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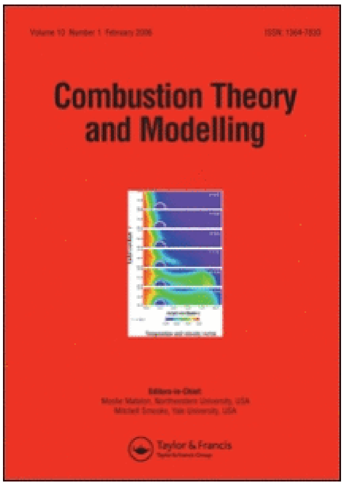

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High order effects in one step reaction sheet jump conditions for premixed flames

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Abstract
The differences need to be understood between the leading order jump conditions, often assumed at a flame sheet in combustion theory, and the actual effect of a one step chemical reaction governed by Arrhenius kinetics. These differences are higher order in terms of a large activation temperature analysis and can be estimated using an asymptotic approach. This paper derives one order of asymptotic correction to the leading order jump conditions that are normally used for describing premixed laminar combustion, providing additional contributions that are due to curvature, flow through the flame sheet and the temperature gradient into the burnt gas. As well as offering more accurate asymptotic results, these can be used to estimate the errors that are inherent in adopting only the leading order version and they can point towards major qualitative changes that can occur at finite activation temperatures in some cases. Applied to steady non-adiabatic flame balls it is found that the effect of a non-zero temperature gradient in the burnt gas provokes the most serious deficiency in the asymptotic approach.

1. Introduction

When studying premixed flames modelled using a thermally sensitive, exothermic, one step reaction of the form

$$\nu_{\mathrm{F}}\mathrm{F} + \nu_{\mathrm{X}}\mathrm{X} \rightarrow \mu_{\mathrm{P}}\mathrm{P}$$

a common practice in combustion theory is to assume that all chemistry takes place on an infinitesimally thin free boundary or sheet of reaction, at a rate that depends sensitively only on temperature. Leading order matched asymptotic studies of an 'inner' reaction region, for large activation temperature, provide the main justification for this. Many illustrative examples are available in the literature (see, e.g. [1–5]).

The use of a reaction sheet and its appropriate assumptions lead to considerable simplification in the model and its analysis, while remaining physically relevant; the restriction of one step reactions to relatively thin regions in premixed flames and an associated thermal sensitivity of the rate at which reactants are consumed and heat is released do seem to mimic the most important practical features of the chemistry in many situations.

However, it is also important to ascertain what effects are being neglected when adopting this form of model, even if only to confirm that the effects are small. This paper examines the jump conditions that may be used across a flame sheet, as a systematic asymptotic reduction from a one step Arrhenius description of the chemistry for large activation temperature. At higher order, the jump conditions depend on curvature, speed of gas flow through the sheet and the normal temperature gradient into the burnt gases at the sheet.

In cases where the activation temperature is large enough and the temperature at the reaction sheet undergoes any form of change, either in time or as parameters change, these higher order effects are likely to be relatively unimportant, qualitatively and quantitatively, providing only minor corrections. However, in cases where there is very little change in temperature at the reaction sheet, other influences may play a more significant role. It is informative, anyway, to clarify the roles that curvature, propagation speed and conduction into the burnt gas must play in modifying the jump conditions usually encountered at a reaction sheet.

Stability analyses, leading for example to the Sivashinsky equation [6], or describing the stability of flame balls [7–10], often involve a dispersion relation that covers two orders of magnitude in a perturbation parameter. Both orders are generated by the leading order jump conditions at a flame sheet, because of their extreme sensitivity to temperature changes at the sheet. Stability boundaries, for Lewis numbers or other parameters, are then determined asymptotically [6]. With one further order of correction to the jump conditions sharper refinements, at least, are made possible in determining stability boundaries.

While the use of jump conditions at a reaction sheet is extremely useful in examining analytically the structure, stability and behaviour of various forms of premixed flames, their use is less practical in carrying out numerical simulations of flames with one step chemistry. The numerical implementation of a moving free-boundary problem, at which specific boundary conditions need to be satisfied, is much more problematic than the simulation of a chemical process that is spatially distributed, even if it only happens on a relatively small, but not infinitesimally small, spatial scale. The activation temperature used in any one numerical simulation must also then be fixed and, necessarily, finite. Unexpectedly large differences can then arise between leading order asymptotic predictions for large activation temperature, based on using jump conditions, and numerical observations at finite activation temperatures [10]. Higher order corrections to the jump conditions can be used to estimate these differences.

In this paper, we derive one order of asymptotic correction to the jump conditions that are normally used in describing laminar premixed combustion. As an illustration of their utility, the improved jump conditions are applied to a model for steady, non-adiabatic flame balls [7–10]. It is found that the higher order jump conditions successfully predict a qualitative disagreement with the leading order results at moderate values of the Zeldovich number, as has been observed numerically [10].

In fact, at reduced Zeldovich numbers, the numerical solutions deviate much more strongly from the leading order results, although the higher order jump conditions successfully predict the onset and basic nature of the disagreement. At the Zeldovich numbers for which major changes occur, the higher order predictions are, of course, no longer accurate and any continued overall similarity with the numerics may be simply fortuitous. However, the leading order predictions are not even qualitatively correct at this stage and they offer no hint about their own loss of validity. The higher order corrections do, at least, predict their own downfall.

This finding helps to demonstrate some of the subtlety that is inherent in the one step chemical model at large but finite Zeldovich numbers. Knowing some of the nuances of its asymptotic expression, in the form of improved jump conditions, can help towards identifying and understanding the ways in which solutions differ from their limiting form at infinite Zeldovich number as well as, simply, providing more accurate asymptotic results.

2. Model

2.1. One step Arrhenius model

A dimensionless low Mach number model, that describes the one step decomposition reaction F → P using an Arrhenius rate law, can be written as

$$\begin{aligned} &F_t + \mathbf{v}\cdot\nabla F = \frac{\nabla^2 F}{\mathrm{Le}} - D\,\omega \\ &T_t + \mathbf{v}\cdot\nabla T = \nabla^2 T + Q D\,\omega - \ell \\ &\omega = F\exp\left(\frac{\theta}{T_*} - \frac{\theta}{T}\right) \end{aligned} \tag{1}$$

in which Le is the Lewis number of the reactant F, the scaled mass-fraction of which is represented by $F(t,\mathbf{x})$; Q is a heat release parameter; ℓ represents the rate of radiative heat loss; T_* is a suitably chosen dimensionless reference temperature, that is typically close to the maximum value that can be estimated for the absolute temperature $T(t,\mathbf{x})$; and the velocity $\mathbf{v}(t,\mathbf{x})$ is a solution of the Navier–Stokes equations

$$\begin{aligned} &\rho_t + \mathbf{v}\cdot\nabla\rho + \rho\nabla\cdot\mathbf{v} = 0 \\ &\rho(\mathbf{v}_t + \mathbf{v}\cdot\nabla\mathbf{v}) + \nabla p = \rho\,\mathrm{Pr}\nabla\cdot\left(\nabla\mathbf{v} + (\nabla\mathbf{v})^{\mathrm{T}}\right) - \rho\,\tfrac{2}{3}\,\mathrm{Pr}\nabla(\nabla\cdot\mathbf{v}) + \mathbf{g} \end{aligned} \tag{2}$$

in which Pr is the Prandtl number and $\mathbf{g}$ is a dimensionless gravitational vector. These equations are linked to the combustion equations (1) via the dimensionless form of the isobaric ideal gas law $\rho T = 1$, since the density $\rho(t,\mathbf{x})$ then changes as the temperature changes. Relatively small variations in absolute pressure, of the order of the square of the Mach number, are represented by the dimensionless hydrodynamic pressure $p(t,\mathbf{x})$. Only if the ideal gas law is replaced by a constant density model, $\rho \equiv 1$, is the velocity made to be independent of any temperature changes [1–4]. Selecting values for T_* and the Damköhler number D has the effect of fixing the scalings of time t and space $\mathbf{x}$.

2.2. Reaction sheet model

When the dimensionless activation temperature θ is large enough, the reaction rate term ω is commonly considered only to act at an infinitesimally thin reaction sheet, or interface, $\mathbf{\Gamma} \subset \mathbb{R}^3$. The effect of the chemistry is then entirely summarized by jump conditions applied to F and T, across the interface, which can be expressed most simply if we rewrite the Damköhler number as $D = \delta^2\theta^2 Q^2/(2\mathrm{Le}T_*^4)$, fully parameterized by δ. The model then typically takes the form

$$\begin{aligned}
&\left.\begin{aligned} F_t + \mathbf{v}\cdot\nabla F &= \frac{\nabla^2 F}{\mathrm{Le}} \\ T_t + \mathbf{v}\cdot\nabla T &= \nabla^2 T - \ell \end{aligned}\right\} \qquad \text{for } \mathbf{x} \notin \mathbf{\Gamma} \\
&\left.\begin{aligned} [T] = [F] = F = 0 \\ \frac{[F_n]}{\mathrm{Le}} = -\frac{[T_n]}{Q} = \Omega \end{aligned}\right\} \qquad \text{at } \mathbf{x} \in \mathbf{\Gamma} \\
&\Omega = \delta \exp\left(\tfrac{1}{2}\left(\frac{\theta}{T_*} - \frac{\theta}{T}\right)\right)
\end{aligned} \tag{3}$$

(sometimes known as the delta function model [1]) in which $\partial_n = \hat{\mathbf{n}}\cdot\nabla$ is the normal gradient, or spatial derivative in the direction to which a normal unit vector $\hat{\mathbf{n}}$ points. The brackets $[\cdot]$ denote the usual jump in value, across the sheet $\mathbf{\Gamma}$, of the contents of the brackets, being the value on the side of the interface to which $\hat{\mathbf{n}}$ points minus the value on the opposite side. It is useful to think of $\hat{\mathbf{n}}$ as pointing in the direction of propagation of the interface or, more generally, towards increasing values of F, and this convention will be adopted in this article, although the leading order jump conditions stated in (3) remain the same for either of the two possible directions in which $\hat{\mathbf{n}}$ can point.

The jump conditions in (3) state that T and F are continuous across the interface where the reactant concentration F must also have the value zero. The jumps in T_n and F_n are linearly related to each other and take a value that is determined by the nonlinear function Ω, which depends only on the temperature at the interface to leading order as $\theta \to \infty$. Since θ is large, they are also a highly sensitive function of temperature. On the one side of the flame sheet the chemistry must be in equilibrium, $\omega = 0$, which is ensured in equations (1) by having $F \equiv 0$. On the other side, the temperature is typically far enough below T_* for the value of ω to be transcendentally small as $\theta \to \infty$ (i.e. ω is smaller than any negative power of θ, as $\theta \to \infty$). Neglecting the reaction rate ω when $\mathbf{x} \notin \mathbf{\Gamma}$ is therefore reasonable when θ is large enough.

According to the model (3), premixed flames of various types (laminar flames or flame-balls) are thus envisaged as having a structure in which all chemical reaction is concentrated at a narrow sheet, the interface $\mathbf{\Gamma}$, with reactant concentration and temperature determined by inert advective, diffusive and radiative processes away from the sheet. The approximation has been demonstrated to work well in examples that are far too numerous to cite in this article (see [1–4]).

2.3. *Jump in pressure*

It is worth noting that the Navier–Stokes equations (2) are not valid if there is any jump in the gradient of density across a flame sheet, as would be the case if, for example, the isobaric ideal gas law $\rho T = 1$ were to hold, as well as the reaction sheet model (3). In particular, if we consider changes across the interface that occur on a very small scale, measured by ε, and if density, velocity and length are rescaled such that

$$\rho = \bar{\rho} + \varepsilon\varrho, \qquad \mathbf{v} = \bar{\mathbf{v}} + \varepsilon\boldsymbol{v}, \qquad \mathbf{x} = \mathbf{\Gamma} + \varepsilon\hat{\mathbf{n}}\eta$$

then the Navier–Stokes equations (2) have the local form for order one values of η

$$(\bar{\mathbf{v}}\cdot\hat{\mathbf{n}})\varrho_\eta + \bar{\rho}\boldsymbol{v}_\eta\cdot\hat{\mathbf{n}} = \mathrm{O}(\varepsilon), \qquad p_\eta = \bar{\rho}\,\tfrac{4}{3}\mathrm{Pr}\boldsymbol{v}_{\eta\eta}\cdot\hat{\mathbf{n}} + \mathrm{O}(\varepsilon).$$

These equations are easily integrated to provide the additional jump conditions across the interface

$$[\mathbf{v}] = [\rho] = 0, \qquad [p] = \tfrac{4}{3}\mathrm{Pr}\,\rho[\mathbf{v}_n]\cdot\hat{\mathbf{n}} = -\tfrac{4}{3}\mathrm{Pr}(\mathbf{v}\cdot\hat{\mathbf{n}})[\rho_n] \tag{4}$$

which need to be applied when solving the Navier–Stokes equations. In particular, pressure and the normal derivative of velocity are both discontinuous if there is a jump in the gradient of density at the interface.

3. Reaction sheet jump conditions

We will now use a singular perturbation approach, based on the limit $\theta \to \infty$, to obtain and to generalize the jump conditions presented in (3), as a systematic asymptotic approximation to the continuum model (1). We shall start with the effect of temperature alone, under conditions in which the temperature gradient in the burnt gas is zero, and move on to examine weak effects of curvature, flow, non-zero temperature gradient in the burnt gas and unsteady behaviour.

3.1. Temperature dependence of the jump conditions

A complete description of the temperature dependence in the jump conditions at the reaction sheet can be arrived at by selecting a particular form for the model problem (1) in which only reactive and diffusive effects are present.

For this, we can consider a completely stationary, one-dimensional problem, with no radiative heat loss and no flow, in which $F = F(x)$ and $T = T(x)$, only. Taking $F(-\infty) = 0$ and $T(-\infty) = \bar{T}$, where the value of $\bar{T}$ is within order θ^{-1} of T_*, equations (1) then reduce to the model equations and boundary conditions for F and T

$$\frac{F''}{\mathrm{Le}} = -\frac{T''}{Q} = DF\exp\left(\theta\frac{T-T_*}{T_*T}\right) \qquad \lim_{x\to-\infty} F = 0, \quad \lim_{x\to-\infty} T = \bar{T}. \tag{5}$$

It can be noted that $(F, T) \equiv (0, \bar{T})$ provides an exact, constant solution, corresponding to complete chemical equilibrium. Also, the reaction rate term becomes transcendentally small as $\theta \to \infty$ when $F > 0$ and $T < \bar{T} - \mathrm{O}(\theta^{-1}\ln\theta)$. The second derivatives are then negligibly small so that F and T become linear in x to exponential orders of accuracy. The change in slope between the constant and linear forms of behaviour (when $F = 0$ and when $F > 0$) is, of course, determined by the reaction-rate term.

Since $F \geqslant 0$ it follows that F, and also T, monotonically approach their values at $x = -\infty$. Thus $F'(-\infty) = T'(-\infty) = 0$ and the first equality in (5) can be integrated exactly to give $T = \bar{T} - QF/\mathrm{Le}$. Writing $f(x) = \theta QF(x)/(\mathrm{Le}\bar{T}^2)$ it follows that

$$f'' = \tfrac{1}{2}k^2 f\exp\left(\frac{-f}{1-f\bar{T}/\theta}\right) \qquad \text{with} \qquad k^2 = 2\mathrm{Le}D\exp\left(\frac{\theta(\bar{T}-T_*)}{T_*\bar{T}}\right)$$

so that, since f' is zero when $f = 0$, we can find that

$$\begin{aligned}\frac{f'^2}{k^2} &= \int_0^f z\exp\left(\frac{-z}{1-z\bar{T}/\theta}\right)\mathrm{d}z = \int_0^{f/(1-f\bar{T}/\theta)} \frac{se^{-s}}{(1+s\bar{T}/\theta)^3}\,\mathrm{d}s \\ &= \int_0^f ze^{-z}\left(1 - z^2\frac{\bar{T}}{\theta} - \left(z^3 - \tfrac{1}{2}z^4\right)\frac{\bar{T}^2}{\theta^2} - \left(z^4 - z^5 + \tfrac{1}{6}z^6\right)\frac{\bar{T}^3}{\theta^3} + \mathrm{O}(\theta^{-4})\right)\mathrm{d}z \\ &= 1 - (1+f)e^{-f} - 6\left(1 - \left(1 + f + \tfrac{1}{2}f^2 + \tfrac{1}{6}f^3\right)e^{-f}\right)\frac{\bar{T}}{\theta} + \mathrm{O}(\theta^{-2})\end{aligned}$$

in which the second integral has an exact, but lengthy and uninformative, expression in terms of exponential integral functions. When f is strictly of order θ, or $F \neq \mathrm{o}(1)$, as $\theta \to \infty$, this exact solution for f'^2/k^2 defines a constant value for f', to exponential orders of accuracy.

Approximating this constant to algebraic orders (by expanding the exponential for large values of θ, as indicated in the equations above, and finally letting $f \to \infty$) gives

$$\frac{f'^2}{k^2} = 1 - 6\frac{\bar{T}}{\theta} + 36\frac{\bar{T}^2}{\theta^2} - 240\frac{\bar{T}^3}{\theta^3} + \mathrm{O}(\theta^{-4}) = \left(1 + 6\frac{\bar{T}}{\theta}\right)^{-1} + \mathrm{O}(\theta^{-3})$$

after continuing the procedure in a straightforward way to the order of θ^{-3}.

The values of F' and T' when $F > \mathrm{O}(\theta^{-1}\ln\theta)$ can therefore be estimated as

$$\begin{aligned}\frac{F'}{\mathrm{Le}} = -\frac{T'}{Q} &= \frac{\bar{T}^2}{Q\theta}\sqrt{2\mathrm{Le}D}\exp\left(\frac{\theta}{2}\frac{\bar{T} - T_*}{T_*\bar{T}}\right)\frac{1 + \mathrm{O}(\theta^{-3})}{\sqrt{1 + 6\bar{T}/\theta}} \\ &= \delta\exp\left(\frac{\theta}{2}\frac{\bar{T} - T_*}{T_*\bar{T}}\right)\frac{\bar{T}^2/T_*^2}{\sqrt{1 + 6\bar{T}/\theta}} + \mathrm{O}(\theta^{-3})\end{aligned} \tag{6}$$

after setting $D = \delta^2\theta^2 Q^2/(2\mathrm{Le}T_*^4)$, as before in equations (3). By noting that T_* needs to be chosen to be within order θ^{-1} of typical values of $\bar{T}$, for order one changes in gradient to arise in the dimensionless scalings adopted in the model, it follows that the ratio $\bar{T}^2/T_*^2$ must be asymptotically close to unity. Now extrapolating the resulting straight-line asymptotic solutions for F and T, valid when $F > \mathrm{O}(\theta^{-1}\ln\theta)$, back to a point where the solution for F would extrapolate to zero, and that for T would extrapolate to $\bar{T}$, this point identifies a location where the overall changes in F, T and their gradients should behave as if they satisfied the jump conditions given in (3), in which the errors are asymptotically of the order of θ^{-1}, as $\theta \to \infty$. Using the final expression given in (6) would reduce the errors to the order of θ^{-3}, provided there were no other factors, apart from temperature, influencing the jump conditions.

3.2. Curvature, flow and unsteady effects at the flame sheet

It is informative now to examine some of the other factors that are being neglected in adopting the flame sheet model (3). For this purpose, let us consider the slightly more simplified version of the model (1)

$$\begin{aligned} &F_t + \mathbf{v}\cdot\nabla F = \frac{\nabla^2 F}{\mathrm{Le}} - \tfrac{1}{2}\frac{\beta^2\omega}{\mathrm{Le}} \\ &\mathsf{T}_t + \mathbf{v}\cdot\nabla\mathsf{T} = \nabla^2\mathsf{T} + \tfrac{1}{2}\frac{\beta^2\omega}{\mathrm{Le}} - l \\ &\omega = F\exp(\beta(\mathsf{T} - 1)) \end{aligned} \tag{7}$$

arrived at: by setting $D = \theta^2 Q^2/(2\mathrm{Le}T_*^4) = \frac{1}{2}\beta^2/\mathrm{Le}$, so that δ is set to unity; by rescaling temperature such that $T = T_* + Q(\mathsf{T} - 1)$, so that $T = T_*$ when $\mathsf{T} = 1$ (note the change in font); by defining $l = \ell/Q$; and by linearizing the exponent in the reaction rate term about the rescaled temperature $\mathsf{T} = 1$, such that

$$\frac{\theta}{T_*} - \frac{\theta}{T} = \frac{\theta Q(\mathsf{T} - 1)}{T_*^2(1 + Q(\mathsf{T} - 1)/T_*)} \sim \beta(\mathsf{T} - 1) \qquad \text{with } \beta = \frac{\theta Q}{T_*^2}.$$

This defines the Zeldovich number β, which is typically large because θ is large. For an adiabatic premixed flame, the Zeldovich number can be written in terms of dimensional quantities as

$$\beta = \frac{T_{\mathrm{A}}(T_{\mathrm{f}} - T_0)}{T_{\mathrm{f}}^2} \tag{8}$$

where T_{A} is the dimensional activation temperature of the one step reaction; T_{f} is the dimensional flame temperature, equivalent to the dimensionless temperature T_*; and T_0 is the dimensional unburnt temperature.

The only real change from the model (1), is the linearization of the exponent in the reaction rate term. Equation (6) can be used to show that this makes no significant difference, to leading order, for changes in the flame temperature $\bar{T}$, from the value T_*, that are of the order of θ^{-1}. The jump conditions that arise using the full form of the Arrhenius exponent, in the absence of effects due to curvature, propagation and temperature gradients in the burnt gas, have already been found in equation (6) and will be brought back into account later on. However, the linearization does, for now, simplify the analysis of the chemistry. This will prove very useful since we are about to introduce greater complexity by considering a more general geometry for the flame sheet in a more general flow field.

In particular, let us seek the asymptotic structure that a reaction region would have when the flame sheet $\boldsymbol{\Gamma}$ is curved, when there is a non-uniform flow field and when temperatures in the reaction region are close to the value $\mathsf{T} = 1$. For simplicity, we can take the problem to be two dimensional, generalizing the results to three dimensions later on, so that, at some moment in time t, the sheet can be taken to lie at $y = Y(t, x)$. By rotating coordinates and shifting the origin, in time as well as in space, to any point of interest at any moment, we can set both Y and Y_x to be zero at $x = t = 0$. We can then focus on the reaction region around $x = y = t = 0$, knowing that this could represent any point on the interface at any moment in time.

By allowing the reference frame to move at a constant speed, the flow field can be described, near $x = y = t = 0$, using

$$\mathbf{v} = (\sigma_0 x + \sigma_1 y + a_1 t,\ \sigma_2 x - \sigma_0 y + a_2 t).$$

In particular, the reference frame is taken to move with the same velocity as the fluid at the origin, at the moment when $t = 0$, so that $\mathbf{v} = \mathbf{0}$ when x, y and t are all zero. If there is a constant and uniform rate of strain, then σ_0 represents a diverging strain rate in the x-direction, converging in the y-direction, while σ_1 and σ_2 represent shear components of the rate of strain tensor. The vector (a_1, a_2) is related to the acceleration of the flow field at $x = y = t = 0$. More generally, σ_0, σ_1, σ_2, a_1 and a_2 are of order one and may vary with space and time for order one values of x, y and t, and may be generalized even further to include any effects of thermal expansion that might be inherent in the model as well as variations in three spatial dimensions.

If we now introduce a new variable r to measure distances in the y-direction ahead of the sheet, such that

$$r = y - Y(t, x)$$

then the equations (7) for F and T can be transformed to the non-orthogonal reference frame of (t, x, r), using

$$\begin{aligned} &\partial_y \mapsto \partial_r, \qquad \partial_x \mapsto \partial_x - Y_x \partial_r \\ &\partial_{xx} + \partial_{yy} \mapsto (1 + Y_x^2)\partial_{rr} - Y_{xx}\partial_r + \partial_{xx} - 2Y_x\partial_{rx} \\ &\partial_t + (\mathbf{v} \cdot \nabla) \mapsto \partial_t - Y_t \partial_r + \boldsymbol{v} \cdot (\partial_x, \partial_r) \end{aligned}$$

in which

$$\boldsymbol{v} = \big(\sigma_0 x + \sigma_1 (Y{+}r) + a_1 t, (\sigma_2{-}\sigma_0 Y_x)x - (\sigma_0{+}\sigma_1 Y_x)(Y{+}r) + (a_2{-}a_1 Y_x)t\big).$$

This gives

$$\big(\partial_t - V\partial_r + \boldsymbol{v} \cdot (\partial_x, \partial_r)\big)F = \big((1 + Y_x^2)\partial_{rr} - Y_{xx}\partial_r + \partial_{xx} - 2Y_x\partial_{rx}\big)\frac{F}{\mathrm{Le}} - \tfrac{1}{2}\frac{\beta^2\omega}{\mathrm{Le}}$$

$$\big(\partial_t - V\partial_r + \boldsymbol{v} \cdot (\partial_x, \partial_r)\big)\mathsf{T} = \big((1 + Y_x^2)\partial_{rr} - Y_{xx}\partial_r + \partial_{xx} - 2Y_x\partial_{rx}\big)\mathsf{T} + \tfrac{1}{2}\frac{\beta^2\omega}{\mathrm{Le}} - l$$

in which we have written $V = Y_t$ to denote the vertical speed of movement of the interface $y = Y(t, x)$, at any fixed value of x. At the point $x = 0$ and time $t = 0$, at which $Y_x = 0$ and $\mathbf{v} = \mathbf{0}$, it represents the normal propagation speed of the interface relative to the medium. By eliminating ω, we obtain the equation for the enthalpy

$$\big(\partial_t - V\partial_r + \boldsymbol{v}\cdot(\partial_x, \partial_r)\big)(\mathsf{T}+F) = \big((1+Y_x^2)\partial_{rr} - Y_{xx}\partial_r + \partial_{xx} - 2Y_x\partial_{rx}\big)\left(\mathsf{T}+\frac{F}{\mathrm{Le}}\right) - l$$

which shows that non-equidiffusive effects, for which $\mathrm{Le} \neq 1$, will tend to redistribute the dimensionless enthalpy, $h = \mathsf{T} + F$.

Broadly speaking, we can imagine that a thin region of reaction forms part of a flame structure of some type, in which temperature and reactant concentration vary by order one over length and time scales (measured by x, y and t) that are of order one. Thus the interface $y = Y(t, x)$ might have a curvature of order one and be moving at a speed $V = Y_t$ that is of order one. Because of the sensitive temperature dependence that has been identified for the jump in gradient across the reaction sheet, which in turn should not change abruptly, the temperature at the sheet should not vary by more than the order of β^{-1}. To be consistent with this limitation, the temperature in the burnt gas (where values of F must be transcendentally small) should also only vary by the order of β^{-1} over length and time scales that are of order one; in turn, for this to be so, temperature gradients in the burnt gas cannot exceed the order of β^{-1} and the value of the heat-loss term l must also be of order β^{-1}, at most.

Thus, without delving any more deeply into the nature of the burnt gas, where reactivity is transcendentally close to equilibrium ($F = 0$), we can simply assume the following asymptotic nature of the burnt region near the flame sheet, when r is small and negative

$$\begin{array}{lll} \mathsf{T} = \bar{\mathsf{T}} + r\bar{\mathsf{T}}_r + \mathrm{O}(r^2\epsilon) & \text{and} & F = \bar{\mathrm{o}}(\epsilon) \\ \text{with } \bar{\mathsf{T}}(t, x) = 1 + \mathrm{O}(\epsilon) & \text{and} & \bar{\mathsf{T}}_r(t, x) = \mathrm{O}(\epsilon) \end{array}$$

as $\epsilon \to 0$, with $\epsilon \ll -r \ll 1$. For the sake of more efficient notation, we have defined $\epsilon = \beta^{-1}$ and we use the symbol $\bar{\mathrm{o}}(\cdot)$ to denote something that is transcendentally small. That is, $F = \bar{\mathrm{o}}(\epsilon) \iff F = \mathrm{o}(\epsilon^\nu)$, as $\epsilon \to 0$, for any power $\nu \in \mathbb{R}$. Thus we ensure that the temperature gradient is small in the burnt region, behind the reaction region, and that the temperature at the reaction sheet deviates only slightly from the temperature $\mathsf{T} = 1$. If we now rescale to focus attention on the region where significant chemical activity takes place, such that

$$F = \epsilon \mathrm{Le} f, \qquad \mathsf{T} = 1 - \epsilon\phi, \qquad r = \epsilon\eta$$

then, in order to match with the properties in the burnt region, the conditions that must be satisfied in the reaction region as $\eta \to -\infty$ are

$$\begin{array}{lll} \phi = \bar{\phi} + \epsilon\eta\bar{\phi}_r + \mathrm{O}(\epsilon^2\eta^2) & \text{and} & f = \bar{\mathrm{o}}(\epsilon) \\ \text{with } \bar{\mathsf{T}} = 1 - \epsilon\bar{\phi} & \text{and} & \bar{\mathsf{T}}_r = -\epsilon\bar{\phi}_r \end{array} \tag{9}$$

as $\epsilon \to 0$, with $1 \ll -\eta \ll \epsilon^{-1}$. We can assume that $\bar{\phi}(t, x)$ and $\bar{\phi}_r(t, x)$ are both of order one as $\epsilon \to 0$.

The governing reactive–diffusive equations describing the region of chemical reaction now become

$$(1 + Y_x^2) f_{\eta\eta} - \tfrac{1}{2} f \mathrm{e}^{-\phi} + \epsilon(\mathrm{Le}V - Y_{xx}) f_\eta = 2\epsilon Y_x f_{\eta x} + \mathrm{Le}\left(\epsilon^2 f_t + \boldsymbol{v}\cdot(\epsilon^2 f_x, \epsilon f_\eta)\right) - \epsilon^2 f_{xx}$$

$$(1 + Y_x^2)\phi_{\eta\eta} - \tfrac{1}{2} f \mathrm{e}^{-\phi} + \epsilon(V - Y_{xx})\phi_\eta = 2\epsilon Y_x \phi_{\eta x} + \epsilon^2\phi_t + \boldsymbol{v}\cdot(\epsilon^2\phi_x, \epsilon\phi_\eta) - \epsilon^2\phi_{xx} - \epsilon l$$

with

$$\boldsymbol{v} = \big(\sigma_0 x + \sigma_1(Y+\epsilon\eta) + a_1 t, (\sigma_2 - \sigma_0 Y_x)x - (\sigma_0 + \sigma_1 Y_x)(Y+\epsilon\eta) + (a_2 - a_1 Y_x)t\big).$$

The terms $\mathrm{Le}\epsilon^2 f_t$ and $\epsilon^2\phi_t$ show that a very fast time scale, of the order of $t = \mathrm{O}(\epsilon^2)$, is present in the problem. Any transient effects, that there may be, will asymptote quickly towards a stable quasi-steady evolution, over this short time scale. On the longer time scale $t = \mathrm{O}(1)$, over which curvature, velocity and other terms may vary by order one, these equations make it clear that the quasi-steady behaviour is then only affected at the order of ϵ^2, at most, by the terms containing f_t and ϕ_t.

If we therefore focus on solving for the quasi-steady forms for f and ϕ, we need only consider the solution at the instant $t = 0$. No generality is lost in doing this since, as already outlined, any point on the interface and any time can be mapped to $x = y = t = 0$. Moreover, at $t = 0$, when coordinates have been chosen to set $Y_x(t, 0) = 0$, we have

$$Y = \tfrac{1}{2}x^2 Y_{xx}(0,0) + \mathrm{O}(x^3), \qquad Y_x = xY_{xx}(0,0) + \mathrm{O}(x^2).$$

Thus, taking Y_{xx} to be of order one, and only considering values of x that are of order ϵ (i.e. considering only the region close to $x = 0$), we have that $\boldsymbol{v} = \mathrm{O}(\epsilon)$. The convective terms containing $\boldsymbol{v}$ are then at most of order ϵ^2. Since we also find that Y_x is of order ϵ, all terms containing Y_x are then at most of order ϵ^2 and, finally, because l is at most of order ϵ, the heat loss term ϵl is also no greater than the order of ϵ^2.

Ignoring terms that are of order ϵ^2 or smaller now gives

$$\begin{aligned} f_{\eta\eta} - \tfrac{1}{2} f e^{-\phi} + \epsilon(\mathrm{Le}V{+}2\kappa)f_\eta &= \mathrm{O}(\epsilon^2) \\ \phi_{\eta\eta} - \tfrac{1}{2} f e^{-\phi} + \epsilon(V + 2\kappa)\phi_\eta &= \mathrm{O}(\epsilon^2) \end{aligned}$$

in which we have used the expression for the mean curvature of the interface in three dimensions, $\kappa = -Y_{xx}/2$, at $x = 0$, to write the equation in terms of the more generic parameter κ rather than Y_{xx}; a flame that is concave when viewed from the unburnt gas is taken to have negative curvature. Both $V(t, x)$ and $\kappa(t, x)$ can be taken to be locally constant, changing very little on the small time and length scales that we are considering around the point $x = y = 0$ at the time $t = 0$.

Eliminating $\frac{1}{2} f e^{-\phi}$ gives the enthalpy equation

$$\phi_{\eta\eta} - f_{\eta\eta} + \epsilon V(\phi_\eta - \mathrm{Le} f_\eta) + 2\epsilon\kappa(\phi_\eta - f_\eta) = \mathrm{O}(\epsilon^2)$$

which can be solved iteratively, along with the matching conditions (9), to obtain the expression for ϕ in terms of f

$$\phi = \bar{\phi} + f + \epsilon\eta\bar{\phi}_r + \epsilon V(\mathrm{Le} - 1)\int_{\bar{\eta}}^{\eta} f\,\mathrm{d}\eta + \mathrm{O}(\epsilon^2) \tag{10}$$

where $\eta = r/\epsilon = \bar{\eta}(\epsilon) < 0$ defines a position at which f and f_η are at most of order ϵ^2. As will be seen later, we can find such positions with $\bar{\eta} = -\mathrm{O}(|\ln\epsilon|)$. The precise choice of $\bar{\eta}$ is not important, provided $\epsilon\bar{\eta}(\epsilon) = \mathrm{o}(1)$ as $\epsilon \to 0$.

Substituting for ϕ now gives

$$f_{\eta\eta} = \tfrac{1}{2} f e^{-\bar{\phi}-f} + \epsilon V(1 - \mathrm{Le}) f_{\eta\eta}\int_{\bar{\eta}}^{\eta} f\,\mathrm{d}\eta - \epsilon\bar{\phi}_r\,\eta f_{\eta\eta} - \epsilon(2\kappa{+}\mathrm{Le}V) f_\eta + \mathrm{O}(\epsilon^2)$$

after using the leading order result, $f_{\eta\eta} = \frac{1}{2} f e^{-\bar{\phi}-f} + \mathrm{O}(\epsilon)$, to eliminate some occurrences of $\frac{1}{2} f e^{-\bar{\phi}-f}$. Multiplying by $2f_\eta$, integrating, by parts where necessary, and using the matching conditions (9), now leads to the expression for f_η in terms of f

$$\begin{aligned} f_\eta^2 = e^{-\bar{\phi}}(1 - (1{+}f)e^{-f}) &+ \epsilon V(1 - \mathrm{Le})\left(f_\eta^2\int_0^f \frac{f}{f_\eta}\,\mathrm{d}f - \int_0^f f f_\eta\,\mathrm{d}f\right) \\ &- \epsilon\bar{\phi}_r\left(\eta f_\eta^2 - \int_0^f f_\eta\,\mathrm{d}f\right) - 2\epsilon(2\kappa{+}\mathrm{Le}V)\int_0^f f_\eta\,\mathrm{d}f + \mathrm{O}(\epsilon^2). \end{aligned} \tag{11}$$

The sign of f_η must be positive, so that $f_\eta = e^{-\bar{\phi}/2}\sqrt{1-(1+f)e^{-f}} + \mathrm{O}(\epsilon)$ to leading order.

When f is small, it follows that $f_\eta \sim f\, 2^{-1/2}e^{-\bar{\phi}/2}$ and hence $f \sim \bar{f}\exp(\eta\, 2^{-1/2}e^{-\bar{\phi}/2})$ for some suitable function $\bar{f}(t,x)$. Solving for η, this demonstrates that $f = \mathrm{O}(\epsilon^2)$ where $\eta = -2^{1/2}e^{\bar{\phi}/2}|\ln\epsilon^2| + \mathrm{O}(1)$, so that $\bar{\eta}$ can be chosen such that $-\bar{\eta} \geqslant 2^{3/2}e^{\bar{\phi}/2}|\ln\epsilon| + \mathrm{O}(1)$.

Also since $f_\eta \sim e^{-\bar{\phi}/2}\sqrt{1-(1+f)e^{-f}}$, each of the integrals in the expression (11) can be reformulated as follows

$$\begin{aligned}
\int_0^f f_\eta\,\mathrm{d}f &= e^{-\bar{\phi}/2}\int_0^f \sqrt{1-(1+z)e^{-z}}\,\mathrm{d}z + \mathrm{O}(\epsilon) \sim e^{-\bar{\phi}/2}\,(f-\varpi)\\
\int_0^f ff_\eta\,\mathrm{d}f &= e^{-\bar{\phi}/2}\int_0^f z\sqrt{1-(1+z)e^{-z}}\,\mathrm{d}z + \mathrm{O}(\epsilon) \sim e^{-\bar{\phi}/2}\left(\tfrac{1}{2}f^2-\varpi_1\right)\\
\int_0^f \frac{f}{f_\eta}\,\mathrm{d}f &= e^{\bar{\phi}/2}\int_0^f \frac{z}{\sqrt{1-(1+z)e^{-z}}}\,\mathrm{d}z + \mathrm{O}(\epsilon) \sim e^{\bar{\phi}/2}\left(\tfrac{1}{2}f^2+\varpi_2\right).
\end{aligned} \tag{12}$$

Unfortunately, none of these integrals seem to have simple expressions in terms of elementary functions, but they do each approach the asymptotic forms shown on the right exponentially quickly as f becomes large. Noting that

$$\begin{aligned}
\varpi_1+\varpi_2 &= \int_0^\infty z(1-\sqrt{1-(1+z)e^{-z}})\mathrm{d}z + \int_0^\infty\left(\frac{z}{\sqrt{1-(1+z)e^{-z}}}-z\right)\mathrm{d}z\\
&= 2\int_0^\infty (1+z)\frac{\frac{1}{2}ze^{-z}}{\sqrt{1-(1+z)e^{-z}}}\,\mathrm{d}z\\
&= 2\left[(1+f)\sqrt{1-(1+f)e^{-f}} - \int_0^f \sqrt{1-(1+z)e^{-z}}\,\mathrm{d}z\right]_0^\infty\\
&= 2(1+\varpi)
\end{aligned}$$

the constants given in these formulae are seen to be related by $1+\varpi = \frac{1}{2}(\varpi_1+\varpi_2)$.

Hence, although the formula (11) cannot be evaluated easily beyond its leading order, it does approach the simple asymptotic behaviour for large values of f or η:

$$\begin{aligned}
f_\eta &= e^{-\bar{\phi}/2} + \epsilon V(1-\mathrm{Le})(1+\varpi) - \tfrac{1}{2}\epsilon\bar{\phi}_r e^{-\bar{\phi}/2}\,\eta\\
&\quad + \epsilon\left(\tfrac{1}{2}\bar{\phi}_r - 2\kappa - \mathrm{Le}V\right)(f-\varpi) + \mathrm{O}(\epsilon^2, e^{-f}).
\end{aligned}$$

Thus, while the formula (11) matches with properties expected in the burnt region for large negative values of η, where $f \to 0$, it is also linked with the region of ‘preheating’, where $f \gg 1$, through this asymptotic result for large positive values of η and f. Moreover, because the asymptote is approached exponentially quickly as f grows, it can be integrated, iteratively, to provide the asymptotic behaviour of f at large η:

$$\begin{aligned}
f &= \left(e^{-\bar{\phi}/2} + \epsilon V(1-\mathrm{Le})(1+\varpi) - \epsilon\left(\tfrac{1}{2}\bar{\phi}_r - 2\kappa - \mathrm{Le}V\right)\varpi\right)\\
&\quad \times\left(\eta - \epsilon(2\kappa+\mathrm{Le}V)\tfrac{1}{2}\eta^2\right) + \mathrm{O}(\epsilon^2, e^{-\phi}).
\end{aligned}$$

In arriving at this result, a function of integration has been chosen such that the solution has the value zero at $\eta = 0$. This amounts to *defining* the location of the path $y = Y(t,x)$, or $\eta = r = 0$, such that the extrapolation of this *asymptotic form* for f at large values of η, back to $\eta = 0^+$, gives the value zero for f. This is equivalent to a ‘closure assumption’ in defining the interface, as discussed in [5]. Equation (10) now takes on the asymptotic form for large values of η

$$\phi = \bar{\phi} + f + \epsilon\bar{\phi}_r\eta + \epsilon V(\mathrm{Le}-1)e^{-\bar{\phi}/2}\tfrac{1}{2}\eta^2 + \mathrm{O}(\epsilon^2, e^{-\phi})$$

and these equations provide the necessary far-field asymptotic behaviour of the solution in the reaction region for matching with a region of ‘preheating’ where $F > 0$ and $f \gg 1$.

The matching requirement that arises for $F(t, x, r)$ and $\mathsf{T}(t, x, r)$, in this preheating region as $r \to 0^{+}$, therefore takes on the form

$$\begin{aligned}\frac{F}{\mathrm{Le}} &= \bar{\mathsf{T}} - \mathsf{T} + r\bar{\mathsf{T}}_r + \tfrac{1}{2}r^2(1-\mathrm{Le})V e^{(\bar{\mathsf{T}}-1)/(2\epsilon)} + \mathrm{O}(\epsilon^2, r^3)\\ &= \left(e^{(\bar{\mathsf{T}}-1)/(2\epsilon)} + \epsilon(1+\varpi-\mathrm{Le})V + \left(\tfrac{1}{2}\bar{\mathsf{T}}_r+2\epsilon\kappa\right)\varpi\right)\left(r - \tfrac{1}{2}r^2(\mathrm{Le}V+2\kappa)\right) + \mathrm{O}(\epsilon^2, r^3).\end{aligned}$$

Alternatively, the requirement for matching of the derivatives F_r and T_r is

$$\begin{aligned}\frac{F_r}{\mathrm{Le}} &= \left(e^{(\bar{\mathsf{T}}-1)/(2\epsilon)} + \epsilon(1+\varpi-\mathrm{Le})V + \left(\tfrac{1}{2}\bar{\mathsf{T}}_r+2\epsilon\kappa\right)\varpi\right)(1-r(\mathrm{Le}V+2\kappa)) + \mathrm{O}(\epsilon^2, r^2)\\ \bar{\mathsf{T}}_r - \mathsf{T}_r &= \left(e^{(\bar{\mathsf{T}}-1)/(2\epsilon)} + \epsilon(1+\varpi-\mathrm{Le})V + \left(\tfrac{1}{2}\bar{\mathsf{T}}_r+2\epsilon\kappa\right)\varpi\right)(1-r(V+2\kappa)) + \mathrm{O}(\epsilon^2, r^2).\end{aligned}$$

When evaluated in the inner asymptotic limit, as $r \to 0^{+}$, these outer asymptotic solutions provide jumps in the values of the gradients, experienced by the outer asymptotic variables F and T, across an apparent flame sheet at $r = 0$, accurate to order ϵ, as $\epsilon \to 0$.

We have therefore shown that a more complete form of the gradient jump conditions, that arises at a flame sheet for the model (7), is

$$\begin{aligned}\frac{[F_n]}{\mathrm{Le}} &= -[\mathsf{T}_n]\\ &= e^{\beta(\mathsf{T}-1)/2} + \frac{(1+\varpi-\mathrm{Le})}{\beta}V + \varpi\left(\tfrac{1}{2}\mathsf{T}_n^{-} + \frac{2\kappa}{\beta}\right) + \mathrm{O}\big(\beta^{-2}, \mathsf{T}_n^{-2}\big) \qquad (13)\end{aligned}$$

as $\beta \to \infty$, where V is the normal propagation speed of the sheet relative to the medium, κ is the mean curvature of the sheet and T_n^{-} is the normal gradient of temperature on the burnt side, taken to be of the order of β^{-1}, or at least small in value. Any strain rate and shear rate in the flow, that change the fluid velocity by order one over lengths that are of the order of the thickness of the preheating region, can only affect the jump conditions at the flame sheet by an amount of the order of β^{-2}. Unsteadiness, radiative heat loss and transverse diffusion along the flame sheet also affect the jump conditions at or below this order of magnitude.

3.3. *High order flame sheet model*

More generally, we can now deduce the corresponding higher order jump conditions that arise from the full one step Arrhenius model (1) in terms of the dimensionless absolute temperature T of equations (1) or (3). To do this we must first invert the scaling that defined T in section 3.2 by rescaling such that $\mathsf{T} = 1+(T-T_*)/Q$. As before, we now set the Damköhler number to $D = \delta^2\theta^2 Q^2 \big/ (2\mathrm{Le}T_*^4)$, or equivalently $D = \delta^2\frac{1}{2}\beta^2/\mathrm{Le}$, which reintroduces its parameterization by δ; this is equivalent to rescaling the time and space variables of the previous section such that $t = \delta^2 t'$, $\mathbf{x} = \delta\mathbf{x}'$ and $\mathbf{v} = \mathbf{v}'/\delta$. After dropping the primes and using the result (6) to compensate for the linearization of the Arrhenius exponent in (7), we then arrive at the higher order flame sheet representation of the original model (1)

$$\begin{aligned}&\left.\begin{aligned}F_t + \mathbf{v}\cdot\nabla F &= \frac{\nabla^2 F}{\mathrm{Le}}\\ T_t + \mathbf{v}\cdot\nabla T &= \nabla^2 T - \ell\end{aligned}\right\}\qquad \text{for }\ \mathbf{x}\notin\boldsymbol{\Gamma}\\ &\left.\begin{aligned}&[T] = [F] = F = 0\\ &\frac{[F_n]}{\mathrm{Le}} = -\frac{[T_n]}{Q} = \Omega\end{aligned}\right\}\qquad \text{at }\ \mathbf{x}\in\boldsymbol{\Gamma}\\ &\Omega = \delta\exp\left(\frac{\theta}{2}\frac{T-T_*}{T_*T}\right)\frac{T^2/T_*^2}{\sqrt{1+6T/\theta}}\\ &\qquad + \frac{1+\varpi-\mathrm{Le}}{Q}V\frac{T^2}{\theta} + \frac{\varpi}{Q}\left(\tfrac{1}{2}T_n^{-} + 2\kappa\frac{T^2}{\theta}\right) + \mathrm{O}\big(\theta^{-2}, T_n^{-2}\big)\end{aligned}\qquad (14)$$

in which the propagation speed of the sheet relative to the flow is $V = (\partial_t \boldsymbol{\Gamma} - \mathbf{v}) \cdot \hat{\mathbf{n}}$, where $\hat{\mathbf{n}}$ is a unit normal to $\boldsymbol{\Gamma}$ pointing towards increasing values of the reactant F. The mean curvature can be defined as $\kappa = \frac{1}{2}\nabla \cdot \hat{\boldsymbol{n}}$, being positive if $\boldsymbol{\Gamma}$ is convex when viewed from the unburnt gas. The constant ϖ is defined by an asymptotic relation in equations (12) for large f, having the integral expression and numerical value

$$\varpi = \int_0^\infty (1 - \sqrt{1 - (1{+}z)e^{-z}}\,)\,\mathrm{d}z = 1.344\,045\,68\ldots$$

when calculated to nine significant figures.

The most significant generalization in this model from the model (3) is not the addition of terms that depend weakly on temperature, since these are dominated by the sensitivity of the chemistry to small changes in temperature at leading order, but the appearance of a relatively weak dependence of the jump conditions on propagation speed V, mean curvature κ and weak conduction into the burnt gas via T_n^-. Increasing the normal propagation speed V increases the jump in gradients, for Lewis numbers $\mathrm{Le} < 1 + \varpi \approx 2.344$; increasing the mean curvature κ increases the jump in gradients. A weak conductive heat loss into the medium behind the flame, for which $T_n^- > 0$, also causes a weak increase in the jump in gradients. These are all weak effects, being of the order of θ^{-1} if $T_n^- = \mathrm{O}(\theta^{-1})$. Increasing the temperature T at the flame sheet has a considerably stronger effect. Temperature changes would typically have to be very small, of the order of θ^{-2}, in order for these additional dependences to have an effect that is similar in magnitude to the effect of changing temperature.

In most practical situations therefore, when the activation temperature is large enough, this extension of the flame sheet conditions to higher orders is unlikely to do more than add minor corrections to the simpler leading order model. There may be circumstances, however, in which temperature changes are genuinely very small and thus the higher order effects may be important. At any rate, for large but finite activation temperatures, they offer asymptotic estimates of the corrections to any leading order results.

4. Finite activation temperature in flame balls

In situations where the dimensionless activation temperature is finite in value, even though it may be reasonably large, the expression for Ω in (14) provides a means of estimating the errors that would arise through adopting only the leading order temperature dependence of the jump conditions at a flame sheet. It is informative to examine these higher order effects in the context of a model for non-adiabatic, spherically symmetric flame balls (as studied in [7–10], for example).

In [10], numerical solutions found using an Arrhenius chemical model with linearized exponent, of the type used in the model (7), were compared with asymptotic solutions based on the corresponding leading order jump conditions. It was found that inordinately large Zeldovich numbers, of the order of $\beta = 100$, were needed to obtain reasonably good comparisons. It is generally thought that realistic values of the Zeldovich number are of the order of ten, or at most twenty, and that these are adequate to justify the use of leading order asymptotics. The numerical comparisons in [10] indicate that this assumption may be far from adequate for non-adiabatic flame balls, modelled using one step chemistry with radiative heat losses from the hot burnt gases. It would appear that higher order effects could play a major role in these calculations at moderately large values of β.

In a steady adiabatic flame-ball [2], the increase in the temperature T above ambient is Q/Le. Thus, if we adopt the scaling $T = T_* + Q(\mathcal{T} - 1)/\mathrm{Le}$, set $T_* = T_\mathrm{a} + Q/\mathrm{Le}$ and select $\delta = 1/\mathrm{Le}$, thereby fixing the scalings of the time and space variables, a suitable model [10] takes the form

$$\left.\begin{aligned}\mathrm{Le}F_t &= F_{rr} + 2r^{-1}F_r\\ T_t &= T_{rr} + 2r^{-1}T_r - \beta^{-1}b\,\mathrm{H}(R-r)\end{aligned}\right\}\quad \text{for } r \neq R$$
$$[T] = [F] = F = 0, \qquad [F_r] = -[T_r] = \Omega \quad \text{at } r = R^{\pm}$$
$$\Omega = e^{\beta(T-1)/2} + \frac{(1+\varpi - \mathrm{Le})}{\beta}R_t + \tfrac{1}{2}\varpi\, T_r^{-} + 2\frac{\varpi}{\beta R} + \mathrm{O}(\beta^{-2}, T_r^{-2}) \tag{15}$$
$$|F| < \infty, \quad |T| < \infty, \qquad \lim_{r\to\infty} F = 1, \quad \lim_{r\to\infty} T = 0$$

in which the Zeldovich number is now defined as $\beta = Q\theta/(\mathrm{Le}T_*^2) \gg 1$, still having the same expression as (8) in terms of appropriate dimensional quantities. The variable r now represents a radial coordinate in a spherically symmetric structure, taking $F = F(t, r)$ and $T = T(t, r)$, with $r \geqslant 0$. A reaction sheet is taken to exist at the spherical surface, $r = R(t)$, leading to the jump conditions shown in (15), in which it is simplest, and sufficient to illustrate the role played by higher order terms, to consider only the linearized Arrhenius exponent as obtained in (13). This also corresponds to the problem studied numerically in [10]. For a sphere the mean curvature is $\kappa = 1/R$.

The problem is non-adiabatic because the parameter b represents a constant rate of heat loss, that is limited to the burnt gases where $r < R$ by the Heaviside function $\mathrm{H}(R-r)$. Other forms of heat loss are examined in [7–10]. Our purpose here is to illustrate the effect of higher order jump conditions rather than to study flame-balls more comprehensively, so that we shall examine only this simple model for heat loss. Taking F and T to be bounded ensures that there is no source of reactant or heat at the origin, while the conditions at infinity fix the nondimensionalization such that $T(t, \infty) = T_{\mathrm{a}}$, with F scaled against the value found for the reactant mass-fraction at very large distances from the flame ball.

Details of the model and its physical origins can be found in [7–10], where it is studied in the asymptotic limit $\beta \to \infty$ using only the leading order expression $\Omega = e^{\beta(T-1)/2}$ to determine the jumps in gradient at the flame sheet. The equivalent problem, in which an Arrhenius rate law with linearized exponent is used to model the chemistry, instead of jump conditions, is arrived at by adding $\frac{1}{2}\beta^2\omega$ to the right-hand side of the equation for temperature T and subtracting $\frac{1}{2}\beta^2\omega$ from the equation for reactant F, taking $\omega = Fe^{\beta(T-1)}$ as in equations (7).

Details of the numerical solution of the latter form of the problem as well as the solution of the problem using leading order jump conditions are available in [10]. Suffice it to say that numerical versions of the heat loss term and the flame-ball radius are taken to be

$$\beta^{-1}b\,\mathrm{H}(R-r) \approx \frac{b/\beta}{1 + (\beta Fr)^4} \qquad \text{and} \qquad R \approx \int_0^{\infty} \frac{\mathrm{d}r}{1 + (\beta Fr)^4} \tag{16}$$

respectively. When β is large, the quotient in these expressions acts like a switch on passing through the region of reaction, where $\beta Fr \approx 1$. It therefore approximates the Heaviside function $\mathrm{H}(R-r)$, which it approaches as $\beta \to \infty$, while also having the advantage, for numerical purposes, of being continuously differentiable. To avoid the cold boundary difficulty, the expression $\omega = Fe^{\beta(T-1)}$ for the reaction rate is modified to set ω to zero in the neighbourhood of $T \approx \beta^{-1}$, by redefining it as

$$\omega = \max\left\{F\left(e^{\beta(T-1)/3} - e^{1-\beta(2T+1)/3}\right)^3,\ 0\right\}. \tag{17}$$

This formula is also continuously differentiable, while giving $\omega \equiv 0$ for $T \leqslant \beta^{-1}$ and being transcendentally close to $Fe^{\beta(T-1)}$ when $T > \mathrm{O}(\beta^{-1})$, for large values of β. The precise details of the way in which the cold boundary difficulty is circumvented appear to have practically no effect on the results that will be presented later, provided ω remains virtually unchanged for $T > \mathrm{O}(\beta^{-1})$.

The numerical problem was solved in two ways. As described in [10], one method used the transformation $r = \beta \tan(x/\beta)$ so that only a finite domain $0 \leqslant x \leqslant \frac{\pi}{2}\beta$ needed discretizing. A standard finite difference approach, with Newton iteration and continuation was then found to produce converged results, with 8000 evenly spaced grid points in x, even when taking $\beta = 100$. The other method was based on using the standard software package COLSYS [11] which is well suited to solving this type of boundary value problem. When using a large enough domain in r, this package was found to produce solutions that were numerically indistinguishable from the solutions obtained using the method already outlined and reported in [10].

Steady flame balls, for which $\partial_t \equiv 0$, are described by the solution of (15)

$$F = \begin{cases} 1 - \dfrac{R}{r} & : r \geqslant R \\ 0 & : r \leqslant R \end{cases} \qquad T = \begin{cases} \dfrac{\bar{T}R}{r} & : r \geqslant R \\ \bar{T} + b\dfrac{(r^2 - R^2)}{6\beta} & : r \leqslant R \end{cases} \tag{18}$$

which is accurate to order β^{-1}. The condition $[F_r] = -[T_r]$ then provides an expression for the temperature at the flame sheet, $\bar{T} = 1 - \frac{1}{3}bR^2/\beta + \mathrm{O}(\beta^{-2})$, using which, the condition $[F_r] = \Omega$ provides an expression relating the steady radius R and the heat-loss parameter b, which gives, when written to order β^{-1}

$$R\mathrm{e}^{-bR^2/6} = 1 - \frac{\varpi}{\beta}\left(2 + \tfrac{1}{6}bR^2\right) \tag{19}$$

or $R = (1 - \varpi(2 + \nu)/\beta)\,\mathrm{e}^{\nu}$ with $b = 6\nu/R^2$ when parameterized in terms of $\nu = bR^2/6$. The resulting curves relating R and b for various values of β are presented in figure 1.

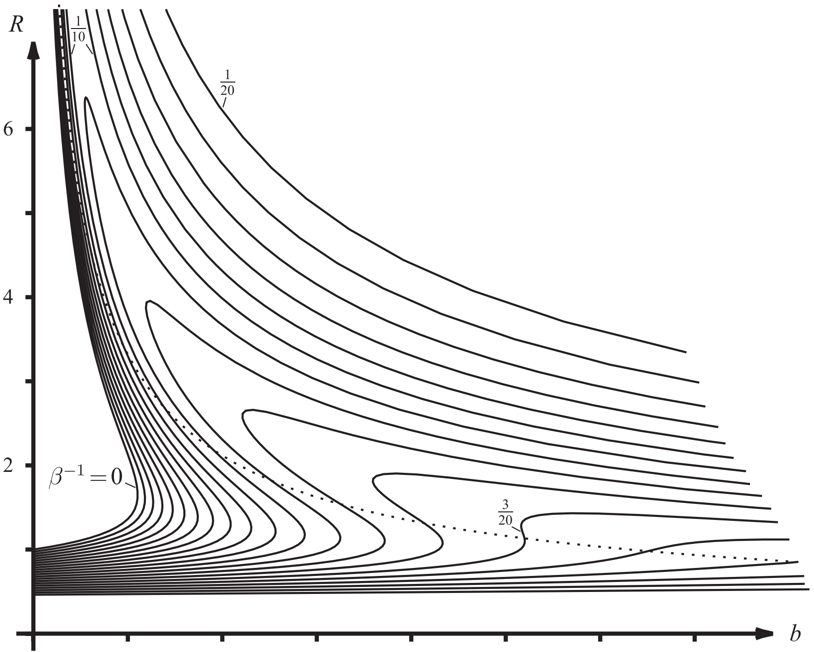

Figure 1. Variation of the steady flame-ball radius R with heat loss b at various Zeldovich numbers β, according to the higher order asymptotic formula (19). The dotted curve marks the path on which $bR^2 = \beta$.

If no account had been taken of the higher order terms, we would have found $R = e^{bR^2/6}$, the same result as is given by (19) in the limit as $\beta \to \infty$. As described by the curve marked $\beta^{-1} = 0$ in figure 1, this provides a value of $R = 1$ when $b = 0$, with values of R increasing with b until a turning point is reached at $(b, R) = (3/e, e^{1/2})$, after which R increases without bound as b decreases towards zero.

For large but finite values of β, the deviation of the flame-ball radius R, from this behaviour as $\beta \to \infty$, becomes of order one when $bR^2 = \mathrm{O}(\beta)$. The branch of solutions for which R is large is therefore particularly strongly affected by the higher order terms. In fact, as seen in figure 1, the formula (19) provides a second turning point, for large enough values of β, after which values of b increase again and R, ultimately, decreases as b increases. For large enough values of β, the solution therefore appears to predict three solutions over some interval of values of the heat loss parameter b. For values of β below about seven, the two turning points merge and there is then only one value of R for any value of b.

However, the asymptotic assumptions leading to equation (19) are not valid when $bR^2 = \mathrm{O}(\beta)$ and so, at this stage, the higher order behaviour seen in figure 1 cannot be expected to be predicted reliably. The dotted line in the figure corresponds to the path where $bR^2 = \beta$ and as bR^2 increases towards this path, as well as for larger values of R and b, significant differences are likely to arise between an accurate numerical simulation and the higher order asymptotic prediction in (19). Relevant numerical solutions are shown in figure 2 for values of β between 20 and 36 and for $\beta = 100$. The numerical results at $\beta = 36$ can be

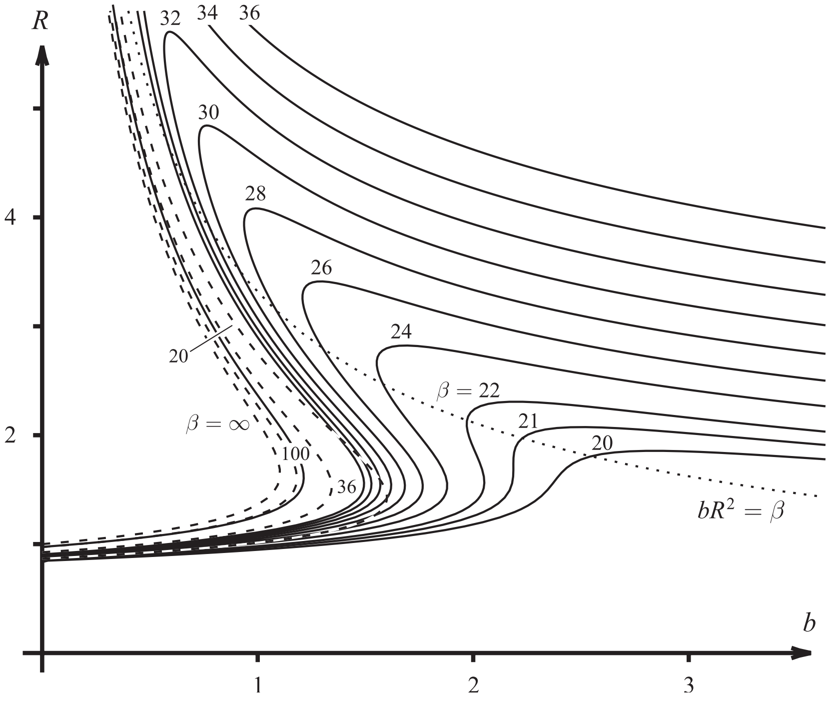

Figure 2. Variation with the heat loss parameter b of the numerically calculated steady flame-ball radius R, using Arrhenius kinetics with a linearized temperature exponent to model the chemistry, for Zeldovich numbers ranging from $\beta = 20$ to 36 and for $\beta = 100$. The dashed curves show the variation predicted by the higher order asymptotic formula (19), for selected values of β, and the dotted curve marks the path, on any asymptotic result, at which $bR^2 = \beta$.

seen to deviate fairly significantly from the higher order asymptotic result, particularly at larger values of R, while the results for $\beta = 20$ deviate so much that the turning point is already lost altogether.

These large differences all appear where $bR^2 = \mathrm{O}(\beta)$ in the asymptotic solution (19); values of T_r^- are then no longer very small making the higher order asymptotic predictions unreliable. Nevertheless, the overall qualitative trend, albeit predicted by assuming that the values of T_r^- are small, is certainly correct and the higher order terms successfully predict their own ultimate downfall. The main physical reason for this seems to be because increased values of T_r^-, brought about by increasing the heat-loss term b, actually strengthen the reaction in the reaction sheet. The greater effect found in the numerical solutions arises because the asymptotic solutions underpredict the effect of T_r^- when it is no longer very small. Both the leading order and the higher order asymptotics are no longer reliable at this stage.

This example is most significant in showing that, for flame balls modelled using one step chemistry, qualitatively and quantitatively important deviations from solutions based on the usual leading order description of jump conditions at a flame sheet can arise, even for what are generally considered to be relatively large values of the Zeldovich number. The numerical results indicate that as β decreases below about twenty one, the intermediate branch of solutions (part of which may be stable under some circumstances [7–10]) disappears altogether. Values of about ten, which are often thought to be adequate because of the exponential sensitivity of reaction-rates to changes in temperature, appear to be far too low for the leading order asymptotic description to apply.

5. Conclusions

The higher order jump conditions (14) provide a generalization from the usual leading order jump conditions (3) assumed at a reaction sheet in combustion theory. As well as allowing for more accurate asymptotic predictions, they provide at least one means of testing the validity of leading order solutions and of predicting where and when they are likely to change significantly.

In the case of flame balls with constant heat loss in the burnt gases [7–10], modelled as in (16) and (17) for finite values of the Zeldovich number β, numerical calculations have been seen to differ quite dramatically from the leading order asymptotic results at Zeldovich numbers that are still moderately large. The higher order asymptotic results prove very useful in examining the onset of and reasons for this deviation. They reveal that the main factor is the role of heat conduction from the flame sheet into the burnt gases, as represented by the temperature gradient immediately on the burnt side of the flame sheet. Increasing this gradient, for example through increasing the rate of radiative heat loss from the burnt gas or through increasing the size of the flame ball (which increases the volume from which heat is radiated), also increases the overall strength of the reaction in the flame sheet, with numerical solutions predicting a stronger effect than the asymptotic analysis when the temperature gradient into the burnt gas is no longer very small.

This finding raises some interesting questions about the modelling of flame balls and the current state of the art in describing flame balls and other types of laminar flame, using one step chemistry. As well as providing an asymptotic extension, to higher order, that is interesting in its own right, the formula (14) provides one tool for testing and addressing these questions.

In the case of flame balls, modelled using one step chemistry, the most obvious question is: ‘how large can the Zeldovich number realistically be?’ It is often assumed that the exponential in the Arrhenius rate law exaggerates the thermal sensitivity of the reaction rate so much that the Zeldovich number does not need to be much greater than about ten to mimic the results of leading order activation temperature asymptotics, at least qualitatively. Fixed values of β between about eight and sixteen are often adopted in numerical simulations of laminar flames.

One approach that can be used to estimate an appropriate value for the Zeldovich number is to compare the rate at which the speed of a planar flame changes with adiabatic flame temperature (obtained using calculations based on more detailed chemical models of hydrocarbon flames) with the same rate as predicted by leading order asymptotics for one step chemistry. This often leads to 'effective' one step Zeldovich numbers of about ten to fifteen, except for flames in mixtures that are near the limit of flammability. In such mixtures, very large effective Zeldovich numbers have been predicted (see, e.g. [12]).

Although the same approach cannot be applied directly to flame-balls, the experimental evidence does suggest that stable flame balls require low Lewis numbers of a lean reactant at concentrations that are below the flammability limit for planar laminar flames. This is at least consistent with the idea that an equivalent (or effective) Zeldovich number could be unusually large. Under such circumstances the predictions of leading order one step asymptotics do seem to be reasonable, even if the requisite Zeldovich numbers might have to be significantly greater than about twenty for the underlying Arrhenius kinetic model to offer qualitatively similar predictions.

In fact recent studies offer more insight into the likely size of an effective one step Zeldovich number for flame balls. It has been found [13] that the more general chain-branching and recombination model for chemistry [2, 4]

$$\begin{array}{lcl} \mathrm{F}+\mathrm{Z}\rightarrow 2\mathrm{Z} & : & k_{\mathrm{B}} = A_{\mathrm{B}}e^{-T_{\mathrm{B}}/T} \\ \mathrm{Z}+\mathrm{M}\rightarrow \mathrm{P}+\mathrm{M} & : & k_{\mathrm{C}} = A_{\mathrm{C}} \end{array} \tag{20}$$

in which Z represents an energetic intermediate species, also provides a useful and qualitatively accurate asymptotic description for stable flame balls, as the activation temperature of the branching reaction T_{B} tends to infinity. Moreover, it is also found [13, 14] that these asymptotic results agree very well with numerical results, even when the corresponding Zeldovich number of the branching reaction β_{B} is as low as about five.

Furthermore, in cases where $\beta_{\mathrm{B}} \rightarrow \infty$ and where the concentration of the intermediate species Z is found to be significant only in a relatively narrow part of an overall flame structure (i.e. for relatively large flame balls), an inner asymptotic analysis can be used to eliminate the intermediate species from the problem [14]. The outer asymptotic model that remains, for the dimensionless fuel concentration F and the absolute temperature T, is then almost identical to the one step model in (14), with the sole difference that Ω (i.e. the value of $[F_n]/\mathrm{Le}$ and $-[T_n]/Q$) is then given, very simply, by

$$\Omega = \mu(T - T_{\mathrm{c}}) \qquad \text{with} \qquad T > T_{\mathrm{c}} \tag{21}$$

where μ is a constant related to the non-dimensionalization and T_{c} represents a form of dimensionless 'crossover temperature' [4, 13, 14]. This formula for Ω can be compared and contrasted with its leading order one step equivalent

$$\Omega = \delta e^{\beta(T-T_*)/2}.$$

In both cases, the temperature T is evaluated at the flame sheet.

For any flame structure to be the same (when calculated using both of these expressions for Ω) and for the structure to have the same linear stability properties, all that is needed is that, at the temperature found at the flame sheet, both of the relations

$$\mu(T - T_{\mathrm{c}}) = \delta e^{\beta(T-T_*)/2} \qquad \text{and} \qquad \mu\frac{\mathrm{d}}{\mathrm{d}T}(T - T_{\mathrm{c}}) = \delta\frac{\mathrm{d}}{\mathrm{d}T}e^{\beta(T-T_*)/2}$$

must be satisfied, which easily leads to the formula for an effective Zeldovich number of a one step reaction

$$\beta = \frac{2}{T - T_\mathrm{c}} \qquad \text{with} \qquad T > T_\mathrm{c} \tag{22}$$

where T is evaluated at the flame sheet. Thus, in cases where the flame sheet temperature is arbitrarily close to the crossover temperature T_c, the chain branching and recombination kinetic model (20) predicts an arbitrarily large effective one step Zeldovich number. This tends to confirm the argument that one step asymptotic studies for flame balls, or any flames near the limit of flammability, can indeed, quite realistically, assume very large values for β.

However, a little caution should be exercised because the effective Zeldovich number is not then constant during any unsteady evolution in which the temperature at the flame sheet changes, or from any one steady solution to another as, for example, changes in a heat loss parameter or in the composition of the mixture alter the temperature. Also, some difference in the details of the expression (22) for β can be anticipated at finite values of β_B although the general character of the formula should remain. Another limitation might be that the formula (21) should, more strictly, be compared with the higher order formula for Ω given in equations (14). Alternatively, as this article has helped to demonstrate, it would seem that the effective value of β needs to be at least greater than about 25 to describe stable spherical flame balls modelled with one step chemistry and heat loss from the burnt gas.

Finally, while demonstrating that arbitrarily large effective one step Zeldovich numbers *can* arise, which helps to reinforce all of the flame ball literature that is based on one step kinetics, the arguments that lead to (22) do not show that all stable flame balls sustained by chain branching and recombination kinetics *must* have a large effective one step Zeldovich number. On the contrary, it has been found that the model (20) can lead to stable asymptotic solutions, as $\beta_\mathrm{B} \to \infty$, for combinations of heat loss and the Lewis numbers of F and Z, in which the conditions that give rise to the formula (21) are simply not satisfied [13]. Indeed, even when they do seem to be satisfied, stable solutions for flame balls with branching kinetics can be found in which an effective one step Zeldovich number would appear to be noticeably below about twenty [13–15],

The one step kinetic model for the chemistry of laminar flames, along with the use of leading order asymptotic analysis based on a large activation temperature, has a distinguished record in the theory of combustion. However, there is always room for closer scrutiny of any model. The higher order asymptotic formulae for the jump conditions at a reaction sheet, that have been derived in this article, provide one means of examining flames, with one step kinetics, beyond only leading order asymptotics. They should also prove valuable for studying flames in many other contexts beside the particular example of a stationary spherical flame ball, that has been examined here.

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References

[1] Buckmaster J D and Ludford G S S 1982 *Theory of Laminar Flames* (Cambridge: Cambridge University Press)

[2] Zeldovich Ya B, Barrenblatt G I, Librovich V B and Makhviladze G M 1985 *The Mathematical Theory of Combustion and Explosions* (New York: Consultants Bureau)

[3] Williams F A 1985 *Combustion Theory* (California: Benjamin/Cummings)

[4] Liñán A and Williams F A 1993 *Fundamental Aspects of Combustion* (New York: Oxford University Press)

[5] Law C K, Chao B H and Umemura A 1992 On closure in activation energy asymptotics of premixed flames *Combust. Sci. Technol.* **88** 59–88

[6] Sivashinsky G I 1983 Instabilities, pattern formation and turbulence in flames *Ann. Rev. Fluid Mech.* **15** 179–99

[7] Joulin G 1985 Point source initiation of lean spherical flames of light reactants: an asymptotic study *Combust. Sci. Technol.* **43** 99

[8] Buckmaster J D, Joulin G and Ronney P 1990 The structure and stability of nonadiabatic flame balls *Combust. Flame* **79** 381–92

[9] Buckmaster J D, Joulin G and Ronney P 1991 The structure and stability of nonadiabatic flame balls: the effects of far-field losses *Combust. Flame* **84** 411–22

[10] Shah A A, Thatcher R W and Dold J W 2000 Stability of a spherical flame ball in a porous medium *Combust. Theory Modelling* **4** 511–34

[11] Ascher U and Petzold L 1991 Projected implicit Runge–Kutta methods for differential–algebraic equations *SIAM J. Num. Anal.* **28** 1097–120

[12] Peters N 1997 Kinetic foundation of thermal flame theory *Prog. Astronautics Aeronautics* **173** 73–91

[13] Dold J W, Weber R O, Thatcher R W and Shah A A 2002 Flame ball with thermally sensitive intermediate kinetics *Combust. Theory Modelling* submitted

[14] Dold J W, Thatcher R W, Omon-Arancibia A and Redman J 2003 From one step to chain branching premixed flame asymptotics *Proc. Combustion Institute* **29** 1519–26

[15] Redman J 2002 Modelling premixed flames with chain-branching chemistry *Undergraduate MMath Project Report* Mathematics Department, UMIST