Asymptotic Theory of Diffusion-Flame Extinction with Radiant Loss from the Flame Zone

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Abstract—Laminar diffusion flames in counterflow configurations such as stagnation-point boundary layers are analyzed by methods of matched asymptotic expansions with large parameters being the temperature sensitivities of the rates of chemical heat generation and radiant heat loss. Formulas are derived defining critical conditions for flame extinction, including influences of radiant loss.

1 INTRODUCTION

An appreciable amount of research has been performed on describing extinction conditions for diffusion flames by use of methods of matched asymptotic expansions, with the ratio of the overall energy of activation for heat release to the thermal energy of the reacting gases taken as a large parameter of expansion. Motivation for this work and literature concerning earlier studies have been discussed by Krishnamurthy, Williams and Seshadri (1976). Newer investigations employing these asymptotic methods have been reviewed recently (Williams, 1981). Objectives include improvement of methods for describing flame extinction in fires and development of procedures for extracting overall rate information, useable in complex fire environments, from laboratory measurements of diffusion-flame extinction in well-controlled environments.

In the work performed by asymptotic methods little attention has been paid to influences of radiant energy loss from the gas on diffusion-flame extinction. One reason for this has been the emphasis on application to small-scale laboratory experiments; estimates and measurements have indicated that radiant losses from the gas are relatively insignificant in such experiments (Seshadri and Williams, 1978). At larger scales, emission of radiation from luminous flame zones, typically as a consequence of the presence of large numbers of particles of hot soot, may exert measurable influences on critical conditions for extinction. Kanury (1975) has suggested that there may be situations in which radiation is the dominant mechanism of energy loss from the reacting gas and that in such cases extinction conditions may best be described on the basis of parameters that differ from those obtained from the existing asymptotic analyses. The present study is intended to provide a step toward the identification of parameters appropriate for describing extinction under conditions such that radiant loss is not negligible.

The problem addressed is that of describing the structure and extinction of the counterflow diffusion flame in the presence of radiant heat loss. It thereby constitutes an extension of the previously published asymptotic analysis of the counterflow diffusion flame for large activation energies (Liñán, 1974). However, to facilitate application to problems having the fuel present in a condensed phase, the analysis is developed within the context of stagnation-point boundary-layer flow. The formulation thus parallels that published previously for this condensed-fuel problem. The result will be a modification of the procedure of Krishnamurthy *et al.* (1976) for extracting overall rate parameters from measurements of diffusion-flame extinction, now accounting for radiant loss from the gas. Influences of radiant loss from the condensed phase will be considered elsewhere (Sohrab and Williams, 1981).

2 FORMULATION

For convenience, the notation is the same as that employed by Krishnamurthy *et al.* (1976) and therefore is not redefined fully here. The Lewis numbers for fuel F and for oxidizer O will be taken to be unity, and the Prandtl number P will be assumed to be constant, as will the product of the density ρ with the coefficient of viscosity μ. Subscripts e and w identify conditions in the oxidizing stream and in the gas at the surface of the condensed fuel, respectively. Primes denote derivatives with respect to the transformed variable η that measures distance normal to the planar surface of the condensed fuel and that is zero at the fuel surface. The conservation equations then become the same as those given by Krishnamurthy *et al.* (1976), except for the occurrence of a radiant loss term in the equation for energy conservation. Only the equations for species and energy conservation will be written here. These are

$$Y_i'' + fY_i' = D_I Y_O Y_F \exp(-T_a/T), \quad i = O, F \qquad (1)$$

and

$$T'' + fT' = -D_I Y_O Y_F \exp(-T_a/T) + F_l(T). \qquad (2)$$

Here f denotes the nondimensional stream function, Y_F the mass fraction of fuel and Y_O that of the oxidizer divided by the stoichiometric mass ratio of oxidizer to fuel. The temperature is TQ/c_p, where Q is the heat released in the gas-phase reaction per unit mass of gaseous fuel consumed, and c_p is the specific heat at constant pressure for the gas, assumed constant herein. The overall activation energy is $R^\circ T_a Q/c_p$, where R° denotes the universal gas constant, and D_I represents Damköhler's first similarity group, the ratio of a flow time (reciprocal of external velocity gradient $k\,du_e/dr$) to a chemical time, defined exactly as in the previous publication (Krishnamurthy *et al.*, 1976). The function $F_l(T)$ is the radiant energy loss per unit volume per second, divided by $\rho Qk\,du_e/dr$.

The boundary conditions for Eqs. (1) and (2) are identical to those employed previously, *viz.*, $T = T_e$, $Y_O = Y_{Oe} \equiv \alpha$ and $Y_F = 0$ at $\eta = \infty$, and $T = T_w$, $Y_O' = -fY_O$, $Y_F' = f(1 - Y_F)$ and $T' = -qf$ at $\eta = 0$, where $q = L/Q$, L being the heat required to gasify a unit mass of fuel (including heat conducted into the interior of the condensed phase and radiant energy loss from the surface of the condensed phase, if they occur). Therefore the only difference from the previous formulation resides in the loss term $F_l(T)$.

For an optically thin gas, the radiant energy loss per unit volume per second is $4\sigma(TQ/c_p)^4/l_P$, where σ denotes the Stefan–Boltzmann constant and l_P is the Planck-mean absorption length (Penner and Olfe, 1968). Therefore the formula

$$F_l(T) = 4\sigma T^4 (Q/c_p)^4/(l_P \rho Qk\,du_e/dr) \qquad (3)$$

may be employed for the loss term. This formula does not take into account radiation absorbed by the gas from the surroundings. Consequently $F_l(T)$ does not vanish as η approaches infinity, unless l_P is made to approach infinity artificially as η goes to infinity. The continuing heat loss that occurs if l_P remains finite causes the problem to be ill-posed, in a manner analogous to that of the problem of the premixed laminar flame with Arrhenius kinetics. Physically, an additional term, negative in sign, belongs on the right-hand side of Eq. (3) to represent absorption of radiation from the surroundings and to cause $F_l(T_e)$ to vanish. This term is not written because it is insignificant within the context of the asymptotic analysis to be performed herein.

Equation (3) shows that if l_P is independent of T then at constant pressure $F_l(T)$ is proportional to T^5. Typically l_P decreases with increasing T in the temperature range of interest here (as a consequence of increases in both equilibrium radiation emitted and nonequilibrium concentrations of emitters), and therefore $F_l(T)$ increases with T more rapidly than T^5. If soot radiation is the dominant contributor to $F_l(T)$, then the loss may be negligible on the oxidizer side of the reaction region, since relatively little soot is found there. A highly simplified model of soot radiation, treating soot particles as independent spherical emitters of radius r_s with emissivities of unity and surface temperatures $T_s Q/c_p$, results in $l_P = (T/T_s)^4/(\pi r_s^2 n_s)$, where n_s is the local number density of soot particles; the decrease of l_P with increasing T on the fuel side of the reaction region then may arise largely from the increase in $n_s r_s^2$, which depends on the chemical kinetics of soot production and particle growth.

The transformation to a new independent variable x, that serves to put the equations in the form studied originally by Liñán (1974) is the same as that given by Krishnamurthy *et al.* (1976), *viz.*,

$$x = -f_w(1+B)^{-1} \int_\eta^\infty \exp(-h)\,d\eta, \qquad (4)$$

where B is a transfer number and

$$h = \int_0^\eta f d\eta.$$

In the transformed variables, Eqs. (1) and (2) become

$$\frac{d^2 Y_i}{dx^2} = \left(\frac{1+B}{f_w}\right)^2 \exp(2h) D_I Y_O Y_F \times \exp(-T_a/T), \qquad i = O, F \tag{5}$$

and

$$\frac{d^2 T}{dx^2} = -\left(\frac{1+B}{f_w}\right)^2 \exp(2h)[D_I Y_O Y_F \times \exp(-T_a/T) - F_l(T)]. \tag{6}$$

In the energetics, it is necessary to account for the radiant heat loss from the gas. This can be done in terms of the function

$$G_l = -\int_0^x \int_0^\eta (d\eta/dx) F_l(T) d\eta\, dx, \tag{7}$$

defined to have boundary values $G_{le}=0$ and $G_{lw}'=0$. The modified definition of the transfer number, taking into account the radiant loss, is

$$B = (a + T_e - T_w - G_{lw} - Y_{Ow})/(q + Y_{Ow}), \tag{8}$$

which is assumed to be positive. Here a, defined in the paragraph preceding that containing Eq. (3), is the ratio of the oxidizer fraction in the approach stream to the stoichiometric oxidizer–fuel ratio. From Eqs. (5), (6) and (7) it is readily shown that

$$T + Y_i + G_l = a_i + b_i x, \qquad i = F, O, \tag{9}$$

where a_i and b_i are constants.

Boundary conditions may be used to evaluate a_i and b_i in a manner paralleling that given by Krishnamurthy *et al.* (1976). It is found that, with

$$\beta = q + T_e - T_w, \tag{10}$$

the results

$$Y_O = a + T_e - T - G_l - (a + \beta - G_{lw})x \tag{11}$$

and

$$Y_F = T_e - T - G_l + (1 - \beta + G_{lw})x \tag{12}$$

are obtained and that as before,

$$x_w = B/(1+B). \tag{13}$$

Thus, x varies from zero in the oxidizer stream to a value between zero and unity at the surface of the condensed fuel.

This formulation achieves a correspondence with the problem of the counterflow diffusion flame for purely gaseous reactants (Liñán, 1974), analogous to the correspondence discussed earlier (Krishnamurthy *et al.*, 1976). The corresponding gaseous problem has gaseous fuel at the boundary $x=1$. If the domain of definition is extended into the range $x_w < x < 1$ by introduction of the requirements that dT/dx and G_l remain constant in this range, then $Y_O=0$, $Y_F=1$ and $T=T_w-q=T_e-\beta$ are obtained as the conditions to be applied at the hypothetical boundary $x=1$, as found previously. Thus, the problem of accounting for radiant heat loss from the gas may equally well be studied within the context of a purely gaseous problem, defined by Eqs. (5) and (6) with the boundary conditions that $Y_O=a$, $Y_F=0$ and $T=T_e$ at $x=0$, and that $Y_O=0$, $Y_F=1$ and $T=T_e-\beta$ at $x=1$.

3 ANALYSIS OF RADIANT HEAT LOSS

Of the various combustion regimes that may occur, attention will be focused on Liñán's (1974) diffusion-flame regime, which is the most important in studies of extinction. From Eqs. (11) and (12) it is seen that

$$Y_F - Y_O = (1+a)x - a, \tag{14}$$

whence setting $Y_F = Y_O = 0$ shows that the location of the flame sheet in the Burke-Schumann limit of infinite D_I is

$$x_f = a/(1+a). \tag{15}$$

In this limit, from Eq. (14) it is seen that $Y_O = a - (1+a)x$ for $x < x_f$ and $Y_F = (1+a)x - a$ for $x > x_f$, so that the jump in the gradient dY_F/dx or dY_O/dx as x increases through the value x_f is $(1+a)$. This shows that the right-hand side of Eq. (5) becomes $(1+a)\delta(x-x_f)$ in this limit, thereby allowing Eq. (6) to be written as

$$\frac{d^2 T}{dx^2} = \left(\frac{1+B}{f_w}\right)^2 \exp(2h) F_l(T) - (1+a)\delta(x - x_f) \tag{16}$$

for $D_I \to \infty$. The first step in the analysis is to consider methods for solving Eq. (16) subject to the boundary conditions that $T=T_e$ at $x=0$ and $T=T_e-\beta$ at $x=1$.

It has been indicated in the previous section that $F_l(T)$ is a strongly increasing function of T. Therefore the dominant contribution of the loss term in Eq. (16) comes from the vicinity of the point of maximum temperature, which physically should be located near $x=x_f$. It will be assumed that if $F_l(T)=0$ then the parameters are such that the maximum temperature occurs at $x=x_f$ instead of at one of the boundaries. Then it follows that for $F_l(T)\neq 0$, the maximum temperature still occurs at $x=x_f$ in the limit $D_I\to\infty$. Let T_f denote the value of T at $x=x_f$. When $F_l(T)\neq 0$ this value will lie below the adiabatic value T_{fa}, obtained when $F_l(T)=0$. Instead of seeking the value of T_f that corresponds to a given magnitude of the radiant loss, it is simpler to specify T_f and to calculate the value of a loss parameter that produces the specified flame temperature.

The sensitivity of $F_l(T)$ to T may be described by $\gamma \equiv d\ln F_l(T)/d\ln T$. If γ is constant, then $F_l(T)$ is proportional to T^γ. Allowance will be made for the possibility of having different functions, γ_F and γ_O, in the fuel and oxidizer regions, $x>x_f$ and $x<x_f$, respectively. It has been indicated that typically γ_i $(i=O, F)$ is large compared with unity. However, within each region γ_i will be assumed to vary slowly with T, *i.e.*, $g_i\equiv d\ln\gamma_i/d\ln T$ will be taken to be of order unity. If γ_{if} denotes the value of γ_i at $T=T_f$ (*i.e.*, at $x=x_f$), then

$$\phi=\gamma_{if}(T_f-T)/T_f \tag{17}$$

is a useful dependent variable. An expansion of $F_l(T)$ in γ_{if}^{-1} for ϕ of order unity may be written in an exponential form that facilitates analysis as

$$F_l(T) = F_{lf}\exp(-\phi)[1+\tfrac{1}{2}(g_{if}-1)\phi^2\gamma_{if}^{-1}+\ldots], \tag{18}$$

where F_{lf} and g_{if} are the values of $F_l(T)$ and of g_i at $T=T_f$. Use of Eq. (18) in Eq. (16) can provide an asymptotic expansion of the solution T for large values of γ_{if}.

If γ_{if} is large, then for $(T_f-T)/T_f$ of order unity $F_l(T)$ is exponentially small and therefore negligible in Eq. (16). Solutions in these outer zones that satisfy the boundary conditions and an anticipated matching condition that $T=T_f$ at $x=x_f$ are

$$T=\begin{cases} T_e+(T_f-T_e)x/x_f & \text{for } x<x_f,\\ T_e-\beta+(T_f-T_e+\beta)(1-x)/(1-x_f) & \text{for } x>x_f.\end{cases} \tag{19}$$

The jump in the slope dT/dx as x increases through x_f, obtained from Eq. (19), is attributable to the sum of the contributions from the delta function in Eq. (16) and from the loss term. A measure of the total energy lost by radiation from the gas therefore is $(1+a)$ plus the jump in the slope. This loss is easily found through use of Eqs. (15) and (19) to be

$$Q_l=(1+a)(1-\beta)(1-\mu_O), \tag{20}$$

where

$$\begin{aligned}\mu_O&=(T_f-T_e)(1+a)/[a(1-\beta)]\\ &=(T_f-T_e)/(T_{fa}-T_e).\end{aligned} \tag{21}$$

The flame-temperature parameter μ_O lies between zero and unity, attaining the latter value when there is no heat loss from the gas. Since $G_{lw}'=0$, it may be seen by virtue of Eq. (7) that Q_l is the value of dG_l/dx at $x=0$. In the first approximation, dG_l/dx is Q_l for $x<x_f$ and 0 for $x>x_f$, whence integration gives, by use of $G_{le}=0$ and of Eq. (15), the formula $G_{lw}=Q_la/(1+a)$. For use only in the outer zones, Eq. (16) may be written as

$$d^2T/dx^2=[Q_l-(1+a)]\delta(x-x_f). \tag{22}$$

While Eq. (20) determines the total radiant loss in terms of T_f, it does not provide either of these parameters in terms of the magnitude F_{lf} of the rate of emission of radiation from the vicinity of the flame. Analyses of the thin zones of radiation emission on each side of the flame sheet are needed for this purpose. In these zones, the stretched variable $\xi=\gamma_{if}(x-x_f)/T_f$ is of order unity. Introduction of ξ and ϕ into Eq. (16) as new variables result, through use of Eqs. (17) and (18), in

$$d^2\phi/d\xi^2=(1+a)\delta(\xi)-l_i\exp(-\phi) \tag{23}$$

at the lowest order in γ_{if}^{-1}, where

$$\begin{aligned}l_i&=[(1+B)f_w]^2\\ &\quad\times[\exp(2h_f)](T_f/\gamma_{if})F_{lf}, \qquad i=O,F.\end{aligned} \tag{24}$$

Here l_i is the ratio of a Bouguer number to a Boltzmann number (Penner and Olfe, 1968) and measures the ratio of the rate of radiant energy emission to the rate of convection of enthalpy, *i.e.*, it may be viewed as the ratio of a characteristic flow time to a characteristic time for emission of radiation. Matching conditions for Eq. (23), obtained from Eq. (19), are that $d\phi/d\xi \to (T_f - T_e + \beta)(1+\alpha)$ as $\xi \to \infty$ and that $d\phi/d\xi \to -(T_f - T_e)(1+\alpha)/\alpha$ as $\xi \to -\infty$.

A first integral of Eq. (23), away from $\xi = 0$, *is* $(d\phi/d\xi)^2 = 2l_i \exp(-\phi) + \text{constant}$. The constant may be evaluated from the matching conditions for $\xi \to \pm\infty$, giving after evaluation of the result at $\xi = 0$,

$$\left.\begin{aligned} \left.\frac{d\phi}{d\xi}\right|_{\xi=0+} &= [2l_F + (T_f - T_e + \beta)^2(1+\alpha)^2]^{1/2}, \\ \left.\frac{d\phi}{d\xi}\right|_{\xi=0-} &= -[2l_O + (T_f - T_e)^2(1+\alpha)^2/\alpha^2]^{1/2}. \end{aligned}\right\} \quad (25)$$

Use of these results in the jump condition at $\xi = 0$ implied by the delta function in Eq. (23) yields

$$\begin{aligned} 1+\alpha &= [2l_O + \mu_O^2(1-\beta)^2]^{1/2} \\ &\quad + [2l_F + \mu_F^2(\alpha+\beta)^2]^{1/2}, \end{aligned} \quad (26)$$

where

$$\begin{aligned} \mu_F &= (T_f - T_e + \beta)(1+\alpha)/(\alpha+\beta) \\ &= (T_f - T_e + \beta)/(T_{fa} - T_e + \beta). \end{aligned} \quad (27)$$

The parameter μ_F is an alternative to μ_O for measuring the departure of T_f from T_{fa}; it too lies between zero and unity.

Equation (26) is the result needed for relating T_f to the rate of radiant loss. It may be solved explicitly for l_i if $l_O = 0$, $l_F = 0$ or $l_O = l_F$. If l_O and l_F are sufficiently small and are known, then T_f may be obtained explicitly from Eq. (26) by expansion; in the first approximation it is found that

$$T_f = T_{fa} - \frac{\alpha}{(1+\alpha)^2}\left(\frac{l_O}{1-\beta} + \frac{l_F}{\alpha+\beta}\right). \quad (28)$$

In practice, often l_O is negligible, and it may then be shown from Eq. (26) that

$$T_f = T_{fa} - \frac{\alpha(\alpha+\beta)}{(1-\alpha^2)} \times \left\{\left[1 + \frac{2l_F(1-\alpha)}{(1+\alpha)(\alpha+\beta)^2}\right]^{1/2} - 1\right\}, \quad (29)$$

which is preferable to Eq. (28) for large values of l_F. Strictly speaking, according to Eq. (24), in Eq. (29) l_F depends on T_f. Since Eqs. (17) and (18) imply that, approximately,

$$F_{lfa} \equiv F_l(T_{fa}) = F_{lf} \exp[\gamma_{Ffa}(1 - T_f/T_{fa})],$$

in Eq. (29) the relationship

$$l_F = l_{Fa} \exp[-\gamma_{Ffa}(1 - T_f/T_{fa})]$$

may be employed, where l_{Fa} is given by Eq. (24) with T_f replaced by T_{fa}, γ_{Ff} replaced by γ_{Ffa} and F_{lf} replaced by F_{lfa}; here l_{Fa}, the loss parameter that would apply if conditions were adiabatic, is known prior to determination of T_f, and Eq. (29) becomes a transcendental equation to be solved for T_f. If this equation is to be solved iteratively, then it is equally convenient to employ Eq. (24) directly in Eq. (29) instead of introducing these further expansions.

It is possible to generate higher-order terms in the expansion of the solution to Eq. (16) in the small parameter γ_{lf}^{-1}, but the analysis will not be pursued here, since the dominant contribution of the radiant loss is that which has been obtained. It may be noted from Eqs. (19) and (25) that within the zone of radiant loss, temperature gradients steepen as the reaction zone is approached. This steepening enhances the tendency for extinction of the reaction to occur. The character of the temperature profiles is illustrated schematically in Figure 1.

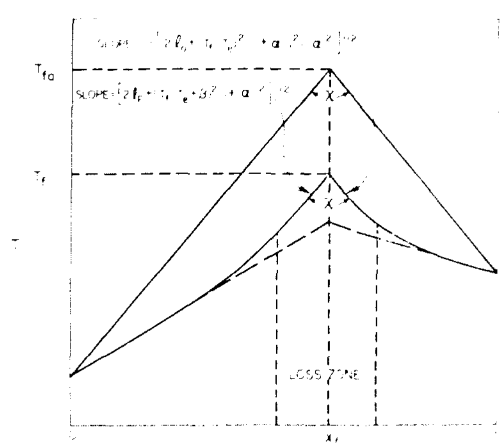

FIGURE 1 Illustration of the character of the results when the reaction zone has a negligible thickness.

4 ANALYSIS OF EXTINCTION

Calculation of extinction necessitates investigation of the structure of the reaction zone. It is known (Liñán, 1974) that the thickness of this zone, in the x coordinate, is of order T_f^2/T_a. Therefore, in view of the definition of ξ, if $T_f^2/T_a \ll T_f/\gamma_{lf}$, then the reaction zone is much narrower than the zone of radiant loss and is embedded within it. If T_f^2/T_a and T_f/γ_{lf} are of the same order of magnitude, then the zones of finite-rate chemistry and of radiant loss coincide and have the same thickness. The remaining possibility, $T_f/\gamma_{lf} \ll T_f^2/T_a$, seldom occurs and is not considered here. The extinction problem will be formulated for T_f^2/T_a of the same order as T_f/γ_{lf} and later will be specialized to $T_f^2/T_a \ll T_f/\gamma_{lf}$, which probably defines the regime of greatest practical interest.

An asymptotic expansion of the solution to Eqs. (5) and (6) is sought for small values of T_f^2/T_a, with $a \equiv \gamma_{lf} T_f/T_a$ of order unity. Here a is the ratio of the temperature sensitivity of the radiant-loss rate to that of the heat-release rate. There are outer zones in which reaction and radiation are negligible and in which the solution for the temperature is given by Eq. (19). These are separated by the inner zone of reaction and radiation, for the analysis of which convenient variables are $y=(x-x_f)(1+\alpha)T_a/T_f^2$, $\theta=(T_f-T)T_a/T_f^2$ and $\psi=(Y_O+T-T_f)T_a/T_f^2$. With these variables, $Y_O=(\psi+\theta)T_f^2/T_a$ and $Y_F=(y+\psi+\theta)T_f^2/T_a$, the latter identity being obtained from Eq. (14) by use of Eq. (15). Substitution of these results and of Eqs. (17) and (18) into Eqs. (5) and (6) produces, to the lowest order in T_f^2/T_a, the pair of differential equations

$$d^2\psi/dy^2 = \lambda \exp(-a\theta) \tag{30}$$

and

$$d^2\theta/dy^2 = \delta(y+\psi+\theta)(\psi+\theta) \times \exp(-\theta) - \lambda \exp(-a\theta), \tag{31}$$

where

$$\lambda = [(1+B)/f_w]^2[\exp(2h_f)] \times (T_f^2/T_a)F_{lf}/(1+\alpha)^2 \tag{32}$$

and

$$\delta = [(1+B)/f_w]^2[\exp(2h_f)](T_f^2/T_a)^3 \times D_I[\exp(-T_a/T_f)]/(1+\alpha)^2. \tag{33}$$

Boundary conditions for Eqs. (30) and (31), obtained from matching, are

$$\left.\begin{aligned} &d\theta/dy \to -d\psi/dy \to T_f - T_e + \beta \\ &\qquad\qquad \text{as} \quad y\to\infty, \\ &-d\theta/dy \to 1 + d\psi/dy \to (T_f-T_e)/\alpha \\ &\qquad\qquad \text{as} \quad y\to-\infty. \end{aligned}\right\} \tag{34}$$

The parameters $\lambda = l_l a/(1+\alpha)^2$ and δ represent a radiant loss-rate parameter and a reduced Damköhler number, respectively. It may be anticipated that, with a fixed, Eqs. (30) and (31) will possess solutions, subject to given boundary gradients in Eq. (34), only if λ and δ lie along a particular curve in the (δ, λ) plane. These curves will extend from a minimum value of δ, say δ_E, which corresponds to extinction, to $\delta=\infty$. For $T_f=T_{fa}$, it is found that the curve is defined by $\lambda=0$, with δ_E given by the analysis of extinction published by Liñán (1974). As T_f decreases from T_{fa} with a, α, β, and T_e fixed, it is expected that along the curve λ will be found to be positive and that δ_E may exceed its value for $T_f=T_{fa}$. The boundary $\delta_E(\lambda)$ of the region in the (δ, λ) plane, generated in this manner, represents an extinction boundary that defines extinction conditions as functions of the rate of radiant heat loss.

In general, solution of the problem defined by Eqs. (30), (31) and (34) requires numerical integration. If λ is small, of order T_f^2/T_a, and a is of order unity, then a perturbation approach may be developed, beginning with the previous solutions (Liñán, 1974) for $\lambda=0$, with $T_f=T_{fa}$ in the first approximation. A limit of greater practical importance appears to be that for small a. In this case, the zone of radiant loss is much thicker than the reaction zone, and the analysis of the preceding section applies to the former. Since it is seen from Eq. (26) that l_l cannot be large if T_f is to exceed the boundary temperatures, it follows that λ must be small when a is small. Therefore, in the reaction zone λ is to be omitted from Eqs. (30) and (31) in the first approximation.

When the reaction zone is narrow compared with the heat-loss zones, Eq. (25) gives the external boundary gradients that through matching provide boundary conditions on θ for solving Eq. (31) in the reaction zone. If

$$m_F = [2l_F/(1+\alpha)^2 + (T_f - T_e + \beta)^2]^{1/2}, \tag{35}$$

then from Eqs. (25) and (26) it is found that these boundary conditions are

$$d\theta/dy \to m_F \quad \text{as} \quad y \to \infty,$$
$$d\theta/dy \to m_F - 1 \quad \text{as} \quad y \to -\infty. \qquad (36)$$

With $\lambda=0$ in the first approximation, the integral of Eq. (30), needed in solving Eq. (31), is $\psi = \psi_f + by$, where ψ_f and b are constants. Here the constant b may be evaluated from matching conditions for $y \to \pm\infty$. In the diffusion-flame regime, these are obtained from $dY_F/dx \to (1+a)^{-1}$ as $x \to x_f^+$, $dY_F/dx \to 0$ as $x \to x_f^-$, $dY_O/dx \to 0$ as $x \to x_f^+$ and $dY_O/dx \to -(1+a)^{-1}$ as $x \to x_f^-$, all of which consistently yield $d\psi/dy \to -m_F$ as $y \to \pm\infty$ when use is made of Eq. (36) and of the definitions of ψ and y. Hence $b = -m_F$. Since Eqs. (11), (15) and (21) and the definition of ψ show that

$$\psi_f = (T_a/T_f)^2[G_{lw}a/(1+a) - G_{lf} + T_{fa} - T_f],$$

a modification in ψ_f of order unity corresponds to a modification in T_f of order T_f^2/T_a. Since the relationships of the preceding section are valid to order unity, not to order T_f^2/T_a, at the present order of approximation there exists freedom in selecting T_f to assure that the convenient value $\psi_f=0$ is obtained. With this selection, $\psi = -m_F y$, and Eq. (31) becomes

$$d^2\theta/dy^2 = \delta(\theta - m_F y)[\theta + (1-m_F)y]\exp(-\theta). \qquad (37)$$

Introduction of the new variables $z = (4\delta)^{1/3}y/2$ and $Z = (4\delta)^{1/3}[\theta + (1-2m_F)y/2]$ converts Eqs. (36) and (37) to

$$\left.\begin{aligned} &d^2Z/dz^2 = (Z+z)(Z-z)\exp\{-(4\delta)^{-1/3} \\ &\qquad\qquad \times [Z + (2m_F - 1)z]\}, \\ &dZ/dz \to 1 \quad \text{as} \quad z \to \infty, \\ &dZ/dz \to -1 \quad \text{as} \quad z \to -\infty. \end{aligned}\right\} \qquad (38)$$

The problem defined by Eq. (38) has been encountered and solved previously (Liñán, 1974); it was found that at extinction

$$\delta_E = (ec/2)(1 - 2c + 1.04c^2 + 0.44c^3) \qquad (39)$$

in an excellent approximation, where

$$c = \begin{cases} m_F & \text{if} \quad m_F \leqslant 1/2 \\ 1 - m_F & \text{if} \quad m_F \geqslant 1/2. \end{cases} \qquad (40)$$

Equation (39) provides the extinction formula for the case of greatest practical interest.

It is of interest to compare Eq. (39) with the results obtained by Krishnamurthy *et al.* (1976) for the diffusion-flame regime, as given by Eqs. (20) and (21) of this previous publication. In view of Eq. (33), it is seen that the results are identical, provided only that the definition of c is modified in accordance with Eqs. (35) and (40). Therefore the earlier results have a direct bearing on the problem of extinction with radiant loss from the gas.

5 EXTINCTION RESULTS

The results obtained here may be cast in a form that enables the parametric results previously published by Krishnamurthy *et al.* (1976) to be employed for calculating overall rate parameters from systematic measurements of extinction, in the presence of radiant loss from the gas. It is necessary first to consider how to obtain flow-field parameters, such as f_w and h_f, from the known parameters of the problem, a, T_e, T_w and q, in the flame-sheet limit. This can be done for large γ_{lf} by making use of the fact that the thickness of the radiant-loss zone then also is narrow. It is seen from Eq. (22) that the effect of the radiant loss then is to reduce the total heat release by the factor $1 - Q_l/(1+a)$. Therefore the flow field is described by the flame-sheet problem with the heat release per unit mass of gaseous fuel consumed given by $Q[1 - Q_l/(1+a)]$. Hence a suitable redefinition of parameters renders the published results applicable.

Since $G_{lw} = Q_l a/(1+a)$, which was indicated after Eq. (21), the transfer number defined in Eq. (8) becomes, in the first approximation,

$$B = [a - aQ_l/(1+a) + T_e - T_w)/q. \qquad (41)$$

With this modified expression for B, the formulas for the remaining parameters, A, C and D, are the same as given by Krishnamurthy *et al.* (1976), *viz.*, $A = (a + T_e - T_w)/(2T_e)$, $C = (B-a)/[(1+a)B]$ and $D = T_w/(2T_e)$. Since A, C and D involve ratios of quantities that have been nondimensionalized by Q, the effectively modified heat release does not alter their formulas. Therefore the only revision needed in the flame-sheet calculation is that given by Eq. (41). Alternative procedures, involving redefinitions of T_e, T_w and q, would be more complex.

If A, B, C and D are known, then a factor F may be obtained from graphs shown by Krishnamurthy *et al.* (1976). This factor contains the flow-field parameters relevant to extinction. In terms of F, it may be shown from the definition of D_I and from Eqs. (33) and (39) that the critical extinction condition may be written dimensionally as

$$\frac{M_F k F C_l T_a{}^3}{\rho_e T_e T_f{}^2}\left(\frac{du_e}{dr}\right)_E = B_{Of}\exp\left(-\frac{T_a}{T_f}\right), \quad (42)$$

where M_F represents the molecular weight of the fuel, B_{Of} denotes the preexponential rate factor for molar oxidizer consumption in a second-order reaction, evaluated at the flame temperature, and $k(du_e/dr)$ is the external normal velocity gradient in the oxidizer stream. Equation (42) is identical to the extinction formula obtained by Krishnamurthy *et al.* (1976) for the diffusion-flame regime, except for the radiant-loss correction factor, C_l, which is given by

$$C_l = \frac{c(1-2c+1.04c^2+0.44c^3)}{c_a(1-2c_a+1.04c_a{}^2+0.44c_a{}^3)}, \quad (43)$$

where according to Eqs. (40) and (35)

$$c = \begin{cases} [2l_F/(1+\alpha)^2+(T_f-T_e+\beta)^2]^{1/2} \quad \text{or} \\ 1-[2l_F/(1+\alpha)^2+(T_f-T_e+\beta)^2]^{1/2} \end{cases} \quad (44)$$

whichever is smaller, and where

$$c_a = \begin{cases} 2A(1-C)/[2(A+D)-1] \\ \qquad \text{if} \quad 2C>1-(2D-1)/2A, \\ [2(AC+D)-1]/[2(A+D)-1] \\ \qquad \text{if} \quad 2C<1-(2D-1)/2A. \end{cases} \quad (45)$$

The loss parameter l_F in Eq. (44) is given by

$$l_i = [ec_a(1+\alpha)^2 F_{lf}]/[2FT_f{}^2\gamma_{if}], \quad i=O, F, \quad (46)$$

where use has been made of Eq. (24) and of the definition of F. It is possible, as in the paper of Krishnamurthy *et al.* (1976), to develop an iterative procedure for improving the accuracy of Eq. (42) if extinction occurs in the premixed-flame regime.

An explicit prescription for calculating rate parameters for extinction, given α, T_e, T_w, q, F_{lf} and γ_{if}, may be stated on the basis of these results. First guess a value of the flame temperature in the presence of radiant loss. Then use Eqs. (20) and (21) to find Q_l and Eq. (41) to find B. Next calculate A, C and D from the formulas given after Eq. (41). Having thus obtained values for the parameters A, B, C and D, employ the published graphs (Krishnamurthy *et al.*, 1976) to obtain the factor F. Then calculate l_O and l_F from Eq. (46), and ascertain whether Eq. (26) is satisfied, using Eqs. (21) and (27) for μ_O and μ_F. Iteratively adjust T_f until this sequence of computations results in Eq. (26) being satisfied. Then calculate C_l by use of Eqs. (43), (44) and (45). Finally, use the procedure described previously by Krishnamurthy *et al.* (1976) to plot an Arrhenius graph from Eq. (42), replacing F by FC_l in that procedure. If radiant losses from the gas are small, then $T_f = T_{fa}$ is a good approximation, and iteration can be avoided. This situation prevails in labroatory experiments (Sohrab and Williams, 1981) and it is found moreover that in these tests the corrections to calculated rate parameters, produced by inclusion of C_l, are negligible.

6 SAMPLE APPLICATION

As an illustrative example, the theory is applied to the diffusion flames above the planar surface of burning poly (methyl methacrylate) (PMMA). The extinction data, involving measurement of U and Y_{O2} at extinction, were reported elsewhere (Sohrab and Williams, 1981). The effects of fuel surface radiation on the parameters q and T_{fa} have been taken into account by procedures described by Sohrab and Williams (1981). Here we wish to examine the influence of gaseous radiation on the predicted kinetic parameters by considering two representative extinction data points. In Table I values of the known flow, fuel and flame parameters U, α, $\hat{T}_e$, $\hat{T}_w$, q and $\hat{T}_{fa}$ are given for the data that are considered. In this table the hats identify dimensional temperatures with units °K; the units of U are cm/s.

In laboratory diffusion flames, radiation occurs primarily from the emission by hot soot particles with a continuous spectrum in the visible and infrared regions. Therefore only the fuel side of the flame needs to be considered; the radiation parameter on the oxidizer side, l_0, is neglected. Typical values of F_{lf} calculated for laboratory flames are exceedingly small, $<10^{-6}$. Unreasonably large losses will be introduced here for purposes of illustration. Therefore the present calculations do not pertain to the real situation. Effects associated with absorption of radiation by the emitting particles or by the fuel surface also are neglected in the present example.

TABLE I

Flow, fuel and flame parameters

α	U	$\hat{T}_e$	$\hat{T}_w$	q	$\hat{T}_{fa}$	l_F	$\hat{T}_f$	C_l	F_{lf}
						0.001	1899	1.19	1.65×10^{-3}
0.09115	44	300	665	0.0954	1908	0.01	1827	1.33	1.46×10^{-2}
						0.1	1405	1.38	6.54×10^{-2}
						0.001	1984	1.18	1.75×10^{-3}
0.09635	75	300	695	0.08875	1994	0.01	1908	1.29	1.51×10^{-2}
						0.1	1463	1.37	7.03×10^{-2}

Since extinction of PMMA flames occurs in the diffusion-flame regime defined by Liñán (1974), the theory can be directly applied; moreover, the iteration described above can be avoided because $l_0=0$. Three different values of l_F are selected to illustrate influences of increasing the radiant loss. These values are used in Eq. (29) to calculate T_f. Then μ_0 is found from Eq. (21) and Q_l from Eq. (20). The transfer number B is next calculated from Eq. (41). The parameters A, C, D, and then F are evaluated in the manner described after Eq. (41). Computation of c and c_a from Eqs. (44) and (45) enables C_l to be obtained from Eq. (43). The Arrhenius graph based on Eq. (42) may then be constructed directly.

Values of the nondimensional radiant loss flux F_{lf} that correspond to the selected values of l_F may be calculated from Eq. (46). For this purpose, the value $\gamma_{Ff}=5$ was selected. Values of F_{lf} obtained in this manner are listed in Table I. Also listed are values calculated for C_l and for the flame temperature. The values of F_{lf} are of the same order of magnitude as those of l_F and exceed realistically estimated values by more than three orders of magnitude. Flames larger than those employed in the laboratory experiments would have larger values of F_{lf}. The flame temperatures in Table I decrease with increasing values of the assumed loss rate as anticipated.

Figure 2 shows the Arrhenius plots that correspond to l_F values of 0, 0.001, 0.01 and 0.1. The calculated kinetic parameters are also given in Figure 2. The activation energies obtained from the data tend to decrease as the loss is increased, and the magnitude of the reaction rate at a fixed flame temperature increases as expected.

7 CONCLUSIONS

This study helps to clarify the nondimensional parameters that are appropriate for describing extinction in the presence of radiant loss from the gas. From Eq. (39), it is evident that the extinction conditions obtained here are best expressed in terms of a Damköhler number of the same type that has been used in the absence of radiant loss. Although Eqs. (35) and (40) show that the factor c depends on the radiant loss, this factor is bounded and cannot be viewed as the primary parameter on the basis of which extinction is to be described. An increase in the loss rate l_F is offset by a decrease in the flame temperature T_f, so as to maintain the balance in Eq. (26). The quantitatively largest effect of radiation occurs through the reduction of T_f on the right-hand side of Eq. (42). Thus, while radiant loss certainly contributes to reduced reaction rates through reduced temperatures, the critical condition for extinction remains defined by a balance between the rate of heat generation in the reaction zone and the rate of conductive loss of heat from that thin zone.

To depart from this fundamental relationship, it is necessary to approach conditions under which each term on the right-hand side of Eq. (31) is large compared with the left-hand side. Under these conditions, there would be an approximate balance of heat release and radiant loss in the reaction zone, and a new parameter, δ/λ, would become relevant. From Eqs. (32) and (33) it is seen that the flow time cancels when this ratio is formed; δ/λ represents the ratio of a characteristic time for emission of radiation to a characteristic time for chemical heat release, a kind of Damköhler

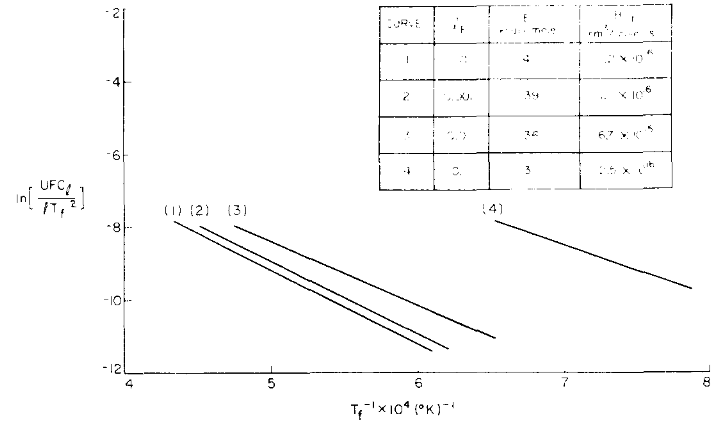

FIGURE 2 Arrhenius plots for various flame radiant losses.

number based on radiant loss. It is known that ignition and extinction phenomena in homogeneous systems can be described by algebraic equations expressing balances similar to the requirement that the right-hand side of Eq. (31) vanishes. However, it is unclear whether the requisite criticality behavior is consistent with Eqs. (30), (31) and (34). Since a θ dependence of the radiation rate weaker than that of the heat-release rate favors the presence of criticality, small values of a would seem to be wanted, but under this condition the analysis that has been presented applies, with conduction overbalancing radiation in the reaction zone.

Description of extinction on the basis of a radiation Damköhler number, δ/λ, if valid at all would seem likely to apply when strong radiant interactions cause gas temperatures to be nearly uniform everywhere, thereby broadening reaction zones to such an extent that Eqs. (30), (31) and (34) are inapplicable. Moreover, in the non-premixed configuration, reaction rates would have to be slow enough to allow extensive molecular mixing to occur prior to reaction. The theory for such a nearly homogeneous system would involve a parameter similar to δ/λ which may achieve a critical value at extinction. Applications might be found in certain industrial furnaces, but relevance to common fire problems seems questionable.

This work has been restricted to optically thin media. Therefore all of the radiant energy emitted is lost from the system. In applications to combustion in planar stagnation-point boundary layers adjacent to condensed fuels, the radiation emitted in the direction of the fuel may be absorbed by the fuel surface. It is straightforward to include this absorption in the parameter q, with the result that, in net, only half of the radiant energy emitted is lost from the system.

In fire configurations of larger scale, absorption of radiation within the gas may influence the temperature field appreciably. For the opposite limit of optically thick media, transport of radiation is diffusive, and Eq. (2) should be replaced by an alternative conservation equation in which the diffusion approximation for radiation is employed. Analysis of extinction on the basis of such a formulation would parallel that of Liñán (1974) in which energy loss occurs by conduction, with the molecular coefficient of thermal conductivity being increased by addition of the coefficient of conductivity for radiation transport, so that an effective Lewis number for the process exceeds unity. The result would be a modified definition of a critical Damköhler number of the second kind for extinction, with the radiation conductivity contributing to the characteristic diffusion time appearing therein. The utility of such a result would be open to question since estimates suggest that in fires the mean-free-path for radiation seldom is short enough to justify the diffusion approximation near the reaction zone.

LIST OF SYMBOLS

- A — $(a+T_e-T_w)/(2T_e)$, energetic parameter
- a — $\gamma_{if}T_f/T_a$
- a_i — arbitrary constant
- B — transfer number, Eq. (41)
- B_O — pre-exponential rate factor for molar oxidizer consumption
- b_i — arbitrary constant
- C — $(B-\alpha)/[(1+\alpha)B]$, flame-location parameter
- C_l — radiant loss correction factor, Eq. (43)
- c — parameter defined in Eq. (44)
- c_a — parameter defined in Eq. (45)
- c_p — specific heat at constant pressure
- D — $T_w/(2T_e)$, surface-temperature parameter
- D_I — Damköhler number
- F — flow factor relating kinetic parameters to extinction conditions
- F_l — radiant energy loss per unit volume per unit time divided by $\rho Qk(du_e/dr)$
- f — nondimensional stream function
- G_l — function defined in Eq. (7)
- g_i — coefficient defining temperature sensitivities of γ_i, $d \ln\gamma_i/d \ln T$
- h — $\int_0^\eta f d\eta$
- k — flow factor (1, two-dimensional; 2, axisymmetric)
- L — effective heat of gasification of fuel
- l — separation distance between fuel surface and oxidizer duct exit
- l_p — Planck-mean absorption length
- l_i — radiant loss parameter, Eqs. (24) and (46)
- M — molecular weight
- m_F — temperature-gradient parameter defined in Eq. (35)
- n_s — local soot-particle number density
- P — Prandtl number
- Q — heat of combustion (fuel–mass based, gaseous fuel to gaseous products)
- Q_l — nondimensional radiant energy loss, Eq. (20)
- q — nondimensional effective heat of gasification for fuel L/Q
- R° — universal gas constant
- r — transverse coordinate
- r_s — soot particle radius
- T — nondimensional temperature
- T_s — nondimensional soot surface temperature
- T_a — nondimensional activation temperature $Ec_p/(R^\circ Q)$
- U — oxidizer stream velocity
- u — tangential velocity
- x — canonical coordinate, Eq. (4)
- x_f — nondimensional flame location, Eq. (15)
- Y_i — stoichiometrically adjusted mass fraction of species i
- Y_{O_2} — free-stream oxygen mass fraction
- y — stretched coordinate $(x-x_f)(1+\alpha)T_a/T_f^2$
- z — $(4\delta)^{1/3}y/2$
- Z — $(4\delta)^{1/3}[\theta+(1-2m_F)y/2]$

Greek Symbols

- α — product of stoichiometric fuel–oxidizer mass ratio with free-stream oxygen mass fraction
- β — effective difference of nondimensional oxidizer and fuel temperature, Eq. (10)
- γ_i — coefficient defining temperature sensitivities of $F_l(T)$, $d \ln F_l/d \ln T$.
- δ — reduced Damköhler number, Eq. (33)
- η — Howarth–Dorodnitsyn variable
- θ — $(T_f-T)T_a/T_f^2$
- λ — radiant loss parameter, Eq. (32)
- μ — coefficient of dynamic viscosity
- μ_i — flame-temperature parameters, Eq. (21), (27)
- ξ — stretched coordinate $\gamma_{if}(x-x_f)/T_f$
- ρ — density
- σ — Stefan–Boltzmann constant
- ϕ — stretched temperature, Eq. (17)
- ψ — $(Y_O+T-T_f)T_a/T_f^2$
- ψ_f — arbitrary constant

Subscripts

- a — activation
- E — extinction
- e — oxidizer stream
- F — fuel
- fa — equilibrium diffusion-flame
- f — adiabatic diffusion-flame
- i — F or O
- O — oxidizer
- w — fuel surface