

Turbulent mean reaction rates in the limit of large activation energies

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Closed-form expressions for the turbulent mean reaction rate and its covariance with the temperature are derived for premixed and non-premixed combustion. The limit of large activation energies is exploited for a chemical reaction rate that, by virtue of coupling functions, depends on the mixture fraction and a non-equilibrium progress variable only. The probability density function (p.d.f.) formulation with an assumed shape of the p.d.f. is used; a beta-function distribution is assumed for the progress variable. The mean reaction rate is expressed in terms of the mean and the variance of the temperature and, for non-premixed combustion, of the mixture fraction. The reaction kinetics are represented by the non-dimensional activation energy and the laminar flame velocity. For non-premixed systems the possibility of local extinction by flame stretch is considered.

1. Introduction

Second-order closure models have received, in spite of their inherent uncertainties, a considerable acceptance in the field of non-reacting flows, but few calculations of reacting flows have yet been performed. One of the reasons for this is, certainly, that the interaction between fluid dynamics and chemistry remains poorly understood. The problem may be split into two parts: the effect of the reaction on the turbulence and that of the turbulence on the reaction. As far as the influence of the reaction-induced density changes upon the flow field is concerned, a considerable simplification of notation is achieved by the use of density-weighted averaging (Libby 1977). However, this does not solve the closure problem. The hypothesis that the turbulence modelling that has been developed for constant-density flows can be carried over to the density-weighted equations has not yet been verified (Bray 1979). In fact, it can be shown that nearly all modelled terms in the equations should depend to some extent on density gradients or on reaction rate (Bilger 1976; Pope 1979; Borghi & Dutoya 1979). It is not yet clear how many additional terms will be needed to account for the influence of the reaction on the flow field.

The present paper focuses on the influence of turbulence on the reaction rate. This problem has been the subject of a survey paper by Libby & Williams (1976) and is discussed in a recent review paper by Bray (1979). Several collections of papers have appeared during recent years that were concerned with the problem. Worth mentioning are: the proceedings of the AGARD Meeting on 'Analytical and Numerical Methods of Investigation of Flowfields with Chemical Reactions, Especially Related

to Combustion' in Liège 1974 (*AGARD Conf. Proc.* 164); the proceedings of the Project Squid Workshop on 'Turbulent Mixing in Non-Reactive and Reactive Flows', Purdue University 1974 (Murthy 1975); the Special Issue on 'Turbulent Reactive Flows' (*Combustion Sci. & Tech.*, vol. 13, 1976); the 15th AIAA Aerospace Science Meeting (Kennedy 1978) and the proceedings of the recent AGARD Meeting on 'Combustor Modeling' in Cologne, 1979 (*AGARD Conf. Proc.* 275).

Briefly, the problem has been attempted by looking at several limiting cases. The early approach was to treat the nonlinearities of the reaction rate in the same way as the convective nonlinearities. The case which was the most often considered is that of a one-step unidirectional reaction $A + B \rightarrow C$ with the reaction rate $\dot{w} = kY_A Y_B$. If the rate coefficient k is a constant the turbulent mean reaction rate can be written $\bar{w} = k(\bar{Y}_A \bar{Y}_B + \overline{Y'_A Y'_B})$ and the problem is closed if an equation for the mean product of the concentration fluctuations is modelled. This approach was also extended to an Arrhenius form of the reaction coefficient, $k = B \exp(-E/T)$, but it breaks down if the ratio of the activation temperature E to the mean temperature is not small (Borghi 1975).

Another limit is related to local chemical equilibrium by the 'fast chemistry' assumption. It applies only to flows that are mixing controlled such as diffusion flames. The assumption is that the reaction is so fast that local chemical equilibrium is reached instantaneously at each point of the flow, i.e. that the characteristic time for the reaction is much shorter than that for the diffusion or the residence time even during fluctuations. The assumption has the advantage that only the mixing field must be calculated by modelled equations for passive scalars.

Recently the turbulent premixed flame has been tackled by Clavin & Williams (1979) from the limit of large-scale fluctuations by considering wrinkled flames in the limit of large activation energies. This approach conserves the structures of the individual fluctuating laminar flamelets and introduces the statistics afterwards. It has the particular advantage that no modelling is required.

The mean turbulent reaction rate can be calculated statistically with the use of a probability density function for the scalars upon which it depends. If the p.d.f. is specified *a priori* the turbulent mean reaction rate can be calculated for any reaction kinetics. Bray & Libby (1976), Bray & Moss (1977), Libby & Bray (1977) and Libby, Bray & Moss (1979) assumed the p.d.f. for the scalar progress variable in a premixed system to consist of two Dirac delta functions at the boundaries and a distribution in between. A weighting factor of the distribution then determines the magnitude of the reaction rate. This factor is not fixed but calculated from a modelled balance equation.

A general way to determine the p.d.f. is the following: If there is enough knowledge about its shape, an n -parameter function can be assumed for the p.d.f. and the parameters can be related to the moments of the random variables. For instance, for a one-dimensional p.d.f. the first n moments determine n parameters. Thus the problem of the turbulent reaction rate is solved once the equations for the moments of those scalars on which the reaction rate depends are modelled. For practical calculations this formulation has the disadvantage that it involves an integration over the reaction rate multiplied by the p.d.f. A closed-form integration would be desirable but except for very simple forms of reaction rate and p.d.f. the integration can only be performed numerically. This is particularly laborious for problems where the reaction rate depends on more than one random variable.

To overcome this difficulty asymptotic limits of the reaction rate can be analysed. For combustion applications a sufficiently realistic limit is that of a one-step reaction with a large activation energy. It is known that the thermal theory of laminar flame propagation (Zeldovich & Frank-Kamenetzki 1938) is based on this limit. Also, by the powerful technique of large activation energy asymptotics many important features of laminar flames have been clarified during recent years. This development was largely stimulated by an early review article by Williams (1971). Highlights of this research were the treatment of the one-dimensional flame propagation (Bush & Fendell 1970; Fendell 1972), the analysis of ignition and extinction in diffusion flames (Liñán 1974), the explanation of the cellular structure of premixed flames by Lewis-number effects (Sivashinsky 1977), the description of the chemical amplification of a gasdynamic disturbance leading to explosion (Clarke 1978, 1979) and the already mentioned analysis of wrinkled turbulent flames by Clavin & Williams. These and other results will be summarized in a forthcoming book by Buckmaster & Ludford (1981).

In this paper the limit of a large activation energy will be exploited to derive closed-form expressions for the turbulent mean reaction rate. From the asymptotic treatment of laminar flames it is known that the reaction takes place in a thin inner layer situated between two chemically inert outer layers. In evaluating the mean turbulent reaction rate in the large activation energy limit, one thus looks for the probability of finding a thin inner layer at a particular point of the turbulent flow field. This view follows from the intuition that the mean reaction rate must be related to the statistics of an oscillating laminar flame surface.

It seems appropriate to close this introduction by reference to the first model for the turbulent mean reaction rate, the 'eddy break-up model' of Spalding (1971*a*). This model relates the mean reaction rate in turbulent shear flows to the velocity gradient $d\bar{u}/dy$ and was successful in predicting confined premixed flames. A careful examination of this flow figuration shows that the model relies on a flame stabilization effect rather than on a specific influence that turbulence has on the reaction rate: As the turbulent flame velocity is the highest where the turbulence intensity is the highest (Damköhler 1940), a flame that propagates against the mean flow in an inhomogeneous turbulence field and that has reached a region of high turbulence intensity will not be able to leave this region and to propagate any further. It is stabilized there and the mean reaction rate is thus greatest in this region. In turbulent shear flows it happens that this is the region where the velocity gradient is the highest. This not only explains the apparent relation between the mean reaction and the velocity gradient, but it also explains the limitations of the model (Spalding 1976).

2. Formulation

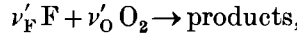
2.1. *The laminar reaction rate*

A most convenient and sufficiently realistic assumption that will be used in this analysis is that of equal diffusivities of heat and matter, $Le_i = 1$, $i = 1, 2, \dots, n$. Furthermore, radiative heat transfer and viscous dissipation are neglected in the energy balance equation and constant pressure is assumed. Then for simple flow geometries (jets, mixing layers, stagnation point flow) there exists a conserved scalar Z to which the element mass fractions and the enthalpy are linearly related. We take Z to be the

fuel element mass fraction or mixture fraction. It varies between $Z = 0$ and $Z = 1$ and obeys the same diffusion equation as a chemically inert species. In Cartesian co-ordinates

$$\rho \frac{\partial Z}{\partial t} + \rho v_\alpha \frac{\partial Z}{\partial x_\alpha} = \frac{\partial}{\partial x_\alpha} \left(\rho D \frac{\partial Z}{\partial x_\alpha} \right). \quad (1)$$

It is assumed that a single chemical reaction between fuel and oxygen is rate determining,



and that the products are related to each other by stoichiometric relations. Then a single progress variable describes the extent to which the reaction is completed. This variable is bounded between its unburnt and its burnt value (index u and b), both depending on the mixture fraction. We take a reduced temperature

$$\Theta = \frac{T - T_u(Z)}{T_b(Z) - T_u(Z)} \quad (2)$$

as the progress variable. The balance equation for the temperature T is

$$\rho \frac{\partial T}{\partial t} + \rho v_\alpha \frac{\partial T}{\partial x_\alpha} = \frac{\partial}{\partial x_\alpha} \left(\rho D \frac{\partial T}{\partial x_\alpha} \right) + \frac{(-\Delta h)}{c_p} \dot{w}. \quad (3)$$

Here the specific heat c_p has been assumed constant for simplicity. As the heat of reaction Δh is negative for combustion reactions, a negative sign has been introduced within brackets.

The chemical reaction term \dot{w} in equation (3) is related to the fuel consumption rate S , which is assumed to have the bimolecular form

$$S = \nu'_F M_F \frac{\dot{w}}{\rho} = \frac{B}{M_O} \rho Y_F Y_O \exp\left(-\frac{E}{T}\right), \quad (4)$$

Here B is the frequency factor and E the large activation temperature. The density ρ is related to the temperature, the pressure and the mass fractions Y_i by the equation of state. Between the temperature and the mass fractions exist the coupling relations

$$\left. \begin{aligned} Y_F &= -\frac{\nu'_F M_F c_p}{(-\Delta h)} (T - T_u(Z)) + Z, \\ Y_O &= -\frac{\nu'_O M_O c_p}{(-\Delta h)} (T - T_u(Z)) + Y_{O\infty}(1 - Z), \end{aligned} \right\} \quad (5)$$

where the M_i 's are the molecular weights and $Y_{O\infty}$ the oxygen mass fraction in the oxidizing stream. With these relations the reaction rate is a function of Z and Θ only. For simplicity the fuel stream has been assumed to consist of pure fuel.

The stoichiometric mixture fraction is derived from (5) and the condition $Y_{Fst} = 0, Y_{Ost} = 0$ as

$$Z_{st} = \frac{Y_{O\infty}}{(\nu'_O M_O / \nu'_F M_F) + Y_{O\infty}}. \quad (6)$$

There the temperature is

$$T_{st} = T_u(Z_{st}) + \frac{Z_{st}(-\Delta h)}{\nu'_F M_F c_p} = T_u(Z_{st}) + \frac{Y_{O\infty}(1 - Z_{st})(-\Delta h)}{\nu'_O M_O c_p}. \quad (7)$$

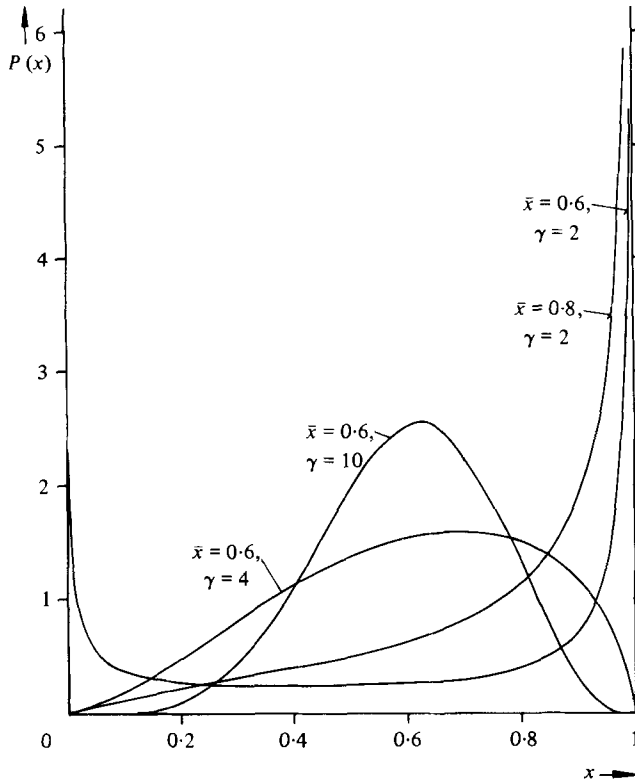


FIGURE 1. Shapes of the beta-function distribution.

In terms of these quantities we have

$$\left. \begin{aligned} Y_F &= -\frac{Z_{st}}{T_{st} - T_u(Z_{st})} (T - T_u(Z)) + Z, \\ Y_O &= -Y_{O\infty} \left\{ \frac{1 - Z_{st}}{T_{st} - T_u(Z_{st})} (T - T_u(Z)) - 1 + Z \right\}. \end{aligned} \right\} \quad (8)$$

These relations will be used in §4.

2.2. The probability distribution

For the present analysis it is essential that the probability density function is continuous and that it is bounded. This reduces the choice to a small number of physically realistic functions. Spalding (1971*b*) has used a sinusoidal p.d.f., whereas Lockwood & Naguib (1975) used a ‘clipped Gauss’ distribution. A triangular p.d.f. has been used by Rhodes & Harsha (1972). These authors commented, however, that a beta function would have been more realistic. The beta-function distribution was used by Richardson, Howard & Smith (1953), Rhodes (1975) and Janicka & Kollmann (1979). The beta-function p.d.f.

$$P(x) = x^{\alpha-1}(1-x)^{\beta-1} \frac{\Gamma(\gamma)}{\Gamma(\alpha)\Gamma(\beta)} \quad (9)$$

has the advantage over the other functions that it allows the two parameters α and β to be expressed in terms of the mean \bar{x} and the variance $\overline{x'^2}$ analytically:

$$\left. \begin{aligned} \alpha &= \bar{x}\gamma, & \beta &= (1-\bar{x})\gamma, \\ \gamma &= \alpha + \beta = \frac{\bar{x}(1-\bar{x})}{\overline{x'^2}} - 1, \\ \alpha, \beta, \gamma &> 0. \end{aligned} \right\} \quad (10)$$

As the maximum of the mean square fluctuation is given by $\bar{x}(1-\bar{x})$ the parameter γ is a measure of the relative mean square fluctuation

$$\frac{\overline{x'^2}}{\overline{x'^2}_{\max}} = \frac{1}{1+\gamma}. \quad (11)$$

The beta function can take quite different shapes, as demonstrated in figure 1. In particular, for large fluctuations it exhibits spikes at the boundaries. This behaviour is attractive for the progress variable in the case of fast chemical reactions, where one expects the material between the unburnt and the burnt state to be rapidly consumed. In this study the beta function distribution will be used for Θ only. In the case of non-premixed combustion only the Θ -dependence of the p.d.f. will be specified. The Z -dependence of the joint p.d.f. remains arbitrary. At this time the influence of intermittency, mixing due to large coherent structures and the influence of the combustion on the p.d.f. of the conserved scalar Z are poorly understood, so that this caution is necessary. Only for the purpose of illustration a beta-function distribution in Z direction will be used.

3. Premixed combustion

This case is simplified by the fact that the mixture fraction is specified, such that the reaction rate depends only on the progress variable. Using Favre's averaging process defined by

$$T = \tilde{T} + T'' \quad (12)$$

with

$$\overline{\rho T} = \bar{\rho} \tilde{T} \quad \text{and} \quad \overline{\rho T''} = 0,$$

the equations for the mean temperature \tilde{T} and the variance $\overline{T''^2}$ may be derived for large Reynolds numbers as

$$\left. \begin{aligned} \bar{\rho} \tilde{v}_\alpha \frac{\partial \tilde{T}}{\partial x_\alpha} &= \frac{\partial}{\partial x_\alpha} (-\bar{\rho} \tilde{v}_\alpha'' \overline{T''}) + \bar{\rho} \frac{(-\Delta h)}{\nu_F' M_F c_p} \tilde{S}, \\ \bar{\rho} \tilde{v}_\alpha \frac{\partial \overline{T''^2}}{\partial x_\alpha} &= \frac{\partial}{\partial x_\alpha} (-\bar{\rho} \tilde{v}_\alpha'' \overline{T''^2}) + 2\bar{\rho} \tilde{v}_\alpha'' \overline{T''} \frac{\partial \tilde{T}}{\partial x_\alpha} - 2\bar{\rho} \tilde{e}_T + 2\bar{\rho} \frac{(-\Delta h)}{\nu_F' M_F c_p} \overline{T'' S''}. \end{aligned} \right\} \quad (13)$$

In these equations the turbulent fluxes $-\tilde{v}_\alpha'' \overline{T''}$ and $-\tilde{v}_\alpha'' \overline{T''^2}$ and the scalar dissipation \tilde{e}_T must be modelled. We restrict our attention here to the modelling of the mean reaction rate \tilde{S} and of the covariance $\overline{T'' S''}$. If the reaction rate is expressed in terms of Θ they are written

$$\tilde{S} = \int_0^1 S(\Theta) \tilde{P}(\Theta) d\Theta, \quad (14a)$$

$$\overline{T'' S''} = \int_0^1 T''(\Theta) S(\Theta) \tilde{P}(\Theta) d\Theta - \tilde{T} \tilde{S}. \quad (14b)$$

We look for approximations to these integrals under the condition that the activation energy is large. In this case the exponential term in equation (4) increases very rapidly as the temperature approaches the maximum temperature T_b . At $T = T_b$, however, either the fuel or the oxidizer mass fraction vanishes so that the reaction rate decreases to zero. For $E \rightarrow \infty$ we thus expect a very steep peak of the reaction rate in the vicinity of T_b .

Following Williams (1975*a*) the small expansion parameter for large activation energy asymptotics is

$$\epsilon = \frac{T_b^2}{E(T_b - T_u)}. \quad (15)$$

With

$$\Theta = 1 - \epsilon y, \quad (16)$$

the density, the concentrations and the Arrhenius term in the reaction rate is written to first order

$$\left. \begin{aligned} \rho &= \rho_b(1 - \epsilon \Omega y), \quad \Omega = \left(\frac{1}{\rho} \frac{\partial \rho}{\partial T} \right)_b (T_b - T_u), \\ Y_F &= Y_{F,b} + \frac{\nu'_F M_F c_p (T_b - T_u)}{(-\Delta h)} \epsilon y, \\ Y_O &= Y_{O,b} + \frac{\nu'_O M_O c_p (T_b - T_u)}{(-\Delta h)} \epsilon y, \\ \exp\left(-\frac{E}{T}\right) &= \exp\left(-\frac{E}{T_b}\right) (1 - \epsilon a y^2) \exp(-y), \quad a = \frac{T_b - T_u}{T_b}. \end{aligned} \right\} \quad (17)$$

Thus the reaction rate is written to first order

$$S = \frac{c_p (T_b - T_u)}{(-\Delta h)} \hat{B}(y + by^2) (1 - \epsilon(\Omega + ya)y) \exp(-y), \quad (18)$$

with

$$\left. \begin{aligned} \hat{B} &= \epsilon \nu'_F M_F B \left(\frac{Y_{F,b} \nu'_O}{M_F} + \frac{Y_{O,b} \nu'_F}{M_O} \right) \rho_b \exp\left(-\frac{E}{T_b}\right), \\ b &= \epsilon \frac{\nu'_O \nu'_F M_O M_F c_p (T_b - T_u)}{(-\Delta h) (Y_{F,b} \nu'_O M_O + Y_{O,b} \nu'_F M_F)}. \end{aligned} \right\} \quad (19)$$

Here the condition $Y_{F,b} Y_{O,b} = 0$ has been used. As both, $Y_{F,b}$ and $Y_{O,b}$ are small within the flammability limits, b has been taken to be of order unity. This is the intermediate case between the limits $b \rightarrow 0$ (first-order reaction far from stoichiometric) and $b \rightarrow \infty$, (second-order reaction at the stoichiometric mixture) which are both covered asymptotically by this formulation (Peters 1979).

The reaction rate S and its zeroth- and first-order expansions are shown in figure 2 for a methane-air flame with $Z = 0.04$ ($\phi = 0.729$), $T_b = 1716$ K, $E = 20000$ K, $T_u = 300$ K, $\rho_u = 1.12$, $\rho = \rho_u T_u/T$, $B = 1.857 \times 10^{17}$ cm³ mol⁻¹ s⁻¹. It is seen that the first-order expansion follows the exact expression, equation (4), very closely even on the left side of the maximum until it overshoots to negative values, while the zeroth-order expansion gives too large values in this region.

The probability density function is expanded as

$$\tilde{P} = \frac{\Gamma(\gamma)}{\Gamma(\alpha)\Gamma(\beta)} (1 - \hat{\alpha}y) (\epsilon y)^{\beta-1}, \quad \hat{\alpha} = (\alpha - 1)\epsilon, \quad (20)$$

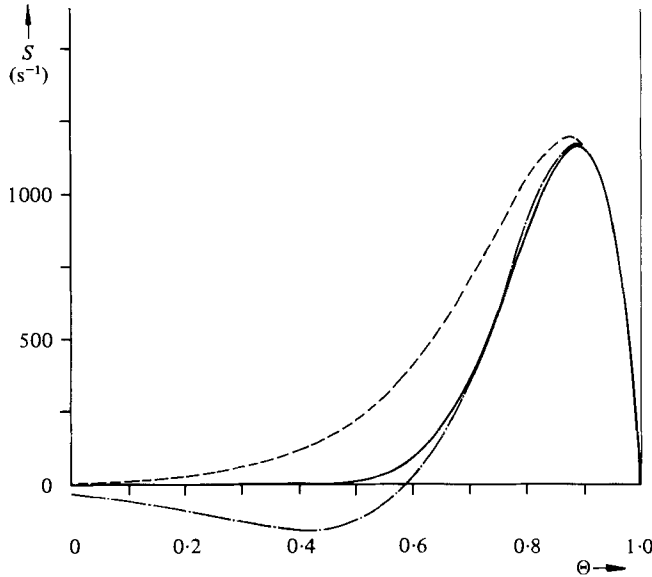


FIGURE 2. The laminar reaction rate. —, exact expression; - - -, zeroth order; ····, first-order expansion.

where $\hat{\alpha}$ is taken to be of order unity to account for the possibility of relatively large values of α .

Substituting equations (18)–(20) into equation (14) and integrating, we obtain in the limit $\epsilon \rightarrow 0$:

$$\begin{aligned}
 S &= \frac{c_p(T_b - T_u)}{(-\Delta h)} \frac{\hat{B}\Gamma(\gamma)\epsilon^\beta}{\Gamma(\alpha)\Gamma(\beta)} \int_0^\infty (1+by)(1-\hat{\alpha}y)y^\beta(1-\epsilon(\Omega+ay)y)\exp(-y)dy \\
 &= \frac{c_p(T_b - T_u)}{(-\Delta h)} \frac{\hat{B}\Gamma(\gamma)\beta\epsilon^\beta}{\Gamma(\alpha)} (1+(\beta+1)(b-\hat{\alpha}-\hat{\alpha}b(\beta+2))+\text{f.o.t.}), \tag{21}
 \end{aligned}$$

where the first-order terms are

$$\text{f.o.t.} = -\epsilon(\beta+1)(\Omega+(\beta+2)(\Omega(b-\hat{\alpha})+a+(\beta+3)(a(b-\hat{\alpha})-\Omega\hat{\alpha}b-(\beta+4)a\hat{\alpha}b))). \tag{22}$$

A simplification of the expression for the mean reaction rate is obtained if \hat{B} is replaced by an expression containing the flame velocity v_u derived in the large activation energy limit for a laminar flame (Peters 1978)

$$\hat{B} = \frac{\rho_u^2 v_u^2 \nu'_F M_F}{2\epsilon\rho_b^2 D_b(1+2b)}. \tag{23}$$

For practical purposes it is noted that the gamma functions appearing in equation (21) may be approximated with sufficient accuracy by the use of Stirling's formula

$$\Gamma(x) = \sqrt{(2\pi)} x^{x-\frac{1}{2}} \exp\left(-x + \frac{1}{12x}\right) \quad \text{for } x \geq 0.2, \tag{24}$$

leading to

$$\frac{\Gamma(\gamma)}{\Gamma(\alpha)} = \gamma^\beta \tilde{\Theta}^{\frac{1}{2}(\beta-\alpha)} \exp\left(-\beta + \frac{1}{12\gamma}\left(1 - \frac{1}{\tilde{\Theta}}\right)\right). \tag{25}$$

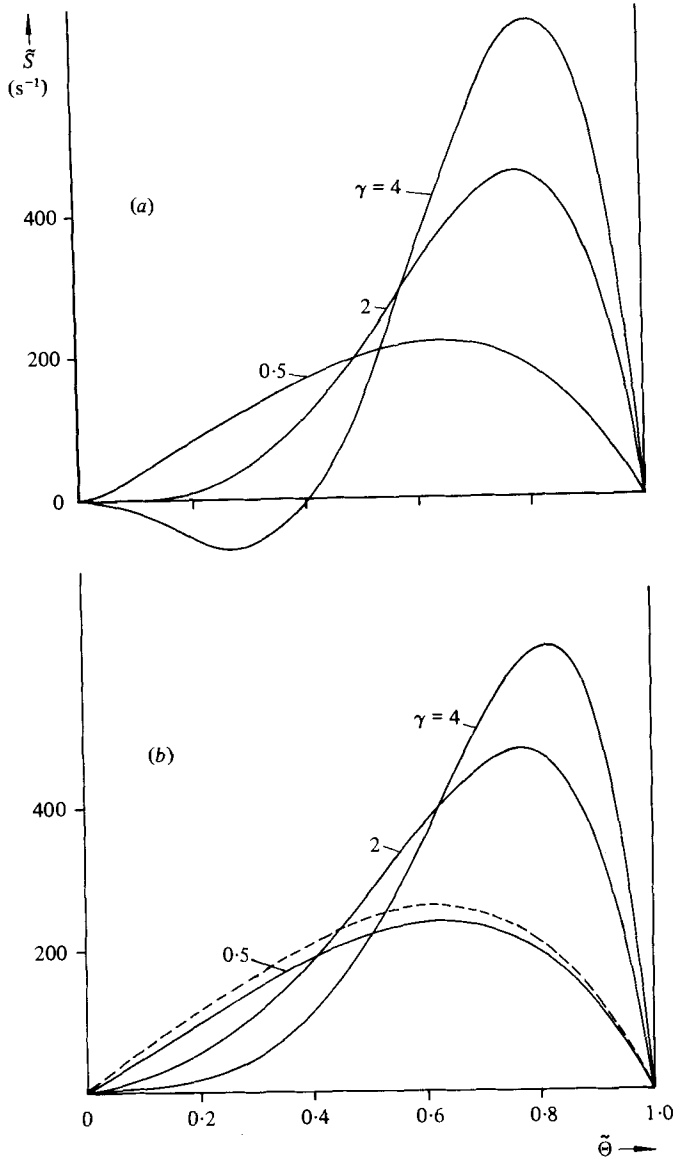


FIGURE 3. The turbulent mean reaction rate. (a) First-order approximation. (b) Exact evaluation; - - -, zeroth-order approximation for $\gamma = 0.5$.

Combination of equations (21), (23) and (25) finally yields the expression

$$\begin{aligned} \tilde{S} = & \frac{c_p(T_b - T_u) \rho_u^2}{(-\Delta h)} \frac{\nu'_F M_F v_u^2}{\rho_b^2 2\epsilon D_b (1 + 2b)} \beta(1 + (\beta + 1)b - \hat{a} - \hat{a}b(\beta + 2) + f.o.t.) \\ & \times \exp\left(-\beta(1 - \ln \gamma - \ln \epsilon) + \frac{1}{12\gamma} \left(1 - \frac{1}{\tilde{\Theta}}\right)\right). \end{aligned} \quad (26)$$

Thus the laminar kinetics are represented by the values of the flame velocity and the activation energy, which are well known for many fuels. Turbulence comes into play via β and γ , which are related to the mean temperature and the temperature

fluctuations. The turbulent mean reaction rate vanishes as it should for $\tilde{\Theta} \rightarrow 0$, $\tilde{\Theta} \rightarrow 1$ and $\gamma \rightarrow 0$. Figure 3 shows a comparison between exact evaluation of \tilde{S} and the asymptotic approximation. It is seen that the zeroth-order result is good for strong fluctuations while the first-order terms give fairly good results up to $\gamma = 4$ if the negative values are dropped.

The limit of strong fluctuations, $\gamma \rightarrow 0$, leads with $\Gamma(x) = \Gamma(x+1)/x$,

$$\frac{\Gamma(\gamma)}{\Gamma(\alpha)} = \frac{\alpha \Gamma(\gamma+1)}{\gamma \Gamma(\alpha+1)} \rightarrow \tilde{\Theta}, \quad (27)$$

$$\text{to zeroth order to } \tilde{S} = \frac{c_p(T_b - T_u) \rho_u^2 \nu_F' M_F v_u^2 (1+b)}{(-\Delta h) \rho_b^2 2\epsilon D_b (1+2b)} \gamma \tilde{\Theta} (1 - \tilde{\Theta}). \quad (28)$$

This expression exhibits basically the same dependence on $\tilde{\Theta}(1-\tilde{\Theta})$ as Bray and Libby's model. It also shows via v_u an explicit dependence on laminar kinetics. Libby & Bray (1980) obtain in a recent paper a linear dependence of S on v_u , while we obtain a quadratic dependence.

The integral in equation (14b) is evaluated in the same way by replacing ρ by ρT . This leads to

$$\widehat{T'' S''} = \tilde{S}_1 (T_b - T_u) - (\tilde{T} - T_u) \tilde{S}, \quad (29)$$

where \tilde{S}_1 has the same form as \tilde{S} with ρ_b replaced by $\rho_b T_b$ and Ω by $\Omega + a$. Then one has to zeroth order

$$\widehat{T'' S''} = (T_b - T_u) (1 - \tilde{\Theta}) \tilde{S}. \quad (30)$$

Very similar expressions were found by Bray (1979) and by Borghi & Dutoya (1979). These authors obtain a constant value $c_m < 1$ rather than the value 1 in the second brackets of equation (30). From the present result it is seen that the constant approaches 1 in the limit of large activation energies.

4. Non-premixed combustion

4.1. General analysis

In this case the reaction rate depends on two quantities, the mixture fraction and the progress variable. The Favre-averaged equations for the temperature and its variance are to be supplemented by the corresponding equations for the mixture fraction. The reaction rate is expressed in terms of Θ and Z by writing

$$\left. \begin{aligned} \rho &= \rho_{st} \left(1 + \Omega \left(\Theta \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} - 1 \right) \right), & \Omega &= \left(\frac{1}{\rho} \frac{\partial \rho}{\partial T} \right)_{st} (T_{st} - T_u(Z_{st})), \\ Y_F &= -Z_{st} \left(\Theta \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} \right) + Z, \\ Y_O &= -Y_{O\infty} \left((1 - Z_{st}) \Theta \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} - 1 + Z \right), \\ -\frac{E}{T} &= -\frac{E}{T_{st}} + \frac{1}{\epsilon} \left(\left(\Theta \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} - 1 \right) - a \left(\Theta \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} - 1 \right)^2 \right), \\ a &= \frac{T_{st} - T_u}{T_{st}}, \end{aligned} \right\} \quad (31)$$

where

$$\epsilon = \frac{T_{st}^2}{E(T_{st} - T_u(Z_{st}))}. \quad (32)$$

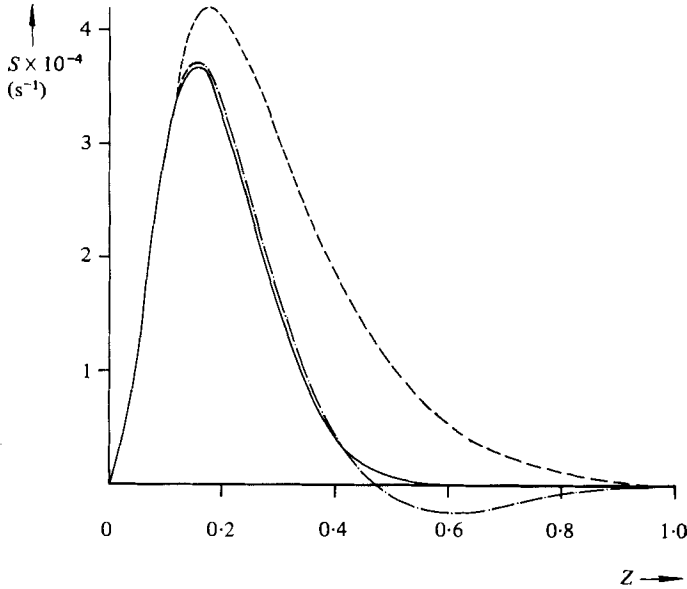


FIGURE 4. The laminar reaction rate for $\Theta = 0.95$. —, exact expression; - - -, zeroth order; - · - · -, expansion equations (36) and (37).

Different expressions for $T_b(Z)$ are valid in the fuel-lean and fuel-rich regions:

$$\left. \begin{aligned} \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} &= \frac{Z}{Z_{st}}, & Z \leq Z_{st}; \\ \frac{T_b(Z) - T_u(Z)}{T_{st} - T_u(Z_{st})} &= \frac{1 - Z}{1 - Z_{st}}, & Z \geq Z_{st}; \end{aligned} \right\} \quad (33)$$

therefore we split the integral for the mean reaction rate into two parts

$$\bar{S} = \int_0^{Z_{st}} \int_0^1 S(\Theta, Z) \bar{P}(\Theta, Z) d\Theta dZ + \int_{Z_{st}}^1 \int_0^1 S(\Theta, Z) \bar{P}(\Theta, Z) d\Theta dZ. \quad (34)$$

Introducing the expansions

$$\Theta = 1 - \epsilon y, \quad Z = Z_{st}(1 + \epsilon \zeta), \quad (35)$$

the concentrations and the product of the density and the exponential term are expanded to first order. The reaction rate is written:

for $Z \leq Z_{st}$,

$$\begin{aligned} S &= \frac{\nu'_F B \rho_{st}}{M_O} Z_{st} Y_{O\infty} \exp\left(-\frac{E}{T_{st}}\right) \epsilon^2 y(1 + \epsilon \zeta) (y(1 - Z_{st})(1 + \epsilon \zeta) - \zeta) \\ &\quad \times (1 - \epsilon y \zeta - \epsilon a(y - \zeta)^2 + \epsilon \Omega(-y + \zeta)) \exp(-y + \zeta); \end{aligned} \quad (36)$$

for $Z \geq Z_{st}$,

$$\begin{aligned} S &= \frac{\nu'_F B \rho_{st}}{M_O} \frac{Z_{st}}{1 - Z_{st}} Y_{O\infty} \exp\left(-\frac{E}{T_{st}}\right) \epsilon^2 \exp\left(-y - \frac{Z_{st}}{1 - Z_{st}} \zeta\right) \\ &\quad \times y(1 - Z_{st}(1 + \epsilon \zeta)) (y(1 - Z_{st}(1 + \epsilon \zeta)) + \zeta) \\ &\quad \times \left(1 + \frac{Z_{st}}{1 - Z_{st}} \epsilon y \zeta - \epsilon a \left(y + \frac{Z_{st}}{1 - Z_{st}} \zeta\right)^2 + \epsilon \Omega \left(-y - \frac{Z_{st}}{1 - Z_{st}} \zeta\right)\right). \end{aligned} \quad (37)$$

This form rather than a truncated second-order expansion is used in order to obtain the best approximation of the reaction rate. For the combustion of methane considered before the reaction rate is plotted over Z for $\Theta = 0.95$ in figure 4. The above expansion follows the exact curve very closely. The maximum of the reaction rate lies on the fuel-rich side rather than at the expansion point Z_{st} . This suggests an expansion of the p.d.f. around Z_m , the value where the reaction has its maximum. This maximum is obtained from the zeroth-order expansion of equation (37) to lie at

$$\zeta_m = \frac{1 - Z_{st}}{Z_{st}} + 1, \quad y_m = \frac{1}{1 - Z_{st}}. \quad (38)$$

Following §2.2 the joint p.d.f. has the form

$$\tilde{P}(\Theta, Z) = \frac{A(Z) \Gamma(\gamma) \Theta^\alpha (1 - \Theta)^\beta}{\Gamma(\alpha) \Gamma(\beta)}, \quad (39)$$

where α , β and $\gamma = \alpha + \beta$ are again functions of Z such that no statistical independence is assumed. By definition $A(Z)$ is identical with the marginal p.d.f.

$$A(Z) = \int_0^1 \tilde{P}(\Theta, Z) d\Theta. \quad (40)$$

We expand equation (39) around $\Theta = 1$ and $Z_m = Z_{st}(1 + \epsilon\zeta_m)$ as

$$\tilde{P} = \frac{A(Z_m) \Gamma(\gamma)}{\Gamma(\alpha) \Gamma(\beta)} (1 - \hat{\alpha}y) (\epsilon y)^{\beta-1}, \quad (41)$$

where for variable $\hat{\alpha}$ and β those at Z_m are used. Since figure 4 shows that the reaction rate has a very steep peak in Z -direction, first-order terms in Z -direction are neglected. Introducing these expansions into equation (34) and integrating, one obtains

$$\begin{aligned} \tilde{S} = & \frac{\nu'_F B \rho_{st} Z_{st} Y_{O_\infty}}{M_O} \exp\left(-\frac{E}{T_{st}}\right) \frac{e^{\beta+3} A(Z_m) \Gamma(\gamma) \beta}{\Gamma(\alpha)} \{Q_0 + (Q_1 - \hat{\alpha}Q_0)(\beta + 1) \\ & + (Q_2 - \hat{\alpha}Q_1)(\beta + 1)(\beta + 2) + (Q_3 - \alpha Q_2)(\beta + 1)(\beta + 2)(\beta + 3) \\ & - \alpha Q_3(\beta + 1)(\beta + 2)(\beta + 3)(\beta + 4)\}, \end{aligned} \quad (42)$$

where the Q_i 's contain terms up to second order that are given in the appendix. Higher-order terms can be neglected. The integral in the covariance of the temperature and the reaction rate

$$\widetilde{T''S''} = \int_0^1 \int_0^1 T(\Theta, Z) S(\Theta, Z) \tilde{P}(\Theta, Z) d\Theta dZ - \tilde{T} \tilde{S} \quad (43)$$

is evaluated in the same way by replacing the density ρ by ρT . This leads to the same form as (41) with ρ_{st} replaced by $\rho_{st} T_{st}$ and Ω by $\Omega + a$. Thus the zeroth-order one has

$$\widetilde{T''S''} = (T_{st} - \tilde{T}) \tilde{S}. \quad (44)$$

The comparison of the numerical evaluation of \tilde{S} with the asymptotic limit, equation (42), is shown in figure 5 and 6. Here $A(Z)$ was assumed to be a beta-function in terms of Z (with parameters α_Z and β_Z). The parameter α_Θ and β_Θ of the beta-function in terms of Θ were set constant, such that statistical independence was assumed for this evaluation. Values of $\tilde{\Theta} = 0.95$ and $\tilde{\Theta} = 0.8$ with $\gamma_\Theta = \alpha_\Theta + \beta_\Theta = 2$, which are typical for diffusion flames, were assigned. The mean reaction rate is plotted over

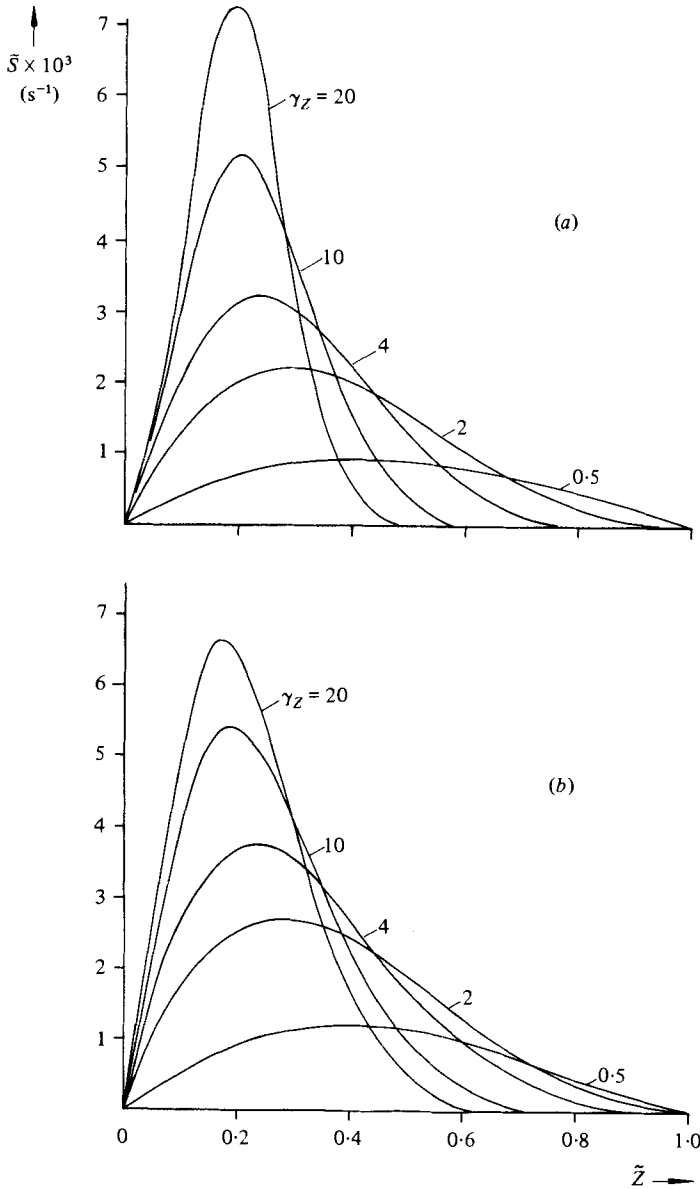


FIGURE 5. The turbulent mean reaction rate for $\tilde{\Theta} = 0.95$. (a) Second-order approximation; (b) exact evaluation.

\tilde{Z} with γ_Z as parameter. It is seen that the overall agreement is satisfactory while the values for $\tilde{\Theta} = 0.8$ are somewhat too low. The broadening of the reaction rate by mixture fraction fluctuations is not as large in the asymptotic value as in the numerical evaluation. This is due to the comparatively poor expansion of the joint p.d.f. in Z -direction.

The question arises: How are the mean and the variance of Θ related to those of T and Z ? Janicka (1979) derived a balance equation for his progress variable r . Its turbulent counterpart has to be modelled, which causes new problems. As an

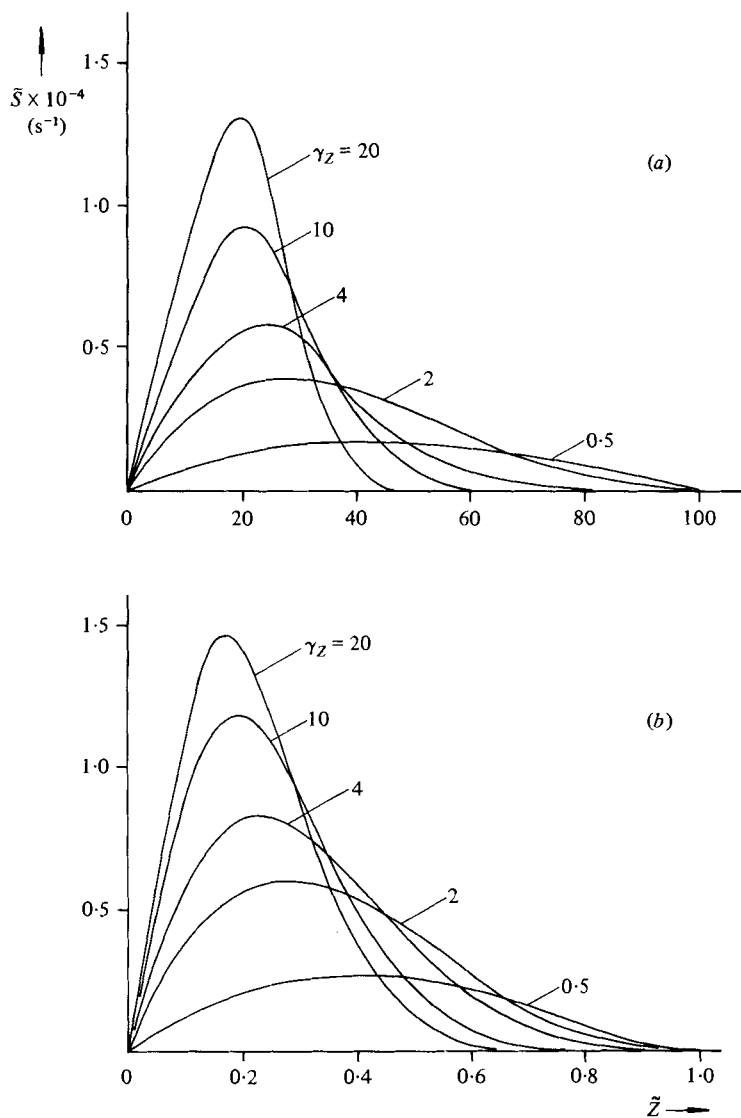


FIGURE 6. The turbulent mean reaction rate for $\tilde{\Theta} = 0.8$. (a) Second-order approximation; (b) exact evaluation.

alternative it is proposed to model the temperature equation in the conventional fashion and to calculate \tilde{T} and $\overline{T^{n_2}}$ from the definitions

$$\left. \begin{aligned} \tilde{T} &= \int_0^1 \int_0^1 T(\Theta, Z) \tilde{P}(\Theta, Z) d\Theta dZ, \\ \overline{T^{n_2}} &= \int_0^1 \int_0^1 T^2(\Theta, Z) \tilde{P}(\Theta, Z) d\Theta dZ - \tilde{T}^2. \end{aligned} \right\} \quad (45)$$

which relate \tilde{T} , $\widetilde{T'^2}$ to $\tilde{\Theta}$, $\widetilde{\Theta'^2}$. The integration over Θ is readily performed:

$$\left. \begin{aligned} \tilde{T} &= \int_0^1 (T_b(Z) - T_u(Z)) \frac{\alpha}{\gamma} A(Z) dZ + \int_0^1 T_u(Z) A(Z) dZ, \\ \widetilde{T'^2} &= \int_0^1 (T_b(Z) - T_u(Z))^2 \frac{\alpha(\alpha+1)}{\gamma(\gamma+1)} A(Z) dZ \\ &\quad + 2 \int_0^1 T_u(Z) (T_b(Z) - T_u(Z)) \frac{\alpha}{\gamma} A(Z) dZ \\ &\quad + \int_0^1 T_u^2(Z) A(Z) dZ - \tilde{T}^2. \end{aligned} \right\} \quad (46)$$

In the special case of statistical independence of Θ and Z with constant α and β the mean and the variance of Θ are given by

$$\left. \begin{aligned} \tilde{\Theta} &= \frac{\tilde{T} - I_1}{I(0, 1)}, \\ \widetilde{\Theta'^2} &= \frac{\widetilde{T'^2} + \tilde{T}^2 - I_2 \tilde{\Theta} - I_3}{I(0, 2)} - \tilde{\Theta}^2, \end{aligned} \right\} \quad (47)$$

where the integrals I_1 - I_3 take the dependence of T_u on Z

$$T_u = Z(T_{u,F} - T_{u,O}) + T_{u,O} \quad (48)$$

into account,

$$\left. \begin{aligned} I_1 &= \tilde{Z}(T_{u,F} - T_{u,O}) + T_{u,O}, \\ I_2 &= 2(T_{u,F} - T_{u,O}) I(1, 1) + 2T_{u,O} I(0, 1), \\ I_3 &= (T_{u,F} - T_{u,O})^2 (\tilde{Z}^2 + \widetilde{Z'^2}) + 2T_{u,O} (T_{u,F} - T_{u,O}) \tilde{Z} + T_{u,O}^2, \end{aligned} \right\} \quad (49)$$

whereas $I(m, n)$ is defined by

$$I(m, n) = \int_0^1 Z^m (T_b(Z) - T_u(Z))^n A(Z) dZ. \quad (50)$$

Thus, once the solution of the system (13) and the corresponding equations for \tilde{Z} and $\widetilde{Z'^2}$ are known at a given point in the flow field, $\tilde{\Theta}$ and $\widetilde{\Theta'^2}$ and thus α and β in equation (42) can be determined.

4.2. Local extinction of diffusion flamelets

Up to this point we have assumed a given shape of the p.d.f. without considering the possibility of extinction. If extinction occurs, at a given extinction temperature $T_q(Z)$, the temperature drops very rapidly to the frozen temperature $T_u(Z)$. Thus we expect the p.d.f. between $T_q(Z)$ and $T_u(Z)$ to be vanishing small. This behaviour can, of course, not be accounted for by a two-parameter shape of the p.d.f. For this reason we will develop an alternative formulation, which takes extinction explicitly into account.

We may picture the turbulent flame as an ensemble of laminar diffusion flamelets. This concept has been introduced by Williams (1975*b*). A laminar diffusion flamelet is defined as a structure in which an asymptotically thin reaction zone is embedded between two convective-diffusive zones at the local instantaneous surface of stoichiometric mixture. In a turbulent shear flow diffusion flamelets are distorted and stretched by the action of vorticity.

Stretching of a flamelet makes the concentration and temperature gradients perpendicular to the flame steeper and increases the heat loss of the reaction zone to its surroundings. If the rate of heat loss is not balanced by the heat production due to chemical reaction, the flamelet will be quenched locally.

Let us assume that the mixture fraction Z is given in the turbulent flow field as a function of space and time by solution of equation (1). Then the surface of stoichiometric mixture can be determined from

$$Z(x_k, t) = Z_{\text{st}}. \quad (51)$$

Let us introduce a co-ordinate system such that one co-ordinate, x_1 , is replaced by the mixture fraction Z , while the others, $Z_2 = x_2$, $Z_3 = x_3$, $\tau = t$ remain unchanged. Applying formally the transformation rules

$$\left. \begin{aligned} \frac{\partial}{\partial t} &= \frac{\partial}{\partial \tau} + \frac{\partial Z}{\partial t} \frac{\partial}{\partial z}, & \frac{\partial Z}{\partial x_1} &= \frac{\partial Z}{\partial x_1} \frac{\partial}{\partial Z}, \\ \frac{\partial}{\partial x_j} &= \frac{\partial}{\partial Z_j} + \frac{\partial Z}{\partial x_j} \frac{\partial}{\partial Z} \quad (j = 2, 3), \end{aligned} \right\} \quad (52)$$

one obtains the temperature equation (3) with $L(Z) = 0$ as

$$\rho \left(\frac{\partial T}{\partial \tau} + v_2 \frac{\partial T}{\partial Z_2} + v_3 \frac{\partial T}{\partial Z_3} \right) - \rho D \left\{ \left(\frac{\partial Z}{\partial x_k} \right)^2 \frac{\partial^2 T}{\partial Z^2} + 2 \frac{\partial Z}{\partial x_2} \frac{\partial^2 T}{\partial Z \partial Z_2} + 2 \frac{\partial Z}{\partial x_3} \frac{\partial^2 T}{\partial Z \partial Z_3} + \frac{\partial^2 T}{\partial Z_2^2} + \frac{\partial^2 T}{\partial Z_3^2} \right\} = \frac{(-\Delta h)}{c_p} \dot{w}. \quad (53)$$

In the asymptotic limit of large activation energies, all terms involving derivatives with respect to τ , Z_2 and Z_3 are of higher order in the thin reaction zone and can be neglected to lowest order. One obtains the temperature equation in the reaction zone as

$$\frac{\partial^2 T}{\partial Z^2} = -2\chi_{\text{st}}^{-1} \frac{(-\Delta h)}{c_p} \left(\frac{\dot{w}}{\rho} \right)_{\text{st}}. \quad (54)$$

Here

$$\chi_{\text{st}} = 2D \left(\frac{\partial Z}{\partial x_k} \right)_{\text{st}}^2 \quad (55)$$

is the instantaneous local scalar dissipation rate, conditioned at stoichiometry. We may look at χ_{st} as the inverse of the characteristic diffusion time. If the ratio of the reaction time to the diffusion time, the second Damköhler number, decreases, quenching occurs. An asymptotic analysis of quenching in laminar counterflow diffusion flames has been given by Liñán (1974). Peters (1980) has cast equation (54) in the same form as the one that governs Liñán's diffusion flame regime to obtain the quenching condition for diffusion flamelets. Essentially, there is a maximum value of χ_{st} , called χ_q , corresponding to a maximum local stretch, at which quenching occurs. In a plot of the temperature profile over Z , obtained from the solution of equation (54), there exists a limiting profile $T_q(Z)$ which corresponds to extinction of the diffusion flamelet. This profile can be calculated from the details of Liñán's analysis. For small values of Z_{st} extinction takes place at the transition from the diffusion flame regime to the premixed flame regime at a premixed temperature

$$T_p = T_{\text{st}} - T_{\text{st}}^2/E, \quad (56)$$

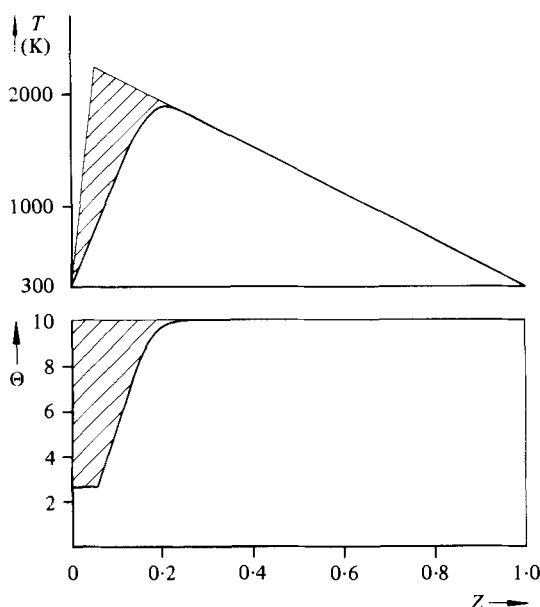


FIGURE 7. Areas of integration with limiting profiles T_q and Θ_q . Shaded area: stable combustion. Blank area: quenched states.

and the profile $T_q(Z)$ is given by

$$\left. \begin{aligned} T_q &= T_p - \frac{Z - Z_p}{1 - Z_p} (T_p - T_u(Z_p)) - (T_{u,F} - T_{u,O}) (Z - Z_p) - \frac{T_{st}^2}{E} y_1(\eta), \\ Z_p &= Z_{st}(1 - \epsilon) + \epsilon, \\ \eta &= \frac{Z - Z_p}{Z_p} \frac{E(T_{st} - T_u(Z_p))}{T_{st}^2(1 - Z_p)} + n_p. \end{aligned} \right\} \quad (57)$$

Here $y_1(\eta)$ is the solution of the inner flame structure in the premixed flame regime. The parameter n_p is in the present case close to 1.344. The inner profile may be approximated by

$$y_1 = \begin{cases} n_p - \eta + B_p \exp(a_p \eta), & \eta \leq n_p, \\ A_p \exp(-a_p \eta), & \eta \geq n_p, \end{cases} \quad (58)$$

where the constants a_p , A_p , B_p are determined from the condition that y_1 and its first and second derivative are continuous at $\eta = n_p$. This leads for $n_p = 1.344$ to $a_p = 0.9262$, $A_p = 1.8745$, $B_p = 0.15547$. This profile is shown for the combustion of methane in air in the T - Z and the Θ - Z plane in figure 7. The shaded area above the limiting profile corresponds to stable burning, the area below it to unstable states which are rapidly extinguished. Thus the quenching changes the joint probability distribution. For a fixed Z quenching results in a temperature $\Theta = \Theta_u(Z)$. With a function $f(\Theta, Z)$ in the form of equation (39) and the definition

$$\tilde{P}_q = \begin{cases} f(\Theta, Z) & \text{for } \Theta \geq \Theta_q(Z), \\ 0 & \text{for } 0 < \Theta < \Theta_q(Z), \end{cases} \quad (59)$$

the joint p.d.f. takes the form

$$\tilde{P}(\Theta, Z) = \tilde{P}_q(\Theta, Z) + \delta(\Theta - \Theta_u(Z)) \int_0^{\Theta_q(Z)} f(\Theta, Z) d\Theta. \quad (60)$$

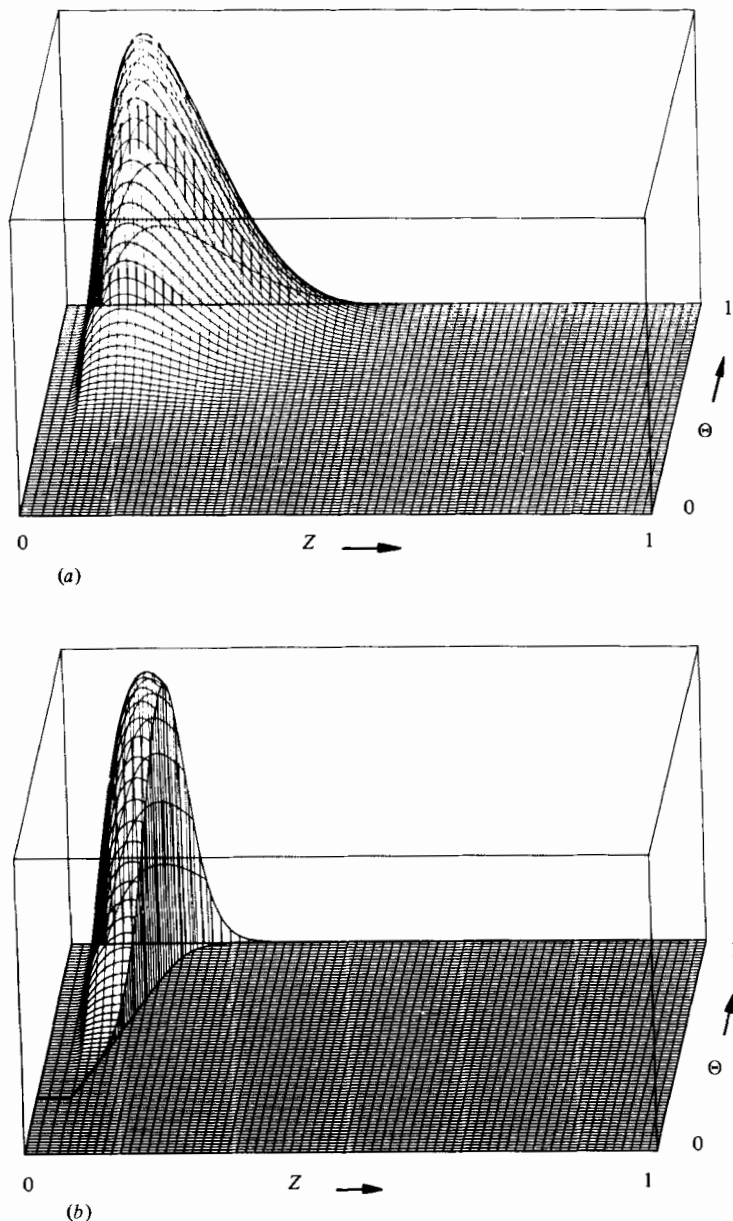


FIGURE 8. Laminar reaction rate over Θ and Z . (a) No quenching; (b) only stable states considered.

Here δ is the Dirac delta function. As the quenched states at $\Theta_q(Z)$ do not contribute to the mean reaction rate the integration has thus to be performed over $\tilde{P}_q(\Theta, Z)$ only,

$$\bar{S} = \int_0^1 \int_0^1 S(\Theta, Z) \tilde{P}_q(\Theta, Z) d\Theta dZ. \quad (61)$$

The entire laminar reaction rate and that part which exists in the stable region of the Θ - Z plane are shown in figure 8. It is seen that nearly half of the total reaction

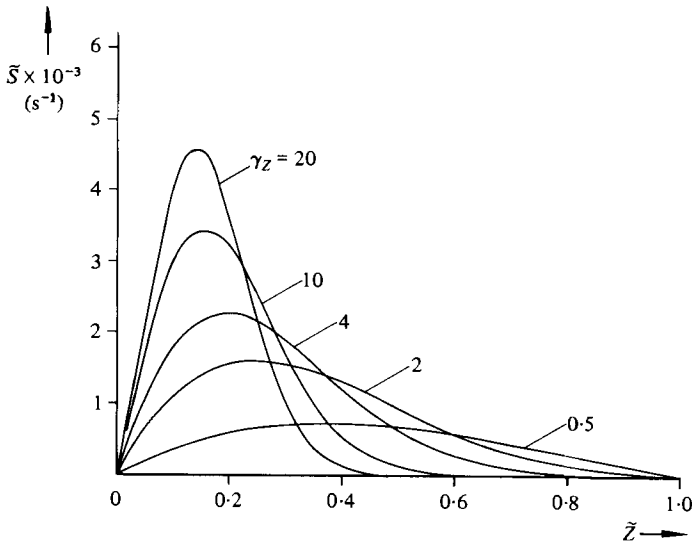


FIGURE 9. Exact evaluation of the turbulent mean reaction rate for stable states only, $\tilde{\Theta} = 0.95$.

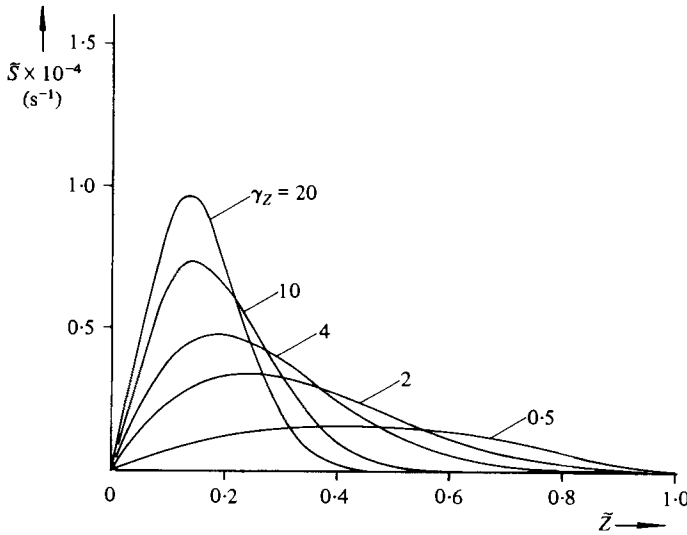


FIGURE 10. Exact evaluation of the turbulent mean reaction rate for stable states only, $\tilde{\Theta} = 0.8$.

falls into the unstable part of the plane. Thus quenching due to flame stretch is an important factor for the evaluation of the mean reaction rate. In figures 9 and 10 the mean reaction rate calculated numerically from equation (61) has been plotted over \bar{Z} for $\tilde{\Theta} = 0.95$ and $\tilde{\Theta} = 0.80$ respectively. These plots are to be compared with those in figures 5 and 6. It is seen that the mean reaction rate calculated from the asymptotic formula, equation (42), lies reasonably between the two limits of zero and entire quenching due to flame stretch.

5. Conclusions

For large activation energies the reaction rate has a sharp peak at a definite location in the probability space. In the asymptotic limit the laminar reaction rate cuts a small part of the p.d.f. out of the integral for the mean turbulent reaction rate. Thus only the value of the p.d.f. at the reaction rate maximum is important for the evaluation. This value depends much on the choice of the p.d.f. and thus the mean reaction rate is very sensitive to this choice. This is in contrast to previous findings (Bray 1979), which state that the p.d.f. does not influence the reaction rate very much.

Physically, the limit of large activation energies results in asymptotically thin flame structures. The present analysis uses this property in the space of the random variables rather than in the physical space: This allows the evaluation of mean turbulent reaction rates. The resulting closed-form expressions can be easily incorporated in turbulent combustion models.

In this study a beta-function p.d.f. was used for the progress variable. The Libby–Bray–Moss model for premixed combustion is hereby recovered as a limiting case for large fluctuations. The beta-function formulation extends the model away from that limit and may thus be regarded as a good choice. In the case of non-premixed combustion the dependence of the joint p.d.f. on Z remained arbitrary. The resulting expression for the turbulent mean reaction rate, equation (42), shows that only one value of the marginal p.d.f., $A(Z_m)$, is needed for the evaluation of the mean turbulent reaction rates.

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Appendix

$$Q_i = \sum_{j=0}^2 \epsilon^j q_{ij},$$

$$q_{00} = Z_{st}^2 + (1 - Z_{st})^2, \quad q_{01} = -(2 + 6a + 2\Omega)q_{00}, \quad q_{02} = (24a + 6\Omega)q_{00},$$

$$q_{10} = Z_{st}(1 - Z_{st}),$$

$$q_{11} = -q_{10}(2 + 2a + \Omega) - q_{00}(4a - 2 + \Omega),$$

$$q_{12} = 2q_{10}(6a + 2\Omega + 1) + 2q_{00}(6a - 3 + \Omega), \quad q_{20} = 0,$$

$$q_{21} = -q_{10}(2a - 1 + \Omega) - q_{00}a,$$

$$q_{22} = 2q_{10}(4a - 2 + \Omega) + 2aq_{00},$$

$$q_{30} = 0, \quad q_{31} = -aq_{10}, \quad q_{32} = 2aq_{10}.$$

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