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Countergradient Diffusion in Premixed Turbulent Flames

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A new theory for premixed turbulent flames normal to the oncoming reactants is developed on the basis of the Bray-Moss-Libby model of premixed combustion and of second-order closure. Gradient transport assumptions are carefully avoided. The final formulation focuses on the intensity of the fluctuations of the velocity component normal to the flame and on the mean flux of product. At low rates of heat release corresponding to small intensities of the density fluctuations, the new theory is in agreement with our earlier theory based on gradient transport. However, as the heat release increases toward values of practical interest, countergradient diffusion, i.e., mean flux in the direction of increasing mean concentration, arises and is attributable to the differential effect of mean pressure gradient on cold reactants and hot products. The implications of these results are discussed.

Nomenclature

c	= progress variable or normalized product concentration
c_m	= coefficient depending on I_n, I_4/I_3
C	= arbitrary constant, see Eq. (34)
F	= nondimensional mean flux of product, see Eq. (26)
I	= nondimensional intensity of velocity fluctuations, see Eq. (26)
I_n	= integral involving $f(c)$ and $w(c)$, $\int_0^1 (w(c)/w_{\max}) f(c)\,dc$
K,L	= auxiliary functions, see Eq. (29)
n	= exponent in $w(c)$
p	= pressure
$P(c),P(u,c),$ $P(u,v,c)$	= probability density functions
$\tilde{q}$	= turbulent kinetic energy, $\frac{1}{2}\overline{\rho u_k'' u_k''}/\bar{\rho}$
S	= nondimensional gradient of mean product, $S=-F$
T	= temperature
u	= x-wise velocity component
v	= velocity component transverse to the reaction zone
w	= volumetric rate of production of product
$w_{\max}$	= maximum volumetric rate of production of product
α,β,γ	= weights applied to modes of entry in the probability density functions, see Fig. 2
β_0,β_1	= parameters entering approximations applicable to $\tilde{c}\sim 0,1$
γ_0,γ_1	= parameters entering approximations applicable to $\tilde{c}\sim 0,1$
κ_1,κ_2	= empirical constants
ν_T	= eddy transport coefficient
ρ	= mass density
τ	= heat release parameter
ϕ_n	= parameter depending on the exponent n in $w(c)$, $n/(n+1)$
$\tilde{\chi}_c,\tilde{\chi}_u,\tilde{\chi}_{uc}$	= mean rates of dissipation
Subscripts	
0	= conditions upstream of the reaction zone
∞	= conditions downstream of reaction zone
p	= conditions in product
r	= conditions in reactants

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Introduction

ALTHOUGH gradient or eddy viscosity models of turbulent transport are widely used in predictions of many types of turbulent flow, this practice is notoriously difficult to justify. If c represents a composition variable, then, according to the gradient transport approximation, the x component of the turbulent flux of c is

$$\overline{\rho u'' c''} = -\bar{\rho}\nu_T \partial\tilde{c}/\partial x \tag{1}$$

where ν_T is an eddy kinematic viscosity, and Favre or mass-weighted averages have been used so that, for example, $c=\tilde{c}+c''=\overline{\rho c}/\bar{\rho}+c''$. The gradient transport approximation requires length and time scales of turbulence to be small in comparison with the corresponding scales of the mean flow, conditions which are rarely satisfied.

If a balance equation is constructed for a component of a turbulent transport flux, then an expression in the form of Eq. (1) may be derived from it. To do so, terms in the balance equation must be equated so that a term representing production due to a mean gradient is set equal to a term describing dissipation either by molecular motion or pressure fluctuation effects. However, experiments in cold, incompressible turbulent flows show that in most circumstances this assumption is not valid. The gradient transport description of turbulence is then purely empirical. Its application to radically different types of flow is an extrapolation which must be viewed with caution.

Turbulent reacting flows are radically different from other turbulent flows. They introduce new similarity such as Damköhler numbers and heat release parameters providing measures of the density changes involved, inclusion of which raises questions concerning the simple dimensional arguments on which so much incompressible turbulent flow modeling is based.[1] It is therefore far from obvious that empiricism

developed in cold flows can be carried over without modification to turbulent combustion. This is particularly true of turbulent transport for the reason that a turbulent flame continually generates large variations in density. Any pressure gradient in the flow can be expected to accelerate preferentially low-density packages, either augmenting or diminishing the turbulent flux depending on the relative orientation of the pressure and density gradients. This effect is absent in constant density turbulent mixing and cannot be described by gradient transport. Bilger[2] makes suggestions along these lines; he considers the influence of pressure-velocity correlation in the conservation equation for turbulent kinetic energy, indicates that a term of the form $\overline{u_k''}(\partial\bar{p}/\partial x_k)$ can either increase or decrease that energy, and discusses qualitatively a shadowgraph of a gaseous diffusion flame which indicates a decrease in energy.

The effect of pressure gradients can be studied through the isothermal mixing of dissimilar gases, although—unlike a flame—the density variations in such mixing tend to be rapidly reduced. However, experiments involving helium-air mixing[3,4] provide evidence of a turbulent flux of helium in a direction opposite to that predicted by gradient transport. Consideration of the pressure gradient effect in these flows provides at least a qualitative explanation of the observed countergradient diffusion, although no quantitative theoretical discussion of this phenomenon is available.

The objective of the present work is to develop a new description of transport processes in premixed turbulent combustion that does not depend upon an assumed gradient transport model. Preliminary work along these lines is provided by Libby,[5] but the present analysis differs in detail and scope; in particular the joint probability density function of velocity and product concentration is exploited more fully here in order to achieve a simpler formulation. Central to our considerations is inclusion of the effect of the mean pressure gradient on the intensity of the velocity fluctuations and on the flux of product. In fact one of the principal findings of the present study is that such effects are important in flames involving significant heat release.

The new description of turbulent transport is based on a balance equation for the turbulent flux of a composition variable. As this flux is a second-order covariance, the method may be described as involving second-order closure. However, it should be noted that gradient transport assumptions, generally employed in the modeling attendant with second-order closure, are avoided. The effects of turbulence upon the combustion thermochemistry are analyzed through the model of Bray and Moss[6] which applies inter alia to flames in intense turbulence and involving fast chemistry. In such circumstances the time-averaged flame zone is made up of packages of fully burned and unburned mixtures separated by narrow combustion zones. The resulting bimodality extends to the joint probability density of velocity and concentration as in Libby and Bray,[7] permitting the turbulent transport to be described in terms of the velocity fields of the burned and unburned packages; this description is the key to the avoidance of any gradient transport assumption in the present work. The result is a complete second-order closure for the turbulent composition flux in which the mean pressure gradient appears explicitly in the Favre-averaged transport equations. For the one-dimensional flames considered here the flame-generated, mean pressure gradient across the flame tends to make the turbulent flux $\overline{\rho u'' c''}$ less negative. In fact, with sufficient heat release it is predicted that this flux changes sign so that countergradient diffusion occurs.‡ The same mechanism also influences the turbulent normal stress component $\overline{\rho u''^2}$.

‡Recent experimental results by J. D. Moss to appear in *Combustion Science and Technology* confirm the existence of extensive regions involving countergradient diffusion in a premixed turbulent flame.

As is characteristic of all turbulence theories, empiricism enters in one form or another. Our earlier work introduces a single parameter involving several empirical constants which in principal could be separately determined, at least for constant density flows. However, this constant must be considered adjustable if agreement with experiment relative to a gross characteristic, turbulent flame speed is to be realized. In the present theory consistent application of the model of the joint probability density function for velocity and product concentration reduces significantly the number of empirical constants available for adjustment. However, we are able to specify arbitrarily a parameter which directly determines the turbulent flame speed. Moreover, the procedure which we adopt insures that both the propagation speed and the thickness of the turbulent flame are consistent with experiment. Thus turbulent transport is studied in a flowfield which is representative of premixed turbulent flames.

Second-order closure applied to turbulent reacting flows is widely discussed in previously published reviews of the subject, but is generally abandoned prior to specific applications. However, a second-order scheme for such flows is carried out by Varma et al.[9] in a study of mixing and reactions in chemical lasers. This investigation involves the modeling and numerical solution of a total of 30 partial differential equations, including equations for the turbulent fluxes of scalar variables similar to those considered here. However, because Reynolds rather than Favre averaging is employed, the mean pressure gradient terms highlighted in the present analysis do not appear explicitly in the turbulent flux equations. Their effects are represented implicitly, at least in the exact but unclosed equations which form the starting point for their study. However, because of the numerical and analytical complexity of the problem studied by Varma et al., it is impossible to identify mean pressure gradient effects in the results which they report.

Borghi and Dutoya[10] also use a balance equation for a turbulent flux as the starting point for their analysis which is concerned with premixed turbulent combustion. However, they do not solve the complete equation but instead assume that the production term equals the sum of two terms representing effects of pressure fluctuations and chemical reactions, respectively. In this way they recover a gradient transport expression similar to Eq. (1) in which the eddy transport coefficient ν_T is a function of the chemical reaction rate. The mean pressure gradient term is not considered.

Because our development is heavily based on previous work[1,5-8] and because space limitations preclude thorough exposition, much of the present description is brief and focuses on the new features related to turbulent transport. Readers not familiar with the present authors' model of premixed turbulent combustion and its previous application to infinite planar flames should refer to Bray and Libby.[8]

Analysis

Figure 1 indicates schematically a normal turbulent reaction zone, the coordinate x and significant quantities entering its description. Note that our analysis applies both to a normal flame as shown and to an oblique flame in which the mean streamline is unconstrained such that the velocity component parallel to the flame front is constant everywhere. The case of an oblique turbulent flame with a constrained streamline will be discussed elsewhere. The following well-known conservation equations involving Favre averaging are needed:

Mass:

$$\frac{\mathrm{d}}{\mathrm{d}x}(\bar{\rho}\tilde{u}) = 0 \tag{2}$$

Momentum:

$$\frac{\mathrm{d}}{\mathrm{d}x}(\bar{\rho}\tilde{u}^2 + \overline{\rho u''^2}) = -\frac{\mathrm{d}\bar{p}}{\mathrm{d}x} \tag{3}$$

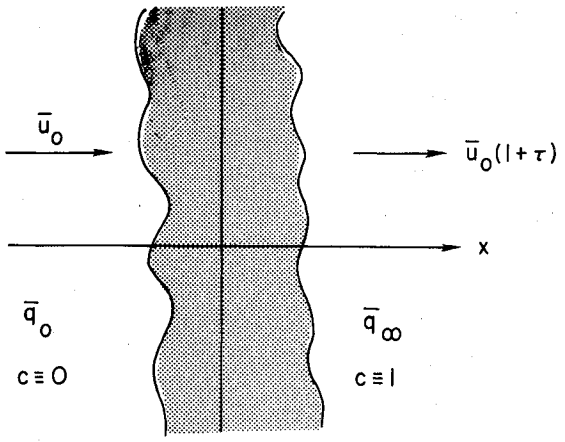


Fig. 1 Schmatic representation of normal turbulent reaction zone including coordinate and key quantities.

Species:

$$\frac{d}{dx}(\bar{\rho}\tilde{u}\tilde{c}+\overline{\rho u''c''}) = \bar{w} \tag{4}$$

Velocity intensity:

$$\frac{d}{dx}\left(\bar{\rho}\tilde{u}\frac{\overline{\rho u''^2}}{\bar{\rho}}+\overline{\rho u''^3}\right)+2\overline{\rho u''^2}\frac{d\tilde{u}}{dx} = -2\overline{u''}\frac{d\bar{p}}{dx}-\bar{\chi}_u \tag{5}$$

Species intensity:

$$\frac{d}{dx}\left(\bar{\rho}\tilde{u}\frac{\overline{\rho c''^2}}{\bar{\rho}}+\overline{\rho u''c''^2}\right)+2\overline{\rho u''c''}\frac{d\tilde{c}}{dx} = 2\overline{c''w}-\bar{\chi}_c \tag{6}$$

Species flux:

$$\frac{d}{dx}\left(\bar{\rho}\tilde{u}\frac{\overline{\rho u''c''}}{\bar{\rho}}+\overline{\rho u''^2c''}\right)+\overline{\rho u''c''}\frac{d\tilde{u}}{dx}$$

$$+\overline{\rho u''^2}\frac{d\tilde{c}}{dx} = -\overline{c''}\frac{d\bar{p}}{dx}+\overline{u''w}-\bar{\chi}_{uc} \tag{7}$$

Transverse velocity intensity:

$$\frac{d}{dx}\left(\bar{\rho}\tilde{u}\frac{\overline{\rho v''^2}}{\bar{\rho}}+\overline{\rho u''v''^2}\right)=0 \tag{8}$$

Several comments regarding these equations are necessary. Molecular transport terms are neglected throughout under the assumption of intense turbulence, i.e., of high-turbulence Reynolds numbers. However, molecular effects enter in the several dissipation terms: $\bar{\chi}_u$, the velocity dissipation; $\bar{\chi}_c$, the scalar dissipation; and $\bar{\chi}_{uc}$, the cross dissipation. All influence of pressure fluctuations is neglected. It is anticipated from modeling applied to constant density flows that the pressure fluctuation terms in these equations lead to only small effects within the narrow turbulent flames considered here. The variable c is a progress variable indicating the extent of reaction in a chemical system of arbitrary complexity[11]; it may also be considered as the mass fraction of product normalized to the value of unity when reaction is complete. We term c the product concentration, but its more general interpretation should be kept in mind.

Equations of State

The state of the gas is expressible in terms of c, namely

$$\rho/\rho_0 = (1+\tau c)^{-1} \tag{9}$$

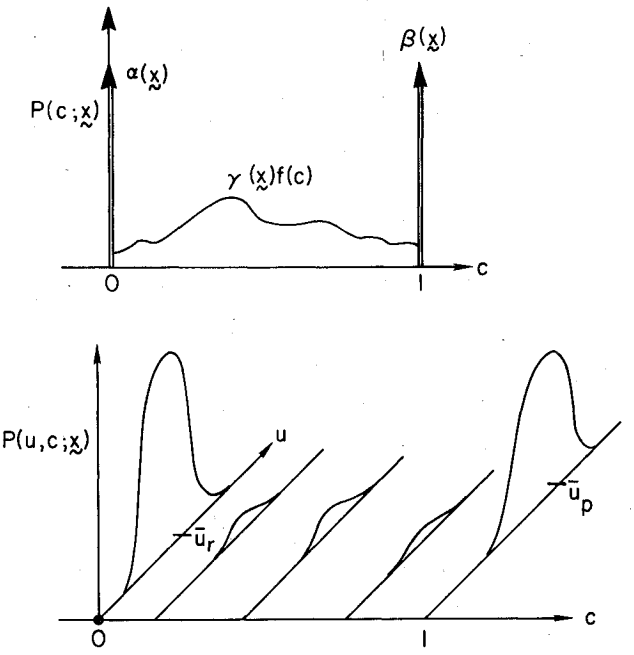


Fig. 2 Probability density functions.

$$T/T_0 = 1+\tau c \tag{10}$$

where τ is a heat release parameter which may be related to the temperature ratio $T_\infty/T_0 = 1+\tau$ from Eq. (10). Note that the variation of pressure is not thermodynamically significant and that the mixture molecular weight is assumed constant.

From Eq. (9) we have

$$\bar{\rho}/\rho_0 = (1+\tau\tilde{c})^{-1} \tag{11}$$

and from Eq. (2)

$$\tilde{u}/\tilde{u}_0 = \rho_0/\bar{\rho} = (1+\tau\tilde{c}) \tag{12}$$

Equations (11) and (12) are used repeatedly in the analysis.

Review of the Bray-Moss-Libby Model

Developing a closed set of equations for the description of the turbulent flame shown in Fig. 1 from Eqs. (2-8) requires the expression of several quantities in terms of the principal dependent variables. We determine these expressions by application of the model of premixed combustion first given in Libby and Bray[7] and subsequently utilized by Libby[5] and Libby and Bray.[12] For completeness we include here a brief description of this extended model.

Figure 2 shows the probability density functions at a particular point within the reaction zone: the single variable $P(c;x)$ and the joint $P(u,c;x)$. Central to the model is the dominance of the entries at $c=0,1$ corresponding to unburned reactants ($c=0$) and fully burned products ($c=1$). If $\alpha(x)$, $\beta(x)$, and $\gamma(x)$ denote the strengths of the three modes of entry corresponding to $c=0,1$ and $0<c<1$, respectively, then $\alpha+\beta+\gamma=1$; the model assumes fast chemistry so that $\gamma \ll 1$.

Several consequences follow: the strengths $\alpha(x)$ and $\beta(x)$ are given by the mean product concentration $\tilde{c}(x)$ according to

$$\alpha = (1-\tilde{c})/(1+\tau\tilde{c}) + O(\gamma)$$

$$\beta = (1+\tau)\tilde{c}/(1+\tau\tilde{c}) + O(\gamma)$$

where we remove the functional dependence on x as no longer being required for clarity. A further consequence is that all correlations involving only state variables are simply related

to $\tilde{c}$.[7] Of greatest utility is the intensity of product fluctuations which is

$$\overline{\rho c''^2}/\bar{\rho} = \tilde{c}(1-\tilde{c}) + O(\gamma) \tag{13}$$

Furthermore, c becomes effectively a conditioning variable, analogous to the intermittency function identifying irrotational and turbulent fluid[13,14] so that, e.g., the statistics of the velocity within reactants alone and within products alone can be expressed in terms of the conditioned probability densities $P(u,0)$ and $P(u,1)$ shown in Fig. 2. Thus, e.g., we introduce

$$\tilde{u}_r \equiv \int_{-\infty}^{\infty} du\, uP(u,0), \quad \tilde{u}_p \equiv \int_{-\infty}^{\infty} du\, uP(u,1) \tag{14}$$

where

$$\int_{-\infty}^{\infty} duP(u,0) = \int_{-\infty}^{\infty} duP(u,1) = 1$$

Note that we use the conventional bar to denote averages involving a single density either ρ_0 or $\rho_\infty = \rho_0/(1+\tau)$. It follows directly from the contributions to $P(u,c)$ that

$$\tilde{u} = (1-\tilde{c})\tilde{u}_r + \tilde{c}\tilde{u}_p + O(\gamma) \tag{15}$$

$$\overline{\rho u'' c''} = \bar{\rho}\tilde{c}(1-\tilde{c})(\tilde{u}_p - \tilde{u}_r) + O(\gamma) \tag{16}$$

Equation (16) is an interesting relationship between the mean flux of the product and the difference between two conditioned velocities. The gradient transport assumption legislates that $\overline{\rho u'' c''} < 0$ and thus that $\tilde{u}_r > \tilde{u}_p$. If it is recalled that with heat release the density of product is less than that of reactant, i.e., that $\rho_p = \rho_\infty = \rho_0/(1+\tau)$ and that $\rho_r = \rho_0$ (see Fig. 1), it may be expected that a small pressure drop across the flame can lead to $\tilde{u}_p > \tilde{u}_r$ and $\overline{\rho u'' c''} > 0$, a result excluded by gradient transport theory.[15] This expectation is indeed confirmed in the present theory.

Equations (15) and (16) permit $\tilde{u}_r$ and $\tilde{u}_p$ to be expressed as

$$\tilde{u}_r \cong \tilde{u} - \overline{\rho u'' c''}/\bar{\rho}(1-\tilde{c}) + O(\gamma)$$

$$\tilde{u}_p \cong \tilde{u} + \overline{\rho u'' c''}/(\bar{\rho}\tilde{c}) + O(\gamma) \tag{17}$$

Closing the Equations

We use ideas related to the conditioned velocities given by Eqs. (17) in the modeling of several terms appearing in Eqs. (2-8). Consider the flux $\overline{\rho u''^2 c''}$, we have from its definition in terms of $P(u,c)$

$$\overline{\rho u''^2 c''}/\bar{\rho} \cong \tilde{c}(1-\tilde{c})(-(\tilde{u}_r - \tilde{u})^2 + (\tilde{u}_p - \tilde{u})^2 - (\overline{u'^2})_r + (\overline{u'^2})_p) + O(\gamma) \tag{18}$$

where

$$(\overline{u'^2})_r = \int_{-\infty}^{\infty} du(u-\tilde{u}_r)^2 P(u,0)$$

$$(\overline{u'^2})_p = \int_{-\infty}^{\infty} du(u-\tilde{u}_p)^2 P(u,1)$$

To proceed further requires an approximate for either of the two conditioned intensities, $(\overline{u'^2})_r$ or $(\overline{u'^2})_p$. Several alternatives are available; here we follow a suggestion of Libby and Bray,[12] simplified slightly to achieve algebraic simplicity, and take

$$(\overline{u'^2})_r = (\overline{u'^2})_0 (\tilde{u}_0/\tilde{u})^2 \tag{19}$$

The equation yielding the unconditioned intensity in terms of the two conditioned intensities gives

$$(\overline{u'^2})_p = ((\overline{\rho u''^2}/\bar{\rho}) - (1-\tilde{c})(\overline{u'^2})_r)/\tilde{c} + O(\gamma) \tag{20}$$

which has the physically correct behavior as $\tilde{c} \sim 0,1$.

The flux term $\overline{\rho u'' c''^2}$ is obtained directly; we have from the definition of this flux in terms of $P(u,c)$ and from Eqs. (17)

$$\overline{\rho u'' c''^2} = (1-2\tilde{c})\overline{\rho u'' c''} + O(\gamma) \tag{21}$$

For the flux term $\overline{\rho u''^3}$ several assumptions can be made. In Libby[5] it is argued that $\tilde{u}\overline{\rho u''^2} \gg \overline{\rho u''^3}$ and this flux is neglected completely. For consistency we assume here that $P(u,0)$ and $P(u,1)$, the conditioned probability density functions, are nearly Gaussian so that $(\overline{u'^3})_r = (\overline{u'^3})_p = 0$. As a consequence

$$\overline{\rho u''^3}/\bar{\rho} \cong (1-\tilde{c})(3(\tilde{u}_r - \tilde{u})(\overline{u'^2})_r + (\tilde{u}_r - \tilde{u})^3) + \tilde{c}(3(\tilde{u}_p - \tilde{u})(\overline{u'^2})_p + (\tilde{u}_p - \tilde{u})^3) + O(\gamma) \tag{22}$$

and the flux in question can be expressed by means of Eqs. (17), (19), and (20) in terms of $\tilde{c}$ and $\overline{\rho u'' c''}$.

The final flux term is $\overline{\rho u'' v''^2}$ in Eq. (8); for its treatment we must contemplate the multivariable probability density function $P(u,v,c;x)$ dominated by entries at $c=0,1$, i.e., by contributions from reactants and products. We find a consistent calculation leads to the conclusion that this flux is negligible and thus that Eq. (8) implies $\overline{\rho v''^2}/\bar{\rho} = \text{const}$, i.e., the intensity of the transverse velocity component is unaltered by the flame.

We next treat the mean values of the fluctuating quantities multiplying the $d\bar{p}/dx$ terms; we find for the thermochemical system described by Eqs. (9) and (10) that

$$\overline{u''} = \tau\overline{\rho u'' c''}/\rho_0$$

$$\overline{c''} = \tau\overline{\rho c''^2}/\rho_0 = \tau\tilde{c}(1-\tilde{c})/(1+\tau\tilde{c}) + O(\gamma) \tag{23}$$

As will be seen an explicit model for the chemical source term $\bar{w}$ in Eq. (4) is not required unless the spatial structure of the flame is to be calculated.

The first of two further terms describing direct chemical effects, namely, $\overline{c'' w}$ in Eq. (6), is given by the Bray-Moss model as

$$\overline{c'' w} = \bar{w}(c_m - \tilde{c}) \tag{24}$$

where c_m is the ratio of two moments involving $f(c)$ and $w(c)$. The second of such terms, $\overline{u'' w}$ appearing in Eq. (7), is expressed with little approximation by consideration of the joint probability density functions $P(u,c)$; details are given in Libby.[5] In brief a linear distribution for the mean velocity in the burning mode between $\tilde{u}_r$ and $\tilde{u}_p$ and a normal distribution about that mean are assumed. Moreover, the chemical kinetic behavior is described as in Libby et al.,[1] i.e., as $w \propto w_{\max} c^n (1-c^{n-1})/(1+\tau c)$ where n is an integer, $n>2$, and $w_{\max}$ is the maximum value of $w(c)$. With these assumptions it is then a straightforward matter to find

$$\overline{u'' w} = -\bar{w}(\overline{\rho u'' c''}/\bar{\rho}(1-\tilde{c}))(1-\varphi_n/\tilde{c}) \tag{25}$$

where $\varphi_n = n/(1+n)$. Here we take $n=5$ so that $c_m = 0.833$.[1]

Finally, we consider the dissipation terms; we shall find that the scalar dissipation $\bar{\chi}_c$ is not required in the present analysis so attention focuses in the velocity dissipation $\bar{\chi}_u$ and the cross-dissipation $\bar{\chi}_{uc}$. Up to this point in the analysis a model to describe the nature of the time-dependent chemical reaction within the reaction zone has been essentially unnecessary since it enters only in the coefficient c_m in Eq. (24). However, Libby

and Bray[12] show that consistent application of the laminar flamelet model of the reaction zone, a model which pictures chemical reaction concentrated at laminar flame surfaces carried and strained by large-scale turbulence, leads to models of dissipation significantly different from those adopted without question from the phenomenology of constant density turbulence. Here we follow the implications of this laminar flamelet model and take

$$\tilde{\chi}_{uc} = \kappa_1 \tilde{w}\overline{\rho u'' c''}/\tilde{\rho}\tilde{c}(1-\tilde{c})$$

$$\tilde{\chi}_u = \kappa_2 \tilde{w}(\overline{\rho u'' c''}/\tilde{\rho}\tilde{c}(1-\tilde{c}))^2$$

where κ_1 and κ_2 are empirical constants.

Before discussing the consequences of the approximations leading to closure introduced in this subsection, it is worth pointing out that the Bray-Moss-Libby model of premixed combustion combined with simple, mechanistic assumptions readily avoids gradient transport approximations and leads to physically attractive models. Only the modeling of the conditioned intensities as in Eq. (19) involves somewhat separate considerations; however, in the course of the study presented here we have used several different models for this intensity without altering the results in any significant way.

The Final Equations

The numerical treatment of the equations which result from the use of these models is facilitated if the coordinate x is replaced as the independent variable by the mean product concentration $\tilde{c}$. Then Eqs. (5) and (7) determine the intensity $\overline{\rho u''^2}$ and the flux $\overline{\rho u'' c''}$, respectively; Eqs. (3) and (4) are used to eliminate the pressure gradient $(d\bar{p}/dx)$ and the chemical source term $\bar{w}$, respectively. Bray[11] shows that for the thermochemical system treated here Eqs. (3) and (6) lead to a direct algebraic relation between $\bar{w}$ and $\tilde{\chi}_c$; as $\bar{w}$ has been eliminated, Eq. (6) is not required here.

It is convenient to introduce nondimensional variables; thus, we let

$$I \equiv \overline{\rho u''^2}/\rho_0 \tilde{u}_0^2, \quad F = \overline{\rho u'' c''}/\rho_0 \tilde{u}_0$$

$$K = \overline{\rho u''^2 c''}/\rho_0 \tilde{u}_0^2, \quad L = \overline{\rho u''^3}/\rho_0 \tilde{u}_0^3 \tag{26}$$

There are special values of I of interest, namely, $I_0 \equiv (\overline{\rho u''^2})_0/\rho_0\tilde{u}_0^2 = (\overline{u'^2})_0/\tilde{u}_0^2$ (the relative-mean-square intensity in the flow approaching the flame) and $I_\infty = (\overline{\rho u''^2})_\infty/\rho_0\tilde{u}_0^2 = (\overline{u'^2})_\infty(1+\tau)/[\tilde{u}_0(1+\tau)]^2$, within a factor of $(1+\tau)$ of the relative-mean-square intensity in the flow downstream of the flame.

In terms of these variables and with $\tilde{c}$ as the independent variable, Eqs. (5) and (7) become

$$((1+\tau\tilde{c})I)' + L' + 2\tau I = 2\tau F(\tau + I') - \kappa_2(1+F')((1+\tau\tilde{c})F/\tilde{c}(1-\tilde{c}))^2 \tag{27}$$

$$((1+\tau\tilde{c})F)' + K' + \tau F + I = \tau\tilde{c}(1-\tilde{c})/(1+\tau\tilde{c})(\tau + I') - (1+F')F((1+\tau\tilde{c})/(1-\tilde{c}))(1-\varphi_n/\tilde{c}) - \kappa_1(1+F')F(1+\tau\tilde{c})/\tilde{c}(1-\tilde{c}) \tag{28}$$

In these equations we have neither expanded the terms nor rearranged them so that identification term-by-term with their primitive counterparts is readily possible.

The auxiliary functions K and L are as follows:

$$K = (1+\tau\tilde{c})\tilde{c}(1-\tilde{c})\left[\frac{1}{\tilde{c}}\left(\frac{I}{1+\tau\tilde{c}} - \frac{I_0}{(1+\tau\tilde{c})^4}\right) + (1-2\tilde{c})\left(\frac{F}{\tilde{c}(1-\tilde{c})}\right)^2\right] \tag{29}$$

$$L = (1+\tau\tilde{c})^2 F\left[\frac{3}{\tilde{c}}\left(\frac{I}{1+\tau\tilde{c}} - \frac{I_0}{(1+\tau\tilde{c})^4}\right) + (1-2\tilde{c})\left(\frac{F}{\tilde{c}(1-\tilde{c})}\right)^2\right]$$

The Boundary Conditions

At $\tilde{c}=0$, $I=I_0$ and at $\tilde{c}=1$, $I=I_\infty$. The obvious conditions on F, namely $F=0$ for $\tilde{c}=0,1$, must be sharpened because of the repeated appearance in the equations of the quotients $(F/\tilde{c})$, $F/(1-\tilde{c})$. Guidance is provided by Eq. (4) which in nondimensional form becomes

$$1 + F' = (\tilde{w}/\rho_0\tilde{u}_0)(d\tilde{c}/dx) \tag{30}$$

Now the right side is nonnegative which implies that $F' \geq -1$. Thus physically acceptable solutions require that this inequality be respected throughout the entire 0-1 range of $\tilde{c}$ including the end points.§

It is clear that we wish to specify too many boundary conditions for the order of the equations. We find that I_∞ must be considered an eigenvalue given as part of the solution but that I_0 is at our disposal. In Libby et al.[1] the data on turbulent flame speed $\tilde{u}_0$ from a wide variety of experiments are shown to be well-correlated by¶

$$(\tilde{u}_0/u_\ell) - 1 = 1.14(\tilde{q}_0^{1/2}/u_\ell) \tag{31}$$

where u_ℓ is the relevant laminar flame speed and $\tilde{q}_0$ is the Favre-averaged turbulent kinetic energy in the flow upstream of the flame, i.e., $\tilde{q} = \frac{1}{2}\rho\overline{u_k'' u_k''}/\bar{\rho}$ and $\tilde{q}_0 = 3/2(\overline{u'^2})_0$, provided the turbulence in the approaching stream is assumed to be isotropic. From the definition of I_0 it is thus easy to find that

$$(\tilde{u}_0/u_\ell) \cong (\tilde{u}_0/u_\ell) - 1 = \left(\frac{2}{3I_0}\right)^{1/2}(\tilde{q}_0^{1/2}/u_\ell) \tag{32}$$

so that if $I_0 = 0.51$ the present theory yields a turbulent flame speed in agreement with experiment.

The behavior of the solutions at the ends of the 0-1 range is obtained from appropriate expansions. We find for $\tilde{c} \sim 0$

$$I \sim (I_0 - I_\infty)(1+\beta_0\tilde{c}) + I_\infty$$

$$F \sim \gamma_0\tilde{c} \tag{33}$$

When these assumed forms are substituted into Eqs. (27) and (28) and the constant terms collected, we find two algebraic equations for β_0 and γ_0. We have no difficulty in selecting the roots which smoothly fair into the numerical solutions away from $\tilde{c}=0$.

Similar expansions apply to $\tilde{c} \sim 1$; where we find an additional acceptable solution so that

$$I \sim (I_0 - I_\infty)(\beta_1(1-\tilde{c}) + BC(1-\tilde{c})^{\alpha_1}) + I_\infty$$

$$F \sim \gamma_1(1-\tilde{c}) + C(1-\tilde{c})^{\alpha_1} \tag{34}$$

§It is interesting to note in this context that the "cold-boundary" problem, i.e., the inapplicability of the equations in the immediate vicinity of $\tilde{c}=0$, would manifest itself by $F' < -1$ as $\tilde{c} \sim 0$. None of our solutions exhibit this behavior and we therefore conclude that in contrast with our earlier theory the present approach is devoid of the "cold-boundary" problem.

¶It must be noted that there are some data yielding a parabolic dependence of the flame speed ratio on $(\tilde{q}_0^{1/2}/u_\ell)$. Our parameter I_0 can be selected to yield agreement with these data but the evidence for the linear dependence given by Eq. (31) seems more substantial and is at least provisionally preferred.

Again substitution into Eqs. (27) and (28) and collection of the constant terms yields nonlinear algebraic equations for β_1 and γ_1. Similarly, the terms proportional to $(1-\tilde{c})^{\alpha_1-1}$, provided $1<\alpha_1<2$ are the next terms in the series, yield α_1 and B. The quantity C is arbitrary. The restriction on α_1 is satisfied by all the solutions presented here. It is this second set of solutions which permits I_0 to be specified. There are no corresponding solutions in the neighborhood of $\tilde{c}\sim 0$.

The Numerical Procedure

The solutions of Eqs. (27) and (28) subject to the conditions given by Eqs. (33) and (34) are obtained as follows: The values of the parameters β_0 and γ_0 depend only on I_0 and other fixed quantities. Thus an integration to the right starting in the neighborhood of $\tilde{c}\sim 0$ is carried to an intermediate value of $\tilde{c}$, typically $\tilde{c}=0.9$. A second integration with assumed values of I_∞ and C is initiated from the neighborhood of $\tilde{c}\sim 1$ and continued to the same intermediate value. An error measure is provided by the discontinuities in I and F given by the two integrations and permits the values of I_∞ and C to be systematically altered; we use quasilinearization of both the differential equations and the boundary conditions to do so.

Solutions are characterized by values of τ and the two empirical constants, κ_1 and κ_2. The discussion in Libby and Bray[12] leads to the conclusion that $\kappa_1\cong\kappa_2$; moreover, estimates suggest that these two constants are of order unity. Accordingly, we take $\kappa_1=\kappa_2=1$ but note that the present results are insensitive to reasonable variations in this value.

Comparison with the Gradient Transport Theory

It is of interest to compare the predictions of the present analysis to those of our earlier theory involving gradient transport. We have already indicated that the turbulent flame speed implied by the value of I_0 can be made the same as that predicted in the earlier theory. In addition we can compare the ratio of the turbulent kinetic energies downstream and upstream of the reaction zone. Our earlier calculation yields $\tilde{q}_\infty/\tilde{q}_0$ which from the definitions of I_0 and I_∞ and the deduction from Eq. (8) of constant intensity $\rho\overline{v''^2}/\bar{\rho}$ is

$$\frac{\tilde{q}_\infty}{\tilde{q}_0}=\frac{(1+\tau)I_\infty+2I_0}{3I_0} \tag{35}$$

The ratio of turbulent kinetic energies is denoted Q_∞ in our previous studies.

A more instructive point of comparison relates to the distribution of the flux of product; a quantity entering our gradient transport theory is S where

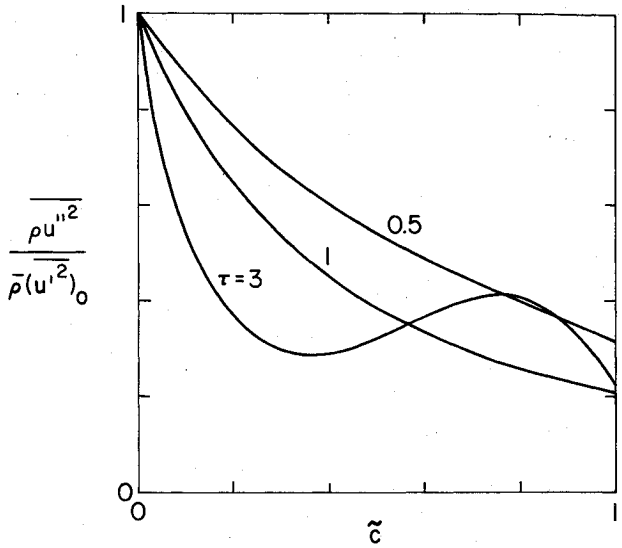
$$S=\frac{\bar{\rho}\nu_T}{\rho_0\tilde{u}_0}\frac{d\tilde{c}}{dx}=-\frac{\overline{\rho u''c''}}{\rho_0\tilde{u}_0}=-F \tag{36}$$

Thus we can compare $-S$ with F. It follows from our discussion of the absence of a "cold-boundary" problem and from our imposition of conditions at values of $\tilde{c}$ arbitrarily close to $\tilde{c}=0$ that the two theories cannot be in close agreement as $\tilde{c}\sim 0$.

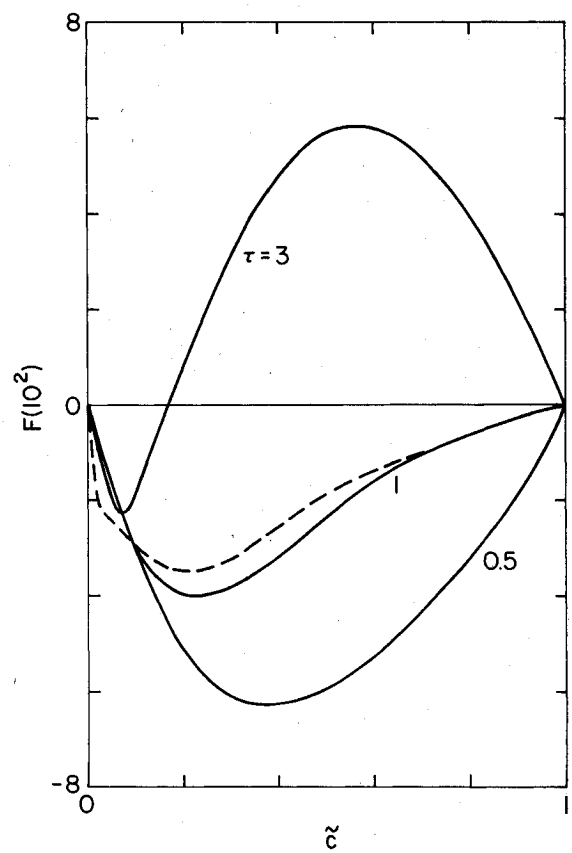
The spatial distributions of the flow variables within the reaction zone are given in each theory by Eq. (30) which readily leads to

$$x=x_0+\rho_0\tilde{u}_0\int_{c_0}^{\tilde{c}}((1+F')/\tilde{w})^{-1}d\tilde{c} \tag{37}$$

The finding of Bray[11] mentioned earlier, namely that $\tilde{w}$ and $\tilde{\chi}_c$ are simply related, permits models for either of these two quantities to be used in Eq. (37). In most of the reaction zone $|F'|\ll 1$ so that the distributions $x(\tilde{c})$ are essentially the same in the two theories. It is worth noting that there is no necessity for a turbulence length scale in the present work; an intractable problem is therefore avoided.



a) Distribution of intensity and velocity fluctuations.



b) Distributions of mean flux of product.

Fig. 3 Solutions:—present theory, – – – gradient transport theory $\tau=1$, from Libby and Bray.[7]

Presentation and Discussion of Results

Results

We present results for a range of τ from 0.5 to three corresponding to low and moderate rates of heat release. In Fig. 3a we show the distribution with respect to $\tilde{c}$ of the velocity intensity in terms of $\rho\overline{u''^2}/\bar{\rho}(\overline{u'^2})_0$, the ratio of local intensity to that in the stream upstream of the flame. For low rates of heat release the velocity intensity monotonically decreases across the flame, reflecting the effect of dilatation associated with the term $\rho\overline{u''^2}(d\tilde{u}/dx)$ in Eq. (5), an effect pointed out in our earlier studies.[8] However, for higher heat

release the monotonic decrease is destroyed and an increase in intensity occurs in the high-temperature regions of the flame. This effect is due to the pressure gradient term $\overline{u''}(d\bar{p}/dx)$ in Eq. (5); thus we see that interaction of the pressure gradient with the density inhomogeneities counteracts dilatation.**

Figure 3b gives the corresponding distributions of the mean flux of product and the predictions of our earlier gradient transport theory for $\tau=1$. We see that the new theory yields a distribution of mean flux of product in remarkable agreement with that of the previous theory, remarkable in view of the different genesis of the two theories and of the different conditions imposed at the upstream edges of the flame. As τ increases regions of counterdiffusion of product, i.e., $F>0$, arise. In fact for $\tau=3$ the mean flux is in the direction of increasing mean product throughout the flame except for a region near the upstream edge. That this effect is due to the pressure gradient term $\overline{c''}(d\bar{p}/dx)$ in Eq. (7) and that the increase in intensity shown in Fig. 3a is due to the corresponding pressure gradient term can be readily established by repeating the calculations with the pressure gradient terms suppressed; the results show negative values of the mean flux F and a monotonic decrease in the intensity of the velocity fluctuations throughout the flame.

The increasing influence of mean pressure gradient as τ increases is clarified by examination of the pressure term in Eq. (28); this term is $-\tau\tilde{c}(1-\tilde{c})(\tau+I')/(1+\tau\tilde{c})$ on the left side of the equation. Except in the region in the neighborhood of $\tilde{c}\sim 0$, $|I'|\ll 1$ so that this term is roughly $-\tau^2\tilde{c}(1-\tilde{c})/(1+\tau\tilde{c})$ and must be considered a pure forcing term with a maximum value of approximately $-\frac{1}{4}\tau^2/(1+\frac{1}{2}\tau)$ in the central portion of the flame, a value which increases with τ. For the cases considered here we have examined the distribution of the various contributions to the equation for $F(\tilde{c})$; space limitations prohibit a discussion of these results. Suffice it to state that for $\tau=3$ the pressure gradient effect is balanced principally by diffusion and convection.

The predicted ratio of the turbulent kinetic energies downstream and upstream of the reactions zone as given by Eq. (35) does not vary greatly over the range considered in the present numerical results. For $\tau=1$ we find $\tilde{q}_\infty/\tilde{q}_0=0.74$ which compares with 0.81 given by our earlier theory.[8] However, the new theory contains the influence of pressure gradient which diminishes the effect of dilatation so that for $\tau=3$ we again find $\tilde{q}_\infty/\tilde{q}_0=0.74$, whereas the earlier theory predicts 0.66. Of more interest than these differences is the predicted anisotropy of the velocity intensities downstream of the flame. As shown in Fig. 3a a significant reduction in the intensity of the x-wise velocity fluctuations occurs across flame with moderate degrees of heat release, whereas the intensities of the two transverse components are unaltered. As a consequence "pancake" turbulence effectively exists downstream of such flames with the return to isotropy presumably occurring considerably further downstream under the influence of pressure fluctuations.

It is illuminating to consider the distribution of the conditioned velocities $\tilde{u}_r$ and $\tilde{u}_p$; accordingly, Fig. 4 shows the distributions of these quantities in the form $(\tilde{u}_r)/\tilde{u}-1$ and $(\tilde{u}_p)/\tilde{u}-1$ for $\tau=3$ and indicates large differences in the conditioned and unconditioned velocities. We see, for example, that near the downstream edge of the reaction zone the conditioned velocity within reactants is predicted to be roughly 20% less than the unconditioned mean. In the central region of the flame, the mean velocity within products is indicated to be roughly 10% higher than the unconditioned mean. These values are sufficiently large to be measured by application of the methods of conditioned sampling to premixed turbulent flames in the laboratory. In the cold region of the flame $\tilde{u}_r>\tilde{u}_p$ as called for by gradient transport.

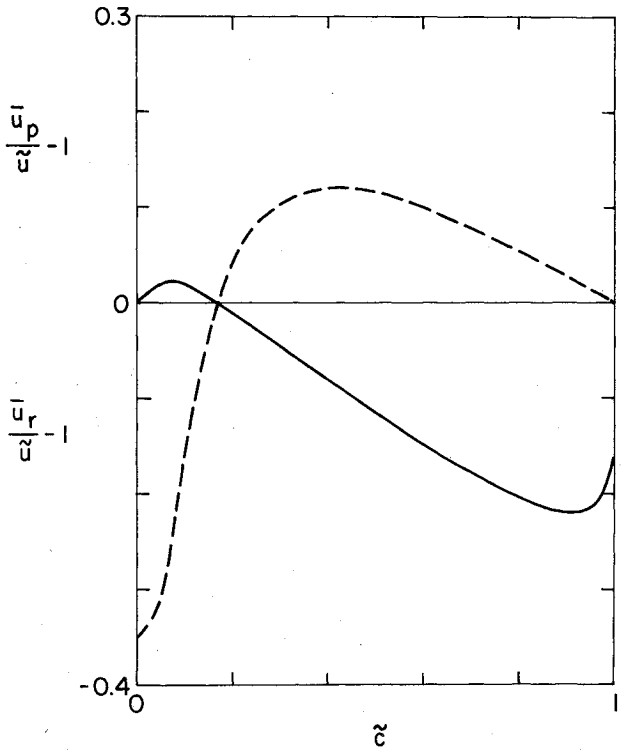


Fig. 4 Distributions of conditioned velocity, $\tau=3$:—$(\tilde{u}_r/\tilde{u})-1$, – – – $(\tilde{u}_p/\tilde{u})-1$.

Discussion

In view of these results the widely accepted arguments regarding the role of pressure in turbulent reacting flows call for re-examination. These arguments are usually as follows (see, e.g., Bray and Libby[8]): The pressure drop across a turbulent flame depends on the square of the Mach number of the upstream flow and is therefore small and certainly thermodynamically negligible. Thus the mean momentum equation is frequently considered to yield the pressure after the mean velocity and density are calculated. However, the equations for the moments and correlations of fluctuating quantities, e.g., the flux $\overline{\rho u'' c''}$, involve balances among relatively small quantities with the consequence that the mean pressure gradient, unimportant relative to mean values, becomes significant. The arguments of Bilger[2] and those relative to the observed countergradients of helium in experiments on helium-air mixing[3,4] are consistent with this view.

The important consequence of these results, both conceptually and also for the development of predictive methods for variable density turbulence, is that gradient transport approximations can be qualitatively incorrect. In the flows studied here the mean pressure decreases in the direction of decreasing density and differentially accelerates high-temperature, low-density products relative to the cold, high-density reactants; countergradient diffusion results. There are other circumstances in which the hydrodynamics of the flow can result in decreases in the mean pressure in the direction of increasing density. In these cases differential acceleration may augment and possibly dominate the mean fluxes so that again transport is not proportional to the related gradients of mean quantities.

Similar considerations apply to all mean turbulent fluxes. To illustrate this point suppose that instead of using the extended Bray-Moss-Libby model to describe the flux term $\overline{\rho u'' c''}$ appearing in the conservation equation for $\overline{\rho u'' c''}$,

**For values of $\tau\gtrsim 5$ we find the pressure gradient effect overtakes the dilatation so that $(\overline{\rho u''^2}/\bar{\rho})_\infty/(\overline{u'^2})_0>1$; thus our original discussion[8] of the influence of heat release on turbulence applies to low and modest degrees of heat release and must be modified for large values of τ. Details will be given elsewhere.

i.e., in Eq. (7), we develop a conservation equation for this flux and invoke some physical ideas regarding the dominant terms in this equation in order to make a useful estimate for this flux. There would arise in the equation for $\rho u''^2 c''$ the term $\overline{u''c''}(d\bar{p}/dx)$. For our aerothermochemical system $\overline{u''c''} \equiv \rho \overline{u''c''}/\rho_0 \quad [(1+\tau\tilde{c}) + (1-2\tilde{c})] + O(\gamma)$ with the consequence that a significant term proportional to $\tau^2 F [(1+\tau\tilde{c}) + (1-2\tilde{c})]$ and increasing in importance with heat release must be taken into account. The implication from this consideration is that the assumption of a gradient transport form for the flux term in question, i.e., for $\rho \overline{u''^2 c''} \propto -\partial \rho \overline{u''c''}/\partial x$ is suspect. Note in this regard that our model for this flux implicitly contains the pressure gradient effect via the conditioned intensities $(\overline{u'^2})_r$, $(\overline{u'^2})_p$ [see Eqs. (18) and (19)].

We thus consider that the results presented here constitute a warning as to the dangers of carrying over the empiricism developed in constant density flows to variable density flows in general and to turbulent reacting flows with significant heat release in particular.

An important but so far unsolved problem is to predict the effects of an externally imposed pressure gradient or buoyancy force on gross properties, such as the turbulent flame speed, which are accessible to experiment. The present work, in which we study turbulent transport in a flowfield representative of premixed turbulent flames, involves a priori specification of the flame speed via the parameter I_0. Modifications to the formulation are therefore required before flame speed variations can be predicted; work to this end is in progress.

When countergradient diffusion prevails over extensive regions of the reaction zone, the question arises as to the physical mechanism stabilizing the flame. From a mathematical point of view specification of boundary conditions at $\tilde{c}=1$ effectively legislates that a flame exists. Moreover, our selection of a turbulent flame speed via I_0 assures that our solutions correspond to observable flames. From a physical point of view we postulate that the flame is stabilized by the region of upstream transport of combustion product (i.e., $F<0$) which is always predicted near the cold boundary.

Finally, it is interesting to consider the terms which incorporate the effect of mean pressure gradient on turbulent transport when conventional averaging is used. This interest arises because physical effects are manifested differently in the equations employing Favre and conventional averages. Varma et al.[9] provide in an Appendix the conservation equations for turbulent multicomponent systems involving chemical reaction and conventional averaging. In the equation for the correlation $\overline{u_i' u_j'}$ there arise the two sets of terms $(\overline{\rho' u_k'}) D/Dt(\tilde{u}_j) + (\overline{\rho' u_j'}) D/Dt(\tilde{u}_k)$. When the appropriate mean momentum equations are substituted for the second factor in each set, the products of density-velocity correlations and mean pressure gradient appear along with other terms, and the mean pressure gradient effects we find important appear explicitly.

Conclusions

Transport processes in a simple premixed turbulent flame are studied by means of a model of premixed combustion and a second-order closure which carefully avoids the assumption of gradient transport. The product concentration plays the role of a conditioning variable and leads to mechanistically attractive estimates for the terms calling for modeling. Where necessary the laminar flamelet model for the chemical reaction is invoked. As a consequence the formulation results in a minimum of empirical constants; one such constant is directly related to the turbulent flame speed and is therefore chosen to assure agreement with this important gross property of turbulent flames. The other two constants relate to two dissipation terms and are estimated.

Comparison is made with the predictions of our earlier theory based on gradient transport. For low rates of heat release the new theory is in qualitative and remarkable quantitative agreement with earlier calculations; and the mean flux of product is in the direction of decreasing mean concentration. However, as the extent of heat release increases and thus the intensity of the density fluctuations increases, the differential effect of the mean pressure drop across the flame on the cold, high-density reactants and on the hot, low-density products gives rise to countergradient diffusion, i.e., to mean flux of product in the direction of increasing concentration. Thus gradient transport predicts a flux in the wrong direction. There are significant implications of this result for the prediction of variable density turbulence in general and turbulent reacting flow in particular.

It is also found that the mean pressure gradient influences the intensity of the velocity fluctuations and counteracts the reduction of that intensity by dilatation. As a consequence for the moderate degrees of heat release considered here ($\tau \leq 3$) the turbulent kinetic energy is not reduced by the heat release to the extent predicted by our earlier theory. Isotropy of the velocity fluctuations within the turbulent flame is not assumed but rather the turbulence downstream of a flame with significant heat release is predicted to be highly anisotropic. This result may also be expected to have serious implications for the development of predictive methods.

Note Added to Proof: Since completion of this paper in 1979, the theory has been developed further. Particularly interesting has been comparison with the experimental results of Moss (CST, *22*, 115 (1980)) which correspond to $\tau = 6.5$, a value sufficiently large so that the turbulent kinetic energy *increases* across the reaction zone. Comparison requires Eq. (19) to be replaced by an empirical relation and altered values of κ_1 and κ_2. With these changes "encouraging" agreement between theory and experiment is achieved. In addition extension of the theory to the case of oblique flames with constrained mean streamlines has been completed. Publication of these developments is in progress.

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