(s + \frac{\partial^2\alpha}{\partial\eta^2}\right)\frac{\partial\theta_{\rm L}}{\partial\xi} = \left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{\partial^2\theta}{\partial\eta^2}\right) \tag{4}$$

for the perturbation, with the boundary conditions

$$\begin{aligned} \theta &= 0 \quad (\xi \to -\infty), \\ \theta &= 0 \quad (\xi \to +\infty). \end{aligned}$$

r is obtained from the solution of (4) by using the nearly isobaric conditions ($M \ll 1$)

$$r_{\rm L} + r = \left[1 + \frac{(\theta_{\rm L} + \theta)\gamma}{1 - \gamma}\right]^{-1},\tag{5}$$

to give for the steady-state solution

$$r_{\rm L} = \begin{cases} \left[1 + \frac{e^{\xi}\gamma}{1-\gamma} \right]^{-1} & (\xi < 0), \\ 1-\gamma & (\xi > 0), \end{cases}$$

and $r = -(\gamma/(1-\gamma))r_{\rm L}^2\theta$ for the perturbation.

Outside the reaction zone, species conservation gives

$$\psi_{\rm L} = \begin{cases} 1 - e^{{\rm L}\xi} & (\xi < 0), \\ 0 & (\xi > 0), \end{cases}$$

and

$$r_{\rm L} \frac{\partial \psi}{\partial \tau} + \frac{\partial \psi}{\partial \xi} + \left(s + \frac{1}{L} \frac{\partial^2 \alpha}{\partial \eta^2}\right) \frac{\partial \psi_{\rm L}}{\partial \xi} = \frac{1}{L} \left(\frac{\partial^2 \psi}{\partial \xi^2} + \frac{\partial^2 \psi}{\partial \eta^2}\right), \tag{6}$$
$$\psi = 0 \quad (\xi \to -\infty, \quad \xi > 0).$$

with

In (4) and (6) the thermal and molecular diffusivities have been considered as temperature-independent. The Lewis number L is defined by $L = D_{\rm th}/D_{\rm mol}$, where $D_{\rm mol}$ is the molecular diffusivity of the reactive species that is in deficit, so that it controls the reaction rate.

Following Joulin & Clavin (1979), whenever the reduced activation energy β of the exothermal reaction is large, the chemical production is located at $\xi = 0$ in a thin reaction zone of thickness d/β and this production term induces jump conditions for the space derivative of ψ and θ :

$$\left[\frac{\partial\theta}{\partial\xi} + \frac{1}{L}\frac{\partial\psi}{\partial\xi}\right]_{\xi=0^{-}}^{\xi=0^{+}} = 0, \quad \frac{\partial\theta}{\partial\xi}\Big|_{\xi=0^{-}} = \frac{1}{2}\beta\theta(\xi=0), \tag{7}$$

 $\psi(\xi=0)=0$ defines the origin $(\xi=0)$ and $\theta(\xi=0)=O(1/\beta)$. These relations are valid asymptotically in the limit $\beta \to \infty$ up to $O(1/\beta)$ with the restriction $1-1/L = O(1/\beta)$. This is not too restrictive for gaseous mixtures where the Lewis number is close to one. In this case by inspection of (4) and (6), the difference $\psi - \theta$ appears to be of order $1/\beta$ and $\theta(\epsilon=0)$ is seen to be proportional to 1-1/L in such a way that the relevant parameter of order unity in the limit $\beta \to \infty$ is $l = \beta(1-1/L)$. In the following, unless explicitly specified, all the quantities should be understood as being O(1) in the limit as $\beta \to \infty$. Furthermore, at order ϵ^2 , as will be explained later, (4) and (6) reduce to quasi-planar and quasi-steady equations, $\partial\theta/\partial\tau$, $\partial^2\theta/\partial\eta^2$, $\partial\psi/\partial\tau$, $\partial^2\psi/\partial\eta^2 = o(\epsilon^2)$, and thus can be solved very easily in terms of s and $\partial^2\alpha/\partial\eta^2$. Then, by prescribing the boundary condition (7), one obtains an integral relation for $s(\xi)$ which, at this order ϵ^2 reduces to

$$\int_{-\infty}^{0} \left(s + \frac{\partial^2 \alpha}{\partial \eta^2}\right) e^{\xi'} d\xi' = \frac{1}{2} l \left[\int_{-\infty}^{0} s(\xi') \left(1 + \xi'\right) e^{\xi'} d\xi' - \frac{\partial^2 \alpha}{\partial \eta^2} \right].$$
(8)

At this stage let us outline the method. As soon as v is known inside the flame thickness d, a direct integration of (3) gives the variation of s through d, and (8) provides its upstream value $(\xi \to -\infty)$. Then, s being completely known inside the flame thickness, (2) gives the flame velocity relative to the upstream flow as well as the modification to the longitudinal gas velocity U inside d. For the planar solution where $s_{\rm L} = 1$ and $U_{\rm L}(\xi) = 1/r_{\rm L}$ one has

$$U_{\rm L}(\xi) = \begin{cases} 1 + e^{\xi} \frac{\gamma}{1 - \gamma} & (\xi < 0), \\ 1 & (9a) \end{cases}$$

$$\left\{\frac{1}{1-\gamma} \qquad (\xi > 0). \right.$$
(9b)

The non-dimensional pressure, i.e. the ratio of the pressure to its value in the fresh mixture, is denoted by $1 + M^2p$. p is given by solving the longitudinal momentum equation, which, when the viscous diffusivities are supposed temperature-independent, can be written as

$$r_{\rm L}\frac{\partial U}{\partial \tau} + \left(s + P\frac{\partial^2 \alpha}{\partial \eta^2}\right)\frac{\partial U_{\rm L}}{\partial \xi} + \frac{\partial U}{\partial \xi} = -r\frac{gd}{u_{\rm L}^2} - \frac{\partial p}{\partial \xi} + P\left(\frac{\partial^2}{\partial \xi^2} + \frac{\partial^2}{\partial \eta^2}\right)U + P'\frac{\partial}{\partial \xi}\left(\frac{\partial U}{\partial \xi} + \frac{\partial v}{\partial \eta}\right),\tag{10}$$

with p = 0, U = 0 as $\xi \to -\infty$.

The pressure in the steady-state solution is given by the corresponding equation for $p_{L}(\xi)$:

$$\frac{\partial U_{\rm L}}{\partial \xi} = -r_{\rm L} \frac{gd}{u_{\rm L}^2} - \frac{\partial p_{\rm L}}{\partial \xi} + (P + P') \frac{\partial^2 U_{\rm L}}{\partial \xi^2},\tag{11}$$

with $p_{\rm L} = 0$, $U_{\rm L} = 1$ as $\xi \rightarrow -\infty$: *P* and *P'* are the Prandtl numbers based on the first and second coefficients of viscosity respectively.

v is given by the transverse component of momentum conservation:

$$r_{\rm L}\frac{\partial v}{\partial \tau} + \frac{\partial v}{\partial \xi} = -\frac{\partial p}{\partial \eta} + \frac{\partial \alpha}{\partial \eta}\frac{dp_{\rm L}}{d\xi} + P\left(\frac{\partial^2}{\partial \xi^2} + \frac{\partial^2}{\partial \eta^2}\right)v + P'\frac{\partial}{\partial \eta}\left(\frac{\partial U}{\partial \xi} + \frac{\partial v}{\partial \eta}\right) - P'\frac{\partial \alpha}{\partial \eta}\frac{\partial^2 U_{\rm L}}{\partial \xi^2}$$
(12)

with v = 0 as $\xi \to -\infty$.

The momentum equations (10) and (12) are valid everywhere in the gas flow, and, since they do not contain the chemical-production term, the jump conditions through

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the reactive zone concerning p, U and v are obtained directly on integration over this zone. Because U and v are continuous on the reactive zone, it is found that

$$[p]_{\xi=0_{-}}^{\xi=0_{+}} = (P+P') \left[\frac{\partial U}{\partial \xi}\right]_{\xi=0_{-}}^{\xi=0_{+}},$$
(13a)

$$[p_{\mathbf{L}}]_{\boldsymbol{\xi}=\boldsymbol{0}_{-}}^{\boldsymbol{\xi}=\boldsymbol{0}_{+}} = (P+P') \left[\frac{\partial U_{\mathbf{L}}}{\partial \boldsymbol{\xi}} \right]_{\boldsymbol{\xi}=\boldsymbol{0}_{-}}^{\boldsymbol{\xi}=\boldsymbol{0}_{+}}, \tag{13b}$$

$$-\left(\frac{\partial \alpha}{\partial \eta}\right) \left[\frac{\partial U_{\rm L}}{\partial \xi}\right]_{\xi=0_{-}}^{\xi=0_{+}} = \left[\frac{\partial v}{\partial \xi}\right]_{\xi=0_{-}}^{\xi=0_{+}}.$$
 (13c)

The system of linear equations (3), (4), (6), (10), (12) with the jump conditions (7) and (13) is solved by perturbation in $\epsilon = d/\Lambda$, up to the second order ϵ^2 , by using, as explained in §2, a two-scale method. All the transverse derivatives $\partial/\partial \eta$ are by definition of order ϵ . Inside the flame the longitudinal derivative $\partial/\partial \xi$ is of order unity. Outside the flame, θ and ψ are constant (at the dominant order in the asymptotic limit $\beta \to \infty$), and, the spatial scale in the variation of the flow field being Λ , the spatial derivatives, including $\partial/\partial \xi$, are of order ϵ . Furthermore, as is shown by the analyses of Darrieus and Landau, the reduced flow perturbation has an intensity of order ϵ and the evolution is on a reduced timescale of order $1/\epsilon$, $\partial/\partial t = O(\epsilon)$. Thus, according to (3), $\partial s/\partial \xi = O(\epsilon^2)$ inside the flame, and, according to (4) and (6), $\theta = O(\epsilon^2)$, $\psi = O(\epsilon^2)$. So, as was anticipated in writing (8), the equations (4) and (6) appear to be quasi-steady and quasi-planar at order ϵ^2 . Additionally, one introduces G = O(1), defined by

$$\frac{gd}{u_{\rm L}^2} = \epsilon G. \tag{14}$$

The reason is that the critical wavelength Λ_c is predicted by a phenomenological analysis to be given by $\Lambda_c = \pi u_L^2/(1-\gamma)g$, which greatly exceeds the flame thickness d as soon as $u_L > 5$ cm/s. Furthermore, the final result will show that for very slow-burning flames $u_L < 5$ cm/s, and for the diffusive properties of the usual reactive components the front is always stable, so that (14) can be used in the stability analysis without loss of generality.

3. Analysis

The flame position is developed in powers of ϵ :

$$\alpha = \alpha_0(Y, T) + \epsilon \alpha_1(Y, T) + o(\epsilon)$$
(15)

where $Y = \epsilon \eta$, $T = \epsilon \tau$. In addition to the reactive zone of thickness d/β , which is completely described by the jump conditions (7) and (13), three more zones are considered: the thermal diffusive zone of thickness d, and, outside, the two hydrodynamical zones, denoted by the subscripts $-\infty$ and $+\infty$ for the fresh mixture and the burnt gas respectively (see figure 1). In these two hydrodynamical zones of thickness Λ the temperature and the concentration are constant, and the solutions are written in the form

$$\begin{aligned} u_{\pm\infty} &= \epsilon U_{\pm\infty}(X, Y, T), \quad v_{\pm\infty} = \epsilon V_{\pm\infty}(X, Y, T), \\ p_{\pm\infty} &= \epsilon P_{\pm\infty}(X, Y, T), \quad s_{\pm\infty} = \epsilon S_{\pm\infty}(X, Y, T), \\ \psi_{\pm\infty} &= \theta_{\pm\infty} = 0, \end{aligned}$$

$$(16)$$

where $X = \epsilon x$, $Y = \epsilon \eta$, $T = \epsilon \tau$. Further, $r_{\rm L} = \psi_{\rm L} = 1$, $\theta_{\rm L} = 0$ and $\partial p_{\rm L} / \partial X = -G$ in the upstream zone $-\infty$, and $r_{\rm L} = 1 - \gamma$, $\psi_{\rm L} = 0$, $\theta_{\rm L} = 1$ and $\partial p_{\rm L} / \partial X = -(1 - \gamma)G$ in the downstream zone $+\infty$.



FIGURE 1. The streamlines associated with a wrinkled front. In the region convex towards the fresh mixture, $U_{-\infty} < 0$. In order to restore a relative velocity equal to the local flame velocity $u_{\rm L}$, the front moves toward the fresh mixture, producing the hydrodynamical instability. However, in this region the transverse convection induced by the deflection of the streamlines enhances the transverse heat diffusion, thus decreasing the local flame velocity, enhances also the stabilizing effect of the diffusion.

The approximation (5) associated with (16) implies that $r_{\pm\infty} = 0$, and thus these two zones are simply controlled by incompressible fluid mechanics:

$$(2) \Rightarrow S_{+\infty} = (1 - \gamma) \left(U_{+\infty} - \frac{\partial \alpha}{\partial T} \right), \quad S_{-\infty} = \left(U_{-\infty} - \frac{\partial \alpha}{\partial T} \right), \quad (17a, b)$$

$$(3) \Rightarrow \frac{\partial U_{+\infty}}{\partial X} + \frac{\partial V_{+\infty}}{\partial Y} = 0, \quad \frac{\partial U_{-\infty}}{\partial X} + \frac{\partial V_{-\infty}}{\partial Y} = 0, \quad (17c, d)$$

$$(10) \quad \left((1-\gamma)\frac{\partial U_{+\infty}}{\partial T} + \frac{\partial U_{+\infty}}{\partial X} = -\frac{\partial P_{+\infty}}{\partial X} + \epsilon P\left(\frac{\partial^2 U_{+\infty}}{\partial X^2} + \frac{\partial^2 U_{+\infty}}{\partial Y^2}\right), \quad (17e)$$

$$\begin{pmatrix} (10) \Rightarrow \\ \frac{\partial U_{-\infty}}{\partial T} + \frac{\partial U_{-\infty}}{\partial X} = -\frac{\partial P_{-\infty}}{\partial X} + \epsilon P \left(\frac{\partial^2 U_{-\infty}}{\partial X^2} + \frac{\partial^2 U_{-\infty}}{\partial Y^2} \right),$$
 (17*f*)

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$$\int (1-\gamma)\frac{\partial V_{+\infty}}{\partial T} + \frac{\partial V_{+\infty}}{\partial X} = -\frac{\partial P_{+\infty}}{\partial Y} - \frac{\partial \alpha}{\partial Y} (1-\gamma) G + \epsilon P \left(\frac{\partial^2 V_{+\infty}}{\partial X^2} + \frac{\partial^2 V_{+\infty}}{\partial Y^2}\right), \quad (17g)$$

$$(12) \Rightarrow \left\{ \frac{\partial V_{-\infty}}{\partial T} + \frac{\partial V_{-\infty}}{\partial X} = -\frac{\partial P_{-\infty}}{\partial Y} - \frac{\partial \alpha}{\partial Y} G + \epsilon P \left(\frac{\partial^2 V_{-\infty}}{\partial X^2} + \frac{\partial^2 V_{-\infty}}{\partial Y^2} \right).$$
(17*h*)

Using the normal-modes decomposition

$$\alpha = \mathscr{C}e^{iKY+\Sigma T}, \quad U_{\pm\infty} = \mathscr{U}_{\pm\infty}(X) e^{iKY+\Sigma T}, \quad V_{\pm\infty} = \mathscr{V}_{\pm\infty}(X) e^{iKY+\Sigma T}, \\ P_{+\infty} = -(1-\gamma)\alpha G + \mathscr{P}_{+\infty}(X) e^{iKY+\Sigma T}, \quad P_{-\infty} = -\alpha G + \mathscr{P}_{-\infty}(X) e^{iKY+\Sigma T},$$
 (18)

and assuming bounded values everywhere for $\Sigma > 0$, the solution of the system (17) can be written in the following form:

$$\begin{aligned} \mathscr{P}_{+\infty}(X) &= \mathscr{P}_{+\infty}(0) \, e^{-KX}, \quad \mathscr{P}_{-\infty}(X) = \mathscr{P}_{-\infty}(0) \, e^{+KX}, \\ \mathscr{U}_{+\infty}(X) &= \mathscr{A} \exp\left\{\frac{1 - [1 + 4\epsilon P((1 - \gamma)\Sigma + \epsilon PK^2)]^{\frac{1}{2}}}{2\epsilon P}x\right\} + \frac{K}{(1 - \gamma)\Sigma - K} \, \mathscr{P}_{+\infty}(0) \, e^{-KX}, \\ \mathscr{U}_{-\infty}(X) &= -\frac{K}{K + \Sigma} \, \mathscr{P}_{-\infty}(0) \, e^{KX}, \quad \mathscr{V}_{+\infty}(X) = \frac{i}{K} \frac{\partial}{\partial X} \, \mathscr{U}_{+\infty}(X), \quad \mathscr{V}_{-\infty}(X) = i \mathscr{U}_{-\infty}(X) \end{aligned}$$
(19)

These linear solutions are valid at all orders of the development in powers of ϵ . Notice that there is another root on the $-\infty$ side that does not appear in (19) because it corresponds to an exponential decrease on the x-scale of the preheated zone of the flame.

The four constants of integration, \mathscr{C} , \mathscr{A} and $\mathscr{P}_{\pm\infty}(0)$, have to be determined afterwards. In order to carry out this determination, the flame structure has to be investigated to provide the jumps through the flame thickness, $\mathscr{P}_{+\infty}(0) - \mathscr{P}_{-\infty}(0)$, $\mathscr{U}_{+\infty}(0) - \mathscr{V}_{-\infty}(0)$. This will be done up to the second order in the development in powers of ϵ .

In the third zone, corresponding to the preheated zone of the flame, the solutions are written in the following form:

$$U = \epsilon U_{-\infty}(\epsilon x, Y, T) + \epsilon \hat{u}_1(\xi, Y, T) + \epsilon^2 \hat{u}_2(\xi, Y, T) + o(\epsilon^2), \qquad (20a)$$

$$v = e V_{-\infty}(ex, Y, T) + e \hat{v}_1(\xi, Y, T) + e^2 \hat{v}_2(\xi, Y, T) + o(e^2),$$
(20b)

$$p = \epsilon P_{-\infty}(\epsilon x, Y, T) + \epsilon \hat{p}_1(\xi, Y, T) + \epsilon^2 \hat{p}_2(\xi, Y, T) + o(\epsilon^2), \qquad (20c)$$

$$p_{\rm L} = p_{\rm L0}(\xi) + \epsilon p_{\rm L1}(\xi, \, Y, \, T), \tag{20d}$$

$$s = \epsilon S_{-\infty}(\epsilon x, Y, T) + \epsilon \hat{s}_1(\xi, Y, T) + \epsilon^2 \hat{s}_2(\xi, Y, T) + o(\epsilon^2), \tag{20e}$$

$$\theta = \epsilon^2 \hat{\theta}_2(\xi, Y, T) + o(\epsilon^2), \quad \psi = \epsilon^2 \hat{y}_2(\xi, Y, T) + o(\epsilon^2), \quad (20f, g)$$

where $\hat{\theta}_2(\xi = 0) = O(\beta^{-1})$, and thus should be considered as zero. Furthermore, all the quantities $eU_{-\infty}(ex)$, $eV_{-\infty}(ex)$, $eS_{-\infty}(ex)$, $eP_{-\infty}(ex)$ have to be developed in powers of e.

It will be shown in the following that, when $\xi \to -\infty$, all the quantities tagged with the circumflex \sim go to zero like $\xi^n e^{\xi}$. Thus the expressions (20) valid in the preheated zone are automatically matched with expressions (16) corresponding to the upstream hydrodynamical region labelled by $-\infty$.

The solution of (3) gives directly $\hat{s}_1 = 0$,

$$(21\,a)$$

$$\hat{s}_2 = \frac{\partial V_{-\infty}}{\partial Y} \int_{\xi}^{-\infty} (r_{\mathrm{L}}(\xi') - 1) \, d\xi' + \int_{\xi}^{-\infty} r_{\mathrm{L}}(\xi') \frac{\partial \hat{v}_1}{\partial Y}(\xi') \, d\xi'.$$
(21b)

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When the first orders of s given by (21) are introduced in (8) one obtains, at the order ϵ , $S_{-\infty,0}(X=0) = 0$ for $\xi \to -\infty$, which gives, according to (15) and (17b).

$$\frac{\partial \alpha_0}{\partial T} = U_{-\infty,0}(X=0).$$
(22)

Then (2) and (10) give

$$\hat{u}_1 = 0, \quad \hat{p}_1 = 0.$$

According to (11),

$$\frac{\partial p_{\mathbf{L}0}}{\partial \xi} = -\frac{\partial U_{\mathbf{L}}}{\partial \xi} + (P+P')\frac{\partial^2}{\partial \xi^2} U_{\mathbf{L}}.$$

Then, using (9), a direct integration of (12) provides the expression for \hat{v}_1 :

$$\hat{v}_1 = -\left[\partial \alpha_0 / \partial Y\right] \frac{\gamma}{1 - \gamma} e^{\xi},\tag{23}$$

where the last jump condition of (13) has been used to determine the constant of integration. When the result (23) is introduced in (21), an expression for \hat{s}_2 is obtained, and (8) with (20) leads to the expression for $S_{-\infty}(X=0)$. One obtains

$$S_{-\infty 1}(X=0) = \frac{\mathscr{L}}{d} \left[\frac{-\partial^2 \alpha_0}{\partial Y^2} + \frac{\partial U_{-\infty,0}}{\partial X} \Big|_{X=0} \right],$$
(24*a*)

$$\hat{s}_{2}(\xi=0) = \left[-\frac{\partial U_{-\infty,0}}{\partial X} \Big|_{X=0} + \frac{\partial^{2} \alpha_{0}}{\partial Y^{2}} \right] \ln \frac{1}{1-\gamma}, \qquad (24b)$$

where

$$\frac{\mathscr{L}}{d} = \frac{1}{\gamma} \ln \frac{1}{1-\gamma} + \frac{1}{2}\beta \left(1 - \frac{1}{L}\right) \frac{1-\gamma}{\gamma} D(\gamma), \qquad (25a)$$

with

$$D = \int_0^{\gamma/(1-\gamma)} dx \, x^{-1} \ln (1+x). \tag{25b}$$

Recalling that $\hat{\theta}_2(\xi = 0) = O(\beta^{-1})$, (2) specifies the value of $\hat{u}_2(\xi = 0)$:

$$\hat{u}_2(\xi=0) = \frac{\gamma}{1-\gamma} S_{-\infty 1}(X=0) + \frac{1}{1-\gamma} \hat{s}_2(\xi=0).$$
(26)

The integration of the order ϵ^2 of (10) yields the value of \hat{p}_2 :

$$\begin{split} \hat{p}_{2}(\xi=0) &= -2\frac{\gamma}{1-\gamma}S_{-\infty1}(X=0) - \frac{2}{1-\gamma} \left(\frac{\partial^{2}\alpha_{0}}{\partial Y^{2}} - \frac{\partial U_{-\infty,0}}{\partial X}\Big|_{X=0}\right) \ln\frac{1}{1-\gamma} \\ &+ \frac{\gamma}{1-\gamma} \left(1-P-P'\right) \frac{\partial^{2}\alpha_{0}}{\partial Y^{2}} + \left(\ln\frac{1}{1-\gamma}\right) \frac{\partial U_{-\infty,0}(X=0)}{\partial T} + \left(P+P'\right) \frac{\partial \hat{u}_{2}}{\partial \xi}\Big|_{\xi=0} \end{split}$$

and the integration of (12) provides the value of $\hat{v}_2(\xi = 0)$:

$$\hat{\vartheta}_{2}(\xi=0) = -\frac{\gamma}{1-\gamma} \frac{\partial \alpha_{1}}{\partial Y} + P\left(\frac{\partial V_{+\infty,0}}{\partial X} - \frac{\partial V_{-\infty,0}}{\partial X}\right)_{X=0} + \left[\frac{\partial V_{-\infty,0}(X=0)}{\partial T} + \frac{\partial^{2}\alpha_{0}}{\partial T \partial Y} + G\frac{\partial \alpha_{0}}{\partial Y}\right] \ln \frac{1}{1-\gamma}, \quad (28)$$

where the jump conditions (13) through the reactive zone have been used to determine the constant of integration.

4. Results and discussion

The definitions (20) and the results (22)-(28) specify the boundary and jump conditions across the flame up to the order ϵ^2 , providing the necessary information for the solution of the hydrodynamical problem (17), (19). To begin with, let us consider the modification of the flame velocity obtained from (22), (24*a*) by using the definitions (16) and (17*b*):

$$\frac{\partial \alpha}{\partial t} = u_{-\infty}(X=0) - s_{-\infty}(X=0),$$

$$s_{-\infty}(X=0) = \frac{\mathscr{L}}{d} \left(\frac{\partial u_{-\infty}}{\partial x} \Big|_{x=0} - \frac{\partial^2 \alpha}{\partial y^2} \right) + o(\epsilon^2),$$
(29)

associated with the following modification $T_{\rm b} - T_{\rm f}$ of the temperature of combustion:

$$(T_{\rm b} - T_{\rm f})/(T_{\rm f} - T_{-\infty}) = + \left(1 - \frac{1}{L}\right) \left(\frac{1 - \gamma}{\gamma}\right) D(\gamma) \left(\frac{\partial u_{-\infty}}{\partial x}\Big|_{x=0} - \frac{\partial^2 \alpha}{\partial y^2}\right) + o(\epsilon^2), \quad (29')$$

where \mathscr{L}/d and $D(\gamma)$ are given by (25a, b).

Equations (29) and (29') are valid in the limit $\beta \to \infty$ for $1 - L = O(b^{-1})$, and correspond to the linearized version of the result obtained in I. $u_{-\infty}(X=0)$ represents the convective effect upon the flame movement produced by the modification of the upstream gas flow induced by the front wrinkling. This was the only effect retained in the early analyses of Darrieus (1938) and Landau (1944), where $s_{-\infty}$ was set equal to zero. $s_{-\infty}(X=0)$ represents the change of the front velocity relative to the upstream gas flow produced by the modification of the flame structure involved in the front wrinkling. Such a modification was first introduced by Markstein (1951) through a phenomenological relation

$$\frac{\partial \alpha}{\partial t} = u_{-\infty} - \frac{\mathscr{L}}{R}, \quad \frac{\mathscr{L}}{d} > 0, \tag{30}$$

where 1/R is the curvature of the front, which, in the weakly curved case, is approximated by $-(\partial^2 \alpha / \partial y^2) d^{-1}$. Except for a supplementary term $\partial u_{-\infty} / \partial x|_{x=0}$, which will be commented on later in the paper, (29) is similar to (30), and provides the expression of the 'Markstein phenomenological length' \mathscr{L} . This modification of the flame structure is produced by the transverse diffusive flux of mass and energy appearing when the front is wrinkled, and is also affected by the corresponding transverse convective flux produced by the deflection of the streamlines. This last effect disappears when the gas expansion is neglected, $\gamma = 0$, as in the purely 'diffusive-thermal' model first studied by Barenblatt *et al.* (1962). In this case, the hydrodynamical effects also disappear completely, $u_{-\infty}(X) = 0$, and the equation (29) for the evolution of the front reduces to a simple diffusion equation

$$\frac{\partial \alpha}{\partial t} = \frac{\mathscr{L}}{d} \frac{\partial^2 \alpha}{\partial y^2},\tag{31}$$

with a 'Markstein length'

$$\frac{\mathscr{L}}{d} = 1 + \frac{1}{2}\beta\left(1 - \frac{1}{L}\right) \tag{32}$$

given by (25a) for $\gamma = 0$. These results, valid in the limit $\beta \to \infty$ for $l \equiv \beta(1-1/L) = O(1)$, are in agreement with that of Barenblatt *et al.* (1962) and can be easily interpreted from a physical point of view (Clavin 1982). The term 1 in (32) represents the contribution of the thermal relaxation of a wrinkled front associated with a constant value of the maximum of the temperature of combustion. The second

term on the right-hand side of (32) describes the effect of the modification of the flame velocity induced by the change of the temperature of combustion resulting in the competitive diffusion processes of heat and mass developed in the transverse direction by the front wrinkling. Further analytical studies of the 'diffusive-thermal' model have been recently carried by Sivashinsky (1977a) and by Joulin & Clavin (1979) to get a better understanding of the dynamical properties of the front. One of the motivations for these works was that, according to (32), the diffusion coefficient \mathscr{L}/d in (31) may easily change sign for the diffusive properties of the usual rich reactive mixture, providing an interpretation of the instability threshold for the appearance of the cellular structures. But, when the transverse convection is taken into account, the result (25a) shows that \mathcal{L}/d can no longer easily reach zero, and indeed is strictly positive even for the lightest limiting component involved in the usual reactive mixtures (an exception is possible for hydrogen). Thus, in agreement with the assumption (30) used by Markstein but contrary to the prediction of the 'diffusivethermal model', the modification of the flame structure by the front wrinkling produces, for the usual hydrocarbon reactive mixtures, a stabilizing effect ($\mathscr{L}/d > 0$). Nevertheless, this 'diffusive-thermal model' is still interesting, because it allows us to investigate the diffusive phenomena in the complete range of wavenumbers (the assumption $\epsilon \ll 1$ is not necessary). Thus, for example, an interesting diffusive mechanism of instability leading to travelling waves has been pointed out by Sivashinsky (1977a) and Joulin & Clavin (1979) for lean mixtures of heavy hydrocarbons.

A comparison between (29) and (30) reveals that, as shown previously in the study of Eckhaus (1961), it is not only the curvature of the front that is involved in the modification of the flame structure, but also the quantity $\partial u_{-\infty}/\partial x$, which is related to the inhomogeneities of the upstream flow. This effect has been known for a long time to exist, and it was referred to as the 'Karlowitz effect' in the traditional literature of combustion (see e.g. Lewis & von Elbe 1961). Eckhaus (1961) interprets (29) as the curvature of the front relative to the deformation of the streamlines. The recent result by Clavin & Joulin (1982), concerning the nonlinear case of finite amplitudes of deformation, provides the precise formulation of this effect, which is shown to be more complex.

It is clear from (29) that the complete description of the dynamical properties of the front requires the solution of the purely hydrodynamical problem for determining the modification of the upstream flow field $u_{-\infty}(X)$, $v_{-\infty}(X)$. This was tentatively undertaken by different early analytical studies, for example those of Eckhaus (1961), Chu & Parlange (1962), Maxworthy (1962) and Istratov & Librovich (1966). However, in the absence of a systematic technique to solve the structure of the wrinkled flame, these analytical studies lead to results that are contradictory and not completely satisfactory. For example, they do not reduce, in the limit $\gamma = 0$, to the rigorous result expressed by (31) and (32). More interesting is the work of Sivashinsky (1977b), but this is exact only for small values of γ . There is also an intermediate study by Lazarev & Pleshanov (1980), in which an incomplete analysis of the flame structure is carried out in such a way that the solution requires the introduction of a phenomenological relation such as (30), but in which the 'Karlowitz effect' is overlooked. More relevant are the results obtained by Markstein (1964) in his early phenomenological analysis, in which the study of the flame structure is not addressed at all, but is replaced by relevant phenomenological relations on the flame front considered as a surface of discontinuity. On both sides of the front, the flow is treated as in the pioneering works

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of Darrieus (1938) and Landau (1944) in the inviscid and incompressible approximation.

Here, the problem is completely solved analytically for small values of $\epsilon = d/\Lambda$ up to order ϵ^2 , but for an arbitrary gas expansion γ and for arbitrary Prandtl numbers. It is proved that the problem is reduced, effectively, to solving for an incompressible flow in the two hydrodynamical zones, with the boundary and jump conditions determined by the analysis of the flame structure of §3. In fact, Markstein (1964) used a phenomenological relation similar to (29), which is proved here and in I to be exact to order ϵ^2 , but the jump conditions used for the pressure and the flow field were incomplete.

Thus (26) gives the jump condition, through the flame thickness, for the longitudinal flow velocity:

$$u_{+\infty}(0) - u_{-\infty}(0) = \left[\frac{\gamma}{1-\gamma}\frac{\mathscr{L}}{d} - \frac{1}{1-\gamma}\ln\frac{1}{1-\gamma}\right] \left[\frac{\partial u_{-\infty}}{\partial x}\Big|_{x=0} - \frac{\partial^2 x}{\partial y^2}\right] + o(\epsilon^2).$$
(33)

The first term $[\gamma/(1-\gamma)] \mathcal{L}/d$ corresponds to the modification of the longitudinal velocity associated with a constant value $eS_{-\infty}(X=0)$ of the longitudinal mass flux across the flame. This term is the only one appearing in the semi-phenomenological analysis of Markstein (1964) (see also Zel'dovich *et al.* 1980). But an additional term appears in (33), corresponding to the modification \hat{s} of s produced by the transverse convection due to the deflection of the streamlines through the flame thickness.

Equation (27) lead to the modification of the pressure through the flame thickness:

$$p_{+\infty}(0) - p_{-\infty}(0) = -2\frac{\gamma}{1-\gamma} \left[\frac{\mathscr{L}}{d} - \frac{1}{\gamma} \ln \frac{1}{1-\gamma}\right] \left[\frac{\partial u_{-\infty}}{\partial x}\Big|_{x=0} - \frac{\partial^2 \alpha}{\partial y^2}\right] + \frac{\gamma}{1-\gamma} (1-P-P')\frac{\partial^2 \alpha}{\partial y^2} + (P+P') \left[\frac{\partial u_{+\infty}}{\partial x}\Big|_{x=0} - \frac{\partial u_{-\infty}}{\partial x}\Big|_{x=0}\right] + \left(\ln \frac{1}{1-\gamma}\right) \frac{\partial u_{-\infty}(0)}{\partial t} + o(\epsilon^2).$$
(34)

The first term corresponds to the linearized Bernoulli law associated with (33). Only a part of this term was retained in the phenomenological analysis mentioned above. Among the additional terms, $[\gamma/(1-\gamma)]\partial^2 \alpha/\partial y^2$, which produces an effect similar to surface tension, will be seen to be one of the most important. According to (18), the jump condition associated with $\epsilon \mathscr{P}_{\pm \infty}$ is obtained by adding $-\epsilon \gamma G \alpha$ to (34). Finally, (23) and (28) specify the jump condition for the transverse velocity:

$$v_{+\infty}(0) - v_{-\infty}(0) = -\frac{\gamma}{1-\gamma} \frac{\partial \alpha}{\partial y} + P\left(\frac{\partial v_{+\infty}}{\partial x} - \frac{\partial v_{-\infty}}{\partial x}\right)_{x=0} + \left[\frac{\partial v_{-\infty}(X=0)}{\partial t} + \frac{\partial^2 \alpha}{\partial t \partial y} + G\frac{\partial \alpha}{\partial y}\right] \ln \frac{1}{1-\gamma} + o(\epsilon^2).$$
(35)

When the solutions (19) are introduced into (29), (33)–(35), the four constants of integration \mathscr{C} , \mathscr{A} , $\mathscr{P}_{\pm}(0)$ are given by the solution of four homogeneous linear equations. In order to obtain a non-trivial solution the corresponding 4×4 determinant must be zero. This leads to the following dispersion relation:

$$A(k)\sigma^{2} + B(k)\sigma + C(k) = 0,$$
(36)

where $\sigma = \epsilon \Sigma$, $k = \epsilon K$, and where the coefficients A, B, C are given by

$$A = (2-\gamma) + \gamma \left[\frac{\mathscr{L}}{d} - \frac{1}{\gamma} \ln \frac{1}{1-\gamma}\right] k, \qquad (37a)$$

$$B = 2k + \left[\frac{2}{1-\gamma}\frac{\mathscr{L}}{d} - \frac{2}{1-\gamma}\ln\frac{1}{1-\gamma}\right]k^2,$$
(37b)

$$C = \frac{\gamma}{1-\gamma} k \left\{ \frac{gd}{u_{\rm L}^2} (1-\gamma) - k \left[1 + \epsilon G(1-\gamma) \left(\frac{\mathscr{L}}{d} - \frac{1}{\gamma} \ln \frac{1}{1-\gamma} \right) \right] + k^2 \left[1 + \frac{2+\gamma}{\gamma} \frac{\mathscr{L}}{d} - \frac{2}{\gamma} \ln \frac{1}{1-\gamma} \right] \right\}.$$
 (37c)

Equations (36) and (37) correspond to the three first orders of the expansion in ϵ of the exact dispersion relation.

The first point of interest is that the Prandtl numbers P and P' do not appear in the final result (36), (37). This is surprising, because they are present at all the stages of the calculation, and the cancellation mechanism is by no means trivial and has not yet received a simple physical interpretation. For example, the viscous effects inside the thermal diffusive zone, described by the term

$$-rac{\gamma}{1-\gamma}(P+P')rac{\partial^2lpha}{\partial y^2}$$

of (34), are exactly counterbalanced in the last term C of the dispersion relation (36) by the viscous effects inside the reactive zone described by (13a, b), which induces the term

$$(P+P')\left[\frac{\partial u_{+\infty}}{\partial x}\Big|_{x=0}-\frac{\partial u_{-\infty}}{\partial x}\Big|_{x=0}\right]$$

of (34). Thus, when the diffusivities are supposed to be temperature independent, the dynamical properties of the flame front are found to be not affected at all by the viscous effects. This result answers the open question concerning the influence of the viscosity, which was found to be stabilizing by some authors and destabilizing by others (see Markstein 1964, 1970).

It is interesting to rewrite the coefficient C of (36) in the following form:

$$C = \frac{\gamma}{1 - \gamma} k \left[\frac{1}{2} k_{\rm c} - k \left(\delta - \frac{k}{k_{\rm n}} \right) \right], \tag{38}$$

gravity hydrodynamics diffusion

where k_{c} varies with the flame speed according to

$$k_{\rm c} = 2(1-\gamma) \, \frac{gd}{u_{\rm L}^2},\tag{39}$$

and where k_n depends on the equivalence ratio of the reactive mixture through the Lewis number L appearing in \mathcal{L}/d (25),[†]

 $[\]dagger$ Far from the stoichiometric composition, L is defined with the binary molecular diffusivity of the limiting component (oxygen in the rich mixtures, fuel in the lean ones) in the neutral component (nitrogen in the air). As the analysis of Joulin & Mitani (1981) has shown, for mixtures close to stoichiometric composition, L appears to be a function of the equivalence ratio (ratio of the amount of fuel to the amount of oxygen) such that L varies continuously from its value based on the oxygen for rich mixtures to its value based on the fuel for lean mixtures.

Stability limits of laminar premixed flames

$$\delta = 1 + \frac{gd}{u_{\rm L}^2} (1 - \gamma) \left(\frac{\mathscr{L}}{d} - \frac{1}{\gamma} \ln \frac{1}{1 - \gamma} \right),$$

$$k_{\rm n} = \left[1 + \frac{2 + \gamma}{\gamma} \left(\mathscr{L}/d \right) - \frac{2}{\gamma} \ln \left(\frac{1}{1 - \gamma} \right) \right]^{-1}$$
(40)

where the second term of δ can be easily neglected compared with unity because its numerical value is of order 10^{-2} in the range of interest for the flame velocity $(u_{\rm L} > 5 \text{ cm/s})$. For ordinary reactive mixtures $k_{\rm n}$ is positive, making the diffusion a stabilizing mechanism.

When the acceleration of gravity is not considered, the dispersion relation (36), (37)reduces to an expression similar to the one obtained in the inviscid approximation by the phenomenological work of Markstein (1964), but where supplementary terms, involving ln $(1-\gamma)^{-1}$, appear in A and in B. Furthermore, the expression (40) for k_n is slightly different from $k_n = \{[(2+\gamma)/\gamma] \mathcal{L}/d\}^{-1}$ obtained by Markstein. The additional terms, involving $\ln (1-\gamma)^{-1}$, come from the modification of the longitudinal mass flux s associated with the transverse convection produced, inside the flame thickness, by the deflection mechanism of the streamlines. The supplementary term 1 in the denominator of (40) comes from the additional 'surface-tension effect' already mentioned in (34). Without the acceleration of gravity, $k_{\rm c} = 0$, and there is always a range of unstable wavenumbers located around zero. This can be clearly understood because, as the pioneering works of Darrieus (1938) and Landau (1944) have shown, the hydrodynamical instability produces in (38) a k^2 term. The stabilizing diffusive effects being associated with a k^3 term, they cannot overcome the hydrodynamical instability near k = 0. As Markstein (1970) pointed out himself, this cannot explain the existing experimental results, which show that the cellular structures fade away at an intrinsic size independent of the dimension of the burner.

Because of a mechanism similar to the gravity waves, the acceleration due to gravity in flames propagating downwards induces a positive k term in C. Thus the criterion of stability given by C > 0 can possibly be satisfied for all wavelengths provided the acceleration due to gravity and the diffusive effects are strong enough. Thus, infinite planar fronts of slow-burning flames associated with a limiting component sufficiently heavy are predicted to be stable when they propagate downwards.

The critical stability condition is given by

$$k_{\rm n} = 2k_{\rm c} \tag{41}$$

where k_c and k_n are given by (39) and (40) respectively.

For $k_n > 2k_c$ the instability occurs only for a finite range of wave vectors centred on k_c and vanishing at the critical condition (41). This unstable domain is limited by the two marginal wavenumbers k_{\pm} given by

$$k_{\pm} = \frac{1}{2}k_{\rm n} \pm k_{\rm c} \left(1 - \frac{2k_{\rm c}}{k_{\rm n}}\right)^{\frac{1}{2}}.$$
(42)

Using (39), (40) and (25) the critical stability condition is presented in figure 2 on the plane (u_L, L) for different values of γ . These curves are universal and can be used for any flame. The parameter L is directly related to the equivalence ratio, and the flame velocity u_L is controlled in the experimental situation by the composition of the reactive mixture, including the dilution but also the equivalence ratio. For each specific reactive mixture, the transcription of the limits of stability on the usual plane (equivalence ratio, dilution) is straightforward as soon as the dependence of the burning velocity of the mixture studied is known.



FIGURE 2. The limit of stability is plotted on the plane $(l = \beta(1 - 1/L), u_L)$ for two values of $\rho D/\rho_{-\infty}$ corresponding to the fresh mixture, 0.2 cm²/s, and the burned gases, 0.4 cm²/s. This last curve is expected to be the more relevant because the reaction zone is at the temperature of the burned gases. The effect of the temperature dependence of ρD is described by Clavin & Garcia (1982). The two figures correspond to different values of the gas expansion, $\gamma = 0.75$ and 0.8, which are realistic for conventional hydrocarbon flames in the range of flame velocities considered. For hydrocarbons heavier than oxygen, the lean mixtures correspond roughly to higher values of l, l > 1, and the rich to l < 1.

The present analysis is limited to the range of small values of k. According to (39), the critical value k_c is effectively found to be small as soon as u_L is large enough $(u_L > 5 \text{ cm})$. For lower flame velocities the experimental values of k_n corresponding to the usual reactive mixtures are expected to be smaller than the corresponding value of $2k_c$. Such flames are thus predicted to be stable at any composition. Thus the limits of stability predicted by the present study is found to cover all the range of the usual reactive mixtures.

5. Concluding remarks

The analytical study developed here provides a rigorous description of the coupling between the hydrodynamical and diffusive effects occurring in the dynamical properties of premixed flame fronts. The corresponding description of the limits of stability is in good qualitative agreement with the existing experimental data (Markstein 1951, 1964; Markstein & Somers 1953). As for the quantitative comparison with these data, the main defect of the model is the use of an over-simplified chemical kinetic description, which impairs the prediction with an insufficient precision of the effective Lewis number L corresponding to each composition of the actual reactive mixture used in the experiments. Nevertheless, one can obtain a good idea of the order of magnitude of this number by looking at the values of the different diffusivities of the reactive mixture. Let us for example consider the case of a propane-oxygen mixture highly diluted with nitrogen. At the normal conditions, the thermal diffusivity of the nitrogen is $D_{\rm th} \approx 0.19 \, {\rm cm^2/s}$ and the binary molecular diffusion coefficients of propane and oxygen in nitrogen are $D(C_3H_8, N_2) \approx 0.11 \text{ cm}^2/\text{s}$ and $D(O_2, N_2) \approx 0.22 \text{ cm}^2/\text{s}$ respectively (Fristrom & Westenberg (1965)). As the activation energy E of propane is about 55 kcal/mol (Kaskan 1957), β is close to 15 for the flame temperature of 1500 K, and the corresponding values of the quantity $l = \beta(1 - 1/L)$ appearing in (14) are $l(C_3H_8) = 6.3$ and $l(O_2) = -2.4$.

Thus, when the composition varies from lean to rich, one can expect that the effective value of $l \equiv \beta(1-1/L)$ appearing in the expression (25) for \mathcal{L}/d varies from 6.3 to -2.4. In fact one can expect that, for most of the time, the influence of the chemical kinetics enhances the stability of the fronts and causes a slight shift of these limits towards higher values. This is due to the fact that the extremely mobile intermediate species involved in chemical reaction attain, as a rule, maximum concentration at the reaction zone. Since the flame velocity increases with their concentration, the molecular diffusivity of these species is expected to reinforce the stabilizing influence of the heat diffusion. Nevertheless, for the usual hydrocarbons, to attain experimental values of l larger than 10 seems unlikely. Thus the results shown in figure 2 demonstrate that freely propagating planar flames with a *laminar* velocity $u_{\rm L}$ larger than 17 cm/s cannot be stabilized in a uniform flow. On the other hand, it seems very difficult to encounter values of l smaller than -3 (except for hydrogen). Hence our theory predicts that slow flames with laminar velocity $u_{\rm L}$ smaller than 5 cm/s should be stable for all compositions. Thus, the instability threshold should be experimentally observable between 5 cm/s $< u_{\rm L} < 17$ cm/s, at a fuel-rich composition for the lower values of $u_{\rm L}$ and at a lean composition for higher values of $u_{\rm L}$. Furthermore, independently of chemical-kinetics effects, the size of cells at the bifurcation is predicted by (39) and (42) to vary from 0.5 cm to 3.5 cm, as the square of the laminar velocity of propagation u_1^2 . It must be emphasized that these considerations apply only to the size of the cells at the critical threshold of the instability, and not to the evolution of this size by the nonlinear effects occurring beyond the threshold.

It should be also noted that for the rich mixtures close to the instability threshold, the temperature is predicted by (29') and (19) to be higher in the parts of the front concave towards the burnt gases. According to the result of our analysis, this is particularly the case at the threshold for cell formation on flame fronts with slow enough laminar velocity ($u_{\rm L} < 12$ cm/s, cf. figure 2).

In the usual hydrocarbon mixtures, highly diluted with nitrogen, these low velocities correspond to situations that are not very far from the thermal-extinction limit (~ 8 cm/s). According to Joulin & Clavin (1979), one may expect that close to

the extinction limit, the above predictions could be affected by the heat losses. The model used in the present paper can be made more realistic by introducing such effects as the influence of heat losses, the temperature variation of the diffusivities, or a more detailed kinetic scheme. The corresponding development is tractable and will be published in the near future (see Clavin & Garcia 1982; Clavin & Nicoli 1983). We have deliberately omitted such complications here, in order to emphasize the important basic phenomena.

In conclusion, when the expansion of the gas is taken into account in adiabatic flames, the diffusive effects appear to be stabilizing $(\mathscr{L}/d, k_n > 0)$ at all compositions for the usual hydrocarbons, and thus cannot be responsible for the cellular structures of the planar fronts. Nevertheless, very light limiting components such as hydrogen require the more-detailed study quoted above. An exception is also still possible for very heavy limiting components, which can possibly produce, as in solid combustion, spinning waves (cf. the discussion in §4). Apart from these extreme cases, which are beyond the scope of the present study, the cellular structures appearing on planar fronts of usual hydrocarbon adiabatic flames are shown to be described by the competition between the hydrodynamical instability and two stabilizing phenomena: acceleration due to gravity and diffusive effects. The corresponding threshold is predicted to be experimentally observable for ordinary hydrocarbon flames propagating downwards with a flame speed between 5 cm/s and 17 cm/s.

The authors are greatly indebted to G. Searby, A. Liñan and A. K. Oppenheim for fruitful discussions. This work was supported in part by the C.N.R.S. under A.T.P. 3966 and by the D.R.E.T. under grant no. 80/434.

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