On Closure of Activation–Energy Asymptotics in the Premixed–Flame Regime

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The present analysis deals with the controversy, surrounding the premixed–flame regime of activation-energy aymptotics, which was raised by Law et al. claiming that an additional constraint is required to uniquely determine the flame structure. The activation-energy asymptotics procedure which employs the distinguished limit of the premixed–flame regime is derived step by step to point out the origin of the controversy. It is shown by directly comparing the flame structures obtained by different closure assumptions that the premixed–flame region indeed leads to assign solution to the flame structure and no extra physical constraint is necessary to close the problem.

Key Words : Activation–Energy Asymptotics, Premixed–Flame Regime, Closure

1. Introduction

The nature of high nonlinearities in chemical reaction rates is the origin of difficulties in analyzing the flame structures. The high nonlinearities require not only larger computational and experimental resources, but also more sophisticated techniques to utilize those resources. As a simplified model chemistry to the complicated chemical kinetic mechanisms, global reaction steps, employing the Arrhenius–type reaction rates with large activation energies, have often been found to be useful, particularly in describing the phenomena arising from interactions of the transport mechanisms with the chemical reactions, such as extinction, ignition and stability problems. By adopting the Zel'dovich number, which is a measure of activation energy compared with thermal energy and often is denoted by β, as a large expansion parameter, the thin reaction zone, in which the high nonlinearity is dominant, is integrated in a stretched coordinate system to give an accurate estimate of the overall reaction rate. This mathematical procedure is known as activation –energy asymptotics (AEA).

The fundamental concept of AEA goes to back to the earlier works of Zel'dovich, Frank–Kamenetskii and Semenov (1980) in description of the problems stemming from thermal explosion, flame propagation and so on. However, it was only after publication of the work by Liñá (1974) that AEA became a popular tool for flame analyses. Among several distinguished limits, appearing in his diffusion flame analyses, is the premixed flame regime, in which concentration of a deficient reactant (rate-controlling species) in the reaction zone is of order β^{-1} and concentrations of abundant reactants are of order unity. Because of the similarity of the premixed–flame-regime structure to the premixed–flame structures, the premixed–flame regime was widely adopted to analyze both diffusion flames and premixed flames.

However, the recent publication by Law, Chao and Umemura (1993) questioned that the premixed–flame regime did not yield a unique solution to the flame structure, and claimed that an additional physical constraint, that was equivalent to position the reaction sheet at the center of reaction by forcing the first-order moment of the reaction rate to vanish, was required to properly

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close the problem. This is so called the closure controversy. After heated debates among the asymptoticians, a short erratum was published admitting that the Liñán's approach to the premixed-flame regime of AEA was indeed a correct method and that an additional constraint is unnecessary to close the problem. However, the essence of the debates was not known to public, who may be unaware of the very existence of the closure controversy. Therefore, the danger of getting misconception that the premixed-flame regime analysis is incomplete is still present.

It is the purpose of this paper to clarify that, irrespective of the closure scheme, the premixed-flame regime of AEA provides a unique solution to the flame structure. In order to point out the origin of the closure controversy, the AEA procedure employing the distinguished limit of the premixed-flame regime will be derived step by step. It will be shown by directly comparing the flame structures obtained by different closure assumptions that the premixed-flame regime indeed leads to a unique solution to the flame structure.

2. Conservation Equations

In order to simplify our presentation, the conservation equation, which was previously employed by Liñán (1974) in his original paper, is adopted as the starting equation. Then the conservation equation for the temperature field is

$$\frac{d^2 T}{dz^2} + z \frac{dT}{dz} = -Da\, Y_O Y_F \exp(-T_a/T) \quad (1)$$

with the boundary conditions

$$T = T_\infty \text{ as } z \to \pm\infty \quad (2)$$

The boundary temperatures as $z \to \pm\infty$ are set to be equal in order to minimize the parameters appearing in the analysis while not losing all the essences of the presentation. Here T is the temperature nondimensionalized by the characteristic temperature $Q/c_p Y_{F-\infty}$, obtained from the heat release per unit mass of fuel Q, the specific heat c_p, and the fuel mass fraction $Y_{F-\infty}$ in the fuel stream. The distance normal to the mixing layer z, measured from the stagnation plane, has been made nondimensional with the characteristic mixing length $\sqrt{D/a}$, where D is the diffusion coefficient, that is assumed to identical to all species (including thermal energy) present in the flame, and a is the rate of strain exerted on the thin mixing layer adjacent to the stagnation place. The fuel stream approaches from $z = -\infty$, and the oxidizer stream from $z = \infty$ where the oxidizer mass fraction is $Y_{O\infty}$. The Damköhler number $Da = B\nu Y_{F-\infty}/a$ is obtained from the frequency factor B and the stoichiometric mass ratio ν of oxidizer to fuel. T_a is the nondimensional activation energy. The nondimensional fuel and oxidizer mass fractions Y_F and Y_O are obtained by division by $Y_{F-\infty}$ and $\nu Y_{F-\infty}$ respectively.

The analysis can be further simplified by introducing a coordinate transformation into the mixture fraction coordinate given by

$$x = \frac{1}{2}\text{erfc}(z/\sqrt{2}) \quad (3)$$

Then, Eq. (1) becomes

$$\frac{d^2 T}{dx^2} = -2\pi \exp(z^2)\, Da\, Y_O Y_F \exp(-T_a/T) \quad (4)$$

Since all the diffusion coefficients are equal, *i.e.* Lewis numbers are unity, the coupling relations exist as

$$\begin{aligned} Y_O + T &= a(1-x) + T_\infty \\ Y_F + T &= x + T_\infty \end{aligned} \quad (5)$$

Once a profile of T is given, the corresponding profiles of Y_O and Y_F can be found from the above coupling relations. Since the reaction term does not appear in the conservation equations of the coupling functions $T + Y_F$ and $T + Y_O$, the above coupling functions are valid even in the reaction zone.

The boundary conditions are rewritten to be

$$T = T_\infty \text{ at } x = 0, \ 1 \quad (6)$$

The nondimensional fuel and oxidizer concentration are, 1 and 0 at $x = 1$, and 0 and a at $x = 0$. Since the fuel is assumed to be abundant compared with the oxidizer, $a \ll 1$. Because of the symmetry of flame structures with unity reaction order, we only need to consider the case of $a \ll 1$.

3. Flame Structure Analysis

In the premixed-flame regime, two transport zones are separated by a thin reaction zone. Then the leading-order reaction-sheet solution arises from the double limit of infinitely large Damköhler number and Zel'dovich number. In this limit, the reaction zone is effectively approximated by a reaction sheet. The reaction-sheet, through which the fuel leaks by an amount of order unity, separates the frozen region in the oxidizer side and the equilibrium region in the fuel side.

The effect of finite-rate chemistry, *i.e.* finite Damköhler number, is taken into account by using AEA in which the Zel'dovich number is now assumed to be a finite large number. For each location of the reaction sheet, the corresponding Damköhler number can be found from matching of the outer transport-zone solution to the inner reaction-zone solution. If the physico-chemical parameters, such as B, T_a, Q and others, are known, the strain-rate parameter a can be obtained from the definition of the Damköhler number.

3.1 Reaction-sheet solution

In the limit of infinitely large activation temperature, the reaction zone becomes an infinitely thin reaction sheet, located at $x = x_p$, which will be posteriorly determined by the value (large value) of the Damköhler number. The temperature profile, corresponding to the reaction-sheet location x_p is given by

$$T^- = T_\infty + (T_p - T_\infty)\frac{x}{x_p}, \qquad 0 < x < x_p$$
$$T^+ = T_\infty + (T_p - T_\infty)\frac{1-x}{1-x_p}, \quad x_p < x < 1 \tag{7}$$

with the reaction-sheet temperature

$$T_p = T_\infty + a(1 - x_p) \tag{8}$$

where the superscripts + and - denote the fuel side and oxidizer side of the flame, respectively.

In addition, the coupling relations yield the oxidizer and fuel profiles

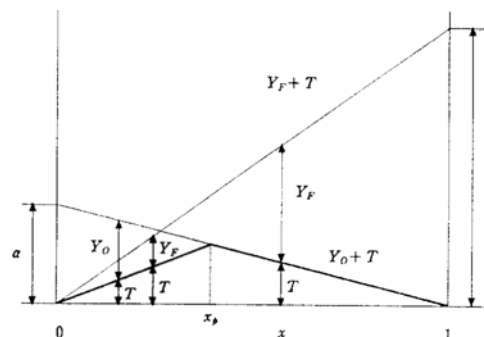$$Y_o^- = a\left(1 - \frac{x}{x_p}\right), \quad Y_o^+ = 0$$

Fig. 1 Asymptotic temperature and concentration distributions for premixed-flame regime.

$$Y_F^- = Y_{Fp}\frac{x}{x_p},$$
$$Y_F^+ = 1 + (Y_{Fp} - 1)\frac{1-x}{1-x_p} \tag{9}$$

where the fuel concentration Y_{Fp} is obtained from the coupling relation in the second of Eq. (5) to be

$$Y_{Fp} = (1 + a)x_p - a \tag{10}$$

The schematic diagram of the leading order flame structure is shown in Fig. 1, where the fuel and oxidizer concentrations are found from the distances of the coupling functions from the temperature profile.

3.2 Activation energy asymptotics

In order to obtain the single valued relation $Da = Da(x_p ; \beta)$ for a finite value of β, the first-order perturbations for the temperature and reactant concentrations are considered in the outer transport zones and in the inner reaction zone. Defining the Zel'dovich number to be

$$\beta = \frac{T_a}{T_p^2} \tag{11}$$

the perturbed temperature profile in the outer transport zone is given by

$$T^- = T_\infty + (T_p - T_\infty)\frac{x}{x_p} - \beta^{-1}\delta^-\left(\frac{x}{x_p}\right)$$
$$T^+ = T_\infty + (T_p - T_\infty)\frac{1-x}{1-x_p} - \beta^{-1}\delta^+\left(\frac{1-x}{1-x_p}\right) \tag{12}$$

where the upstream (oxidizer side) and downstream (fuel side) displacements δ^- and δ^+ are yet to be determined from matching with the

solution to the inner reaction–zone analysis.

On the other hand, temperature in the inner reaction zone is also expanded from the value of the reaction-sheet solution as

$$T^{in} = T_p - \beta^{-1}(\theta + m\xi) \qquad (13)$$

where the stretched coordinate is defined as

$$\xi = A\beta(x - x_p) \qquad (14)$$

Here the nondimensional heat loss parameter m, which measures the ratio of the downstream heat loss to the chemical energy influx, and the scale factor A are given by

$$m = x_p, \quad A = a/x_p \qquad (15)$$

The coupling relations yield the inner expansions of the fuel and oxidizer concentrations in the form,

$$Y_O = \beta^{-1}\theta + \cdots, \quad Y_F = Y_{Fp} + \cdots \qquad (16)$$

Then the resulting conservation equation for the inner layer becomes

$$\frac{d^2\theta}{d\xi^2} = A\theta e^{-(\theta + m\xi)}$$
$$\theta \rightarrow \delta^+ \qquad \text{as } \xi \rightarrow \infty \qquad (17)$$
$$\theta + \xi \rightarrow \delta^- \text{ as } \xi \rightarrow -\infty$$

where the boundary conditions are obtained by matching with solution to the outer transport zone, given in Eq. (12), and the reduced Damköhler number Λ is defined to be

$$\Lambda = 2\pi \exp(z_p^2) \frac{T_p^4}{T_a^2} \frac{x_p^2}{a^2} Da\, Y_{Fp}\, e^{-T_a/T_p} \qquad (18)$$

Three unknown parameters, namely Λ, δ^- and δ^+, exist in the above equation, while there are only two boundary conditions. Therefore, the solution can be found only after specifying a value for one of the three parameters, most conveniently δ^-. Then the corresponding value for Λ can be calculated by numerical integration, thereby resulting in the value of Da by using Eq. (18). For many people, this prespecifying procedure is indeed understood as a closure procedure. In the previous practices, Liñán (1974) set the upstream displacement δ^- to be identically zero, while Joulin and Clavin (1979) assumed that $\delta^- = \delta^+$.

As an alternative solution method, which is much simpler than the above formulation, the inner stretched coordinate is translated by an amount of λ in the form

$$\zeta = \xi + \lambda \qquad (19)$$

with a constraint that

$$\Lambda e^{m(\theta + \zeta) - \infty} e^{-m\delta^-} = \frac{1}{2} \qquad (20)$$

Then the inner governing equation recovers the Liñán's canonical form (1974)

$$2\frac{d^2\theta}{d\zeta^2} = \theta e^{-(\theta + m\zeta)}$$
$$\theta_\zeta \rightarrow 0 \qquad \text{as } \zeta \rightarrow \infty \qquad (21)$$
$$\theta_\zeta \rightarrow -1 \text{ as } \zeta \rightarrow -\infty$$

This problem now has a unique solution for a given value of m, which is already prescribed in the reaction-sheet solution. Then the values of θ (∞) and $(\theta + \zeta)$ $(-\infty)$ are obtained as parts of the solution. Variations of $\theta(\infty)$ and $(\theta + \zeta)$ $(-\infty)$ with m are already given by Liñán and the functional approximations are also given in his paper (1974).

Revisiting the matching conditions in Eq. (17), we find

$$\theta_\infty = \delta^+$$
$$\lambda = (\theta + \zeta)_{-\infty} - \delta^- \qquad (22)$$

From Eq. (20), the Damköhler number is now given by

$$Da = \frac{\exp(-z_p^2)}{4\pi} \frac{T_a^2}{T_p^4} \frac{a^2}{x_p^2} Y_{Fp}^{-1} e^{T_a/T_p} e^{-m(\theta + \zeta) - \infty} e^{m\delta^-} \qquad (23)$$

As seen from the above equation, we may find different values of Da by choosing different values of the upstream displacement δ^-, that is, depending on the choice of closure schemes, the premixed flame regime analysis gives different values of Da. This nature appears to be the origin of the so-called closure controversy that an additional constraint needs to be introduced in order to avoid the nonuniqueness of Da for a given flame structure. However, it should be kept in mind that we should not confine ourselves to inner and outer solutions but rather concentrate on composite solution of the flame structure. It will be demonstrated in the next section that Eq. (23) in fact yields a unique value of the Damköhler number for the composite flame structures, that are uniformly identical throughout the flow
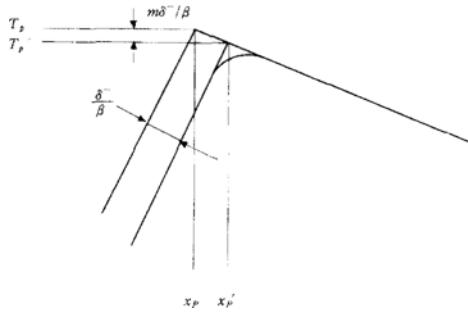
Fig. 2 Temperature profiles for two different representations of an identical flame structure.

field.

4. Criteria for Uniqueness

Since the Zel'dovich number is the large parameter of expansion, the temperature profiles, which are uniformly identical in order β^{-1} throughout the whole flame, must have an identical value of the Damköhler number in order unity.

In order to prove the above statement, let us consider two different representations of an identical flame structure, illustrated in Fig. 2. The first case corresponds to the flame configuration, such that the reaction–sheet is located at x_p with the upstream displacement set to be δ^-. Then the reaction–sheet temperature T_p, the outer temperature profiles and the heat loss parameter m are given by

$$T_p = T_\infty + \alpha(1 - x_p)$$

$$T^- = T_\infty + (T_p - T_\infty)\frac{x}{x_p} - \frac{\delta^-}{\beta}\left(\frac{x}{x_p}\right) + O(\beta^{-2})$$

(24)

$$T^+ = T_\infty + (T_p - T_\infty)\frac{1-x}{1-x_p} - \frac{\delta^+}{\beta}\left(\frac{1-x}{1-x_p}\right)$$
$$+ O(\beta^{-2})$$

$$m = x_p$$

The second case corresponds to the reaction–sheet located at x_p' at which the upstream perturbations are seen to vanish, i.e. zero upstream displacement, by choosing x_p' to be

$$x_p' = x_p\left(1 + \frac{\delta^-}{\beta\alpha}\right)$$

(25)

Then, T_p', the outer temperature profiles and the

heat loss parameter m' are respectively given by

$$T_p' = T_\infty + \alpha(1 - x_p) - \frac{x_p\delta^-}{\beta} = T_p - \frac{m\delta^-}{\beta}$$

$$T^- = T_\infty + (T_p' - T_\infty)\frac{x}{x_p'} + O(\beta^{-2})$$

$$= T_\infty + (T_p - T_\infty)\frac{x}{x_p} - \frac{\delta^-}{\beta}\left(\frac{x}{x_p}\right) + O(\beta^{-2})$$

(26)

$$T^+ = T_\infty + (T_p' - T_\infty)\frac{1-x}{1-x_p} - \frac{\delta^+}{\beta}\left(\frac{1-x}{1-x_p'}\right)$$
$$+ O(\beta^{-2})$$

$$= T_\infty + (T_p - T_\infty)\frac{1-x}{1-x_p} - \frac{\delta^+}{\beta}\left(\frac{1-x}{1-x_p}\right)$$
$$+ O(\beta^{-2})$$

$$m' = x_p' = x_p + \frac{x_p\delta^-}{\beta\alpha}$$

where the superscript $'$ denotes the second case.

Since m and m' differ only by an amount of $O(\beta^{-1})$, the inner reaction–zone structures are identical at $O(\beta^{-1})$. The outer structures are also identical at $O(\beta^{-1})$ as seen from Eq. (24) and (26). Therefore, the above two flame configurations are uniformly identical at $O(\beta^{-1})$ throughout the whole flame. Calculating the Damköhler numbers for each of the flame configurations,

$$Da = \frac{\exp(-z_p^2)}{4\pi}\frac{T_a^2}{T_p^4}\frac{\alpha^2}{x_p^2}Y_{Fp}^{-1}e^{Ta/Tp}e^{-m(\theta+\zeta)-\infty}e^{m\delta^-}$$

$$Da' = \frac{\exp(-z_p'^2)}{4\pi}\frac{T_a^2}{T_p'^4}\frac{\alpha^2}{x_p'^2}Y_{Fp}'^{-1}e^{Ta/Tp'}e^{-m'(\theta+\zeta)-\infty}$$

where the apparent differences between Da and Da' at order unity appear only from the factors $e^{Ta/Tp}e^{m\delta^-}$ and $e^{Ta/T'p}$. Since $T_p' = T_p - \beta^{-1}m\delta^-$, expansions of these factors up to order β^{-1} lead the leading–order ratio of Da to Da' to become

$$\frac{Da}{Da'} = \frac{e^{Ta/Tp}e^{m\delta^-}}{e^{Ta/Tp'}} = \frac{e^{Ta/Tp}e^{m\delta^+}}{e^{Ta/(Tp-m\delta^-/\beta)}} = 1 + O(\beta^{-1})$$

(28)

which clearly shows that the two Damköhler numbers are the same at order unity.

Therefore, the AEA adopting the premixed–flame regime yields a unique Damköhler number for flame configurations, which are uniformly identical throughout the whole flame. The so–called closure problem does not exist, and the additional constraint is by no means necessary to obtain the correct Damköhler number. If the leading-order reaction-sheet locations are set to

be equal, while employing different closure schemes, the resulting composite flame structures differ at order β^{-1}, so that different values of the Damköhler number at order unity must be obtained.

In view of Eq. (23), the calculation for Da is algebraically simplest if the upstream displacement is chosen to be zero, which was used in the original paper of Liñán (1974). The Damköhler number can be calculated without additional considerations for δ^-. In fact, Liñán recommended, though not necessary, to place the reaction -sheet at the location, at which extrapolation of the frozen oxidizer profile vanisheed. By unambiguously defining the reaction-sheet location in such a manner, the subsequent confusion on the resulting $Da - x_p$ curve may be avoided. However, it is worthy of note that different assumptions on the upstream displacement are able to shift the $Da - x_p$ curves by an amount of $O(\beta^{-1})$ in each direction.

5. Concluding Remarks

Employing a simplified flame model, the present analysis has shown that the premixed-flame regime of AEA yields a unique solution to the flame structure regardless of choice of the closure schemes. The presentation does not imply that the analysis by Law et al. is wrong. Their analysis is still valid in calculating the leading-order Damköhler number though a redundant constraint was introduced. However, the present analysis disprove the claim that their analysis is the only correct approach to the premixed-flame regime of AEA. Moreover, it should be noted that the accuracy of their analysis is limited to order β^{-1} because of introduction of the redundant constraint, and that their analysis cannot be carried out into the higher orders.

Related to the premixed-flame regime, the so -called fast-time instability of the inner layer equation in Eq. (21) is the rather outstanding

problem. The linear stability analysis by Peters (1978) showed that the inner equation became unstable if the heat loss parameter m became positive, i. e. the flame suffered a heat loss to the equilibrium side (the fuel side in this analysis). However, we often experimentally find premixed flame structures with downstream heat loss. It is believed that there exists a narrow region of positive m of order β^{-1}, in which the inner equation is stable. The analysis to find the critical value of m, at order of β^{-1}, at which the reaction-zone structure is marginally stable, is yet to be performed in order to bound the range of applicability of the premixed-regime adopted here. However, it is not still clear how the fast-time instability will affect the whole flame structure including the outer transport zone structures. Clarification of the problem will provide better use of the premixed-flame regime for the future flame structure analysis.

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