



Laminar flame propagation with volumetric heat loss and chain branching–termination reactions

B. H. CHAO

Department of Mechanical Engineering, University of Hawaii, Honolulu, HI 96822, U.S.A.

and

C. K. LAW

Department of Mechanical and Aerospace Engineering, Princeton University,
 Princeton, NJ 08544, U.S.A.

(Received 23 December 1992 and in final form 13 April 1993)

Abstract—The steady propagation of the planar laminar premixed flame in the doubly-infinite domain, with chain branching–termination reactions and weak volumetric heat loss, is studied using activation energy asymptotics. Two flame propagation regimes are identified and analyzed: a fast recombination regime in which the recombination reaction occurs in a steady-state manner with the high-activation-energy branching reaction in an overall reaction region which is much thinner than that of diffusion, and an intermediate recombination regime in which the recombination region is much thicker than the branching region but much thinner than the diffusion region. The analysis yields the characteristic dual solution, extinction turning point flame response, and shows that the flame propagation speed is reduced to $e^{-1/2}$ of the adiabatic value for both reaction mechanisms. The generality of this limit flame speed is noted.

1. INTRODUCTION

A CLASSICAL model problem for the study of premixed flame extinction is that of Spalding [1], who analyzed the one-dimensional freely-propagating flame with temperature sensitive one-step overall reaction and radiative heat loss. By letting these processes assume power-law temperature variations, with the exponents being 11 and 4 for the reaction rate and heat loss rate such that the former is more temperature sensitive than the latter, the analysis yields an extinction turning point, at which the normalized flame speed $\tilde{s}_{f,ex} = s_{f,ex}/s_f^0$ is 0.504, where s_f is the flame speed and s_f^0 the adiabatic, laminar flame speed, and the subscript 'ex' designates the extinction state. A more rigorous analysis [2] of the Spalding problem by using activation energy asymptotics and for weak, $O(\varepsilon)$ conductive heat loss subsequently yielded $\tilde{s}_{f,ex} = e^{-1/2}$, which is about 0.607, and a suitably-scaled heat loss rate $\tilde{L} = e^{-1}$ at the state of extinction, where ε is the reciprocal of the nondimensional activation energy to be defined later. The same result was shown to also hold [3] for a general $O(\varepsilon)$ volumetric heat loss function.

Recently, Sibulkin and co-workers [4–6] reported numerical solutions of transient planar and outwardly-propagating lean methane/air flame, allowing for radiative heat loss but with constant transport properties and one-step overall reaction. It was shown that at the limit of propagation \tilde{s}_f again assumed a value around 0.6. Lakshmisha *et al.* [7] and Law

and Egolfopoulos [8] extended these calculations to include variable properties and detailed chemistry, and further demonstrated the near-constancy of this value. Compared to previous analytical studies, these computational results are significant because they indicate the possibility that at the extinction limit the flame speed is always reduced to about 60% of its adiabatic value, being very insensitive both to the reaction mechanism, whether it is one-step or detailed, and to the loss mechanism, whether it is conductive or radiative, as long as it is $O(\varepsilon)$ and volumetric in nature.

The objective of the present study is to provide a fairly general proof of the above possibility based on activation energy asymptotics. Recognizing that previous analytical studies mostly involve a one-step large activation energy reaction, which obviously cannot capture the inherently-important chain branching and termination nature of realistic reaction schemes, we shall employ the Zel'dovich–Liñán two-step mechanism [9, 10], which consists of a branching reaction and a competing recombination (termination) reaction. This is believed to be the simplest representation of the chain nature of realistic reaction mechanisms. A general, $O(\varepsilon)$ volumetric heat loss function will be used in the analysis.

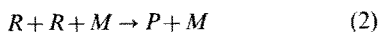
The system to be analyzed is formulated in the next section. It will be shown that there are two situations of interest, which respectively involve fast and intermediate recombination reaction rates. These two situations are separately analyzed in Sections 3 and 4.

NOMENCLATURE

a, b, c	integration constants	γ	Euler's constant
B	pre-exponential factor	δ	small parameter representing the relative thickness of the recombination region to the preheat region
c_p	specific heat at constant pressure	ε	small parameter of expansion, defined as \bar{T}_{ad}^2/\bar{E}
D	mass diffusion coefficient	η	compressed coordinate defined as $\varepsilon\bar{x}$
Da	Damköhler number	Θ	temperature perturbation in the recombination region
E	activation energy	θ	temperature perturbation in the branching region
f	mass flux through the flame or the flame propagation rate	Λ	burning rate eigenvalue
f^∞	flame propagation rate in the adiabatic limit	λ	thermal conductivity
F	rate controlling reactant (fuel)	ζ	stretched coordinate defined as \bar{x}/ε
H	temperature-dependent volumetric heat loss function	ρ	density
H_v	Heaviside function defined after equation (A24)	ϕ	fuel concentration perturbation in the branching region
L	heat loss coefficient	Ψ	radical concentration in the recombination region
L_C	conductive heat loss coefficient	ψ	radical concentration in the branching region
L_R	radiative heat loss coefficient	ω	reaction rate function.
Le	Lewis number		
M	third body of collision needed for recombination		
P	combustion product		
p	pressure		
q_F	heat of combustion per unit mass of fuel		
R	radical species		
R^∞	universal gas constant		
s_f	flame speed		
s_f^0	adiabatic, laminar flame speed		
T	temperature		
T_{ad}	adiabatic flame temperature		
T_f	leading order flame temperature		
W	molecular weight		
x	spatial coordinate attached to the flame front		
Y	mass fraction.		
Greek symbols		Subscripts	
Γ	parameter defined after equation (A20)	ex	the extinction state
		F	fuel
		M	third body of collision needed for recombination
		R	radical species
		$-\infty$	quantities at the upstream ambience.
		Superscripts	
		- , +	quantities in the upstream and downstream of the branching region
		n	pressure exponent
		~	dimensionless quantities.

2. FORMULATION

The two-step branching-recombination reaction mechanism adopted in this study is given by



where F , R , P and M respectively denote the rate controlling reactant (say the fuel), a radical species, the combustion product, and a third body of collision needed for recombination. In this mechanism, reaction (1) represents the two-body, high activation energy, branching reaction that has a small heat of combustion and produces more radicals than it consumes when reacted with the fuel. Equation (2) is the three-body, low activation energy, highly exothermic, termination reaction that combines the radicals to

form combustion products and generate heat. For simplicity we shall therefore assume that the branching reaction is thermally neutral while the termination reaction has zero activation energy. If we further assume that the rates of these two reaction steps, ω_j , vary with the first order of each reactant, then they can be respectively expressed for reactions (1) and (2) as

$$\omega_1 = B_1 p^{n_1} \frac{Y_F}{W_F} \frac{Y_R}{W_R} \exp(-E/R^\infty T), \quad (3)$$

$$\omega_2 = B_2 p^{n_2} \left(\frac{Y_R}{W_R} \right)^2 \frac{Y_M}{W_M}, \quad (4)$$

where T is the temperature, Y_i and W_i the mass fraction and molecular weight of species i , p the pressure.

B_j and n_j the pre-exponential factor and pressure exponent of reaction j , E the activation energy of the branching reaction, and R° the universal gas constant. The mass fraction of the third body, Y_M , can be considered to be a constant.

The physical problem under study is the steady propagation of the one-dimensional premixed flame in the doubly-infinite domain with an arbitrary temperature-dependent volumetric heat loss function $H(T)$. The governing equations and boundary conditions are given by

$$f c_p \frac{dT}{dx} - \frac{d}{dx} \left(\lambda \frac{dT}{dx} \right) = q_F W_F \omega_2 - \varepsilon H(T), \quad (5)$$

$$f \frac{dY_F}{dx} - \frac{d}{dx} \left(\rho D_F \frac{dY_F}{dx} \right) = -W_F \omega_1, \quad (6)$$

$$f \frac{dY_R}{dx} - \frac{d}{dx} \left(\rho D_R \frac{dY_R}{dx} \right) = W_R (\omega_1 - 2\omega_2), \quad (7)$$

$$x \rightarrow -\infty: T \rightarrow T_{-\infty}, \quad Y_F \rightarrow Y_{F,-\infty}, \quad Y_R \rightarrow 0, \quad (8a)$$

$$x \rightarrow \infty: \frac{dT}{dx} \rightarrow 0, \quad \frac{dY_F}{dx} \rightarrow 0, \quad \frac{dY_R}{dx} \rightarrow 0, \quad (8b)$$

where x is the spatial coordinate attached to the flame front, f the mass flux through the flame, ρ the density, c_p the specific heat at constant pressure, q_F the heat of combustion per unit mass of fuel consumed, λ the thermal conductivity, and D_i the mass diffusion coefficient of species i . The quantities λ , c_p and ρD_i are assumed to be constants. It is appropriate to concentrate on only $O(\varepsilon)$ heat loss because it represents the limiting situations of the extinction of weakly-burning flames. Clearly a flame will extinguish with $O(1)$ heat loss if it extinguishes with $O(\varepsilon)$ heat loss.

It is worth noting that equation (8b) implies $T \rightarrow T_\infty$, $Y_F \rightarrow 0$, and $Y_R \rightarrow 0$ at $x \rightarrow \infty$, for which T_∞ is the adiabatic flame temperature if the system is adiabatic and $T_\infty = T_{-\infty}$ in the presence of heat loss. Thus both of these conditions are applicable and yield the same result. Equations (8b) are adopted because they are more general. For the case of moderate (i.e. $O(1)$) heat loss in the downstream of the branching region, equations (8b) are still valid except $Y_F \neq 0$ at $x \rightarrow \infty$.

Introducing the nondimensional quantities

$$\begin{aligned} \tilde{T} &= \frac{2T}{q_F Y_{F,-\infty}/c_p}, & \tilde{Y}_F &= \frac{Y_F}{Y_{F,-\infty}}, \\ \tilde{Y}_R &= \frac{Y_R/W_R}{Y_{F,-\infty}/W_F}, & \tilde{f} &= \frac{f}{f^\circ}, & \tilde{x} &= \frac{x}{\lambda/(f^\circ c_p)}, \\ \tilde{E} &= \frac{2E/R^\circ}{q_F Y_{F,-\infty}/c_p}, & \tilde{H} &= \frac{2H\lambda}{(f^\circ)^2 c_p q_F Y_{F,-\infty}}, \\ Le_i &= \frac{\lambda/c_p}{\rho D_i}, & \varepsilon &= \frac{\tilde{T}_{ad}^2}{\tilde{E}}, \\ Da_1 &= \frac{B_1 \lambda p^{n_1} Y_{F,-\infty}}{(f^\circ)^2 c_p W_F}, \end{aligned}$$

$$Da_2 = \frac{2B_2 \lambda p^{n_2} Y_{F,-\infty} Y_M}{(f^\circ)^2 c_p W_F W_M},$$

where Le_i is the Lewis number of species i , Da_j the Damköhler number of the j th reaction, $f^\circ(Le_F)$ the flame propagation rate in the adiabatic limit, and $\tilde{T}_{ad} = 1 + \tilde{T}_{-\infty}$ the adiabatic flame temperature, the governing equations are nondimensionalized to

$$\tilde{f} \frac{d\tilde{T}}{d\tilde{x}} - \frac{d^2\tilde{T}}{d\tilde{x}^2} = Da_2 \tilde{Y}_R^2 - \varepsilon \tilde{H}(\tilde{T}), \quad (9)$$

$$\tilde{f} \frac{d\tilde{Y}_F}{d\tilde{x}} - \frac{1}{Le_F} \frac{d^2\tilde{Y}_F}{d\tilde{x}^2} = -Da_1 \tilde{Y}_F \tilde{Y}_R \exp(-\tilde{E}/\tilde{T}), \quad (10)$$

$$\begin{aligned} \tilde{f} \frac{d\tilde{Y}_R}{d\tilde{x}} - \frac{1}{Le_R} \frac{d^2\tilde{Y}_R}{d\tilde{x}^2} &= \\ Da_1 \tilde{Y}_F \tilde{Y}_R \exp(-\tilde{E}/\tilde{T}) - Da_2 \tilde{Y}_R^2, & (11) \end{aligned}$$

$$\tilde{x} \rightarrow -\infty: \tilde{T} \rightarrow \tilde{T}_{-\infty}, \quad \tilde{Y}_F \rightarrow 1, \quad \tilde{Y}_R \rightarrow 0, \quad \tilde{H} \rightarrow 0, \quad (12a)$$

$$\tilde{x} \rightarrow \infty: \frac{d\tilde{T}}{d\tilde{x}} \rightarrow 0, \quad \frac{d\tilde{Y}_F}{d\tilde{x}} \rightarrow 0, \quad \frac{d\tilde{Y}_R}{d\tilde{x}} \rightarrow 0, \quad \tilde{H} \rightarrow 0. \quad (12b)$$

This system will be solved by activation energy asymptotics.

Depending on the rate of the recombination reaction relative to that of the branching reaction, three flame propagation regimes can be identified. In a fast recombination regime both reaction rates are of the same order so that the reactions occur in the same thin reaction region. In an intermediate recombination regime the recombination rate is much slower than the branching rate but is much faster than the diffusion rate. Consequently the recombination region is much thicker than the branching region but much thinner than the preheat region. Finally, in a slow recombination regime the recombination rate is either comparable to or slower than the diffusion rate such that the thickness of the recombination region is either of the same order of or larger than that of the preheat region.

In the next two sections, the fast and intermediate recombination regimes will be analyzed sequentially. The slow recombination regime will not be analyzed because it represents very weak chemical systems which are not likely to be of interest to combustion.

3. FAST RECOMBINATION REGIME

In this regime, the rates of the recombination and branching reactions are of the same order so that both reactions occur in the same thin reaction region. Thus the radicals are consumed to produce heat almost immediately after they are generated. This implies that the concentration of the radicals is basically independent of time and is very small, and hence the steady

state approximation, $dY_R/dt \rightarrow 0$, is applicable. Figure 1 shows the species and temperature profiles of such a flame.

With the application of the steady state approximation, setting equation (11) to zero readily yields

$$\tilde{Y}_R = \frac{Da_1}{Da_2} \tilde{Y}_F \exp(-\tilde{E}/\tilde{T}). \quad (13)$$

The vanishing of the LHS of equation (11) is justified by the small radical concentration. Substituting \tilde{Y}_R into the reaction rate terms of equations (9) and (10), the problem is then reduced to a one-step reaction with a rate

$$\frac{Da_1^2}{Da_2} \tilde{Y}_F^2 \exp(-2\tilde{E}/\tilde{T}),$$

which is second order with respect to \tilde{Y}_F , and has an effective Damköhler number Da_1^2/Da_2 and an effective activation energy $2\tilde{E}$. Compared with previous asymptotic studies (see, for example, refs. [2, 3]) which adopted a one-step first-order overall reaction, the difference in the mathematical aspect of the two problems is only the reaction order. It is therefore of interest to study that, apart from the obvious difference in the physical interpretation of these two problems, in terms of the chain branching and termination reactions through modifications of the effective Damköhler number and activation energy, what additional effect a simple change in the reaction order can produce. Due to the similar nature of the asymptotic derivation with previous studies, only the key steps will be shown below.

In the outer, chemically inert regions, there is no branching reaction in the upstream region because the flow temperature is low and the reaction has a high activation energy. In the downstream region branching is terminated because of complete fuel consumption. The recombination reaction does not exist in the outer regions due to complete radical consumption. Thus the outer solutions for \tilde{Y}_F are

$$\tilde{Y}_F^- = 1 - (b_0 + \epsilon b_1 + \dots) \exp(Le_F \tilde{f} \tilde{x}), \quad (14)$$

$$\tilde{Y}_F^+ = 0, \quad (15)$$

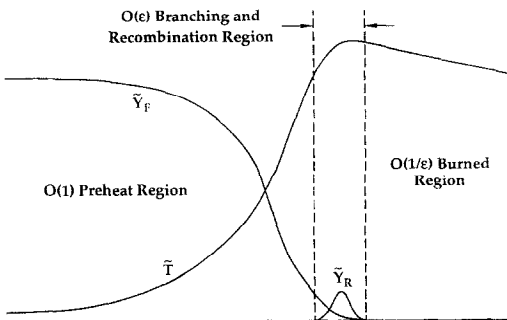


FIG. 1. Schematic of the flame structure for the fast recombination regime. \tilde{Y}_R has been magnified for clarity.

while \tilde{T} can be expanded as $\tilde{T}^\pm = \tilde{T}_0^\pm + \epsilon \tilde{T}_1^\pm + \dots$. This yields

$$\tilde{T}_0^- = \tilde{T}_{-,\infty} + a_0 \exp(\tilde{f} \tilde{x}), \quad (16a)$$

$$\tilde{f} \tilde{T}_1^- - \frac{d\tilde{T}_1^-}{d\tilde{x}} = - \int_{-\infty}^{\tilde{x}} \tilde{H}(\tilde{T}_0^-) d\tilde{x}, \quad (16b)$$

$$\tilde{f} \frac{d\tilde{T}_0^+}{d\eta} = -\tilde{H}(\tilde{T}_0^+). \quad (17)$$

In the above the superscripts $-$ and $+$ respectively denote quantities in the upstream preheat region and downstream burned region, while a_i and b_i the integration constants to be determined. Note that in the downstream region, a compressed coordinate $\eta = \epsilon \tilde{x}$ is used because the temperature decreases gradually from the flame temperature to the ambient temperature due to the heat loss.

In the inner, chemically reactive region, the coordinate is stretched as $\xi = \tilde{x}/\epsilon$ while \tilde{T} and \tilde{Y}_F are expanded as

$$\tilde{T} = \tilde{T}_1 - \epsilon \theta_1 - \epsilon^2 \theta_2 + \dots, \quad (18)$$

$$\tilde{Y}_F = \epsilon \phi_1 + \epsilon^2 \phi_2 + \dots. \quad (19)$$

Substituting equations (18) and (19) into equations (9), (10) and (13), and expanding, we obtain the inner equations

$$\frac{d^2 \phi_1}{d\xi^2} = \Lambda \phi_1^2 \exp \left[-2 \left(\frac{\tilde{T}_{ad}}{\tilde{T}_1} \right)^2 \theta_1 \right], \quad (20)$$

$$\frac{d^2 \theta_1}{d\xi^2} - \frac{1}{Le_F} \frac{d^2 \phi_1}{d\xi^2} = 0, \quad (21)$$

$$\frac{d^2 \theta_2}{d\xi^2} - \frac{1}{Le_F} \frac{d^2 \phi_2}{d\xi^2} - \tilde{f} \left(\frac{d\theta_1}{d\xi} - \frac{d\phi_1}{d\xi} \right) = 0, \quad (22)$$

where

$$\Lambda = \epsilon^2 Le_F \frac{Da_1^2}{Da_2} \exp \left(-2 \frac{\tilde{E}}{\tilde{T}_1} \right). \quad (23)$$

Matching the inner and outer solutions yields the relations

$$a_0 = \tilde{T}_1 - \tilde{T}_{-,\infty}, \quad \tilde{T}_0^+(\eta=0) = \tilde{T}_1, \quad b_0 = 1, \quad (24)$$

and the matching conditions to solve equations (20) to (22),

$$\theta_1(\xi \rightarrow -\infty) = -\tilde{T}_1^-(\tilde{x}=0) - \tilde{f}(\tilde{T}_1^- - \tilde{T}_{-,\infty}) \xi, \quad (25a)$$

$$\theta_1(\xi \rightarrow \infty) = -\tilde{T}_1^+(\eta=0), \quad (25b)$$

$$\left(\frac{d\theta_2}{d\xi} - \tilde{f} \theta_1 \right)_{\xi \rightarrow -\infty} = \left(\tilde{f} \tilde{T}_1^- - \frac{d\tilde{T}_1^-}{d\tilde{x}} \right)_{\tilde{x} \rightarrow 0} = - \int_{-\infty}^0 \tilde{H}(\tilde{T}_0^-) d\tilde{x}, \quad (26a)$$

$$\frac{d\theta_2}{d\xi}(\xi \rightarrow \infty) = - \frac{d\tilde{T}_0^+}{d\eta}(\eta \rightarrow 0) = \frac{\tilde{H}(\tilde{T}_1)}{\tilde{f}}, \quad (26b)$$

$$\phi_1(\xi \rightarrow -\infty) = -b_1 - Le_F \tilde{f} \xi, \quad (27a)$$

$$\phi_1(\xi \rightarrow \infty) = 0, \tag{27b}$$

$$\left(\frac{1}{Le_F} \frac{d\phi_2}{d\xi} - \tilde{f}\phi_1 \right)_{\xi \rightarrow -\infty} = 0, \tag{28a}$$

$$\frac{d\phi_2}{d\xi}(\xi \rightarrow \infty) = 0, \tag{28b}$$

where equations (27a, b) also imply

$$\frac{d\phi_1}{d\xi}(\xi \rightarrow -\infty) = -Le_F \tilde{f}, \tag{29a}$$

$$\frac{d\phi_1}{d\xi}(\xi \rightarrow \infty) = 0. \tag{29b}$$

Equations (16b) and (17) have been applied in obtaining the second relation of equations (26a, b).

Integrating equations (21) twice and (22) once subject to the matching conditions in equations (25)–(28) yields

$$\tilde{T}_r = 1 + \tilde{T}_{-\infty} = \tilde{T}_{ad}, \tag{30}$$

$$\theta_i = \frac{\phi_1}{Le_F} - \frac{\tilde{L}}{2\tilde{f}^2}, \tag{31}$$

where

$$\tilde{L} = 2 \left[\tilde{H}(\tilde{T}_{ad}) + \tilde{f} \int_{-\infty}^0 \tilde{H}(\tilde{T}_0^-) d\tilde{x} \right]. \tag{32}$$

It can be readily demonstrated, by re-defining $\tilde{x} = \tilde{f}\tilde{x} = x/[\lambda/(fc_p)]$, that \tilde{L} is independent of \tilde{f} .

Equation (30) shows that the leading order flame temperature is the adiabatic flame temperature, which is reasonable because the heat loss is assumed to be $O(\epsilon)$. Substituting equation (31) into equation (20) and integrating the resulting equation once subject to the boundary conditions in equations (27a, b) and (29a, b), we obtain an expression which determines the flame propagation rate,

$$\tilde{f}^2 \exp\left(\frac{\tilde{L}}{\tilde{f}^2}\right) = \frac{Le_F}{2} \Lambda. \tag{33}$$

In the adiabatic limit, $\tilde{H} \equiv 0$ and $\tilde{f} = 1$. We then have $(Le_F \Lambda/2) = 1$ for the laminar flame propagation rate f° . Equation (33) then becomes

$$\tilde{f}^2 \exp\left(\frac{\tilde{L}}{\tilde{f}^2}\right) = 1. \tag{34}$$

The quantities \tilde{T}_r^\pm and b_1 are not of interest and hence will not be solved.

Equation (34), of course, is exactly the same as that of, say, Joulin and Clavin [2] who considered conductive heat loss and a first-order reaction. Repro-

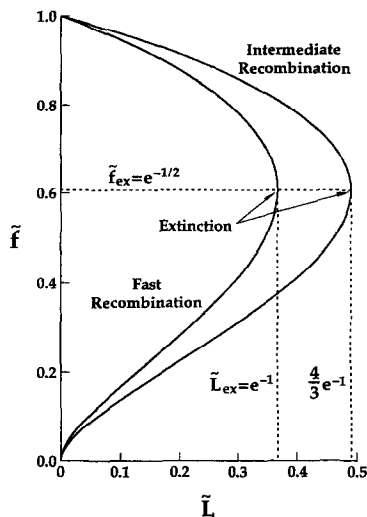


FIG. 2. Normalized flame propagation rate \tilde{f} as a function of the heat loss parameter \tilde{L} for the fast and intermediate recombination regimes.

ducing equation (34) in Fig. 2, it is seen that there exists a maximum value of \tilde{L} , \tilde{L}_{ex} , above which no solution exists. For $\tilde{L} < \tilde{L}_{ex}$, there are two solutions for each \tilde{L} , although it is well established that only the upper branch gives the stable solution. This critical state is then defined as that of extinction, being characterized by

$$\tilde{L}_{ex} = e^{-1} \quad \text{and} \quad \tilde{f}_{ex} = e^{-1/2}. \tag{35}$$

4. INTERMEDIATE RECOMBINATION REGIME

In this regime the rate of the recombination reaction is much slower than that of the branching reaction but much faster than the diffusive-convective transport rate. Consequently the recombination region is much thicker than the branching region but much thinner than the transport region. By defining a second small parameter δ to describe the characteristic thickness of the recombination region, where $\epsilon \ll \delta \ll 1$, the flame structure then includes an $O(\epsilon)$ branching region sandwiched by $O(\delta)$ recombination regions, which in turn are embedded within an $O(1)$ upstream preheat region and an $O(1/\epsilon)$ downstream burned region. Since the exothermic recombination reaction continues subsequent to completion of the branching reaction, temperature will also continuously increase until heat loss becomes dominant. The species and temperature profiles are shown in Fig. 3.

By separately analyzing the five regions and performing the requisite matching, which is presented in the Appendix, the flame speed response is now given by

$$\tilde{f}^{4/3} \exp\left(\frac{\tilde{L}}{2\tilde{f}^2}\right) = 1. \tag{36}$$

Comparing equations (36) and (34), we can see that

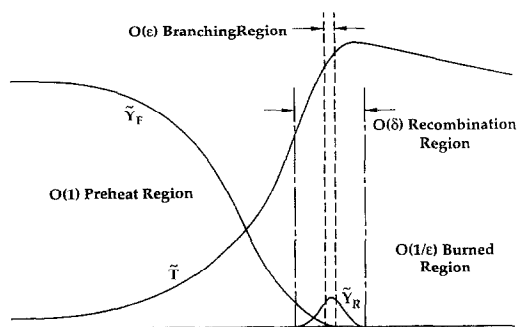


FIG. 3. Schematic of the flame structure for the intermediate recombination regime.

their functional expressions are similar although the branching and recombination reactions occur at different length scales. Thus the behavior of \tilde{f} as a function of \tilde{L} is qualitatively similar to that of the fast recombination regime, as shown in Fig. 2. The flame extinction limit is now described by

$$\tilde{f}_{\text{ex}} = e^{-1/2}, \quad \tilde{L}_{\text{ex}} = (4/3)e^{-1}. \quad (37)$$

It is interesting to note that although the reaction kinetics adopted in this regime are different from those of Section 3, the flame still extinguishes at $\tilde{f} = e^{-1/2}$, albeit at a different \tilde{L}_{ex} . Combining this result with those obtained from the analysis by adopting a one-step overall reaction [2] and the numerical studies [4–8], it may be suggested that extinction is achieved when the inherent nonadiabaticity of the system reduces the flame propagation rate to about $e^{-1/2}$ or 0.607 of its adiabatic value. The fact that a large heat loss is needed to extinguish the flame in the intermediate recombination regime as compared to that in the fast recombination regime is also reasonable because the overall reaction rate is less temperature sensitive for the slower recombination rate.

5. CONCLUDING REMARKS

In this study, we have analyzed the extinction limit of the planar premixed flame with $O(\varepsilon)$ volumetric heat loss by adopting a two-step, branching and termination kinetics. Based on the relative rates of these two reactions, the fast and intermediate combination situations are studied. The results show that the flame always extinguishes when the flame propagation rate is reduced to $e^{-1/2}$ of its adiabatic value, which is consistent with previous analytical results with one-step reaction and numerical results with detailed chemistry.

The above analysis is based on a general heat loss function H . Two loss mechanisms that are usually specified are conduction and radiation. For a linear heat loss function, $H = L_C(T - T_{\infty})$ where L_C is the heat loss coefficient. We then have $\tilde{L} = (4\lambda L_C)/(f^{\infty} c_p)^2$. For radiative loss, $H = L_R(T^4 - T_{\infty}^4)$ and we have

$$\tilde{L} = \frac{2\lambda L_R}{(f^{\infty} c_p)^2} \left(\frac{q_F Y_{F,\infty}}{2c_p} \right)^3 \left(\frac{5}{4} + \frac{16}{3} \tilde{T}_{\infty} + 9\tilde{T}_{\infty}^2 + 8\tilde{T}_{\infty}^3 \right).$$

Finally, we note that the influence of the reactant concentration on flame extinction is primarily through the factor $(f^{\infty})^2$ in the definition of \tilde{H} . Thus as the fuel concentration becomes either leaner or richer, the decrease in f^{∞} would lead to a corresponding increase in \tilde{H} . It is therefore clear that the present result also predicts the existence of concentration limits at the heat loss rate \tilde{L}_{ex} , beyond which steady flame propagation is not possible. Such limits have been identified as the flammability limits [1, 4, 5, 7, 8].

Acknowledgements—This study was supported by the Air Force Office of Scientific Research and the Office of Basic Energy Sciences of the Department of Energy. We thank Dr John K. Bechtold for stimulating discussions.

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APPENDIX: DERIVATION FOR THE INTERMEDIATE RECOMBINATION REGIME

All the solutions are expanded with respect to the two small parameters ε and δ . Thus in the outer, transport regions away from the recombination region, $\tilde{Y}_R^{\pm} = 0$, while \tilde{Y}_F^{\pm} is given by equation (15) and

$$\tilde{Y}_F^{\pm} = 1 - \{ [b_0 + O(\delta)] + \varepsilon [b_1 + O(\delta)] + O(\varepsilon^2) \} \exp(Le_F \tilde{f} \tilde{x}). \quad (A1)$$

The solution of \tilde{T}^\pm can be obtained by first expanding it in the form

$$\tilde{T}^\pm = [\tilde{T}_0^\pm + \delta\tilde{T}_1^\pm + O(\delta^2)] + \varepsilon[\tilde{T}_2^\pm + O(\delta)] + O(\varepsilon^2), \quad (\text{A2})$$

which is then substituted into equation (9) to yield equations (16a), (17), and

$$\tilde{T}_1^- = a_1 \exp(\tilde{f}\tilde{x}), \quad (\text{A3})$$

$$\tilde{f}\tilde{T}_2^- - \frac{d\tilde{T}_2^-}{d\tilde{x}} = - \int_{-\infty}^{\tilde{x}} \tilde{H}(\tilde{T}_0^-) d\tilde{x}. \quad (\text{A4})$$

The compressed coordinate $\eta = \varepsilon\tilde{x}$ is again used in the burned region such that all the terms in \tilde{T}^+ are functions of η .

In the $O(\delta)$ recombination region, the coordinate is stretched as $\zeta = \tilde{x}/\delta$. There is no branching reaction in this region because of its high activation energy. Thus the outer solution of \tilde{Y}_F is still applicable. Moreover, only $O(\delta)$ variations are possible for \tilde{T}^\pm and \tilde{Y}_R^\pm so that their expansions are

$$\tilde{T}^\pm = [\tilde{T}_{r,0}^\pm - \delta\Theta_1^\pm - \delta^2\Theta_2^\pm + O(\delta^3)] - \varepsilon[\Theta_3^\pm + \delta\Theta_4^\pm + O(\delta^2)] + O(\varepsilon^2), \quad (\text{A5})$$

$$\tilde{Y}_R^\pm = [\delta\Psi_1^\pm + \delta^2\Psi_2^\pm + O(\delta^3)] + \varepsilon[\Psi_3^\pm + \delta\Psi_4^\pm + O(\delta^2)] + O(\varepsilon^2). \quad (\text{A6})$$

Substituting equations (A5) and (A6) into equations (9) and (11), but with the branching reaction frozen, and expanding, we obtain for \tilde{Y}_R^\pm ,

$$\frac{1}{Le_R} \frac{d^2\Psi_i^\pm}{d\zeta^2} = 2^{i-1}\Lambda_2\Psi_1^\pm\Psi_i^\pm, \quad i = 1, 2, \quad (\text{A7})$$

$$\frac{1}{Le_R} \frac{d^2\Psi_{i+2}^\pm}{d\zeta^2} - \tilde{f} \frac{d\Psi_i^\pm}{d\zeta} = 2\Lambda_2[\Psi_1^\pm\Psi_{i+2}^\pm + (i-1)\Psi_i^\pm\Psi_{i+1}^\pm], \quad i = 1, 2, \quad (\text{A8})$$

where $\Lambda_2 = \delta^3 Da_2$ is the reduced Damköhler number for the termination reaction. The local coupling functions are given by

$$\frac{d^2}{d\zeta^2} \left(\Theta_i^\pm - \frac{\Psi_i^\pm}{Le_R} \right) = 0, \quad i = 1, 2, \quad (\text{A9})$$

$$\frac{d^2}{d\zeta^2} \left(\Theta_{i+2}^\pm - \frac{\Psi_{i+2}^\pm}{Le_R} \right) - \tilde{f} \frac{d}{d\zeta} \left(\Theta_i^\pm - \Psi_i^\pm \right) = 0, \quad i = 1, 2. \quad (\text{A10})$$

The volumetric heat loss is not important in the above equations because the recombination regions are still very thin, of $O(\delta)$. Each of the above equations needs to be solved separately in the upstream and downstream of the branching region.

Matching the solutions in the recombination region with the outer diffusive-convective regions yield $a_0 = \tilde{T}_{r,0} - \tilde{T}_{-\infty}$, $\tilde{T}_0^+(\eta=0) = \tilde{T}_{r,0}$, and the conditions to solve equations (A7)–(A10),

$$\Psi_i^\pm(\zeta \rightarrow \pm\infty) = 0, \quad i = 1, 2, 3, 4, \quad (\text{A11})$$

$$\Theta_i^+(\zeta \rightarrow \infty) = -\tilde{T}_1^+(\eta=0), \quad i = 1, 2, \quad (\text{A12})$$

$$\frac{d\Theta_4^+}{d\zeta}(\zeta \rightarrow \infty) = -\frac{d\tilde{T}_0^+}{d\eta}(\eta=0) = \frac{\tilde{H}(\tilde{T}_{r,0})}{\tilde{f}}, \quad (\text{A13})$$

$$\Theta_1^-(\zeta \rightarrow -\infty) = -a_1 - \tilde{f}(\tilde{T}_{r,0} - \tilde{T}_{-\infty})\zeta, \quad (\text{A14})$$

$$\Theta_2^-(\zeta \rightarrow -\infty) = -\tilde{T}_2^-(\tilde{x}=0), \quad (\text{A15})$$

$$\left(\frac{d\Theta_4^-}{d\zeta} - \tilde{f}\Theta_2^- \right)_{\zeta \rightarrow -\infty} = \left(\tilde{f}\tilde{T}_2^- - \frac{d\tilde{T}_2^-}{d\tilde{x}} \right)_{\tilde{x}=0} = - \int_{-\infty}^0 \tilde{H}(\tilde{T}_0^-) d\tilde{x}. \quad (\text{A16})$$

Equations (17) and (A4) have respectively been used in deriving the second relation of equations (A13) and (A16).

Solving equations (A7) and (A8) subject to the matching conditions in equation (A11), we obtain

$$\Psi_1^\pm = 4/(\Gamma\zeta + c_1^\pm)^2, \quad (\text{A17})$$

$$\Psi_2^\pm = \Gamma c_2^\pm / (\Gamma\zeta + c_1^\pm)^3, \quad (\text{A18})$$

$$\Psi_3^\pm = \frac{1}{\Gamma\zeta + c_1^\pm} \left[\frac{c_3^\pm}{(\Gamma\zeta + c_1^\pm)^2} + \frac{4Le_R\tilde{f}}{5} \right], \quad (\text{A19})$$

$$\frac{d\Psi_4^\pm}{d\zeta} = \frac{\Gamma}{(\Gamma\zeta + c_1^\pm)^3} \left[\frac{c_4^\pm}{\Gamma\zeta + c_1^\pm} - \frac{3}{2} \frac{c_2^\pm c_3^\pm \Gamma}{(\Gamma\zeta + c_1^\pm)^2} - \frac{1}{5} c_2^\pm Le_R \tilde{f} \right], \quad (\text{A20})$$

where c_i^\pm are the integration constants to be determined and $\Gamma = (2Le_R\Lambda_2/3)^{1/2}$.

Applying equations (A17)–(A20) to equations (A9) and (A10), then solving the resulting equations subject to the matching conditions (A12)–(A16), we obtain

$$\Theta_1^\pm = \frac{1}{Le_R} \frac{4}{(\Gamma\zeta + c_1^\pm)^2} - [\tilde{f}(\tilde{T}_{r,0} - \tilde{T}_{-\infty})\zeta - a_1]H_v(-\zeta) - \tilde{T}_1^+(\eta=0)H_v(\zeta), \quad (\text{A21})$$

$$\Theta_2^\pm = \frac{1}{Le_R} \frac{\Gamma c_2^\pm}{(\Gamma\zeta + c_1^\pm)^3} - \tilde{T}_2^-(\tilde{x}=0)H_v(-\zeta) - \tilde{T}_2^+(\eta=0)H_v(\zeta), \quad (\text{A22})$$

$$\frac{d^2\Theta_3^\pm}{d\zeta^2} = \frac{8}{(\Gamma\zeta + c_1^\pm)^3} \left[\frac{c_3^\pm \Lambda_2}{(\Gamma\zeta + c_1^\pm)^2} + \Gamma\tilde{f} \left(\frac{6}{5} - \frac{1}{Le_R} \right) \right] - \tilde{f}^2(\tilde{T}_{r,0} - \tilde{T}_{-\infty})H_v(-\zeta), \quad (\text{A23})$$

$$\frac{d\Theta_4^\pm}{d\zeta} = \frac{1}{(\Gamma\zeta + c_1^\pm)^3} \left[\frac{\Gamma}{Le_R} \frac{c_4^\pm}{\Gamma\zeta + c_1^\pm} - \Gamma\tilde{f}c_2^\pm \left(\frac{6}{5} - \frac{1}{Le_R} \right) - \frac{c_2^\pm c_3^\pm \Lambda_2}{(\Gamma\zeta + c_1^\pm)^2} \right] + \frac{\tilde{H}(\tilde{T}_{r,0})}{\tilde{f}} H_v(\zeta) - \left[\tilde{f}\tilde{T}_2^-(\tilde{x}=0) + \int_{-\infty}^0 \tilde{H}(\tilde{T}_0^-) d\tilde{x} \right] H_v(-\zeta), \quad (\text{A24})$$

where H_v is the Heaviside function with $H_v(\zeta) = 1$ for $\zeta > 0$ and 0 for $\zeta < 0$. In the above only the solutions required for matching with those in the branching region are presented. Because $(d\Psi_1^-/d\zeta)_0 \geq 0$ and $(d\Psi_1^+/d\zeta)_0 \leq 0$, we must have $c_1^- < 0$ and $c_1^+ > 0$.

In the thin branching region, the coordinate is stretched as $\xi = \tilde{x}/\varepsilon$ while \tilde{Y}_F and \tilde{Y}_R are expanded as

$$\tilde{Y}_F = \varepsilon(\phi_1 + \dots) + \varepsilon^2(\phi_2 + \dots) + O(\varepsilon^3), \quad (\text{A25})$$

$$\tilde{Y}_R = (\delta\psi_1 + \delta^2\psi_3 + \dots) + \varepsilon(\psi_2 + \dots) + \varepsilon^2(\psi_4 + \dots) + O(\varepsilon^3). \quad (\text{A26})$$

Because the activation energy for the branching reaction is high, variation of temperature can only be $O(\varepsilon)$. Thus \tilde{T} is expanded as

$$\tilde{T} = (\tilde{T}_{r,0} - \delta\tilde{T}_{r,1} + \dots) - \varepsilon \left(\theta_1 + \frac{\varepsilon}{\delta}\theta_2 + \dots \right) - \varepsilon^2(\theta_3 + \dots) + \dots \quad (\text{A27})$$

Although the activation energy is large, it is not extremely high such that the branching reaction can occur in a temperature range smaller than its maximum value by an $O(\delta)$ amount. Substituting equations (A25)–(A27) to equations (9)–(11) and expanding, we obtain the following equations that describe the branching region,

$$\frac{d^2\psi_i}{d\xi^2} = 0, \quad i = 1, 3, \quad (\text{A28})$$

$$\frac{d^2\theta_1}{d\xi^2} = 0, \quad (\text{A29})$$

$$\frac{d^2\theta_2}{d\xi^2} = \Lambda_2\psi_1^2, \quad (\text{A30})$$

$$\frac{d^2\theta_3}{d\xi^2} - \tilde{f} \frac{d\theta_1}{d\xi} = 2\Lambda_2\psi_1\psi_3, \quad (\text{A31})$$

$$\frac{d^2}{d\xi^2} \left(\frac{\phi_1}{Le_F} + \frac{\psi_2}{Le_R} \right) = 0, \quad (\text{A32})$$

$$\frac{1}{Le_F} \frac{d^2\phi_2}{d\xi^2} - \tilde{f} \frac{d\phi_1}{d\xi} + \frac{1}{Le_R} \frac{d^2\psi_3}{d\xi^2} - \tilde{f} \frac{d\psi_2}{d\xi} = 2\Lambda_2\psi_1\psi_3, \quad (\text{A33})$$

$$\frac{1}{Le_F} \frac{d^2\phi_1}{d\xi^2} = \Lambda_1\phi_1\psi_1 \exp \left[- \left(\frac{\tilde{T}_{ad}}{\tilde{T}_{t,0}} \right)^2 \theta_1 \right], \quad (\text{A34})$$

where

$$\Lambda_1 = \varepsilon^2 \delta Da_1 \exp \left(- \frac{\tilde{E}}{\tilde{T}_{t,0} - \delta \tilde{T}_{t,1} + \dots} \right) \quad (\text{A35})$$

is the reduced Damköhler number for the branching reaction.

Matching is performed first by expressing the outer solutions of \tilde{Y}_{\pm}^{\pm} in terms of $\xi = \tilde{x}/\delta$, expanding, and then equating the resulting expression with the solution in the branching region. This gives $b_0 = 1$ and equations (27)–(29). Next, the solutions of \tilde{T}^{\pm} and \tilde{Y}_{\pm}^{\pm} in the recombination region are expressed in terms of $\xi = (\varepsilon/\delta)\xi$ and expanded to yield

$$\tilde{T}_{t,1} = \frac{4}{Le_R(c_1^{\pm})^2} - \tilde{T}_1^{\pm}(\eta = 0), \quad (\text{A36})$$

and the matching conditions

$$\psi_1(\xi \rightarrow \pm\infty) = 4/(c_1^{\pm})^2, \quad (\text{A37})$$

$$\psi_2(\xi \rightarrow \pm\infty) = \Gamma(c_2^{\pm} - 8\xi)/(c_1^{\pm})^3, \quad (\text{A38})$$

$$\psi_3^{\pm}(\xi \rightarrow \pm\infty) = \frac{1}{c_1^{\pm}} \left[\frac{c_3^{\pm}}{(c_1^{\pm})^2} + \frac{4}{5} \frac{Le_R \tilde{f}}{\Gamma} \right], \quad (\text{A39})$$

$$\frac{d\psi_4}{d\xi}(\xi \rightarrow \pm\infty) = \frac{\Gamma}{(c_1^{\pm})^3} \left\{ \frac{c_4^{\pm}}{c_1^{\pm}} - \left[\frac{3}{2} \frac{c_3^{\pm} \Gamma}{(c_1^{\pm})^2} + \frac{1}{5} Le_R \tilde{f} \right] (c_2^{\pm} - 8\xi) \right\}, \quad (\text{A40})$$

$$\theta_1(\xi \rightarrow \pm\infty) = \frac{\Gamma}{Le_R(c_1^{\pm})^3} (c_2^{\pm} - 8\xi) - [\tilde{T}_2^{\pm}(\tilde{x} = 0) + \tilde{f}(\tilde{T}_{t,0} - \tilde{T}_{t,\infty})\xi] H_1(-\xi) - \tilde{T}_2^{\pm}(\eta = 0) H_v(\xi), \quad (\text{A41})$$

$$\theta_2(\xi \rightarrow \pm\infty) = 2\Lambda_2(4\xi - c_2^{\pm})\xi/(c_1^{\pm})^4, \quad (\text{A42})$$

$$\begin{aligned} \frac{d\theta_3}{d\xi}(\xi \rightarrow \pm\infty) &= \frac{1}{(c_1^{\pm})^3} \left\{ \frac{c_3^{\pm} \Gamma}{c_1^{\pm} Le_R} - \left[\frac{c_3^{\pm} \Lambda_2}{(c_1^{\pm})^2} + \Gamma \tilde{f} \left(\frac{6}{5} - \frac{1}{Le_R} \right) \right] (c_2^{\pm} - 8\xi) \right\} + \frac{\tilde{H}(\tilde{T}_{t,0})}{\tilde{f}} H_v(\xi) \\ &- \left[\tilde{f}^2(\tilde{T}_{t,0} - \tilde{T}_{t,\infty})\xi + \tilde{f} \tilde{T}_2^{\pm}(\tilde{x} = 0) + \int_{-\infty}^0 \tilde{H}(\tilde{T}_0) d\tilde{x} \right] H_v(-\xi). \end{aligned} \quad (\text{A43})$$

In analyzing the branching region, first equations (A28) for ψ_1 and (A32) are solved subject to equations (27), (A37) and (A38) to yield

$$c_1^+ = -c_1^- = 2 \left(\frac{8\Lambda_2}{3Le_R \tilde{f}^2} \right)^{1/6}, \quad (\text{A44})$$

$$\psi_1 = \left(\frac{3Le_R \tilde{f}^2}{8\Lambda_2} \right)^{1/3}. \quad (\text{A45})$$

Substituting equations (A44) into equation (A36), $\tilde{T}_{t,1}$ can be determined if $\tilde{T}_1^{\pm}(\eta = 0)$ is known. Because $\tilde{T}_{t,1}$ represents the temperature at which branching reaction occurs, it is only a function of activation energy and chemical reactivity but does not depend on the heat loss. Thus by knowing that $\tilde{T}_1^{\pm}(\eta = 0) = 0$ and $\tilde{f} = 1$ in the adiabatic limit, we obtain

$$\tilde{T}_{t,1} = \left(\frac{3}{8Le_R \Lambda_2} \right)^{1/3}, \quad (\text{A46})$$

and hence $\tilde{T}_1^{\pm}(\eta = 0) = \tilde{T}_{t,1}(\tilde{f}^{2/3} - 1)$.

Next, by sequentially integrating equations (A28) and (A29) twice, and equations (A30), (A31) and (A33) once, subject to equation (A44) and the proper boundary conditions of (A39)–(A43), we obtain $\tilde{T}_{t,0} = 1 + \tilde{T}_{t,\infty} = \tilde{T}_{ad}$, $c_2^- = c_2^+$, $c_3^- = c_3^+$, $b_1 = -Le_F \tilde{f} c_2^{\pm}/8$, as well as

$$\theta_1 = -\frac{\tilde{f}}{2} \left(\xi - \frac{c_2^+}{8} \right) + \frac{\tilde{L}}{2\tilde{f}^2}, \quad (\text{A47})$$

where \tilde{L} is defined in equation (32).

Finally, substituting equations (A45) and (A47) into equation (A34), and defining the new variables

$$\begin{aligned} \tilde{\phi} &= \frac{\phi_1}{2Le_F}, \\ \tilde{\xi} &= \left[2(2Le_F \Lambda_1)^{1/2} \left(\frac{3Le_R}{\Lambda_2} \right)^{1/6} \right] \left[\tilde{f}^{2/3} \exp \left(\frac{\tilde{L}}{4\tilde{f}^2} \right) \right]^{-1} \\ &\times \exp \left[\frac{\tilde{f}}{4} \left(\xi - \frac{c_2^+}{8} \right) \right], \end{aligned} \quad (\text{A48})$$

the structure equation is obtained,

$$\tilde{\xi}^2 \frac{d^2\tilde{\phi}}{d\tilde{\xi}^2} + \tilde{\xi} \frac{d\tilde{\phi}}{d\tilde{\xi}} - \tilde{\xi}^2 \tilde{\phi} = 0, \quad (\text{A49})$$

$$\begin{aligned} \tilde{\phi}(\tilde{\xi} = 0) &= \ln \left\{ \left[2Le_F \Lambda_1 \left(\frac{3Le_R}{\Lambda_2} \right)^{1/3} \right] \right. \\ &\times \left. \left[\tilde{f}^{4/3} \exp \left(\frac{\tilde{L}}{2\tilde{f}^2} \right) \right]^2 - 2 \ln(\tilde{\xi}/2) \right\}, \end{aligned} \quad (\text{A50})$$

$$\tilde{\phi}(\tilde{\xi} \rightarrow \infty) = 0. \quad (\text{A51})$$

This is a modified Bessel function of order zero whose solution is $\tilde{\phi} = cK_0(\tilde{\xi})$, where c is the integration constant, after equation (A51) is applied. Because $K_0(\tilde{\xi} = 0) = -\ln(\tilde{\xi}/2) - \gamma$, where $\gamma = 0.5772$ is the Euler's constant, upon applying equation (A50) we obtain

$$\left[2Le_F \Lambda_1 \left(\frac{3Le_R}{\Lambda_2} \right)^{1/3} \right] \left[\tilde{f}^{4/3} \exp \left(\frac{\tilde{L}}{2\tilde{f}^2} \right) \right]^2 = \exp(-2\gamma). \quad (\text{A52})$$

In the adiabatic limit, $\tilde{L} = 0$ and $\tilde{f} = 1$, we have

$$\left[2Le_F \Lambda_1 \left(\frac{3Le_R}{\Lambda_2} \right)^{1/3} \right] = \exp(-2\gamma). \quad (\text{A53})$$

Substituting equation (A53) into equation (A52), we obtain the flame response given by equation (36).