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Diffusional-thermal instability of adiabatic flame propagation in dissociation equilibrium

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Effects of product dissociation and preferential diffusion on the structure, propagation and diffusional-thermal instability of the classical one-dimensional laminar flame have been studied by using activation energy asymptotics. Analytical expressions as functions of dissociation and diffusion parameters have been obtained for such bulk flame parameters as the flame temperature reduction and the propagation rate eigenvalue, and for the dispersion relation governing flame stability. Results on flame propagation show that while under most situations the flame speed is reduced due to product dissociation, it can attain values in excess of the non-dissociative limit for highly mobile product species which can preferentially back diffuse to the upstream portion of the reaction zone where they recombine and release the associated recombination heat. Results on flame stability show that it is promoted in the presence of product dissociation which has a moderating influence on the flame temperature fluctuations, and for highly-mobile product species because of the enhanced burning rate and curvature-induced concentration modification.

1. Introduction

The application of activation energy asymptotics has recently made possible the successful analyses of a variety of phenomena related to the stability of laminar flame propagation. Notable among them are the diffusional-thermal stability of adiabatic (Sivashinsky 1977), non-adiabatic (Joulin & Clavin 1979) and stretched (Sivashinsky, Law & Joulin 1982) flames, and the combined diffusional-thermal and hydrodynamic instability of the adiabatic flame (Clavin & Williams 1982; Pelce & Clavin 1982; Matalon & Matkowsky 1984).

In these previous studies the reaction between fuel and oxidizer is assumed to be one-step, irreversible and complete, producing certain products which are taken to be in frozen equilibrium. This leads to the most intense burning possible because of the complete conversion of the available chemical energy to thermal energy. However, since product dissociation invariably occurs to various extents, in realistic situations a certain amount of energy is withheld from the combustion process of interest.

Recently Chao & Law (1988) successfully analysed the propagation of adiabatic and non-adiabatic flames in dissociation equilibrium, adopting the reaction scheme originally proposed by Williams & Peters (1985) for the study of the structure of diffusion flames. This scheme, relevant for hydrocarbon oxidation, can be expressed as

$$\nu_F[\mathrm{F}] + \nu_O[\mathrm{O}] \xrightarrow{k_f} [\mathrm{P}_1] + [\mathrm{P}_2] \overset{K_p}{\rightleftharpoons} [\mathrm{Q}_1] + [\mathrm{Q}_2], \tag{1.1}$$

which states that fuel (F) and oxidizer (O) undergo a one-step irreversible and complete reaction to form products P_1 and P_2, which further partially dissociate into Q_1 and Q_2 such that dissociation equilibrium is maintained between P_1, P_2, Q_1 and Q_2. In (1.1) k_f is the reaction rate constant for the oxidation step, K_p the equilibrium constant for the dissociation–recombination process, and ν_i the stoichiometric coefficient of i although the specific value of unity is assumed for ν_{P_1}, ν_{P_2}, ν_{Q_1} and ν_{Q_2} in order to facilitate the analysis. The work of Chao & Law (1988) yields the interesting result that dissociation alone cannot cause extinction of an adiabatic flame.

In the present study we shall apply reaction scheme (1.1) to assess the effect of dissociation equilibrium on the diffusional-thermal stability of laminar flame propagation. The fundamental interest here arises from the recognition (Joulin & Clavin 1979) that heat loss to the environment promotes the onset of flame-front diffusional-thermal instability. Since product dissociation can be interpreted as an internal 'heat loss' mechanism as far as the total heat budget is concerned, it is relevant to understand the influence of this 'heat loss' mechanism on flame-front stability.

An auxiliary contribution of the present study is the derivation of the basic solution and thereby identification of the structure of the undisturbed flame propagation, in dissociation equilibrium and with preferential diffusion. An especially interesting result that emerges from the analysis is the possibility that the propagation speed of a flame in dissociation equilibrium can actually exceed the corresponding value when dissociation is suppressed.

The formulation, basic solution, stability solution and discussion of results will be presented in the following sections.

2. Formulation

The problem we are studying is the steady propagation of a planar, one-dimensional laminar flame in the doubly-infinite domain. With conventional assumptions, the equations governing the conservation of energy and species concentrations can be expressed as

$$\rho c_p \frac{\partial T}{\partial t} + \rho \boldsymbol{v} c_p \cdot \nabla T - \lambda \nabla^2 T = \nu_F W_F q_F w_r - W_{P_1} q_D w_d, \tag{2.1}$$

$$\rho \frac{\partial Y_F}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_F - \rho D_F \nabla^2 Y_F = -\nu_F W_F w_r, \tag{2.2}$$

$$\rho \frac{\partial Y_O}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_O - \rho D_O \nabla^2 Y_O = -\nu_O W_O w_r, \tag{2.3}$$

$$\rho \frac{\partial Y_i}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_i - \rho D_i \nabla^2 Y_i = W_i (w_r - w_d) \quad (i = P_1, P_2), \tag{2.4}$$

$$\rho \frac{\partial Y_i}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_i - \rho D_i \nabla^2 Y_i = W_i w_d \quad (i = Q_1, Q_2), \tag{2.5}$$

where t is the time, ρ the density, T the temperature, Y_i the mass fraction, W_i the molecular weight, c_p an average specific heat at constant pressure, λ the thermal conductivity, D_i the mass diffusivity, $\boldsymbol{v}$ the flow velocity vector, and

$$q_{\mathrm{F}} = h^0_{\mathrm{F}} + \mu_{\mathrm{O}} h^0_{\mathrm{O}} - \mu_{\mathrm{P}}[h^0_{\mathrm{P}_1} + (W_{\mathrm{P}_2}/W_{\mathrm{P}_1})\, h^0_{\mathrm{P}_2}], \tag{2.6}$$

$$q_{\mathrm{D}} = [(W_{\mathrm{Q}_1}/W_{\mathrm{P}_1})\, h^0_{\mathrm{Q}_1} + (W_{\mathrm{Q}_2}/W_{\mathrm{P}_1})\, h^0_{\mathrm{Q}_2}] - [h^0_{\mathrm{P}_1} + (W_{\mathrm{P}_2}/W_{\mathrm{P}_1})\, h^0_{\mathrm{P}_2}], \tag{2.7}$$

are respectively the heat of reaction per unit mass of fuel oxidized and the heat absorbed per unit mass of P_1 dissociated, where $\mu_{\mathrm{O}} = (\nu_{\mathrm{O}} W_{\mathrm{O}})/(\nu_{\mathrm{F}} W_{\mathrm{F}})$, $\mu_{\mathrm{P}} = W_{\mathrm{P}_1}/(\nu_{\mathrm{F}} W_{\mathrm{F}})$, and ρ, c_p, λ and D_i are assumed to be constants.

For the chemical source terms, we first assume that the oxidation reaction is one step, irreversible and complete, with a rate given by

$$w_{\mathrm{r}} = \left(\frac{B\rho^2}{W_{\mathrm{F}} W_{\mathrm{O}}}\right) Y_{\mathrm{F}} Y_{\mathrm{O}} \exp(-E/T), \tag{2.8}$$

where B is the pre-exponential factor and E the activation temperature. For the dissociation–recombination equilibrium we define an equilibrium constant

$$K_{\mathrm{p}} = \frac{X_{\mathrm{Q}_1} X_{\mathrm{Q}_2}}{X_{\mathrm{P}_1} X_{\mathrm{P}_2}}, \tag{2.9}$$

where X_i is the mole fraction such that the mass fractions Y_i of the product species P_2, Q_1 and Q_2 are related to Y_{P_1} and K_{p} through

$$Y_{\mathrm{P}_2} = \left(\frac{W_{\mathrm{P}_2}}{W_{\mathrm{P}_1}}\right) Y_{\mathrm{P}_1}, \quad Y_{\mathrm{Q}_1} = \left(\frac{W_{\mathrm{Q}_1}}{W_{\mathrm{P}_1}}\right) Y_{\mathrm{P}_1} (K_{\mathrm{p}})^{\frac{1}{2}}, \quad Y_{\mathrm{Q}_2} = \left(\frac{W_{\mathrm{Q}_2}}{W_{\mathrm{P}_1}}\right) Y_{\mathrm{P}_1} (K_{\mathrm{p}})^{\frac{1}{2}}. \tag{2.10}$$

Finally, following Williams & Peters (1985) we approximate K_{p} by

$$\kappa = J \exp(-A/T),$$

where $\kappa = (K_{\mathrm{p}})^{\frac{1}{2}}$ and A and J are positive constants.

Using the above relations, it can be shown that (2.1)–(2.5) can be reduced to the set of source-free equations

$$\left[\rho \frac{\partial Y_{\mathrm{F}}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{F}} - \rho D_{\mathrm{F}} \nabla^2 Y_{\mathrm{F}}\right] - \frac{1}{\mu_{\mathrm{O}}}\left[\rho \frac{\partial Y_{\mathrm{O}}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{O}} - \rho D_{\mathrm{O}} \nabla^2 Y_{\mathrm{O}}\right] = 0, \tag{2.11}$$

$$\begin{aligned}\left[\rho \frac{\partial Y_{\mathrm{F}}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{F}} - \rho D_{\mathrm{F}} \nabla^2 Y_{\mathrm{F}}\right] + \frac{1}{\mu_{\mathrm{P}}}\Bigg\{\left[\rho \frac{\partial Y_{\mathrm{P}_1}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{P}_1} - \rho D_{\mathrm{P}_1} \nabla^2 Y_{\mathrm{P}_1}\right] \\ + \left[\rho \frac{\partial}{\partial t}(\kappa Y_{\mathrm{P}_1}) + \rho \boldsymbol{v} \cdot \nabla (\kappa Y_{\mathrm{P}_1}) - \rho D_{\mathrm{Q}_1} \nabla^2 (\kappa Y_{\mathrm{P}_1})\right]\Bigg\} = 0,\end{aligned} \tag{2.12}$$

$$\begin{aligned}\left[\rho c_p \frac{\partial T}{\partial t} + \rho c_p \boldsymbol{v} \cdot \nabla T - \lambda \nabla^2 T\right] + (q_{\mathrm{F}} - \mu_{\mathrm{P}} q_{\mathrm{D}})\left[\rho \frac{\partial Y_{\mathrm{F}}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{F}} - \rho D_{\mathrm{F}} \nabla^2 Y_{\mathrm{F}}\right] \\ - q_{\mathrm{D}}\left[\rho \frac{\partial Y_{\mathrm{P}_1}}{\partial t} + \rho \boldsymbol{v} \cdot \nabla Y_{\mathrm{P}_1} - \rho D_{\mathrm{P}_1} \nabla^2 Y_{\mathrm{P}_1}\right] = 0,\end{aligned} \tag{2.13}$$

plus an additional equation containing the source terms, which we choose to be (2.2) because of its relative simplicity.

Consider the flame front to be propagating in the $-x$-direction with its constant flame speed u in a flow field which has a constant speed u^0 in the x-direction, and assume for simplicity its otherwise smooth surface to be perturbed only in the z-direction. The location of the flame front can then be defined as

$$x_f(z,t) = x_0 + (u^0 - u)\,t + \delta F(z,t),$$

where x_0 is the unperturbed flame location at $t = 0$, $F(z,t)$ is an implicit flame shape function, and δ the amplitude of the flame-front corrugations. Introducing the non-dimensional quantities

$$\tilde{T} = \frac{T - T_{-\infty}}{T^* - T_{-\infty}}, \quad \tilde{Y}_{\mathrm{F}} = \frac{Y_{\mathrm{F}}}{Y_{\mathrm{F},-\infty}}, \quad \tilde{Y}_{\mathrm{O}} = \frac{Y_{\mathrm{O}}}{\mu_{\mathrm{O}} Y_{\mathrm{F},-\infty}}, \quad \tilde{Y}_{\mathrm{P}_1} = \frac{Y_{\mathrm{P}_1}}{\mu_{\mathrm{P}} Y_{\mathrm{F},-\infty}},$$

$$\tilde{x} = \frac{\rho u c_p}{\lambda}(x - x_f), \quad \tilde{z} = \frac{\rho u c_p}{\lambda} z, \quad \tilde{t} = \frac{\rho u^2 c_p}{\lambda} t, \quad f = \frac{\rho u c_p}{\lambda} F,$$

and the non-dimensional parameters

$$\tilde{A} = A/T^*, \quad \tilde{E} = E/T^*, \quad \alpha = A/E,$$

$$\beta = \frac{T^* - T_{-\infty}}{T^*}, \quad \kappa^* = J\,\mathrm{e}^{-\tilde{A}}, \quad \gamma = \frac{\mu_{\mathrm{P}} q_{\mathrm{D}}}{q_{\mathrm{F}}} \frac{\kappa^*}{\epsilon},$$

where $T^* = (T_{-\infty} + q_{\mathrm{F}} Y_{\mathrm{F},-\infty}/c_p)$ is the adiabatic flame temperature for the non-dissociative flame and the superscript (*) is used to designate this state, the governing equations are transformed to

$$\left[\frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{F}}} \Delta \tilde{Y}_{\mathrm{F}}\right] - \left[\frac{\partial \tilde{Y}_{\mathrm{O}}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{O}}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{O}}} \Delta \tilde{Y}_{\mathrm{O}}\right] = 0, \qquad (2.14)$$

$$\left[\frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{F}}} \Delta \tilde{Y}_{\mathrm{F}}\right] + \left[\frac{\partial \tilde{Y}_{\mathrm{P}_1}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{P}_1}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{P}_1}} \Delta \tilde{Y}_{\mathrm{P}_1}\right]$$
$$+ \kappa^* \left[\frac{\partial}{\partial \tilde{t}}(G\tilde{Y}_{\mathrm{P}_1}) + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial}{\partial \tilde{x}}(G\tilde{Y}_{\mathrm{P}_1}) - \frac{1}{Le_{\mathrm{Q}_1}} \Delta (G\tilde{Y}_{\mathrm{P}_1})\right] = 0, \qquad (2.15)$$

$$\left[\frac{\partial \tilde{T}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{T}}{\partial \tilde{x}} - \Delta \tilde{T}\right] + \left(1 - \frac{\mu_{\mathrm{P}} q_{\mathrm{D}}}{q_{\mathrm{F}}}\right) \left[\frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{F}}} \Delta \tilde{Y}_{\mathrm{F}}\right]$$
$$- \frac{\mu_{\mathrm{P}} q_{\mathrm{D}}}{q_{\mathrm{F}}} \left[\frac{\partial \tilde{Y}_{\mathrm{P}_1}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{P}_1}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{P}_1}} \Delta \tilde{Y}_{\mathrm{P}_1}\right] = 0, \qquad (2.16)$$

$$\frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{t}} + \left(1 - \delta \frac{\partial f}{\partial \tilde{t}}\right) \frac{\partial \tilde{Y}_{\mathrm{F}}}{\partial \tilde{x}} - \frac{1}{Le_{\mathrm{F}}} \Delta \tilde{Y}_{\mathrm{F}} = -\left(\frac{\Lambda}{\epsilon^2 \tilde{Y}_{\mathrm{O},\infty}}\right) \tilde{Y}_{\mathrm{F}} \tilde{Y}_{\mathrm{O}} \exp\left[\frac{1}{\epsilon} \frac{\tilde{T} - 1}{1 + \beta(\tilde{T} - 1)}\right], \qquad (2.17)$$

where $\epsilon = (\beta \tilde{E})^{-1}$, $Le_i = \lambda/(c_p \rho D_i)$ is the Lewis Number,

$$\Lambda = \frac{\epsilon^2 \nu_{\mathrm{F}} B \lambda Y_{\mathrm{O},\infty}\,\mathrm{e}^{-\tilde{E}}}{W_{\mathrm{O}}\, c_p\, u^2} \qquad (2.18)$$

is the burning rate eigenvalue,

$$\Delta = \left[1 + \left(\delta \frac{\partial f}{\partial \tilde{z}}\right)^2\right] \frac{\partial^2}{\partial \tilde{x}^2} + \frac{\partial^2}{\partial \tilde{z}^2} - \delta \frac{\partial^2 f}{\partial \tilde{z}^2} \frac{\partial}{\partial \tilde{x}} - 2\delta \frac{\partial f}{\partial \tilde{z}} \frac{\partial^2}{\partial \tilde{x} \partial \tilde{z}} \qquad (2.19)$$

is the non-Cartesian Laplacian operator, and

$$G = \exp\left[\frac{\alpha}{\epsilon}\frac{\tilde{T}-1}{1+\beta(\tilde{T}-1)}\right]. \tag{2.20}$$

In the above $\beta = O(1)$ and $\tilde{E}$ and $\tilde{A}$ are both large quantities such that $\epsilon \ll 1$ and α, γ are $O(1)$ quantities. The parameters γ and α emerge to be of particular significance, respectively indicating the extent of dissociation and the sensitivity of the dissociation/recombination reactions to temperature variations. Equations (2.14)–(2.17) are to be solved subject to the boundary conditions that,

$$\begin{aligned} \tilde{x} \rightarrow -\infty: &\quad \tilde{T} \rightarrow 0, \quad \tilde{Y}_F \rightarrow 1, \quad \tilde{Y}_O \rightarrow \tilde{Y}_{O,-\infty}, \quad \tilde{Y}_{P_1} \rightarrow 0, \\ \tilde{x} \rightarrow \infty: &\quad \frac{\partial \tilde{T}}{\partial \tilde{x}} \rightarrow 0, \quad \tilde{Y}_F \rightarrow 0, \quad \tilde{Y}_O \rightarrow \tilde{Y}_{O,\infty}, \quad \frac{\partial \tilde{Y}_{P_1}}{\partial \tilde{x}} \rightarrow 0. \end{aligned} \tag{2.21}$$

Fuel-lean flow is assumed in this problem so that there is complete fuel consumption at the flame and the amount of the oxidizer remaining far downstream of the reaction zone is $\tilde{Y}_{O,\infty}$.

The problem is now completely formulated. The methodology and solution for the basic steady state and the linear stability analysis will be outlined in the next two sections; detailed steps in the derivation can be found in Chao (1987). It may also be mentioned that the essential requirement of non-unity Lewis numbers for the diffusional-thermal instability necessitates a somewhat more involved analysis for the steady state basic solutions as compared with that of the unity Lewis number case studied previously (Chao & Law 1988).

3. Steady, planar flame propagation

For the steady propagation of the planar flame, we set $\partial/\partial\tilde{t} \equiv 0$, $\partial/\partial\tilde{x} \equiv \mathrm{d}/\mathrm{d}\tilde{x}$ and $\Delta \equiv \mathrm{d}^2/\mathrm{d}\tilde{x}^2$ in (2.14)–(2.17), which are then solved in the limit of large activation energy, with $\epsilon \ll 1$ and $\alpha = O(1)$.

From studies of non-dissociative flames it is well established that for large activation energy reactions the flame structure consists of a thin chemically-reacting region separating two broad, upstream unburnt and downstream burnt regions in which fuel oxidation effectively does not occur. For the present dissociative flames we have assumed, realistically, that the activation energies for oxidation and the dissociation equilibrium constant are of the same order of magnitude, or $\alpha = O(1)$. Consequently the upstream unburnt region is also dissociative-recombinatively frozen in the presence of $O(1)$ reduction in the flow temperature from the flame temperature. This leads to $G_s \sim 0$ and the decoupling of the governing equations in this region, where the subscript (s) designates the steady solution. In the downstream region all properties are uniform because dissociation equilibrium is attained and there is no source or sink there.

In the thin reaction region convection is unimportant such that the controlling processes are diffusion, oxidation, dissociation and recombination. The temperature in this region is expected to deviate from that of the non-dissociative case by $O(\epsilon)$. Thus if we designate this deviation by $\epsilon\theta_1(\xi)$, where $\xi = \tilde{x}/\epsilon$ is the stretched inner variable, and $\theta_{1\infty}$ the value of $\theta_1(\xi)$ at the downstream boundary of this reaction region, then $\theta_{1\infty}$ can be used as a parameter to indicate the effect of dissociation on the various flame responses.

Thus by separately analysing these three regions and performing the requisite matching (Chao 1987), with the Lewis numbers of the various species being expanded as

$$Le_i = l_{i,0}+\epsilon l_{i,1}+\epsilon^2 l_{i,2}+\dots \quad (i = \mathrm{F},\mathrm{O},\mathrm{P}_1,\mathrm{Q}_1), \tag{3.1}$$

the bulk flame responses can be represented by the following two key relations,

$$\theta_{1\infty}\,\mathrm{e}^{\alpha\theta_{1\infty}} = \gamma \tag{3.2}$$

$$2l_{\mathrm{F},0}\Lambda = \mathrm{e}^{\theta_{1\infty}}\left\{1+\frac{\alpha\theta_{1\infty}}{(1+\alpha)\,l_{\mathrm{Q}_1,0}}\left[\frac{2+\alpha}{1+\alpha}+\frac{\alpha\theta_{1\infty}}{(1+2\alpha)\,l_{\mathrm{Q}_1,0}}\right]\right\}^{-1}. \tag{3.3}$$

Equation (3.2) allows the determination of $\theta_{1\infty}$ for given dissociation–recombination parameters α and γ, while (3.3) gives the burning rate eigenvalue Λ as function of α, γ, $l_{\mathrm{F},0}$ and $l_{\mathrm{Q}_1,0}$. In the limit of no dissociation, $\gamma = 0$ such that $\theta_{1\infty} = 0$ and $\Lambda^* = 1/(2l_{\mathrm{F},0})$. Furthermore, since $\Lambda \sim u^{-2}$, we have

$$\frac{u}{u^*} = \frac{1}{(2l_{\mathrm{F},0}\Lambda)^{\frac{1}{2}}}. \tag{3.4}$$

4. Diffusional-thermal instability

We now study the linear stability of adiabatic flame propagation when it is subjected to small harmonic perturbations with amplitude δ. Concerning the magnitude of δ relative to ϵ, we note that most of the linear stability analyses in flame propagation have adopted the limit $(\epsilon/\delta)\to 0$ as $\epsilon\to 0$, although McIntosh & Clarke (1984) have recently argued for the rationality of the opposite limit, $(\delta/\epsilon)\to 0$ as $\epsilon\to 0$. While this issue is as yet unsettled, we have ascertained that the dispersion relations obtained from separate analyses assuming either limit are the same because of the linearized nature of the analyses. Since the purpose of the present study is not to resolve this issue on the limits, and since the analysis involving the $(\epsilon/\delta)\to 0$ limit is somewhat simpler and more straightforward, the solution sequence as indicated in the following is based on this specific limit.

The analysis involves first perturbing all the flow variables by an amount proportional to δ, and then letting the disturbances be harmonic functions of time and the transverse distance. For example, for the temperature variation we express

$$\tilde{T}(\tilde{x},\tilde{z},\tilde{t}) = \tilde{T}_{\mathrm{s}}(\tilde{x})+\delta\tilde{T}'(\tilde{x},\tilde{z},\tilde{t}), \tag{4.1}$$

and

$$\tilde{T}'(\tilde{x},\tilde{z},\tilde{t}) = \hat{T}(\tilde{x})f(\tilde{z},\tilde{t}), \tag{4.2}$$

where

$$f(\tilde{z},\tilde{t}) = \exp(\omega\tilde{t}+\mathrm{i}k\tilde{z}). \tag{4.3}$$

Substituting these expressions and the steady-state solutions into the governing equations (2.14)–(2.17), expanding, and solving by using asymptotic analysis (Chao 1987), the dispersion relation for flame stability is derived as

$$\begin{aligned}(1-\Gamma)\Bigg\{&\left(1+\frac{\alpha\theta_{1\infty}}{l_{\mathrm{Q}_1,0}}\right)(1-\Gamma)-(1+\alpha\theta_{1\infty})-\Bigg\{(1+\alpha\theta_{1\infty})^2\\ &+4\left(1+\frac{\alpha\theta_{1\infty}}{l_{\mathrm{Q}_1,0}}\right)\left[\omega(1+\alpha\theta_{1\infty})+k^2\left(1+\frac{\alpha\theta_{1\infty}}{l_{\mathrm{Q}_1,0}}\right)\right]\Bigg\}^{\frac{1}{2}}\Bigg\}\\ &-(1+2\omega-\Gamma)\left[\theta_{1\infty}\left(\frac{1}{l_{\mathrm{Q}_1,0}}-1\right)+\frac{l_{\mathrm{F},1}}{\Gamma}\right] = 0,\end{aligned} \tag{4.4}$$

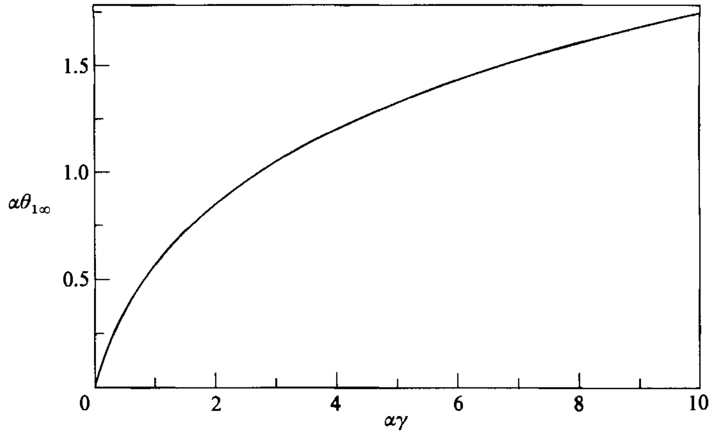

FIGURE 1. Flame temperature reduction parameter, $\theta_{1\infty}$, as function of the dissociation parameters $\alpha(\neq 0)$ and γ.

where
$$\Gamma = [1+4(\omega+k^2)]^{\frac{1}{2}} \tag{4.5}$$

and $\theta_{1\infty}$ is given by (3.2). In obtaining (4.4) it has also been found necessary to require that $l_{F,0} = l_{O,0} = l_{P_1,0} = 1$, implying that the stability analysis is applicable only for near equi-diffusion flames except for the diffusion of the dissociated products. For $l_{Q_1,0} = 1$, the dispersion relation further reduces to the simple expression

$$\frac{1}{2}\left[\frac{l_{F,1}}{1+\alpha\theta_{1\infty}}\right]\left[1-\frac{1+2\omega}{\Gamma}\right] = \Gamma(1-\Gamma). \tag{4.6}$$

Finally, in the non-dissociative limit of $\alpha\theta_{1\infty} \to 0$, the dispersion relation specializes to that of Joulin & Clavin (1979), as expected.

5. Results and discussion

The parameters directly related to product dissociation are α and γ, which respectively indicate the sensitivity of the dissociation/recombination reactions to temperature variations and the extent of dissociation. The parameter $\theta_{1\infty}$, given by (3.2) and plotted in figure 1, represents the reduction of the flame temperature and is the most direct indicator of the effect of dissociation on the various flame responses due to product dissociation. Figure 1 shows that $\theta_{1\infty}$ increases with increasing γ, which is reasonable because for larger γ more thermal energy is withheld through dissociation. Furthermore, since $\alpha\theta_{1\infty}$ increases slower than $\alpha\gamma$, it is clear that $\theta_{1\infty}$ decreases with increasing α because of the increased difficulty of dissociation occurring. It may also be noted that $\theta_{1\infty}$ does not depend on the species Lewis numbers and therefore is not affected by preferential diffusion. The reason is that for unstretched flames preferential diffusion does not affect the flame temperature, and thus has no influence on the extent of dissociation which depends only on temperature.

Equation (3.3) gives the expression of the burning rate eigenvalue Λ as a function of the dissociation parameters and the Lewis number of the fuel, $l_{F,0}$. Since $\Lambda \sim u^{-2}$, we find that the flame speed is affected by the Lewis number of the fuel as $u \sim (l_{F,0})^{\frac{1}{2}}$. Physically, when $l_{F,0}$ is, say, greater than unity, the diffusion length

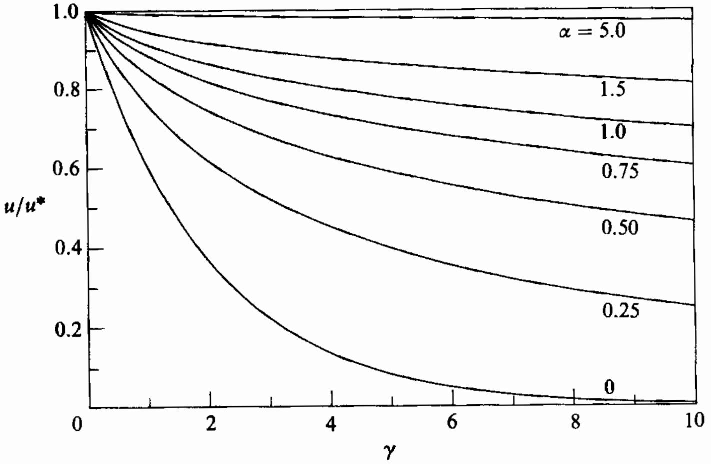

FIGURE 2. Normalized flame speed, u/u^*, as function of the dissociation parameters α and γ, with the diffusion parameter $l_{Q_1,0} = 1$.

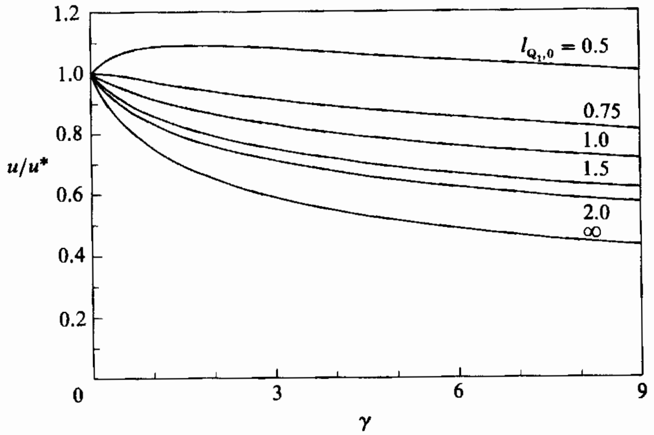

FIGURE 3. Normalized flame speed, u/u^*, as function of the diffusion parameter $l_{Q_1,0}$ and dissociation parameters γ and $\alpha(=1)$.

of the fuel is shorter than that of heat, implying that at the ignition point the slope of the fuel profile is steeper than that of heat. The flame therefore experiences more fuel in the reaction zone and thus burns faster. The converse argument holds for $l_{F,0} < 1$.

Figure 2 shows the normalized flame speed, given by (3.4), as a function of the dissociation parameters for the special case of $l_{Q_1,0} = 1$. It is seen that the flame speed decreases with increasing dissociation strength, i.e. increasing γ and decreasing α, and monotonically approaches zero as $\gamma \to \infty$. Figure 3 shows the effect of the Lewis number of the dissociated products, $l_{Q_1,0}$, on the flame speed, for a fixed value of $\alpha = 1.0$. It is seen that the flame speed is higher for smaller value of $l_{Q_1,0}$ for the same extent of dissociation. Furthermore, in some situations the flame speed may be even higher than the laminar flame speed. This flame-speed enhancement result can be explained as follows.

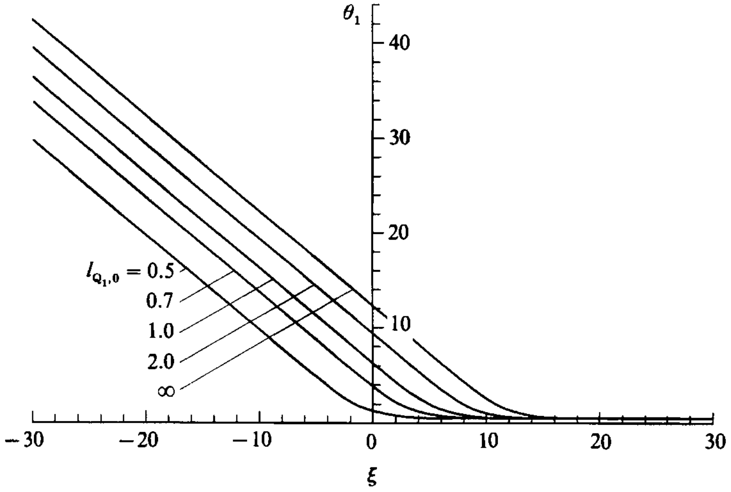

FIGURE 4. Perturbed temperature profiles in the reaction zone for selected values of the diffusion parameter $l_{Q_1,0}$, with fixed dissociation parameters of $\alpha = \gamma = 1$.

Upon production of the dissociated product, part of it is convected downstream towards the burnt side of the flame, while the rest diffuses backwards towards the unburnt side. There is no downstream diffusion because of downstream uniformity. The amount that diffuses backwards recombines to form the oxidation products as they encounter lower temperatures; complete recombination is achieved at the upstream boundary of the reaction zone. Thus by recapturing part of the thermal energy initially withheld by the dissociated products, the burning intensity is increased. Such an increase, however, is not sufficient to compensate for all the dissociative loss if the product (back-) diffuses at rates less than, or comparable to, that of heat, that is if $l_{Q_1,0} \gtrsim 1$. On the other hand if the product does diffuse fairly rapidly, for $l_{Q_1,0}$ sufficiently smaller than unity, then the additional recombinative heat release, plus the fact that this heat release occurs towards the upstream portion of the reaction region which has the most pronounced influence on flame propagation, can indeed boost the flame speeds to values exceeding the corresponding undissociated cases.

The above concept amounts to a spatial redistribution of the heat release rate and thereby modification of the temperature profile in the reaction region. The calculated perturbed temperature profiles in the reaction region, $\theta_1(\xi)$, for $\alpha = \gamma = 1$ (figure 4) confirm this phenomenon. Here we note that although the exact value of θ_1 at any ξ depends on where the origin is located and where ∞ is considered to be, the relative values will not change when shifting the origin. We can therefore see that for smaller values of $l_{Q_1,0}$, θ_1 is smaller before the chemical reaction is completed, implying an overall increase in temperature before the final flame temperature is reached. Also note that for smaller values of $l_{Q_1,0}$, the flame reaches its final temperature earlier than situations involving larger values of $l_{Q_1,0}$, thereby further supporting the fact that the flame burns faster. Finally, as $\gamma \rightarrow \infty$, we still have $u/u^* \rightarrow 0$ because the effect of product dissociation is dominating.

The stability of the steadily propagating flame is determined by assessing whether the disturbance of finite transverse wavenumber, k, grows or diminishes with time. This depends on the sign of the real part of the frequency parameter ω which is

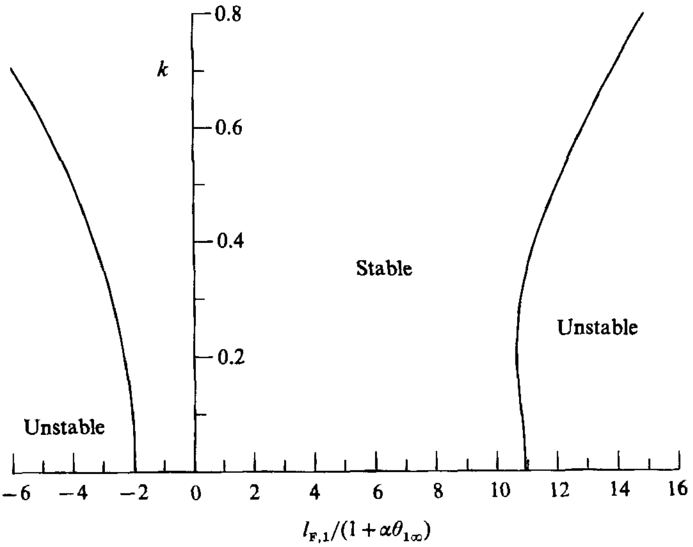

FIGURE 5. Cellular and pulsating stability diagram for flame propagation in dissociation equilibrium, with the diffusion parameter $l_{Q_1,0} = 1$.

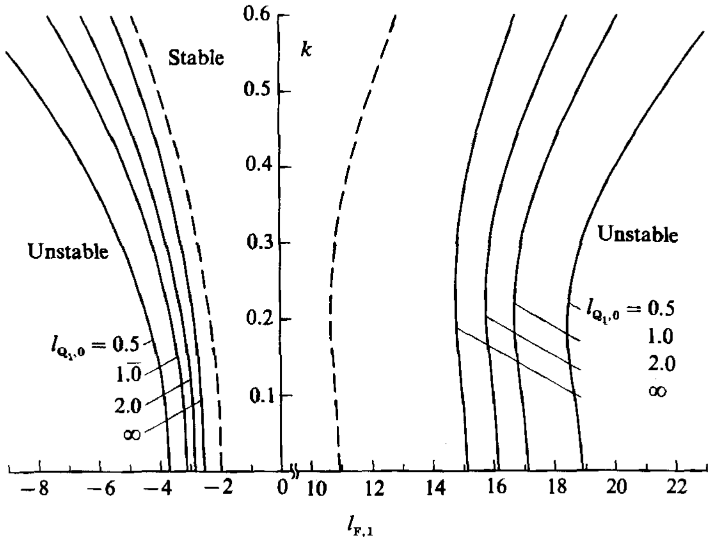

FIGURE 6. Cellular and pulsating stability diagram showing the effect of the diffusion parameter $l_{Q_1,0}$, with fixed dissociation parameters $\alpha = \gamma = 1$. The broken curves are for the non-dissociative flame.

determined from the dispersion relation. We first consider the stability of flames with $l_{Q_1,0} = 1$. The neutral stability curves for both the cellular and pulsating instabilities are shown in figure 5 in the wavenumber versus perturbed Lewis number plane. These curves are determined by setting the real part of ω to zero in (4.6). The stable and unstable regions are also shown in this figure. The results are the same as those of Joulin & Clavin (1979) for the adiabatic non-dissociative flames except that the Lewis number of fuel, $l_{F,1}$, is now reduced by a factor $(1+\alpha\theta_{1\infty})$ because $\alpha\theta_{1\infty}$ is nonnegative. Thus the stable region is widened, implying that product dissociation tends to stabilize the flame. Physically, when disturbances are applied to a steady flame, causing the flame temperature to, say, increase locally, the flame segment will propagate faster and become wrinkled. However, with product dissociation, the increase in the local flame temperature is accompanied by a corresponding increase in the amount of dissociation, which tends to reduce the flame temperature. A similar argument can be extended to the case where the disturbance lowers the local flame temperature, which then leads to recombinative heat release and consequently the tendency for the flame temperature to increase. Thus the effect of the initial disturbance is weakened when allowing for dissociation, implying that dissociation is stabilizing.

To identify the effect of the Lewis number of the dissociated product, (4.4) is used and the results for the neutral stability curves are shown in figure 6 for $\alpha = \gamma = 1$. Again, it is seen that product dissociation has the effect of stabilizing the flame. It is also shown that the stabilizing effect increases with decreasing $l_{Q_1,0}$. There are two reasons for this result. The first is that the undisturbed flame burns stronger for smaller $l_{Q_1,0}$ and therefore is less susceptible to diffusional-thermal instability. The second reason is that when the flame is disturbed, say wrinkled, the coupled effects of flame curvature and back diffusion of the dissociated products is stabilizing. This point can be demonstrated by considering, for example, the segment of the flame which bulges towards the upstream. Then the concentration of the dissociated product is diluted as it back-diffuses towards the flame front. This leads to reduced rate of recombinative heat release and therefore tends to weaken the bulge, causing it to recede and flatten.

6. Concluding remarks

In the present study we have shown that, whilst the dissociation of products, in most cases, lowers the flame propagation rate, the latter can be enhanced if the dissociation products are highly mobile. In such circumstances the products back-diffuse more effectively than heat into the preheat zone, where they recombine and reinforce the rate of liberation of chemical energy within the flame. This is an interesting concept because it demonstrates the possibility of increasing the burning rate by modifying the heat release rate and thereby the temperature profile within the reaction region. That is, since flame propagation is more sensitive to the temperature in the upstream portion, it is advantageous to shift part of the heat release from the downstream to the upstream portion. The agents in effecting the shift in the present case are the dissociated products. We also note that for realistic chemical systems the dissociated products indeed have smaller molecular weights and thereby higher mobilities.

We have also shown that product dissociation has a stabilizing influence on the flame. Thus contrary to the result of Joulin & Clavin (1979) that external heat loss tends to destabilize the flame, 'internal heat loss' in the form of dissociation is recoverable. The recovery process also promotes flame stability.

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