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Some Implications of Recent Theoretical Studies in Turbulent Combustion

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Nomenclature

$a_i(x)$	= coefficients in assumed probability density function, Eq. (14)
D	= molecular diffusion coefficient
$f(\xi;x)$	= interior distribution for the probability density function of ξ
h	= enthalpy
H	= step function
I_i	= intermittency functions
K	= extinction parameter, cf. Eq. (31)
ℓ	= length scale characterizing large eddies
$P(\xi;x), P(\xi, u_i;x)$	= probability density functions, single and multivariate
R_t	= turbulence Reynolds number, $\frac{1}{2}(\rho u_k'' u_k'' / \bar{p})^{1/2} \ell / \nu_0$
$S^e(\xi;x)$	= parameter for ordering reaction rates, cf. Eq. (17)
T	= temperature
u_i	= Cartesian velocity components
\dot{w}_i	= mass rate of production per unit volume of the i th species
$\dot{w}_{ik}^e, \dot{w}_{ikl}^e$	= contribution to the production of the i th trace species from trace species, cf. Eqs. (21) and (22)
x_i	= Cartesian coordinate
Y_i	= mass fraction of the i th species
Z_i	= mass fraction of the i th element
δ	= delta function
ρ	= mass density
ξ	= mixture variable, cf. Eq. (1)
$\chi_{\xi\xi}, \chi_{u_i u_i}, \chi_c$	= dissipation terms
ν	= kinematic viscosity
μ	= viscosity coefficient
<i>Subscripts</i>	
1,2	= conditions in the primary jet and secondary flow, cf. Fig. 1

I. Introduction

THERE are various reasons for continuing interest in turbulent reacting flows. Increasingly stringent requirements of improved efficiency, of reduced emissions, and of the use of alternative fuels in various combustion devices call for improved understanding of the processes involved and for improved predictive capability. Advances in diagnostic techniques¹ for acquiring and analyzing data that is spatially resolved and that leads to new information on the statistics of turbulent reacting flows call for accompanying progress in the related theory. Finally theoretical advances related to the treatment of nonreacting turbulent flows suggest improvements in the theory of turbulent combustion.

As a consequence of this continuing interest a variety of workshops, specialized meetings, etc. are held regularly and a large number of papers on the subject appear in the technical literature.² Difficulties in obtaining perspective on developments in the subject are ameliorated only partially by reviews of work prior to 1976 (Libby and Williams³ and Hill⁴). Jones⁵ provides a more recent survey and a comprehensive, tutorial presentation of the theory will soon appear (Libby and Williams⁶). The purpose of the present paper is to provide an overview of the current status of the theory with emphasis on those aspects requiring additional attention if predictions of practical importance are to be made with confidence. There is an overlap with the discussion of Jones⁵ but the emphasis and details here are different.

One motivation for this review is criticism from some quarters to the effect that turbulent combustion is attracting excessive attention. It is claimed that some important problems in combustion are being neglected because of this attention, that some problems currently studied in turbulent combustion are of no practical importance, and that diagnostic methods remain far from producing useful experimental results. Our effort to make a technical statement of the cases that can and cannot be dealt with confidently in

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turbulent combustion may provide a perspective for judgment of the validity of these objections.

II. General Evaluations and Outline of the Discussion

It is conceded at the outset that predictions of efficiencies and emission characteristics of combustors utilizing granular propellants,⁷⁻⁹ pulverized fuels,¹⁰ or heavy sprays¹¹ will not be achievable from fundamental considerations in the immediate future; the complexities of turbulence in two-phase flows undergoing chemical reaction preclude such achievements. Although fundamental research on turbulence in multiphase systems offers gains both in the long and short term, empiricism will continue to be required to obtain results of practical interest in the short term.¹¹ In view of this situation attention is focused here on turbulent combustion involving a single phase.

Radiative transport is another phenomenon that is difficult to include in fundamental studies of turbulent combustion. It is most likely to be important in multiphase systems and may influence turbulent dynamics in extreme cases. Although procedures are available for estimating the rates of radiant energy loss,^{12,13} methods for making accurate calculations from fundamental considerations for turbulent flows are unavailable. Thus, we exclude radiative transport from our discussion.

Buoyancy and the various effects associated with high Mach numbers introduce additional complications in turbulent reacting flows. The former is important in fires and the latter effects in supersonic combustion. Existing methods for the calculation of reacting flows involving either buoyancy effects or high speeds employ descriptions of the turbulence adopted from nonreacting, low-speed flows, a dubious although perhaps provisionally necessary practice.^{14,15} Again we exclude these effects from consideration.

In spite of the restrictions introduced here many important and interesting problems remain for discussion. Gas-fired combustors as well as liquid-fueled burners employing fuels of high volatility involve the turbulent combustion of gases with small influence from radiative transfer, buoyancy, or high-speed effects. Furthermore, spark-ignition and turbojet engines and stationary power plants employ combustors within which the adopted restrictions apply. It is thus useful to discuss, relative to the turbulent combustion in such systems, the predictions that can and cannot be made with confidence.

It is convenient to categorize the problems connected with turbulent reacting flows into those associated with chemical behavior and with fluid mechanical behavior. These two aspects are treated in separate subsequent sections. For the discussion of the chemical aspects it is useful to identify two limiting cases of nonpremixed and premixed reactants. Our discussion of these two cases largely follows the contributions of Bilger^{16,17} and Bray,^{18,19} respectively. After consideration of the treatment of the chemical source terms, attention is turned to the influences of heat release on the turbulence, e.g., influences associated with density variations. The main conclusion of this latter discussion is that prudence necessitates healthy skepticism regarding the present practice of carrying over to turbulent combustion the models and notions reasonably well established for constant-density turbulence.

Prior to embarking on this review it is of interest to discuss briefly the current approaches to the theory of turbulent combustion.

III. Classification of Theoretical Approaches

Five distinct methods for the prediction of turbulent combustion may be identified.²⁰ Several of the methods employ the moment approach but we make a distinction among them on the basis of the point of view adopted in the

modeling required to achieve a closed set of equations. The most common method involves the classical moment approach in which no special attention is paid to the probability density functions (pdf's) for the various fluid mechanical and thermochemical variables in the flow under consideration. Thus intermittency and definite bounds on some of the variables are largely ignored. Higher moments are modeled by analogy with nonreacting, constant-density turbulence. It is known³ that chemical reaction introduces some severe questions concerning this approach and the following discussions will raise additional questions. Thus application of this method to the problems of greatest practical interest suffers from substantial uncertainties; however, there are flows, e.g., in laboratory situations and in the chemical processing industry, in which the fluctuations of composition and temperature are sufficiently small so that these uncertainties are not significant and, thus, that this first method is applicable. It should be noted that as the activation temperature associated with the operative chemical kinetic rates increases the acceptable level of fluctuations in temperature decreases.

A variant of the first method involves introduction of physically motivated approximations based on single and joint pdf's of appropriate variables. Some fundamental objections are thereby removed and more plausible models achieved. Much of our discussion concerns this method; we shall note the successes achieved to date and the difficulties requiring attention.

In some flows it is possible to avoid modeling hypotheses through the use of formal expansions for values of the parameters determining the fluid mechanical and thermochemical behavior of the flow. The third approach has achieved a few successes and will be discussed later. However, as is to be expected relatively limited classes of problems currently are tractable by such formal procedures.

A fourth method attempts solution of the differential equations for the evolution of the pdf's defining the thermochemical state of the flow. This method is not discussed here since it is admirably assessed by O'Brien²¹ (see also Ref. 22). He concludes that for simple geometries and simple thermochemical systems this approach may provide a useful gage against which methods of greater engineering utility may be carefully assessed.

The final class of methods we identify involves consideration of coherent flow structures within which calculable chemical reaction occurs.²³⁻²⁵ Attention then focuses on the descriptions of the random behavior of these structures. Such approaches are not considered here either because they are not developed yet in generally usable form or because thorough, well-posed mathematical formalisms have not been defined for them.

IV. Chemistry Problem

An idealization of flow configurations of practical interest providing a convenient vehicle for identifying limiting cases of interest is shown schematically in Fig. 1. A primary jet issues smoothly into a secondary stream with the subscripts 1 and 2 identifying quantities in the two streams.

Nonpremixed systems may be typified by considering the primary jet to be fuel and the secondary flow to be oxidizer. For premixed combustion the primary jet may be viewed as the hot products of combustion and the secondary stream as cold, premixed reactants. An intermediate situation arises when the primary jet is fuel and the secondary stream is a fuel-oxidizer mixture. Although calculations based on moment methods for this intermediate case have been published, the fundamental validity of such calculations remains open to question. Although improved methods for this situation may be developed in the near future, our discussion is restricted mainly to the nonpremixed and premixed limits.

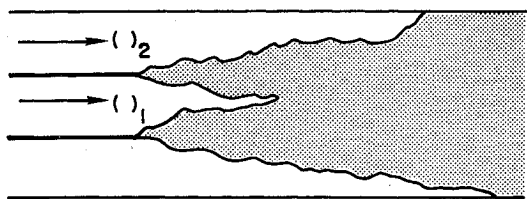


Fig. 1 Schematic representation of an idealized turbulent reacting flow.

If the operative chemical reaction rates are small compared with the rates of mixing, then both nonpremixed and intermediate cases become premixed prior to reaction. This condition has been described as that of a well-stirred reactor. If, moreover, fluctuations in temperature and composition are sufficiently small, the influence of turbulence on chemical reaction is small and simple moment methods suffice. Methods for predicting with confidence turbulent flows with complex kinetic schemes are available for such problems.²⁶ Unfortunately, the restrictions required for their applicability are seldom satisfied in the combustion devices cited earlier.

If some of the operative chemical rates are large compared with the rates of mixing, then the cases of nonpremixed and premixed reactants must be distinguished. For each of these limits advances have been made recently for the treatment of the limiting case of fast chemistry. These advances are now discussed as well as potential extensions to cases involving finite-rate chemistry. Nonpremixed systems are treated first; consideration of premixed systems follows. In these sections attention is focused mainly on the mean rates of chemical production with discussion of fluid mechanical effects deferred.

Nonpremixed Reactants in Chemical Equilibrium

In Fig. 1 let the primary jet be fuel, the secondary flow be oxidizer, and let each be statistically stationary and in thermochemical equilibrium with uniform, nonfluctuating composition and enthalpy. We are concerned with long term behavior and thus with statistically stationary conditions. For thermochemical purposes let the pressure p be spatially uniform and temporally constant; we note that pressure variations of small percentage often are significant for turbulent transport of various quantities but thermochemically unimportant. Finally, describe molecular transport in terms of a single diffusivity with a molecular Lewis number of unity. Large values of the turbulence Reynolds number R_t tend to enhance the utility of this last approximation by making the turbulent transport dominant. It should be noted, however, that because of the strong increase in kinematic viscosity with temperature (roughly as T^2), this Reynolds number may not be large in high temperature portions of a reacting flow and influences of differing molecular diffusivities may be locally significant.

The restrictions on the nature of the two streams pertain only to their thermochemical uniformity and to their being in chemical equilibrium; for example, they may consist of mixtures of fuel and oxidizer with products of previous combustion.

The duct walls shown in Fig. 1 may be sufficiently remote for their presence to be negligible; otherwise the duct must be adiabatic, chemically inactive, and not selectively permeable to different chemical species.

It is remarkable that under these relatively lenient restrictions a simple conserved scalar goes a long way in determining the entire thermochemical state of the gas mixture.^{16,17} In particular, if all chemical reactions are everywhere fast enough to maintain complete chemical equilibrium, then knowledge relative to this scalar provides knowledge relative to the temperature and the concentrations of all species.

This result is demonstrated by observing that the time-dependent equations for the conservation of total enthalpy $h(x,t)$ and of the element mass fractions $Z_i(x,t)$ are identical in form and that the boundary conditions can be made identical by suitable normalization. Specifically, introduce the mixture variable $\xi(x,t)$ defined as

$$\xi = (h - h_2) / (h_1 - h_2) = (Z_i - Z_{i2}) / (Z_{i1} - Z_{i2}) \quad (1)$$

These equations relate h and each Z_i linearly to the variable ξ which, like h and Z_i , is given by a transport equation free of chemical source terms. As a consequence, information on the statistics of ξ immediately yields corresponding statistical information for h and Z_i . This is true irrespective of whether chemical equilibrium prevails in the flow; however, with complete chemical equilibrium Eqs. (1), the equation of state, the linear equations defining Z_i in terms of the species mass fractions Y_i , the equation relating the enthalpy to the temperature and the species mass fractions, and the nonlinear equations of chemical equilibrium determine the values of the density ρ , the temperature T , and the mass fractions Y_i for each value of ξ . Thus statistical information on the scalar ξ in this case extends to ρ , T , and Y_i . In particular, if sufficient statistical information is obtained so that an estimate for the pdf $P(\xi; \mathbf{x})$ can be made, then pdf's for h and Z_i may be constructed. Also, if a joint pdf for ξ and a velocity component u_i , i.e., for $P(\xi, u_i; \mathbf{x})$, is known, then corresponding joint pdf's involving h and Z_i may be obtained. We thus see that knowledge of ξ is in every respect equivalent to knowledge of h and Z_i and that in the case of complete chemical equilibrium this equivalence extends to all thermochemical variables.

Equations for Averages

To illustrate the utility of Eqs. (1) assume that complete chemical equilibrium prevails and that $P(\xi; \mathbf{x})$ is known either from a suitable experiment involving a passive scalar²⁷ or from theoretical estimates. Following Bilger,^{16,17} let $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$, $i = 1, \dots, N$ denote the equilibrium values of ρ , T , and Y_i in a system containing N chemical species for a particular value of ξ . The dependence of these state variables on ξ is exhibited explicitly here although clearly they depend as well on p and on the parameters h_1 , h_2 , Z_{i1} , and Z_{i2} characterizing the two streams.

Every one-point, one-time statistical property of the state variables ρ , T , and Y_i is obtainable from $P(\xi; \mathbf{x})$ under these conditions. Either conventional, i.e., volume weighted, or Favre, i.e., mass weighted, averages may be used; we use standard notation and denote a Favre averaged mean as $(\bar{\cdot})$ with the associated fluctuation as $(\cdot)''$. Thus with $(\bar{\cdot})$ denoting the usual time average, $\bar{u}_k = (\rho u_k) / \bar{\rho}$, $u_k'' = u_k - \bar{u}_k$; similar notation applies to the moments, e.g., $\bar{Y}_i''^2 = (\rho Y_i''^2) / \bar{\rho}$. Illustrative examples of various averaged quantities as determined from $P(\xi; \mathbf{x})$ are

$$(\bar{\rho} \bar{Y}_i)(\mathbf{x}) = \int_0^1 \rho^e(\xi) Y_i^e(\xi) P(\xi; \mathbf{x}) d\xi \quad (2)$$

$$\bar{\rho}(\mathbf{x}) = \int_0^1 \rho^e(\xi) P(\xi; \mathbf{x}) d\xi \quad (3)$$

$$(\bar{\rho} \bar{Y}_i''^2)(\mathbf{x}) = \int_0^1 \rho^e(\xi) Y_i^e(\xi)^2 P(\xi; \mathbf{x}) d\xi - \bar{\rho} \bar{Y}_i^2 \quad (4)$$

$$(\bar{\rho} \bar{Y}_i \bar{Y}_j)(\mathbf{x}) = \int_0^1 \rho^e(\xi) Y_i^e(\xi) Y_j^e(\xi) P(\xi; \mathbf{x}) d\xi - \bar{\rho} \bar{Y}_i \bar{Y}_j \quad (5)$$

$$(\bar{\rho} \bar{T})(\mathbf{x}) = \int_0^1 \rho^e(\xi) T^e(\xi) P(\xi; \mathbf{x}) d\xi \quad (6)$$

Computation of averages of quantities involving the chemical source terms requires special consideration because in a flowing system the mass rate of production per unit volume of the i th species, $\dot{w}_i(x, t)$, is indeterminate at chemical equilibrium; the large rates of reaction in one direction are nearly cancelled by rates in the opposing direction. Accordingly, use must be made of the conservation equation for the i th species, namely,

$$\frac{\partial}{\partial t}(\rho Y_i) + \frac{\partial}{\partial x_k}(\rho u_k Y_i) - \frac{\partial}{\partial x_k}\left(\rho D \frac{\partial Y_i}{\partial x_k}\right) = \dot{w}_i \quad (7)$$

where D denotes the molecular diffusion coefficient. If $Y_i = Y_i^e(\xi)$, then substitution into Eq. (7) leads directly to

$$\dot{w}_i = -\rho D \frac{\partial \xi}{\partial x_k} \frac{\partial \xi}{\partial x_k} \frac{d^2 Y_i^e}{d\xi^2} \quad (8)$$

Equation (8) is compromised only by the approximations related to the two streams and by the assumptions of simplified molecular transport and of chemical equilibrium. Equation (8) may be used for calculating the mean values of \dot{w}_i and of correlations involving \dot{w}_i . (See Bilger²⁸ for an interesting extension of this discussion dealing with the perturbation of species concentrations from a state of chemical equilibrium.)

Equation (8) focuses attention on the importance of the joint pdf of ξ and the magnitude of its gradient in calculating the mean rate of chemical production. In turbulence with moderate to high Reynolds numbers $\partial \xi / \partial x_k \ll \partial \xi'' / \partial x_k$ so that the magnitude of the gradient is related to the instantaneous scalar dissipation of ξ which in Favre variables is

$$\chi_{\xi\xi} \equiv 2D \frac{\partial \xi''}{\partial x_k} \frac{\partial \xi''}{\partial x_k} \quad (9)$$

Thus, in terms of the joint pdf $P(\chi_{\xi\xi}, \xi; x)$ Eq. (8) yields

$$\bar{\dot{w}}_i(x) = -\frac{1}{2} \int_0^1 d\xi \int_0^\infty \rho^e(\xi) \chi_{\xi\xi} \frac{d^2 Y_i^e}{d\xi^2} P(\chi_{\xi\xi}, \xi; x) d\chi_{\xi\xi} \quad (10)$$

The equations introduced here emphasize the importance and usefulness of statistical information on ξ . However, the essential difficulty in calculating the mean rates of chemical production by direct application of Eq. (10) is clearly seen to center on the requirement of information relative to the joint probability density of the dissipation of the fluctuations of the conserved scalar and the scalar itself. Bilger²⁹ has conjectured that $\chi_{\xi\xi}$ and ξ may be uncorrelated so that the simpler problem of two single-variable pdf's is obtained. However, in view of the difficulty of measuring $\chi_{\xi\xi}$ it may not be possible to put this conjecture on a firm basis. (See comment of O'Brien in Ref. 29.) It should be noted that if $\bar{Y}_i(x)$ is known from Eq. (2), the mean rate of production of the i th species can be deduced from the conservation equation for \bar{Y}_i provided the turbulent flux terms $\overline{\rho u_k'' Y_i''}$ are obtained in some fashion.

Background, Applicability, and Observability of the Conserved Scalar

The use of a conserved scalar formally equivalent to ξ in the description of nonpremixed, turbulent reacting flows is not new; it is discussed in an extensive literature (cf. the review in Ref. 3 and Refs. 30 and 31). However, usually the methodology relates to a simple chemical reaction $A + B \rightarrow C$ where the species denoted A is in stream 1, B is in stream 2 and the product C is formed by reaction at a fluctuating flame surface separating the two reactants. In this case there is a critical value of ξ , say ξ_c , such that A is absent for $0 < \xi < \xi_c$ and B is absent for $\xi_c < \xi < 1$. In many applications this utilization of the concept of a conserved scalar with its at-

tendant flame sheet may be reasonably accurate. However, the utility of ξ in equilibrium flows as discussed here is not restricted to such cases,¹⁷ so that more realistic chemical systems can be treated.

As indicated earlier under conditions of chemical equilibrium an appropriate number of linear and nonlinear equations is available to determine for a given flow situation the functions $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$ irrespective of the complexity of the chemical system. Effective techniques exist for solving the applicable equations for the entire range of ξ (Ref. 32). A potential difficulty with respect to condensed phases is worthy of note. Under some flow conditions the equilibrium programs predict the occurrence of condensed phases, solid or liquid or both, thereby compromising our early hypothesis of a single phase system. Significant accumulations of condensed material imply poorly diffusing, possibly radiating components in the mixture; therefore, predicted concentrations of condensed material must be sufficiently small for the conserved scalar approach to be applicable. For the combustion of hydrocarbons in air under a wide variety of conditions of practical interest condensation is acceptably small; moreover, substantial concentrations of solid carbon are predicted only if the mixture is quite rich in fuel. Equilibrium concentrations of liquid water are generally negligible in the range of ξ of interest. Since carbon production seldom proceeds to equilibrium in flames, a useful strategy to be applied when condensed carbon is predicted may be to exercise the option in equilibrium programs ruling out condensation; the result is a metastable equilibrium that may be realistic. From this point of view the conserved scalar approach enjoys wide applicability in the chemical equilibrium description of nonpremixed combustion.

Objections to the use of ξ are sometimes raised on the grounds that it is a synthetic, nonobservable variable. Since Z_i is a linear function of mass fractions of the species containing the i th element, measurement of ξ by means of concentration measurements entails determination of the concentrations of all species that are major contributors to the i th element. Current experimental capabilities³³ allow averages of ξ to be obtained in this way at various spatial locations x but to measure a pdf of ξ through individual species determination is beyond present experimental capability. In this regard it must be noted that if the assumption of simplified molecular transport is accepted as being sufficiently accurate, then measurements of passive scalars can be interpreted in terms of the conserved scalar.

A more immediate approach to the measurement of ξ in reacting flows is to seed stream 1 with suitably fine, permanent particles and to measure the intensity of the light they scatter.²⁷ This provides a direct measurement of the pdf of ξ ; combined with laser-Doppler measurements of the velocity components we are able to obtain the important joint pdf's of ξ and u_j .^{34,35} Questions concerning the accuracy with which ξ is obtained by such techniques may be raised on the grounds that at lower turbulence Reynolds numbers differences in the molecular diffusivities of gas and seed particles become significant while at high Reynolds numbers the particles may not track the gas accurately. However, these questions call for improved experimental techniques and do not in our view vitiate the observability of ξ in combustion systems. Thus we do not consider the objections to the concept of a conserved scalar on grounds of nonobservability to be substantive.

Role of Intermittency in Modeling $P(\xi; x)$

The utility of the pdf of ξ in obtaining the statistics of the state variables as indicated, e.g., by Eqs. (2-6), calls for modeling approaches to its estimation. These are usually based on the conservation equations for the mean and intensity of ξ , i.e., on

$$\frac{\partial}{\partial x_k} (\bar{\rho} \bar{u}_k \bar{\xi} + \overline{\rho u_k'' \xi''}) = 0 \quad (11)$$

and

$$\frac{\partial}{\partial x_k} (\bar{\rho} \bar{u}_k \bar{\xi}''^2 + \overline{\rho u_k'' \xi''^2}) + \overline{\rho u_k'' \xi''} \frac{\partial \bar{\xi}}{\partial x_k} = \bar{\chi}_{\xi\xi} \quad (12)$$

where molecular diffusion is omitted under the assumption of high turbulence Reynolds numbers. After introduction of suitable closure approximations into Eqs. (11) and (12), these equations yield $\bar{\xi}(x)$ and $\bar{\xi}''^2(x)$ which may then be used in estimating $P(\xi; x)$. In this connection it should be noted that Pope³⁶ indicates that the third moment $\overline{\rho \xi''^3}$ may be necessary to construct an adequate pdf. On physical grounds it might be expected that this additional moment plays a significant role, but enthusiasm for its incorporation is tempered by the need for difficult modeling in order to close the conservation equation for $\overline{\rho \xi''^3}$; unless that modeling is sufficiently accurate, improved accuracy relative to the pdf may not result.

Further steps toward modeling $P(\xi; x)$ involve consideration of intermittency. In Fig. 1 let x_1 be the coordinate measured along the centerline of the primary jet and x_2 be the coordinate normal thereto. Upstream of the end of the potential core two types of intermittency occur. For values of x_2 close to the plane of symmetry, ξ is unity for a fraction of the time equal to $(1 - \bar{I}_1)$ where \bar{I}_1 is the intermittency function defined to be zero when $\xi = 1$ and unity otherwise. For values of x_2 near the extremity of the mixing layer between the primary jet and the secondary stream a second intermittency must be considered; in this case ξ is identically zero for a fraction of the time equal to $(1 - \bar{I}_2)$ where \bar{I}_2 is defined to be zero if $\xi = 0$ and unity otherwise. The influence of \bar{I}_2 prevails at all downstream locations until the edge of the jet impinges on the duct bounding the flow. On the contrary, downstream from the end of the potential core ξ is never unity so $\bar{I}_1 = 1$.

In regions of the flow involving intermittency $P(\xi; x)$ must reflect contributions at $\xi = 0, 1$ arising from intermittency. In terms of delta and step functions P may be expressed as

$$P(\xi; x) = \delta(\xi) [1 - \bar{I}_2(x)] + \delta(1 - \xi) [1 - \bar{I}_1(x)] + [\bar{I}_1(x) + \bar{I}_2(x) - 1][H(\xi) - H(1 - \xi)]f(\xi; x) \quad (13)$$

where $f(\xi; x)$ is the pdf in the interior range $0 < \xi < 1$. The normalization

$$\int_0^1 P(\xi; x) d\xi = 1$$

leads through Eq. (13) to

$$\int_0^1 f(\xi; x) d\xi = 1$$

If intermittency is improperly disregarded, then P would be identified with f , the delta function contributions in Eq. (13) being neglected. [Note that the use of "clipped Gaussian" and other distributions that do not account for intermittency but only reflect bounds on ξ are not equivalent to Eq. (13).]

Figure 2 shows schematically the pdf's at various representative locations within our idealized flow of Fig. 1. Situations in which the delta functions play a dominant role are clearly to be expected. In this regard it is interesting to note that there are cases in premixed combustion in which the breakdown analogous to that given by Eq. (13) leads to the details $f(\xi; x)$ being unimportant, $P(\xi; x)$ being dominated by the delta functions at $\xi = 0, 1$.^{18,19}

The strengths of the delta functions needed for the determination of the intermittencies $\bar{I}_1(x)$ and $\bar{I}_2(x)$ are

*Note that our discussion implies that the flow is statistically two-dimensional; there is no conceptual difficulty relative to the case of an axisymmetric configuration.

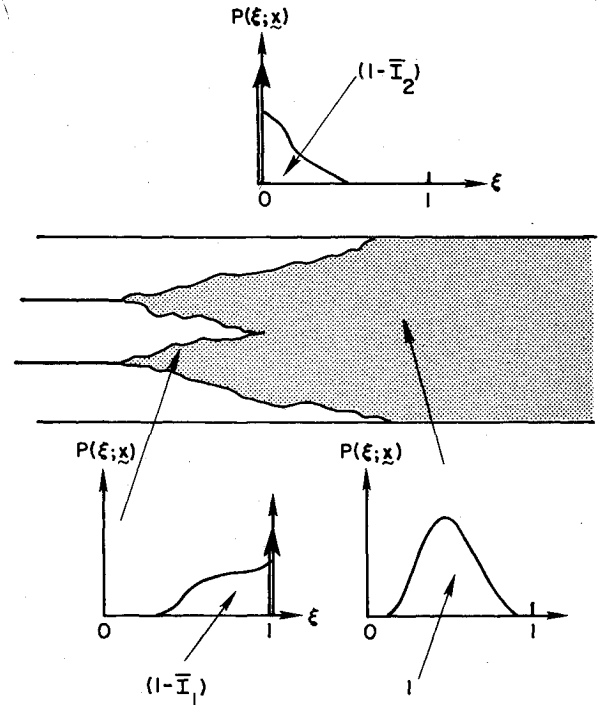


Fig. 2 Typical probability density functions of the conserved scalar in an idealized turbulent reacting flow.

currently best obtained from experiment. In the future the theory of intermittent turbulence (Libby³⁷ and Dopazo³⁸) may be developed so as to apply to more complicated flows of interest in combustion.

Two remarks regarding the delta functions appear appropriate. Experimentally such functions appear as spikes with a structure produced in part by measurement errors and contamination; some judgement is therefore required in interpreting experimentally determined pdf's. Libby et al.³⁹ introduce the notion of a "gate" in the theoretical description of the interior portion of the pdf for the purpose of suitably identifying the delta functions; their theoretical gate is analogous to the use of a signal level by experimentalists in measurements of intermittency.

Equation (13) indicates that with $\bar{I}_1(x)$ and $\bar{I}_2(x)$ known specification of $f(\xi; x)$ serves to determine the form of $P(\xi; x)$ for subsequent use. A two parameter representation of f is consistent with knowledge of the mean and intensity of ξ and is usually considered to provide a suitable pdf of ξ . A variety of functional forms for f have been considered; an example is

$$f(\xi; x) = a_0(x) \exp[-a_1(x)(\xi - a_2(x))^2] \quad (14)$$

where a_0 is related to a_1 and a_2 through the normalization condition. The functions $a_1(x)$ and $a_2(x)$ are related to $\bar{\xi}$ and $\bar{\xi}''^2$ through

$$\bar{\rho} \bar{\xi} = \int_0^1 \rho^e(\xi) \xi P(\xi; x) d\xi \quad (15)$$

$$\bar{\rho} \bar{\xi}''^2 = \int_0^1 \rho^e(\xi) \xi^2 P(\xi; x) d\xi - \bar{\rho} \bar{\xi}^2 \quad (16)$$

A virtue of Eq. (14) is that it can describe a wide variety of distributions. This may be seen as follows. If a_2 lies outside the range $0 < a_2 < 1$, then $f(\xi; x)$ resembles a decaying exponential. If $0 < a_2 < 1$ and a_1 is sufficiently large, then a Gaussian distribution results. Although the numerical effort needed to obtain $a_1(x)$ and $a_2(x)$ from Eqs. (15) and (16) may be significant, in principle these expressions complete the modeling of $P(\xi; x)$.

Knowledge of $P(\xi; \mathbf{x})$ enables the averages such as those given by Eqs. (2-6) to be calculated. However, it does not provide the average values of the source terms or correlations involving the source terms such as $\bar{Y}_i' \bar{w}_i$; joint pdf's are needed for these additional quantities as implied by Eq. (8) or (10). Nor does it give averages involving the velocity components, e.g., those needed to close Eqs. (11) and (12), $\overline{\rho u_i' Y_j'}$ or a velocity-chemical reaction correlation such as $u_i' \bar{w}_i$; these quantities involve joint pdf's for ξ and u_i or for ξ , $\chi_{\xi\xi}$, and u_j . Intermittency plays a further role in the modeling of the joint pdf $P(\chi_{\xi\xi}, \xi; \mathbf{x})$ through delta functions at $\chi_{\xi\xi} = 0$. Nevertheless, extensions of the approach outlined here could provide predictive methods based on fundamental considerations and applicable to the calculation of many properties of turbulent combustion involving nonpremixed reactants maintaining chemical equilibrium. A beginning for the treatment of two-variable pdf's is provided by Janicka and Kollman⁴⁰ but considerable additional work is required to establish confidence in this extension of current practice. An approach exploiting activation energy asymptotics is indicated by Peters et al.⁴¹

Applicability of Chemical Equilibrium

Because of the significant simplifications provided by the assumption of chemical equilibrium throughout the flow, it is important to establish the extent of its applicability. A formal criterion for such applicability at any location \mathbf{x} may be obtained from Eq. (8). Associated with a known chemical kinetic scheme is an expression for \bar{w}_i as a sum of terms, each dependent on the state variables ρ , T , and Y_i . Introduction of the equilibrium approximation allows each term to be evaluated as a function of ξ through the use of the functions $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$. Strict equilibrium at any spatial location \mathbf{x} requires each term so evaluated to be large compared with the right side of Eq. (8) for all $\xi(0 < \xi < 1)$ and for every i ($i = 1, \dots, N$) throughout the entire range of values of $\chi_{\xi\xi}$ present at \mathbf{x} . Here we assume that the specified kinetic scheme includes enough detail to permit the occurrence of equilibrium but excludes redundant alternative paths that are slow.

An equilibrium criterion in this strict form is seldom satisfied in turbulent combustion. Failure of the assumption of chemical equilibrium at the extreme values of ξ is unlikely to be significant because chemical reaction is relatively inconsequential at such values. However, failure of major reactions at values of ξ near those corresponding to the peak temperature would preclude the assumption of complete chemical equilibrium from providing a useful approximation to reality; although present estimates suggest that often such a failure does not occur for flows of practical interest, further work is needed to define better the relevant bounds on conditions under which specific chemical systems are essentially in complete equilibrium.

An approximate criterion for equilibrium in the mean involving the mean scalar dissipation is

$$\bar{w}_{ik}^e(\xi) \gg \frac{1}{2} \rho^e(\xi) \bar{\chi}_{\xi\xi} \left| \frac{d^2 Y_i^e}{d\xi^2} \right| \equiv s_i^e(\xi; \mathbf{x}) \quad (17)$$

which is to be applied for i corresponding to major species and for major reactions k over a range of values of ξ near the peak temperature. Here the subscript k identifies a positive additive contribution to \bar{w}_i ; near equilibrium such a contribution is balanced by a nearly equal, negative contribution from the corresponding reverse reaction. Improved criteria are under investigation.²⁸

Special cases may be identified in which influences of turbulence on chemical behavior may be taken into account without difficulty. Suppose that there are for the chemical system under consideration M reactions identified with the subscripts $k = 1, \dots, M$ and that $\bar{w}_{ik}^e \gg s_i^e$ for some of these reactions while $\bar{w}_{ik}^e \ll s_i^e$ for all other reactions; the inequalities are to apply for all i of interest at all values of \mathbf{x} and ξ .

When conditions permit such a categorization of the operative chemical reactions, those reactions having $\bar{w}_{ik}^e \ll s_i^e$ may be considered effectively frozen. A partial equilibrium results from assigning zero rates to these reactions while those reactions with $\bar{w}_{ik}^e \gg s_i^e$ lead to modified solutions for $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$. If the same relative ordering remains valid for the modified solutions, then the resulting partial equilibrium is given by a consistent analysis. The interpretation to be applied in this case is that the turbulence has turned off some marginal reactions. By this we mean that the time scales associated with small length scales of the turbulence are too short relative to the chemical times of these reactions for the marginal reactions to proceed to a significant extent.

The utility of this approach is limited, of course, to the possibility of categorizing the reactions as indicated by the two sets of inequalities. There may be some reactions for which \bar{w}_{ik}^e is comparable with s_i^e . It should also be noted that $\bar{w}_{ik}^e \ll s_i^e$ is a necessary but not sufficient condition for reaction k to be inoperative; if the mean flow time is sufficiently long, then the reaction associated with this reaction step may eventually occur even though it proceeds to a negligible extent over a turbulent time scale. Although this partial equilibrium approach has not been pursued, there are certain situations in which it may apply, e.g., for the rapid partial oxidation of hydrocarbons to carbon monoxide while the slower reactions that produce carbon dioxide are ineffective.

Except in fuel-rich regions estimates for hydrocarbon-air combustion indicate that complete equilibrium is a reasonable approximation for the major species.¹⁷ Similar consideration applies to hydrogen-air combustion.²⁸ Thus while temperatures and concentrations of major species are often well described by complete equilibrium, concentrations of minor species often are not. Prime examples of such species are soot and the oxides of nitrogen, both of which are produced in small, nonequilibrium concentrations by finite-rate chemical processes. Methods for calculating such nonequilibrium trace species may be formulated with relative simplicity.

Analysis of Nonequilibrium Trace Species

Of the N chemical species in the system under consideration let there be P dominant species in complete chemical equilibrium and $N-P$ trace species whose behavior is determined by finite chemical rates. Arrange the species such that $i = 1, \dots, P$ denotes the dominant species and $i = P+1, \dots, N$ the trace species. Any trace species in chemical equilibrium may be included in the first set.

Since the trace species have a negligible influence on the thermochemistry of the system, in a first approximation we may calculate the complete equilibrium of the species denoted by $i = 1, \dots, P$. In particular $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$, $i = 1, \dots, P$ may be calculated by setting $Y_i = 0$ for $i = P+1, \dots, N$. Perturbations to the resulting solutions due to the trace species may be calculated by introducing small values for Y_{P+1}, \dots, Y_N in the equations yielding $\rho^e(\xi)$, $T^e(\xi)$, and $Y_i^e(\xi)$. Functions which we denote $\rho^f(\xi, Y_{P+1}, \dots, Y_N)$, $T^f(\xi, \dots, Y_N)$, and $Y_i^f(\xi, \dots, Y_N)$, $i = 1, \dots, P$ naturally result and lead to the following expansions:

$$\begin{aligned} \rho &= \rho^e(\xi) + \sum_{k=P+1}^N \rho_k^f(\xi) Y_k \\ T &= T^e(\xi) + \sum_{k=P+1}^N T_k^f(\xi) Y_k \\ Y_i &= Y_i^e(\xi) + \sum_{k=P+1}^N Y_{ik}^f(\xi) Y_k, \quad i = 1, \dots, P \end{aligned} \quad (18)$$

where

$$\rho_k^f(\xi) = \frac{\partial \rho^f}{\partial Y_k}(\xi, Y_{P+1}, \dots, Y_N)$$

with the partial derivatives evaluated with $Y_{P+1} = \dots Y_N = 0$. Similar considerations apply to $T_k^f(\xi)$ and $Y_{ik}^f(\xi)$.

The multivariate pdf $P(\xi, Y_{P+1}, \dots, Y_N; \mathbf{x})$ is now introduced for the purposes of calculating various statistical quantities. Then, for example, from Eqs. (18) we have the expansion

$$\begin{aligned} \bar{\rho}(\mathbf{x}) = & \int_0^1 dY_N \int_0^1 \dots \int_0^1 \left[\rho^e(\xi) + \sum_{k=P+1}^N \rho_k^f(\xi) Y_k \right] P \\ & \times (\xi, Y_{P+1}, \dots, Y_N; \mathbf{x}) d\xi = \int_0^1 \rho^e(\xi) P(\xi; \mathbf{x}) d\xi \\ & + \sum_{k=P+1}^N \int_0^1 Y_k dY_k \int_0^1 \rho_k^f(\xi) P(\xi, Y_k; \mathbf{x}) d\xi \end{aligned} \quad (19)$$

We see that the bivariate pdf's $P(\xi, Y_k; \mathbf{x})$ arise; if Y_k were statistically independent of ξ , then the summation in Eq. (19) would take on the considerably simpler form

$$\sum_{k=P+1}^N \bar{Y}_k \int_0^1 \rho_k^f(\xi) P(\xi; \mathbf{x}) d\xi$$

but such independence seems unlikely to prevail for trace species being produced at rates dependent upon ξ . An example of interest in which such independence may not be expected to prevail is provided by the production of NO, a trace species, at rates dependent on the concentration of oxygen, a concentration directly related to ξ if equilibrium for oxygen is maintained. Equations similar to Eq. (19) and yielding the corrections to the thermochemical state determined by the dominant species, i.e., to $\bar{T}(\mathbf{x})$ and to $\bar{Y}_i(\mathbf{x})$ for $i=1, \dots, P$, as well as corrections to $\bar{w}_i(\mathbf{x})$ for the same sequence of $i=1, \dots, P$ values, depend on these same bivariate pdf's. For the trace species we have

$$\bar{\rho}(\mathbf{x}) \bar{Y}_k(\mathbf{x}) = \int_0^1 Y_k dY_k \int_0^1 \rho^e(\xi) P(\xi, Y_k; \mathbf{x}) d\xi, \quad k=P+1, \dots, N \quad (20)$$

Modeling of the bivariate pdf's would be needed to proceed further along these lines, i.e., to evaluate the mean concentrations of trace species by means of Eqs. (20) and such modeling can be developed.^{40,41} However, the mean production rates of trace species can often be obtained without knowledge of the bivariate pdf's and an alternative approach is therefore indicated. Indeed, since the trace species are assumed to be in nonequilibrium, their average production rates can be calculated without the complexities of Eqs. (8) and (10). In particular, substitution of the various state variables into the operative expressions for \bar{w}_i , $i=P+1, \dots, N$, leads through the use of ρ^f , T^f , and Y_i^f , $i=1, \dots, P$, to well-behaved functions $\bar{w}_i^f(\xi, Y_{P+1}, \dots, Y_N)$ for $i=P+1, \dots, N$. Expansion of this function as indicated by Eq. (18) leads to

$$\begin{aligned} \bar{w}_i = & \bar{w}_i^e(\xi) + \sum_{k=P+1}^N \bar{w}_{ik}^f(\xi) Y_k \\ & + \sum_{k=P+1}^N \sum_{\ell=P+1}^N \bar{w}_{ik\ell}^f(\xi) Y_k Y_\ell, \quad i=P+1, \dots, N \end{aligned} \quad (21)$$

where $\bar{w}_{ik}^f(\xi)$ is defined in a manner similar to $\rho_k^f(\xi)$; and where

$$\bar{w}_{ik\ell}^f(\xi) = \frac{1}{2} (2 - \delta_{k\ell}) \frac{\partial^2 \bar{w}_i^f}{\partial Y_k \partial Y_\ell} (\xi, Y_{P+1}, \dots, Y_N)$$

evaluated at $Y_{P+1} = \dots Y_N = 0$; here δ_k is the Kronecker delta. The double summation, neglected in Eq. (18), is retained in

Eq. (21) to account for the possibility that significant reactions may be second order with respect to trace species.

In the same way Eq. (19) is derived it may be shown from Eq. (21) that

$$\begin{aligned} \bar{w}_i(\mathbf{x}) = & \int_0^1 \bar{w}_i^e(\xi) P(\xi; \mathbf{x}) d\xi \\ & + \sum_{k=P+1}^N \int_0^1 Y_k dY_k \int_0^1 \bar{w}_{ik}^f(\xi) P(\xi, Y_k; \mathbf{x}) d\xi \\ & + \sum_{k=P+1}^N \sum_{\ell=P+1}^N \int_0^1 Y_\ell dY_\ell \int_0^1 Y_k dY_k \\ & \times \int_0^1 \bar{w}_{ik\ell}^f(\xi) P(\xi, Y_k, Y_\ell; \mathbf{x}) d\xi, \quad i=P+1, \dots, N \end{aligned} \quad (22)$$

where we see that trivariate pdf's appear; for $\ell=k$ the trivariate pdf becomes bivariate. When concentrations of trace species exceed their equilibrium values, the second and third terms in Eq. (22) are larger than the first and multivariate pdf's must be considered; however, trivariate pdf's occur only if reactions involving unlike trace species are important. Commonly trace species have concentrations well below equilibrium values and the first term in Eq. (22) provides a good approximation for their rates of production. The resulting formula

$$\bar{w}_i(\mathbf{x}) = \int_0^1 \bar{w}_i^e(\xi) P(\xi; \mathbf{x}) d\xi, \quad i=P+1, \dots, N \quad (23)$$

enables the average production rates of such species to be calculated from the pdf of the conserved scalar. In this respect calculation of the mean rates of production of trace species in nonequilibrium is simpler than that for species in equilibrium since the latter depends on the joint pdf in Eq. (10); according to Eq. (23) the mean rates of production of trace species do not depend on the scalar dissipation.

Comparison of Eqs. (20) and (23) verify our earlier statement that the average production rates of trace species usually can be calculated more easily than their average concentrations. These concentrations are obtained from the mean conservation equations with the chemical source term now considered known and with appropriate modeling applied to the description of turbulent transport. Such modeling is likely to employ methods paralleling those adopted for the corresponding transport of ξ although it should be emphasized that the presence of the source term makes such an approach less convincing for trace species.

Average Production Rates of NO and Soot

In this section we discuss applications of Eq. (23) to the production of NO and soot in turbulent flames. With respect to NO production we note that if the Zeldovich mechanism is adopted, for example, then calculation by means of Eq. (23) is straightforward, provided that oxygen atoms maintain equilibrium with the major species. However, inaccurate prediction will result to the extent that nonequilibrium concentrations of oxygen atoms occur. Since this nonequilibrium often is not negligible, inclusion of such atoms in the set of trace species is called for. However, this would necessitate further the inclusion of other trace species such as OH and H in that same set since the concentrations of oxygen atoms strongly depend on the concentrations of these species. The difficulty that arises is that Eq. (23) is inapplicable for the calculation of these species and the more difficult Eq. (22) must be resorted to. Accordingly, alternative approaches such as that described in the following section may be preferable for investigating the influence of nonequilibrium concentrations of oxygen atoms on NO

production. It remains quite convenient to calculate \dot{w}_i for NO by a formula similar to Eq. (23) but with a modification to \dot{w}_f accounting for nonequilibrium of oxygen atoms.²⁸

The prediction of the mean production rates for soot in turbulent flames is difficult because reliable kinetic information permitting $\dot{w}_f(\xi)$ to be calculated is unavailable. It is known that numerous reactions contribute to soot formation and that in some circumstances soot burnup proceeds simultaneously with soot formation. In this latter case Eq. (23) would be inadequate and Eq. (22) would be needed, although the third term, that involving the double summation may be negligible. It is possible that under suitable conditions Eq. (23) may describe average rates of soot production with acceptable accuracy. However, practical calculations with Eq. (23) require simplified expressions for overall rates of soot production not involving the concentrations of a large number of species. Useful data of this type currently are being obtained.⁴²

Departures from Equilibrium for Dominant Species

When the species determining the thermochemical state of the gas mixture are not in equilibrium, the conserved scalar ξ , although still determining the enthalpy h and the element mass fractions Z_i through Eq. (1), no longer establishes the complete state of the mixture; values of some additional species mass fractions Y_i must also be obtained if ρ , T , and all the dominant Y_i 's are to be calculated. Through reasonable approximations it is often sufficient to introduce just one additional variable that plays the role of a progress variable and that measures the departure of the system from equilibrium. This variable is selected most conveniently as a particular combination of various species mass fractions.^{28,40}

For illustrative purposes it will be assumed here that knowledge of ξ and Y_N is sufficient for determining all state variables. For example, it might be supposed that all Y_i 's except Y_N maintain complete equilibrium. In this case expressions for the state variables in the form $\rho = \rho^f(\xi, Y_N)$, $T = T^f(\xi, Y_N)$, and $Y_i = Y_i^f(\xi, Y_N)$, $i = 1, \dots, N-1$ are obtainable. One-point, one-time statistics for all state variables may then be expressed in terms of the bivariate pdf $P(\xi, Y_N; \mathbf{x})$. For example,

$$(\bar{\rho} \tilde{Y}_i)(\mathbf{x}) = \int_0^1 dY_N \int_0^1 \rho^f(\xi, Y_N) Y_i^f(\xi, Y_N) P(\xi, Y_N; \mathbf{x}) d\xi \quad i = 1, \dots, N-1 \quad (24)$$

$$(\bar{\rho} \tilde{Y}_i^2)(\mathbf{x}) = \int_0^1 dY_N \int_0^1 \rho^f(\xi, Y_N) (Y_i^f(\xi, Y_N))^2 P(\xi, Y_N; \mathbf{x}) d\xi - (\bar{\rho} \tilde{Y}_i^2)(\mathbf{x}), \quad i = 1, \dots, N \quad (25)$$

$$(\bar{\rho} \tilde{T})(\mathbf{x}) = \int_0^1 dY_N \int_0^1 \rho^f(\xi, Y_N) T^f(\xi, Y_N) P(\xi, Y_N; \mathbf{x}) d\xi \quad (26)$$

which are analogous to Eqs. (2), (4), and (6), respectively.

The mean rates of chemical production of the species corresponding to $i = 1, \dots, N-1$ are given by expressions more complicated than Eq. (10) due to the need to replace Eq. (8) by

$$\dot{w}_i = -\rho D \left[\frac{\partial \xi}{\partial x_k} \frac{\partial \xi}{\partial x_k} \frac{\partial^2 Y_i^f}{\partial \xi^2} + 2 \frac{\partial \xi}{\partial x_k} \frac{\partial Y_N}{\partial x_k} \frac{\partial^2 Y_i^f}{\partial \xi \partial Y_N} + \frac{\partial Y_N}{\partial x_k} \frac{\partial Y_N}{\partial x_k} \frac{\partial^2 Y_i^f}{\partial Y_N^2} \right] + \frac{\partial Y_i^f}{\partial Y_N} \dot{w}_N, \quad i = 1, \dots, N-1 \quad (27)$$

Equation (27) implies that joint pdf's involving gradients of ξ and Y_N are required for the evaluation of the mean source term \dot{w}_i . The direct contribution to these mean rates from Y_N

is given by the last term in Eq. (27) and becomes

$$\int_0^1 dY_N \int_0^1 \frac{\partial Y_i^f}{\partial Y_N} \dot{w}_N P(\xi, Y_N; \mathbf{x}) d\xi \quad i = 1, \dots, N-1 \quad (28)$$

where $\dot{w}_N = \dot{w}_N^f(\xi, Y_N)$ is obtainable in terms of ξ and Y_N directly from the rate expression for the N th species. Calculation of the mean rate of production of the N th species is more straightforward, namely by use of the equation

$$\dot{w}_N(\mathbf{x}) = \int_0^1 dY_N \int_0^1 \dot{w}_N^f(\xi, Y_N) P(\xi, Y_N; \mathbf{x}) d\xi \quad (29)$$

The essential difficulty in calculations of this nature is that the bivariate pdf $P(\xi, Y_N; \mathbf{x})$ plays a central role in all statistics. It is appreciably more difficult to make estimates for this pdf than for the single variable pdf of the conserved scalar $P(\xi; \mathbf{x})$. Study has shown that in some cases ξ and Y_N are approximately statistically independent here, and the resulting ξ dependence is relatively weak so that only $P(Y_N; \mathbf{x})$ enters in strong manner.²⁸ Procedures that have been applied along these lines⁴⁰ involve describing $P(\xi, Y_N; \mathbf{x})$ in terms of the mean values of ξ and Y_N and of low moments thereof. Appropriate conservation equations, e.g., that for \tilde{Y}_N , are solved along with Eqs. (11) and (12) to determine the spatial distributions of the parameters in the pdf. Since the significant uncertainties involved in this procedure can be readily appreciated, it is recognized that considerable work remains relative to the treatment from a fundamental point of view of finite rate chemical reactions in nonpremixed systems. Although the problems are difficult, in our view there is promise for significant progress on their resolution in the near future.

Premixed Reactants Undergoing Fast Chemical Reactions

By definition the mixture variable in premixed systems has the same, constant value at each point in the flow with the consequences that it loses its utility and that the definition of ξ as given by Eq. (1) becomes meaningless. Instead, progress variables such as Y_N introduced in the previous section become useful. In flows for which knowledge of one such progress variable determines fully the thermochemical state of the system there evidently exists an analogy between that progress variable and the conserved scalar ξ , although their basic meanings are quite different.

A necessary condition for the density, temperature, and concentrations to be described in terms of one progress variable is that heat losses must be negligible, often a reasonable first approximation. Relative to the flow shown in Fig. 1 this condition requires that the primary jet consist of the products of the complete adiabatic combustion of the reactants in the secondary stream. The previously imposed requirement of adiabatic duct walls must prevail in this case as well. Then under the same simplifying assumptions regarding the pressure and molecular transport properties introduced earlier, the equations for conservation of energy and elements take on the simple forms

$$h = h_1 = h_2 \quad Z_i = Z_{i1} = Z_{i2} \quad (30)$$

Despite the simplicity implied by Eqs. (30) efforts to develop a sound treatment of the chemistry in premixed systems meet greater fundamental difficulties than those in nonpremixed systems. The notion of complete chemical equilibrium is inapplicable because one of the inlet streams, e.g., the secondary stream in Fig. 1, involves a reactant mixture that at best may be assumed to be in a metastable state destroyed when the reactants are heated by the products from previously burned reactants. Conditions of practical interest permitting formal expansions about an equilibrium state seldom prevail. In the most typical situation rates of

reaction are negligibly small in part of the flow and sufficiently rapid to maintain near equilibrium in other parts. The edge of the region at which rates are negligible is the upstream boundary of the turbulent flame that propagates at the turbulent flame speed with respect to the average motion of the oncoming reactants. Although at high turbulence Reynolds numbers turbulent flame speeds depend strongly on the fluid mechanics of turbulent mixing, effects of chemical rates, known to be significant for laminar flames, are found experimentally not to be entirely irrelevant to the propagation rates of turbulent flames.

The primitive moment methods mentioned at the beginning of this section are unlikely to be useful in premixed turbulent combustion. An approximation that has been found to be effective is that of fast chemistry. This terminology has different meanings for premixed and nonpremixed flows; in the premixed case complete equilibrium or near equilibrium is not maintained everywhere but, instead, once reactions begin they proceed rapidly to completion and establish an equilibrium state. This approximation is justified if all the chemical kinetic rates depend strongly on the extent of the reaction, e.g., if the reactions are represented as one-step processes with a value of the overall activation energy large compared with the thermal enthalpy of the burned gas. However, the fast chemistry approximation can apply to complex chemical systems and is not restricted to one-step processes.¹⁸

The fast chemistry approximation has been applied in two ways, through perturbation methods and through approximation of the pdf for a progress variable. These ways are discussed in the following sections.

Developments in Theories of Wrinkled Laminar Flames

Turbulent flames have often been discussed qualitatively as being composed of laminar flames with oscillating, wrinkled shapes imposed by the turbulence. Formal justification for this view and the limits on its validity may be studied by analyzing the structures and motions of such flames in turbulent flows. For the calculation of overall properties such as turbulent flame speeds, realistic complex chemistry may often be reasonably approximated as a one-step reaction with parameters in the rate expression obtained from measurements of the related laminar flame speed. Typically the values for the overall activation energy are large compared with the thermal enthalpies: ratios of energies greater than ten. In the present context the fast chemistry approximation involves treating this ratio as a large parameter while simultaneously assuming that magnitudes of reaction rates are sufficiently large so as to assure chemical reaction somewhere in the flow.

Singular perturbation techniques show that the laminar flame consists of a broad convective-diffusive zone on its upstream face followed by a narrow reactive-diffusive zone.⁴³ This same type of singular perturbation may be applied to turbulent flames if methods are devised to accommodate this laminar flame structure within a description of the turbulence. To date the only method developed along these lines is based on a perturbation for small gradients; all length scales for the turbulence are assumed to be large compared to the convective-diffusive zone of the laminar flame.⁴⁴ Estimates indicate that in a number of situations of interest this length scale assumption is applicable, e.g., in fires and large furnaces.

In addition to clarifying the structure of some turbulent flames these analyses provide formulas for turbulent flame speeds in terms of statistical properties of the turbulence in the unburned mixture. Transverse gradients of the Eulerian displacement† play a dominant role in these formulas⁴⁴ since they effectively determine the shape of the wrinkled laminar

flame. Small differences between molecular diffusivities for heat and species introduce, in addition, influences of strain and curvature of the laminar flames on the predicted turbulent flame speeds.⁴⁵

These latter effects suggest limitations concerning the wrinkled flame view of a turbulent flame. It is known⁴⁵ that small departures of molecular Lewis numbers from unity are predicted to introduce instabilities of planar laminar flames, leading to cellular flames, which are often observed. Moreover, these cellular structures can exist within turbulent flames satisfying the small gradient conditions under which wrinkled flames are anticipated. Thus, perturbation descriptions of turbulent flames should begin with cellular rather than planar geometries if the relevant laminar flame is unstable; analysis along this line appears difficult.

Even when planar laminar flames are stable, local extinction of laminar flames can occur through local flame stretching if the instantaneous strain rates due to the turbulence are too large.⁴⁶ The relevant parameter determining the extent of such extinction is approximately

$$K = \left| \frac{\partial u_k''}{\partial x_k} \right| \frac{\delta_L}{u_L} (1-L) \quad (31)$$

where the subscript k does not imply summation, δ_L is the diffusive thickness of the unstrained laminar flame, u_L the corresponding laminar flame speed, and L the Lewis number, i.e., the ratio of molecular diffusivities of heat and species. Except for the factor involving the Lewis number the parameter K is approximately the product of a strain rate due to turbulence with the residence time within the laminar flame. Values of K large compared with unity assure local extinctions and imply conditions under which the accurate description of turbulent flames on the basis of the wrinkled laminar flame is mathematically difficult. A laminar flame in a turbulent reaction zone may be subjected to large strain rates and thus extinguished for short periods of time; reaction may subsequently take place as those strain rates are reduced sufficiently for chemical reaction to proceed.

There are flow conditions under which K is always small so that small gradient theories may be applied. The attractiveness of such theories is their avoidance of modeling hypothesis, as indicated earlier. Much work remains to be done in extending such theories,⁴⁵ e.g., in accounting for the influences of density changes associated with heat release. There are also flow conditions in which K is not small and in which appropriate expansion parameters do not appear to exist; modeling hypotheses are needed in such cases. Such considerations lead naturally to discussion of the second way of treating premixed combustion, that involving approximations to the pdf of a progress variable.

pdf Approximation for Fast Chemistry

To illustrate the approximation of a pdf for state variables in premixed systems we assume that with Eq. (30) taken into account one progress variable determines ξ , T , and all Y_i 's. This variable, normalized to the zero-one range, has been assigned various symbols including Y (Ref. 44) and c (Ref. 18). Here we employ the notation implied by

$$c = (T - T_0) / (T_\infty - T_0) \quad (32)$$

where T_0 and T_∞ are the temperatures in the reactants and fully burned products, respectively. Although space and time resolved measurements in turbulent flames are notoriously difficult, measurements which can be related to the variable c in premixed turbulent flames can be made. (In this regard, see the measurements recently made by Moss⁴⁷ in a premixed flame.)

For conditions under which the fast chemistry approximation applies the pdf $P(c; \mathbf{x})$ is dominated by delta functions at $c = 0, 1$ at all positions in the flow as illustrated in

†The Eulerian displacement is the time integral of the velocity fluctuations at a fixed point in space and is thus a random vector with zero mean.

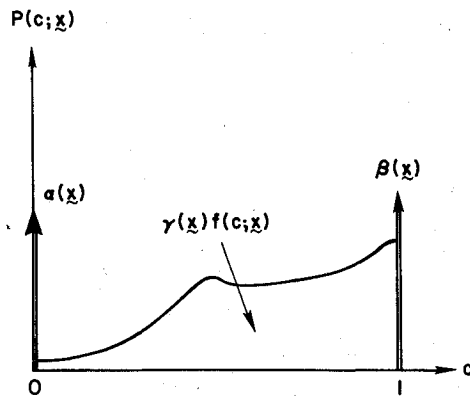


Fig. 3 Schematic representation of the probability density function for a premixed turbulent flame.

Fig. 3. The entries described by the strength of these delta functions correspond, respectively, to reactants and fully burned products. The contribution to $P(c; x)$ from interior values of c , i.e., from $0 < c < 1$, is small but corresponds to times when narrow regions of reaction are present at the spatial location x in question.

Bray¹⁹ presents a laminar flamelet model of the reaction regions in which the classical laminar flame provides a calculation of the interior distribution. Libby and Bray⁴⁸ examine the implications of that model insofar as various dissipation terms and physicochemical notions of premixed turbulent flames are concerned. Bray, et al.^{18,19,39,49,51} exploit the fast chemistry approximation in order to study interaction effects, in some cases with the laminar flamelet model of reaction surfaces employed and in others without reference to a specific model of those surfaces. Of particular interest is the finding of Bray¹⁸ that under appropriate assumptions the description of the thermochemical state in premixed combustion in terms of a single progress variable is applicable to chemical systems of arbitrary complexity.

It should be recognized that the use of the classical laminar flame solutions in providing the interior distribution of $P(c; x)$ involves approximations. Even for a one-reactant chemical system described by a one-step reaction two variables are needed if the Lewis number is nonunity. In addition, the strain associated with the turbulence alters the structure of the laminar flame so that the interior distribution reflects passages of reaction surfaces subject to various rates of strain. However, substantial alterations of the interior distribution may not be significant if the chemistry is sufficiently fast.

Departures from Fast Chemistry in Premixed Systems for Trace Species

It is evident that in general terms the applicability of the fast chemistry approximation in premixed systems depends on a suitable comparison between representative chemical and turbulence times. For dealing with trace species that do not respect the fast chemistry requirements, e.g., NO, methods applicable to premixed systems paralleling those presented earlier exist.⁵² As an example of the goal of such methods a formula such as Eq. (23) may be derived, namely,

$$\dot{w}_i(x) = \int_0^1 \dot{w}_i^s(c) P(c; x) dc \quad (33)$$

where $\dot{w}_i^s(c)$ denotes the rate of production of the i th species on the basis of the temperature and concentrations of the dominant species determined in terms of the progress variable c .

Concluding Remarks Relative to the Chemistry Problem

There are chemistry problems that are not readily treated from fundamental considerations in the current state of

development of the theory of turbulent combustion. Phenomena such as flame holding, blowout, thermal ignition, and total extinction in such combustion are complex in that they involve both reacting and nonreacting flows simultaneously; the conditions leading to the various limiting treatments discussed here do not prevail under these circumstances. As a consequence, accounting for the influences of the properties of the turbulence on these phenomena remains largely empirical. Partially premixed systems in which multiple variables are required to describe the thermochemical state of the gas are not subject to satisfactory treatment at present. Finally, we note that transient combustion such as that occurring in spark ignition engines involves additional problems not discussed here. (See Mellor and Ferguson⁵³ for a perspective on problems of practical interest relative to exploitation of the theory of turbulent reacting flows including a discussion of the internal combustion engine.) This recital of intractable problems suggests on the one hand the limitations of current predictive methods and on the other the richness of chemistry problems in turbulent combustion of both fundamental and practical interest.

V. Fluid Mechanics Problems

Establishing confidence in modeling methods required for the treatment of the fluid mechanical aspects of turbulent reacting flows is more difficult than that for the chemical problems discussed earlier. As a consequence, our discussion in this section will be less extensive; brevity should not be interpreted as implying reduced importance. Rather it implies an absence of substantive information regarding the fundamental treatment of the fluid mechanical problems arising in turbulent reacting flows and the need for considerable research on such problems.

Perturbation methods which, as indicated earlier, provide insight into the chemical behavior in certain turbulent reacting flows have to date dealt with limited fluid mechanical problems; such methods have taken the statistics of the velocity field as prescribed and therefore have avoided the interaction between the chemical and fluid mechanical behavior. Although these methods afford predictions of turbulent transport of species and scalar dissipation independent of modeling assumptions, they make no predictions of statistical quantities involving the velocity field alone. In contrast, moment methods whether excluding or including considerations of the operative pdf's necessitate introduction of modeling assumptions to achieve closure. Methods in which the evolution of pdf's related to the thermochemical state of the system are calculated^{21,22} involve analogous modeling. Recently new difficulties with the modeling traditionally employed in various moment methods have been indicated⁵¹; these difficulties are reviewed here and prospects for improvements are assessed.

Our discussion will employ Favre averaging although these findings apply with some modification when conventional averaging is used. In the conservation equations usually used for the description of turbulent reacting flows four types of terms calling for modeling arise:

- 1) mean stresses such as $\overline{\rho u_i' u_j'}$;
- 2) mean fluxes, e.g., $\overline{\rho u_i' u_j' u_k'}$, $\overline{\rho u_i' Y_j'}$, etc;
- 3) pressure-velocity and pressure-scalar correlations, e.g., $\overline{u_i' \partial p / \partial x_j}$ and $\overline{Y_i' \partial p / \partial x_j}$;
- 4) dissipation terms, e.g., $\overline{\mu (\partial u_i / \partial x_k) (\partial u_j / \partial x_k)}$ and $\overline{\rho D (\partial Y_i / \partial x_k) (\partial Y_j / \partial x_k)}$ where μ is the viscosity coefficient.

Treatments of these various terms in order to achieve closure in predictive methods are generally adaptations with little alteration of those that have previously been more or less successful for turbulent flows without chemical reaction and without density variations. It is thus important to consider whether chemical reaction and variations in density can influence the models introduced in these treatments.

Gradient Transport

Whether closure is effected at the level of the conservation equations for mean quantities, e.g., \bar{u}_i , \bar{Y}_j , etc., or at the level of the equations for intensities, e.g., for $\bar{u}_i'^2$, $\bar{Y}_j'^2$, etc., and for fluxes, e.g., $\bar{u}_i'Y_j'$, etc., as in second-order closure, gradient transport assumptions in one form or another are almost universally used. For example, in the simplest methods the stresses appearing in the mean momentum equations are represented as

$$\overline{\rho u_i' u_j''} = -\bar{\rho} \nu_T \left(\frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial \bar{u}_j}{\partial x_i} \right), \quad i \neq j \quad (34)$$

where ν_T is a turbulent exchange coefficient. In second-order closure a conservation equation for $\overline{\rho u_i' u_j''}$ is included in the system of equations and the flux terms appearing there are represented by means of a gradient transport assumption, for example,⁵⁴ as

$$\begin{aligned} \overline{\rho u_i' u_j' u_k''} = & -(\nu_T / \bar{q}) \left[\overline{\rho u_i' u_k''} \frac{\partial}{\partial x_k} \left(\frac{\overline{\rho u_j' u_i''}}{\bar{\rho}} \right) \right. \\ & \left. + \overline{\rho u_j' u_k''} \frac{\partial}{\partial x_k} \left(\frac{\overline{\rho u_i' u_i''}}{\bar{\rho}} \right) + \overline{\rho u_i' u_k''} \frac{\partial}{\partial x_k} \left(\frac{\overline{\rho u_j' u_j''}}{\bar{\rho}} \right) \right] \end{aligned} \quad (35)$$

where $\bar{q} = \frac{1}{2} \overline{\rho u_k' u_k''} / \bar{\rho}$ is the Favre-averaged turbulent kinetic energy.

There are several disturbing aspects to the use of these gradient transport approximations in reacting flows. Even if the density is constant, it is widely understood that such transport can be justified from a fundamental point of view only if the scale of the largest eddies is small compared with the scale of the gradients of mean quantities. There are many nonreacting shear flows of practical interest in which this condition is not met; nevertheless, the use of gradient transport is found to provide results of sufficient accuracy for many engineering purposes. However, such success does not imply that extension to reacting flows is appropriate.

We suggest some reasons for being skeptical regarding such extension. The perturbation method applicable to wrinkled laminar flames as discussed earlier has been employed to calculate the turbulent transport of reactant in a premixed turbulent flame that is one-dimensional in the mean.⁴⁴ It is found that even with constant density flow, i.e., with heat release negligibly small, if the streamwise Eulerian displacement is statistically stationary, then the mean turbulent flux of reactant changes sign at a point in the interior of the turbulent flame upstream of the mean location of the reactive-diffusive zone by a distance on the order to the thickness of the convective-diffusive zone of the laminar flame. This change of sign is obtained in the first order of the expansion for small gradients; it is brought about without modification of the concentration profile of the laminar flame through transient movement of the laminar flame caused by the turbulence. Essential to the change in sign is the decreased velocity of the reactive-diffusive zone due to positive stretching of the laminar flame.

A change in sign of a mean turbulent species flux such as $\overline{\rho u_i' Y_j''}$ in a region of the flow where $\partial Y_j / \partial x_i$ maintains one sign implies the turbulent exchange coefficient in a gradient transport description of the flux must change sign. Since negative values of an exchange coefficient are inconsistent with both concept and practice in gradient transport, it is evident skepticism regarding the appropriateness of such an approximation in reacting flows is engendered by these findings from the perturbation method.⁴⁴

There are additional reasons for concern about the applicability of gradient transport in reacting flows. In some flows, for example in premixed turbulent flames, the reactions and significant heat release are confined to relatively

narrow regions; such heat release produces large rates of mean strain, considerably larger than those arising in constant density flows, even those usually considered to involve large rates of strain. Such rates make traditional closure models difficult to justify.

Consideration of the influences of pressure gradients provides further cause for questioning gradient transport. If on the basis of large values of the turbulence Reynolds number molecular transport is neglected, then the conservation equation for Reynolds stresses in Favre averages is found to be

$$\begin{aligned} \frac{\partial}{\partial x_k} (\bar{\rho} \tilde{u}_k u_i' u_j'' + \overline{\rho u_i' u_j' u_k''}) = & -\overline{\rho u_i' u_k''} \frac{\partial \tilde{u}_j}{\partial x_k} - \overline{\rho u_j' u_k''} \frac{\partial \tilde{u}_i}{\partial x_k} \\ & - \left(\tilde{u}_i'' \frac{\partial \bar{p}}{\partial x_j} + \tilde{u}_j'' \frac{\partial \bar{p}}{\partial x_i} \right) - \left(\tilde{u}_i'' \frac{\partial \bar{p}'}{\partial x_i} + \tilde{u}_j'' \frac{\partial \bar{p}'}{\partial x_j} \right) - \bar{\chi}_{u_i u_j} \end{aligned} \quad (36)$$

where the dissipation term is

$$\bar{\chi}_{u_i u_j} = 2\mu \frac{\partial u_i'}{\partial x_k} \frac{\partial u_j''}{\partial x_k}$$

and is analogous to the term given by Eq. (9). Except for the terms multiplying the gradient of the mean pressure, each term in this equation can be identified with a corresponding term in the equation for the Reynolds stress $\overline{u_i' u_j'}$ in constant density turbulence. The implication from the presence of these terms for variable density flows is that they describe an interaction between density inhomogeneities and mean pressure gradient, an interaction absent in constant density flows.

To investigate the influence of these terms we neglect variations in molecular weight so that $\rho = \rho_0 T_0 / T$ where the subscript denotes a reference state, e.g., upstream of a turbulent flame. Then we can write

$$\tilde{u}_i'' \frac{\partial \bar{p}}{\partial x_j} = \frac{\overline{\rho u_i' T''}}{\rho_0 T_0} \frac{\partial \bar{p}}{\partial x_j} = -\frac{\overline{\rho u_i' T''}}{\rho_0 T_0} \frac{\partial}{\partial x_k} (\bar{\rho} \tilde{u}_k \tilde{u}_j + \overline{\rho u_k' u_j''}) \quad (37)$$

where we have used the mean-momentum equation. A similar equation is obtained for $\tilde{u}_j'' \partial \bar{p} / \partial x_i$. These terms are seen from Eq. (37) to be as large as the other terms in Eq. (36) and, moreover, they increase with temperature and, therefore, with the extent of heat release within the flame. It may therefore be concluded that, in general, the interaction between pressure gradient and density inhomogeneities is not negligible and can influence the stresses and fluxes that arise in turbulent flames.

We can be more specific regarding the influence of mean pressure gradients; Libby and Bray⁵² show that for premixed turbulent flames orthogonal in the mean to the oncoming reactants the action of the mean pressure gradient on density inhomogeneities produces countergradient diffusion, i.e., negative turbulent exchange coefficients as alluded to earlier. Physically, as may be seen from the Rankine-Hugoniot relations, we know fluid experiences a small decrease in pressure in traversing a flame. Since the lighter, burned gas responds much more readily to such a pressure decrease than do the heavier reactants, there is a tendency for burned packets to be driven downstream toward the fully burned products relative to the unburned packets. Although the pressure drop is proportional to the square of the Mach number based on the turbulent flame speed and is negligible with respect to mean momentum, its effect on turbulent transport is dominant if the heat release is large compared with the initial thermal enthalpy. These results are thus to be anticipated on simple physical grounds and bring into question the applicability of conventional gradient transport in reacting flows with significant heat release.

The influence of mean pressure gradients indicated here extends to the various terms modeled by gradient transport assumptions in second-order closure methods. To appreciate

this influence consider any flux term appearing in the conservation equation for a second-order correlation, e.g., that modeled by gradient transport as in Eq. (35); if we develop the conservation equation for that flux, there will appear therein one or more terms involving the product of the mean pressure gradient and various second-order correlations. Just as these terms in the conservation equation for the second-order correlation yielding the mean flux of product concentration dominate the behavior of that flux, we can expect that pressure gradient terms can significantly influence the behavior of the third-order correlations. The implication is that applying gradient transport assumptions as indicated by Eq. (35) to model the flux terms when significant heat release occurs is suspect.

It is natural to inquire if there is any experimental evidence to support the theoretical findings of Libby and Bray⁵¹; there are two pieces of such evidence. In measurements with extended hot-wire anemometry in flows involving the low-speed turbulent mixing of helium and air, Stanford and Libby⁵⁵ and LaRue and Libby⁵⁶ found countergradient diffusion of helium in regions of high helium concentration, i.e., low mean density. Recently, Moss⁴⁷ reports measurements with light scattering and LDV techniques in a turbulent premixed flame of the progress variable c and the normal velocity component u ; the correlation between these two variables is positive implying countergradient diffusion. Both sets of experiments are difficult and additional measurements are called for but we can conclude that there is evidence to support the physical and theoretical arguments for the occurrence of fluxes in contradiction with conventional gradient transport.

Dissipation Terms

The models representing the various dissipation terms that arise in the conservation equations for reacting flows are usually carried over without significant alteration from those used for constant density turbulence. Thus for example, the scalar dissipation arising in Eq. (12) is generally modeled as

$$\bar{\chi}_{\xi\xi} \propto \bar{\rho} \bar{q}^{1/2} \xi''^2 / \ell \quad (38)$$

where ℓ is a length scale characterizing the large eddies.

There seems to be general confidence in the validity of models for dissipation along the lines of Eq. (38); Jones⁵ notes: "The viscous dissipative correlation... is perhaps the least difficult term to model." However, Libby and Bray⁴⁸ pursue the implications of the laminar flamelet model for reaction at the molecular level within the reaction zone of a premixed turbulent flame. By considering a joint pdf $P(u, c; \mathbf{x})$ where u is the velocity component normal to the flame and c is the progress variable [cf. Eq. (32)], they infer by means of a simple calculation that the scalar dissipation of the fluctuations of c superficially resembles that given by Eq. (38), namely,

$$\bar{\chi}_{cc} \propto \rho_0 u_L \bar{c}''^2 / \ell \quad (39)$$

where u_L is the laminar flame speed previously introduced. Note the replacement of the square root of the turbulent kinetic energy in the conventional model with the laminar flame speed. This result suggests an influence of chemical rates on scalar dissipation.

Bray¹⁸ gives a different but related connection between dissipation and chemical reaction; for premixed reactants undergoing fast chemistry he derives the formula

$$\bar{w} = \bar{\chi}_{cc} / (2c_m - 1) \quad (40)$$

where c_m is the ratio of two successive moments of the interior pdf $P(c; \mathbf{x})$. Equation (40) implies the connection alluded to earlier: a model for one of the two leads directly to the prediction of the other.

So-called cross-dissipation terms such as $\bar{\chi}_{u_{ij}}$ in Eq. (36) for $i \neq j$ frequently are argued to be negligible on the basis of the following reasoning. These and the various other dissipation terms that arise in the conservation equations are dominated by small turbulence scales. At such scales the flow is usually considered locally isotropic and by symmetry the cross dissipation is zero in an isotropic flow. However, the reasoning of Libby and Bray⁴⁸ leads again rather directly to

$$\bar{\chi}_{cu} = \bar{\chi}_{cc} \bar{\rho} u'' c'' / (\bar{\rho} \bar{c} (1 - \bar{c})) \quad (41)$$

which generally cannot be considered negligible.

These considerations raise questions concerning the apparent confidence in the currently accepted models for the several dissipation terms that must be dealt with and imply that direct chemical effects may call for alterations of those models. In our view additional research on the modeling of the dissipation terms within the context of moment methods is required. In this regard, we note the difficulties associated with the experimental determination of dissipation quantities even in nonreacting turbulent flows.

Concluding Remarks Relative to Fluid Mechanical Problems

Clarification of the various difficulties regarding the proper treatment of the fluid mechanical problems arising in turbulent combustion with significant heat release raised in this section requires additional research. Since variation in density is a cause of the interaction between the chemical and fluid mechanical behavior in turbulent combustion, some clarification can be provided by experiments on the nonreactive turbulent mixing of dissimilar gases along the lines indicated in Refs. 55 and 56. Although such experiments are not simple, involving as they do advanced hot-wire anemometry, they provide data difficult to obtain in reacting flows with comparable density inhomogeneities. Furthermore, as capabilities to make detailed measurements in combustion flows increase we can expect improved understanding and improved modeling of the fluid mechanical behavior in such flows to be achieved. In this regard, it must be emphasized that simultaneous measurements of state and velocity variables are essential if the stresses and fluxes which play central roles in predictive methods are to be obtained; detailed data on state variables while valuable are of considerably less interest in the resolution of the difficulties discussed here.

Experiments with the focus indicated in this discussion should be supplemented by appropriate theoretical studies following various lines: formal perturbation techniques applicable to idealized flows, extended direct pdf approaches applied to simple flows, and improved moment analyses incorporating as much physicochemical information as possible.

In concluding this section we note the problem of modeling the various terms involving pressure fluctuations. These terms are generally considered to describe important processes in constant density turbulent flows and extensive analyses have been devoted to their representation in such flows.⁵⁴ Generally, the resultant models are adopted without significant alteration to the analysis of turbulent reacting flows. Our discussion in this section of the importance of interaction between mean pressure and density inhomogeneities and the increased noise associated with turbulent combustion engender skepticism regarding this practice. The experimental difficulties of measuring static pressure fluctuations and their correlations with various other fluid mechanical and thermochemical quantities even in nonreacting flows are well known and indicate that new techniques are needed to provide useful information on the effect of pressure fluctuations in reacting flows. Theoretical research particularly focused on the role and importance of pressure fluctuations in turbulent combustion should be encouraged.

VI. Concluding Remarks

In the previous sections we provide an assessment of the current status of the theory of turbulent reacting flows. In making this assessment we a priori exclude certain important phenomena, those connected with multiphase flows, radiative transfer, and high speed. That exclusion is adopted to make the field of our discussion tractable. (In Ref. 6 we provide a review of, and entries to the literature of, these excluded phenomena.) Our purpose here is to summarize the results of our findings.

We conclude that significant advances have been achieved in the past few years relative to the treatment of the chemical aspects of a wide class of turbulent flows of practical interest. Provided all species, or at least the species dominating the thermochemical state of the gas, respect the notions of chemical equilibrium or fast chemistry the chemical behavior in reacting flows with complex composition and complex mechanisms can be confidently simplified to a significant extent. The behavior of trace species involving finite rate chemistry and embedded in such a flow can, under some circumstances, be treated in a convincing fashion; while in other circumstances, involving trace species-trace species interactions, multivariate pdf's, calling for additional attention, are required.

It is remarkable that despite their dissimilarities the two limiting cases of nonpremixed and premixed reactants under the conditions of fast chemistry involve methodologies for their treatment according to the moment method that are so similar. In both types of flows single variables combined with models for their pdf's lead to convincing descriptions of the thermochemical states involved, although the two variables are interpreted differently. In the circumstances that the species dominating the thermochemical state respect the fast chemistry approximation we find trace species involving finite rate chemistry can be treated in a similar fashion for the two cases.

In addition to the cases of practical interest that can be treated successfully at present, there are flows perhaps of less import but not without interest in which believable results can be generated. These include situations in which the chemistry is slow enough and the intensities of fluctuations of the state variables are sufficiently small for classical moment methods to be applicable even with full chemistry.

By implication in these remarks and explicitly noted in Sec. IV present techniques of analysis fail when the flow and thermochemical situation is such that finite rate chemistry determines chemical behavior. We are unable to deal convincingly from fundamental considerations with either cases involving the relatively slow reaction in nonpremixed flows or those we term intermediate between the nonpremixed and premixed limits. Thus, theory cannot currently contribute complete treatment of a range of practically important phenomena in turbulent combustion connected with flame holding, blowout, thermal ignition, and total extinction. Transient combustion such as occurs in spark-ignition engines also involves special problems not readily treated at present.

We find the fluid mechanical aspects of turbulent reacting flows with degrees of heat release of practical interest are subject to considerable uncertainties. Such uncertainties are probably less severe in nonpremixed flows of practical interest; the fluctuations in density that are responsible for significant uncertainties are considerably smaller in nonpremixed flows. Thus the interaction of such fluctuations with mean pressure gradients should be of smaller importance than found to be the case in premixed turbulent flames. It appears that conventional modeling based on gradient transport is more likely to be satisfactory in nonpremixed flows, at least for conserved scalars. However, the skepticism regarding modeling based on gradient transport as well as the modeling of various dissipation and pressure fluctuation terms recommended here should be maintained even for nonpremixed situations. Such skepticism can be resolved by careful coordination between theory and experiment.

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