

The Implications of the Probability Equations for Turbulent Combustion Models

S. B. POPE

*Imperial College of Science and Technology,
Department of Mechanical Engineering, Exhibition Road, London SW7 2BX*

The physical foundations of combustion models employed in turbulent flow calculation procedures are examined by reference to the transport equation for the joint probability distribution of the scalars characterizing the system. It is shown that, with certain restrictions, an arbitrarily fuelled flame resulting from a one-step irreversible reaction can be characterized by two scalars, the total fuel mass fraction, f , and the mass fraction of products, c . This system includes the special cases of premixed and diffusion flames. The transport equation for the joint probability of f and c is introduced and discussed: it is shown that, in the limit of very rapid reaction, the distribution can be expressed in terms of two single probability distributions and a scalar, A , which represents the mass fraction of activated species. By deriving the transport equations for these quantities and modelling the unknown terms a closure is obtained which is used to assess existing combustion models. Lockwood and Naguib's [9] model for arbitrarily fuelled flames and the model for diffusion flames derived from it [6, 7] are sound in principle but improvements to detail are indicated. An alternative is provided for the eddy-break-up model [1] which is found to be deficient on mathematical and physical grounds.

1. INTRODUCTION

In recent years turbulent combustion models have been developed for incorporation into flow solution algorithms in order to account for the interaction of turbulence and combustion. The most widely used of these models are applicable to situations where the reaction rate is very rapid and consequently the rate of fuel consumption is primarily determined by the turbulent structure. Spalding [1] introduced the "eddy-break-up" model for premixed flames and this has been used to calculate practical combustion situations by Mason and Spalding [2], Khalil, Spalding, and Whitelaw [3], Pope [4], and others. Diffusion flames have been calculated by Gosman and Lockwood [5], Elgobashi and Pun [6], and Lockwood and Naguib [7] using models based on the analysis of Toor [8]. Flames which are neither premixed nor diffusion have been calculated, to a lesser extent, by Khalil, Spalding, and Whitelaw [3] and

by Lockwood and Naguib [9]. All these authors have compared their calculations with measured values which allow an assessment of the accuracy of the models. In addition, the various values of the "constants" appearing in the models used by each investigator indicates the degree of universality of the models. In both accuracy and generality the performance is encouraging although not entirely satisfactory. For example, Khalil *et al.* [3] and Lockwood and Naguib [9] calculated flame temperatures with a 200°K discrepancy and Pope's [4] calculations of bluff-body stabilised flames overestimate the width of the flame by 30%: the values of the eddy-break-up constant used by Mason and Spalding, Khalil *et al.*, and Pope were 0.53, 1.0, and 1.1, respectively, which indicates the lack of universality.

The work reported here represents an attempt to understand and to decrease the lack of accuracy and generality of turbulent combustion models by relating them more rigorously to the transport

equations on which they are based. The probability approach lends itself well to this end as it overcomes the problems associated with the highly nonlinear reaction rate expressions experienced in combustion systems: that is, in a probability formulation, the term relating to chemical reaction is closed.

Hopf [10] introduced the probability distribution functional equation into the study of turbulence and the use of the probability distribution function equation in inert flows has been demonstrated by Lundgren [11, 12], Fox [13, 14], and Ribeiro [15]. This equation, for a scalar embedded in a turbulent flow, has been discussed in a series of works published jointly and severally by Dopazo and O'Brien (notably [16] and [17]) and by Pope [18]. In each of the latter three works, closure approximations for the unknown terms in the probability distribution function equation are suggested and are discussed below.

The formulation of a closure for turbulent reacting flows entails three types of modelling: first, as in inert flows, a turbulence model is required to close the mean momentum equations by determining the Reynolds stresses; second, account must be taken of the influence of density fluctuations; and, third, the effect of the turbulence on the reaction rate must be modelled. The calculations referred to above were performed with a turbulence model (see, e.g., [19]) in which transport equations were solved for the kinetic energy of turbulence, k , and its rate of dissipation, ϵ . This model has proved successful in jet-type flows although there is evidence that it can produce significant errors in bluff-body-stabilized flame calculations [20]. The influence of density fluctuations has usually been ignored in combustion calculations in spite of the fact that density variations of a factor of five are commonplace in practical combustion systems. Bray [21] has considered the effect of density fluctuations on the turbulence and, as shown below, their influence on the scalar fields may be accommodated within the probability approach. The third effect, that of the turbulence on the reaction, forms the main theme of this paper.

In the next section a simplified reacting system, termed an "arbitrarily fuelled flame," is defined

and contains the special cases of premixed and diffusion flames. It is shown that, with certain assumptions, restrictions and approximations, the system can be characterised by two scalars, namely, the mass fraction of combustion products, c , and the total fuel mass fraction, f . In section 3 the transport equation for the joint probability distribution function of c and f is introduced and described and further equations are derived for the case of rapid reaction rates. Possible closure approximations for the unknown terms in these equations are discussed and are used in section 4 to assess existing combustion models and to clarify the assumptions on which they are based. The paper closes with a summary of the more important conclusions.

2. FORMULATION

The probability approach has the advantage of overcoming difficulties associated with nonlinearities at the price of increasing the dimensionality of the problem by the number of dependent variables involved: it is a high price to pay and in order to obtain the best value, the minimum number of dependent variables required to characterize a system should be considered with care. A two-scalar system is studied here, whereas solutions obtained to date [17, 18] have been for systems characterized by a single scalar. In this section, restrictions and approximations are applied sparingly to a general reacting system in order that the resulting two-scalar system be as general and as physically realistic as possible.

The chemical and thermodynamic properties of a reacting system can be characterized by the mass fraction of each chemical species, the enthalpy and the pressure: an equation of state relates these quantities to the density. Both the mass fractions and the enthalpy are assumed to obey the transport equation

$$\rho \frac{\partial \phi_\alpha}{\partial t} + \rho U_i \frac{\partial \phi_\alpha}{\partial x_i} = \frac{\partial}{\partial x_i} \Gamma \frac{\partial \phi_\alpha}{\partial x_i} + S_\alpha(\phi, p), \quad (1)$$

where ρ , U , and p are the density, velocity, and pressure at a position x and time t : ϕ_α represents

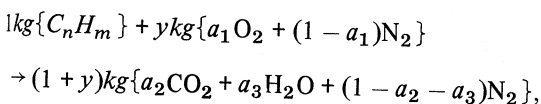
one of the scalars with a source S_α and Γ is a diffusive coefficient. The last term in this equation implies that the source is solely a function of ϕ and p at the point involved: this is always true when ϕ_α represents a mass fraction but, for the enthalpy equation, kinetic heating and radiation must be negligible. The only other assumption embodied in Eq. (1) is that the scalars are transported by simple gradient diffusion. While such an assumption is unrealistic for reacting flows and would undoubtedly lead to error if a laminar flame were to be calculated, the macroscopic properties of high Reynolds number turbulent flames are independent of Reynolds number and consequently the detailed form of the term is unimportant. The specific form is used here as a convenient way to specify the two important properties of molecular transport: namely, that it is only significant at length scales below some size and that the tendency is to mix not to segregate. The further restriction of low Mach number is made in order to remove the dependence of S_α and ρ upon fluctuations in pressure. Thus,

$$S_\alpha(\phi, p) = S_\alpha(\phi, \langle p \rangle) = S_\alpha(\phi) \quad (2)$$

$$\rho(\phi, p) = \rho(\phi, \langle p \rangle) = \rho(\phi) \quad (3)$$

where the brackets, $\langle \rangle$, indicate an ensemble average.

With these restrictions and a knowledge of the velocity field Eq. (1) is a closed set. The number of scalars that need be considered is reduced to two by requiring that the sources, S , be mutually linearly dependent and that the boundary conditions for each scalar be related to either one of two boundary condition specifications by the same linear relation. For this situation Eq. (1) comprises two sets of linearly dependent equations, and consequently, from a knowledge of two independent scalars, all other scalars can be evaluated. The sources of each scalar are linearly related provided that the reactants combine in fixed proportions to produce products of a given composition: for example, for a hydrocarbon fuel burning in air,



where $a_1 = 0.232$ is the fraction of oxygen in air,

$$y = 8(4n + m)/\{a_1(12n + m)\},$$

$$a_2 = 44n/\{12n + m + 8(4n + m)/a_1\},$$

and

$$a_3 = 9m/\{12n + m + 8(4n + m)/a_1\}. \quad (4)$$

In most reacting flow situations, the boundary can be divided into three regions; "inlet," "outlet," and impervious walls. Only the first need concern us since the zero normal gradient boundary condition appropriate to impervious (and, by assumption, adiabatic) walls preserves the linearity of the equations and the parabolic nature of the flow near "outlets" removes the dependence on these conditions. The system considered is termed an "arbitrarily fuelled flame" since no restriction is imposed on the inlet boundary condition for fuel concentration, f_b . However, it is required that the concentration of products be zero at inlet and that the inlet values of enthalpy, h , be linearly related to the fuel concentration. Subject to these restrictions, the mass fractions of each chemical specie and the enthalpy can be related to the mass fraction of products, c , and the synthetic scalar, f , which represents the mass fraction of burnt and unburnt fuel. The relationship between these scalars for a hydrocarbon/air flame is shown in Table 1.

In summary, a one-step reaction taking place in an adiabatic flow in which the Reynolds number is high and the Mach number is low can be characterised by the two scalars, c and f , provided that at inlet the enthalpy is proportional to the mass fraction of fuel and the mass fraction of products is zero. Bluff-body-stabilised flames and both premixed and diffusion jet flames are well represented by this system while furnace flames and the flow in gas-turbine combustion chambers are, in certain circumstances and to varying degrees of approximation, encompassed by the restrictions. Consequently, the detailed experimental data available for the former flows (e.g., [22-24]) can be used to validate the combustion models which can then be used to calculate other flows of practical importance.

TABLE 1
Relationship between Scalars for an Arbitrarily Fuelled Hydrocarbon Flame

ϕ	S_ϕ	Boundary Condition	$\phi(f, c)$
f	0	f_b	f
c	S_c	0	c
m_{fu}	$-S_c/(1+y)$	f_b	$f - c/(1+y)$
m_{O_2}	$-S_c y/(1+y)$	$a_1(1 - f_b)$	$a_1(1 - f) - cy/(1+y)$
m_{N_2}	0	$(1 - a_1)(1 - f_b)$	$(1 - a_1)(1 - f)$
m_{CO_2}	$a_2 S_c$	0	$a_2 c$
m_{H_2O}	$a_3 S_c$	0	$a_3 c$
h	0	$h_1 + h_2 f_b$	$h_1 + h_2 f$

3. PROBABILITY EQUATIONS

Substituting Eqs. (2)-(3) into Eq. (1) written for f and c yields

$$\rho(f, c) \frac{\partial f}{\partial t} + \rho(f, c) U_i \frac{\partial f}{\partial x_i} = \frac{\partial}{\partial x_i} \Gamma \frac{\partial f}{\partial x_i} \quad (5)$$

and

$$\rho(f, c) \frac{\partial c}{\partial t} + \rho(f, c) U_i \frac{\partial c}{\partial x_i} = \frac{\partial}{\partial x_i} \Gamma \frac{\partial c}{\partial x_i} + S(f, c) \quad (6)$$

which, if U_i is known, is a closed set. As is indicated on Fig. 1, paired values of c and f are bounded: both c and f are bounded by zero and unity and the similar bounds on fuel and oxidant mass fractions require that paired values of c and f be within the indicated triangle, i.e., $0 \leq c \leq c_A(f)$. The apices of the triangle correspond to single species—fuel, oxidant, or product—while the bounding lines represent a mixture of two species in the absence of the third: regions inside the triangle correspond to mixtures of all three components.

The instantaneous joint probability distribution of c and f is defined (more to simplify the notation than to imply physical significance) as

$$P(\hat{f}, \hat{c}) \equiv \delta(f - \hat{f}) \delta(c - \hat{c}) \quad (7)$$

and, clearly, is zero outside the triangle of Fig. 1.

δ is the Dirac delta function and the circumflex indicates an independent variable in composition space. The inert mixing of fuel and oxidant is characterized by $c = 0$ and so the ensemble averaged joint probability distribution for this situation is

$$\langle P(\hat{f}, \hat{c}) \rangle = \langle P(\hat{f}) \rangle \delta(\hat{c}). \quad (8)$$

Similarly a homogeneously premixed flame is characterized by a constant value of f (f_0 say) and consequently,

$$\langle P(\hat{f}, \hat{c}) \rangle = \langle P(\hat{c}) \rangle \delta(f_0 - \hat{f}). \quad (9)$$

These two examples serve to illustrate the signifi-

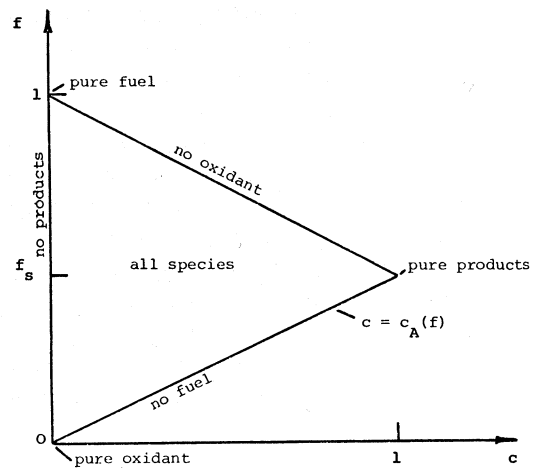


Fig. 1. Sketch of properties in composition space.

cance of different regions of the composition space (\hat{f}, \hat{c}).

An exact transport equation for the averaged joint probability distribution of f and c can be derived from Eqs. (5)–(7), see [18];

$$\begin{aligned} \langle \rho \rangle \frac{\bar{D}}{Dt} \bar{P}(\hat{f}, \hat{c}) &= \frac{\partial}{\partial x_i} \Gamma \frac{\partial}{\partial x_i} \langle P(\hat{f}, \hat{c}) \rangle - \frac{\partial}{\partial x_i} \overline{\langle \rho \rangle U_i'' P''(\hat{f}, \hat{c})} \\ &\quad - \frac{\partial}{\partial \hat{c}} S(\hat{f}, \hat{c}) \langle P(\hat{f}, \hat{c}) \rangle \\ &\quad - \frac{\partial^2}{\partial \hat{c}^2} \left\langle \Gamma \left(\frac{\partial c}{\partial x_i} \right)^2 P(\hat{f}, \hat{c}) \right\rangle \\ &\quad - \frac{\partial^2}{\partial \hat{f}^2} \left\langle \Gamma \left(\frac{\partial f}{\partial x_i} \right)^2 P(\hat{f}, \hat{c}) \right\rangle \\ &\quad - \frac{\partial^2}{\partial \hat{f} \partial \hat{c}} \left\langle \Gamma \frac{\partial f}{\partial x_i} \frac{\partial c}{\partial x_i} P(\hat{f}, \hat{c}) \right\rangle, \end{aligned} \quad (10)$$

where terms involving the density are expressed as Favre averages [25], i.e.,

$$\tilde{\phi}_\alpha \equiv \langle \rho \phi_\alpha \rangle / \langle \rho \rangle, \quad (11)$$

$$\overline{\phi_\alpha'' \phi_\beta''} \equiv \langle \rho (\phi_\alpha - \tilde{\phi}_\alpha) (\phi_\beta - \tilde{\phi}_\beta) \rangle / \langle \rho \rangle, \quad (12)$$

and consequently

$$\bar{P}(\hat{f}, \hat{c}) = \langle P(\hat{f}, \hat{c}) \rho(\hat{f}, \hat{c}) \rangle / \langle \rho \rangle \quad (13)$$

and

$$\frac{\bar{D}}{Dt} \equiv \frac{\partial}{\partial t} + \tilde{U}_i \frac{\partial}{\partial x_i}. \quad (14)$$

The first term on the right-hand side of Eq. (10) represents transport in position-space due to molecular diffusion and is negligible at the high Reynolds number presumed here. The next term represents transport due to turbulent convection

and contains the unknown correlation $\overline{U_i'' P''(\hat{f}, \hat{c})}$.

Dopazo (17) obtained a closure for the analogous term $\langle U_i' P(f) \rangle$, by assuming that the conditional expected value of U_i' , given f , is conditionally Gaussian,

$$\langle U_i' P(\hat{f}) \rangle = \frac{\langle U_i' f' \rangle}{\langle f'^2 \rangle} (\hat{f} - \langle f \rangle) \langle P(\hat{f}) \rangle, \quad (15)$$

where U_i' and f' are the fluctuating components of U_i and f . On the other hand, in an earlier work (18), it was argued that a mixing length approximation is equally applicable to this term as to more familiar terms such as $\langle U_i' f' \rangle$; thus,

$$\langle U_i' P(\hat{f}) \rangle = -B_1 \nu l \frac{\partial}{\partial x_i} \langle P(\hat{f}) \rangle \quad (16)$$

where ν and l are turbulent velocity and length scales and B_1 is a constant. Both models lack experimental support although they both behave correctly for the case of a homogeneous field. The value of the correlation $\langle U_i' f'^n \rangle$ given by each clearly demonstrates the difference between the two models; Eq. (15) gives

$$\langle U_i' f'^n \rangle = \langle U_i' f' \rangle \frac{\langle f'^{n+1} \rangle}{\langle f'^2 \rangle} \quad (17)$$

and Eq. (16) gives

$$\langle U_i' f'^n \rangle = -B_1 \nu l \frac{\partial}{\partial x_i} \langle f'^n \rangle. \quad (18)$$

The mixing length approximation will be employed here for the reason that, in a moment formulation, Eq. (15) does not provide a closure: that is, as can be seen from Eq. (17), in order to determine $\langle U_i' f'^n \rangle$, which appears in the equation for $\langle f'^n \rangle$, the value of $\langle f'^{n+1} \rangle$ must be known. Thus, the term representing turbulent convection in Eq. (10) will be modelled as,

$$\overline{U_i'' P''(\hat{f}, \hat{c})} = -B_1 \nu l \frac{\partial}{\partial x_i} \bar{P}(\hat{f}, \hat{c}) \quad (19)$$

The term in Eq. (10) containing the source is

closed: it causes probability to be transported from lower to higher values of \hat{c} . The final three terms represent transport in composition space due to molecular mixing and they pose the greatest problem in formulating a closure: their effect is to reduce the standard deviation of f and c without affecting their mean values. No satisfactory model for these terms has been found but, as will be shown, a closure for the case of very rapid reaction rates can be obtained with a knowledge of their one-dimensional counterparts,

$$G_f(\hat{f}) \equiv \left\langle \Gamma \left(\frac{\partial f}{\partial x_i} \right)^2 P(\hat{f}) \right\rangle,$$

for which models have been proposed (17, 18). For the moment, the notation,

$$G_{fc}(\hat{f}, \hat{c}) \equiv \left\langle \Gamma \frac{\partial f}{\partial x_i} \frac{\partial c}{\partial x_i} P(\hat{f}, \hat{c}) \right\rangle \quad \text{etc.}, \quad (20)$$

is introduced to allow Eq. (10) to be rewritten as

$$\begin{aligned} \langle \rho \rangle \frac{\tilde{D}}{Dt} \tilde{P}(\hat{f}, \hat{c}) &= \frac{\partial}{\partial x_i} B_1 \nu l \langle \rho \rangle \frac{\partial}{\partial x_i} \tilde{P}(\hat{f}, \hat{c}) \\ &\quad - \frac{\partial}{\partial \hat{c}} S(\hat{f}, \hat{c}) \langle P(\hat{f}, \hat{c}) \rangle - \frac{\partial^2}{\partial \hat{c}^2} G_{cc}(\hat{f}, \hat{c}) \\ &\quad - \frac{\partial^2}{\partial \hat{f}^2} G_{ff}(\hat{f}, \hat{c}) - \frac{\partial^2}{\partial \hat{f} \partial \hat{c}} G_{fc}(\hat{f}, \hat{c}). \end{aligned} \quad (21)$$

The shape adopted by $\langle P(\hat{f}, \hat{c}) \rangle$ for very rapid reaction rates can be deduced both from physical arguments and from its transport equation. For diffusion flames the assumption of fast reactions leads to the conclusion that fuel and oxidant are not copresent (8): the fuel and oxidant streams are separated by a hot flame front and so, at any interface between fuel and oxygen, the temperature is sufficiently high for the reaction rate to reduce the concentration of either specie rapidly to zero. Thus, $\langle P(\hat{f}, \hat{c}) \rangle$ adopts the shape illustrated on Fig.

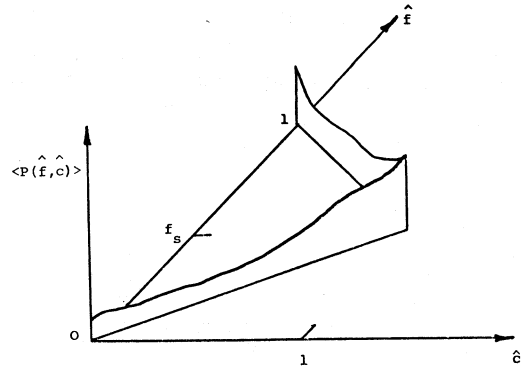


Fig. 2. Sketch of $\langle P(\hat{f}, \hat{c}) \rangle$ for a diffusion flame.

2, being nonzero only along the line $\hat{c} = c_A(\hat{f})$. For premixed flames the assumption of fast reactions can be interpreted to mean (18, 26), that although the incoming fuel/air mixture does not react spontaneously, i.e., $S(\hat{f}, 0) = 0$, a small increase in temperature caused by mixing with combustion products causes the reaction rate to be large. Consequently, in the limit, the fuel/air mixture and combustion products do not coexist and $\langle P(\hat{f}, \hat{c}) \rangle$ tends to the double delta function distribution illustrated on Fig. 3.

These physical arguments can be extended to arbitrarily fuelled flames with the conclusion that fuel, oxidant and products cannot coexist. This conclusion is clearly supported by an examination of the transport equation for $\tilde{P}(\hat{f}, \hat{c})$: as the reaction rate, $S(\hat{f}, \hat{c})$, tends to infinity, the term containing it can only remain finite by $\langle \tilde{P}(\hat{f}, \hat{c}) \rangle$ tending to zero wherever $S(\hat{f}, \hat{c})$ is not identically zero;

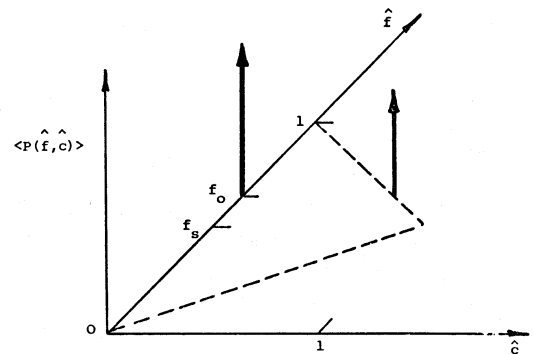


Fig. 3. Sketch of $\langle P(\hat{f}, \hat{c}) \rangle$ for a premixed flame.

that is, in the limit, $\langle P(\hat{f}, \hat{c}) \rangle$ is only nonzero where one of the species is zero. Thus, the joint probability may be expressed as the sum of the three single probabilities representing the absence of one specie,

$$\begin{aligned} \langle P(\hat{f}, \hat{c}) \rangle &= \langle P_1(\hat{f}) \rangle \delta(\hat{c}) + \\ &\quad \text{(no products)} \\ &\quad \langle P_2(\hat{f}) \rangle \delta(\hat{f} - \hat{c} \hat{f}_s) + \\ &\quad \text{(no fuel)} \\ &\quad \langle P_3(\hat{f}) \rangle \delta(\hat{c} - (1 - \hat{f}) / (1 - \hat{f}_s)). \end{aligned} \quad (22)$$

(no oxidant)

A sketch of such a distribution is shown on Fig. 4 where it is seen that $\langle P_2(\hat{f}) \rangle$ and $\langle P_3(\hat{f}) \rangle$ are nowhere nonzero together and consequently they may be summed to form one distribution without loss of information;

$$\langle P_A(\hat{f}) \rangle \equiv \langle P_2(\hat{f}) \rangle + \langle P_3(\hat{f}) \rangle. \quad (23)$$

The subscript "A" stands for "activated species" a term introduced by Lockwood and Naguib [9] to indicate that, in this region, the energy is sufficient for reactions to be rapid.

The problem of formulating a closure for arbitrarily fuelled flames has been reduced to that of determining the two distributions $\tilde{P}(\hat{f})$ and $\tilde{P}_A(\hat{f})$: the latter distribution, which does not integrate to unity, can be expressed in terms of \tilde{A} , the mass fraction of activated species, and the normalised distribution $\tilde{P}_A^*(\hat{f}) \equiv \tilde{P}_A(\hat{f}) / \tilde{A}$. A more rigorous definition of these quantities allows their transport equations to be derived from Equation (21);

$$\tilde{P}(\hat{f}) \equiv \int_0^1 \tilde{P}(\hat{f}, \hat{c}) d\hat{c},$$

$$\tilde{P}_A(\hat{f}) \equiv \lim_{\epsilon \rightarrow 0} \int_{|\epsilon|}^1 \tilde{P}(\hat{f}, \hat{c}) d\hat{c},$$

$$\tilde{A} \equiv \int_0^1 \tilde{P}_A(\hat{f}) d\hat{f},$$

and

$$\tilde{P}_A^*(\hat{f}) \equiv \tilde{P}_A(\hat{f}) / \tilde{A}, \quad (24)$$

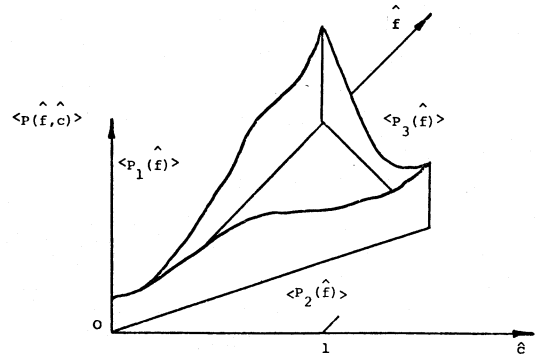


Fig. 4. Sketch of $\langle P(\hat{f}, \hat{c}) \rangle$ for an arbitrarily fuelled flame.

hence,

$$\langle \rho \rangle \frac{\tilde{D}}{Dt} \tilde{P}(\hat{f}) = \frac{\partial}{\partial x_i} B_1 \nu l \langle \rho \rangle \frac{\partial}{\partial x_i} \tilde{P}(\hat{f}) - \frac{\partial^2}{\partial \hat{f}^2} G_f(\hat{f}), \quad (25)$$

$$\langle \rho \rangle \frac{\tilde{D}\tilde{A}}{Dt} = \frac{\partial}{\partial x_i} B_1 \nu l \langle \rho \rangle \frac{\partial \tilde{A}}{\partial x_i} + \lim_{\hat{c} \rightarrow 0^+} \frac{\partial}{\partial \hat{c}} G_c(\hat{c}), \quad (26)$$

and

$$\begin{aligned} \langle \rho \rangle \frac{\tilde{D}}{Dt} \tilde{P}_A^*(\hat{f}) &= \frac{\partial}{\partial x_i} B_1 \nu l \langle \rho \rangle \frac{\partial}{\partial x_i} \tilde{P}_A^*(\hat{f}) - \frac{\partial^2}{\partial \hat{f}^2} G_f(\hat{f}) \\ &\quad - \frac{1}{\tilde{A}} \frac{\partial^2}{\partial \hat{f}^2} \int_{0^+}^1 \{ G_f(\hat{f}) \tilde{P}(\hat{c}) \\ &\quad - G_{ff}(\hat{f}, \hat{c}) \} d\hat{c} \\ &\quad + \frac{1}{\tilde{A}} \lim_{\hat{c} \rightarrow 0^+} \frac{\partial}{\partial \hat{c}} \{ G_{cc}(\hat{f}, \hat{c}) \\ &\quad - \tilde{P}_A^*(\hat{f}) G_c(\hat{c}) \} \\ &\quad + 2B_1 \nu l \langle \rho \rangle \frac{1}{\tilde{A}} \frac{\partial \tilde{A}}{\partial x_i} \frac{\partial}{\partial x_i} \tilde{P}_A^*(\hat{f}) \end{aligned} \quad (27)$$

A notable feature of these equations is that there is no term containing $S(\hat{f}, \hat{c})$: that is, in the limit of fast reactions, the behaviour of $\tilde{P}(\hat{f}, \hat{c})$ is independent of the specific form of the reaction rate. A similar result was obtained for premixed systems [18], in which case \tilde{A} is directly proportional to the mass fraction of combustion products, \tilde{c} . $\tilde{P}(\hat{f})$

is a passive scalar which, as can be seen, is transported in position space by mean flow and turbulent convection and in composition space by viscous mixing. The first three terms in the equation for $\tilde{P}_A^*(\hat{f})$ have similar interpretations while the physical significance of the remaining terms is less clear since they arise from the combined effects of transport of $\tilde{P}_A(\hat{f})$ and \tilde{A} . Equation (26) reveals that \tilde{A} is transported by mean flow and turbulent convection and may be increased by the final term which is by definition nonnegative: the term accounts for the decrease of inactive species by viscous mixing with active species.

The one-dimensional correlations, $G_f(\hat{f})$ and $G_c(\hat{c})$, appearing in the equation have been modelled by Dopazo [17] and Pope [18]. The former author's proposed closure for these terms is based on the assumption that the conditional probability density function of f' at a point x' , given f at x , is conditionally Gaussian. While this may be a good assumption for near Gaussian probability distribution functions, it has been shown [18], that in general such modelling is contrary to several restraints imposed on the exact expression. As a result, the model can, in some circumstances, produce unrealistic results such as negative probability. On the other hand, the proposal of Ref. [18] does not have this defect but its quantitative behaviour is suspect since it does not result in Gaussian probability distributions in homogeneous flows. The modelled term is:

$$G_\phi(\hat{\phi}) = B_2 \phi'' \frac{v}{l} \langle \rho \rangle \times \int_{-\infty}^{\hat{\phi}} \ln \left(1 + \frac{\hat{\phi} - \phi^*}{\phi''} \right) \tilde{P}(\phi^*) d\phi^* \times \int_{\hat{\phi}}^{\infty} \ln \left(1 + \frac{\phi^* - \hat{\phi}}{\phi''} \right) \tilde{P}(\phi^*) d\phi^* \quad (28)$$

which results in the source of activated species being given by

$$S_A \equiv \lim_{\hat{c} \rightarrow 0^+} \frac{\partial G_c}{\partial \hat{c}}(\hat{c}) = B_2 \frac{v}{l} \langle \rho \rangle (1 - \tilde{A}) \times \int_0^1 \ln(1 + \hat{c}/c'') \tilde{P}(\hat{c}) d\hat{c} \quad (29)$$

where

$$\phi'' \equiv \overline{\phi'^2}^{1/2}$$

The remaining unclosed terms are the two-dimensional correlations $G_{ff}(\hat{f}, \hat{c})$ and $G_{cc}(\hat{f}, \hat{c})$. Linear closures for these terms, such as the assumption that the two-point conditional probability is Gaussian, result in the terms in Eq. (27) in curled brackets being zero. A satisfactory closure for $G_{ff}(\hat{f}, \hat{c})$ analogous to Eq. (28) has not been found and so, in spite of the objections to linear modelling mentioned above, the attractive result that two terms in Eq. (27) can be neglected will be maintained.

A closure has now been achieved and will be used, in the next section, to assess existing combustion models: in summary, the transport equations for $\tilde{P}(\hat{f})$, $\tilde{P}_A^*(\hat{f})$ and \tilde{A} are closed by modelling the turbulent convection through Eq. (19), $G_\phi(\hat{\phi})$ through Eq. (28) and neglecting the terms involving the two-dimensional correlations G_{cc} and G_{ff} . From the definition of these three quantities, averaged values of scalars can be evaluated through,

$$\langle \phi(f, c) \rangle = \langle \rho \rangle \int_0^1 \{ \tilde{P}(\hat{f}) - \tilde{A} \tilde{P}_A^*(\hat{f}) \} \phi(\hat{f}, 0) / \rho(\hat{f}, 0) + \tilde{A} \tilde{P}_A^*(\hat{f}) \phi(\hat{f}, c_A(\hat{f})) / \rho(\hat{f}, c_A(\hat{f})) d\hat{f} \quad (30)$$

The modelling of $G_\phi(\hat{\phi})$ is not completely satisfactory but, unlike other proposals, Eq. (28) ensures that the predicted behaviour of $\tilde{P}(\hat{f})$ is qualitatively correct.

4. COMBUSTION MODELS

4.1 Arbitrarily fuelled flames

The closure outlined in the last section contains two integrodifferential equations whose subjects are functions both of position, x , and of composition, \hat{f} . To solve this set of equations for a practical flame would be a considerable task and, bearing in mind the uncertainties in the modelling, the reward might not justify the effort. A more attractive approach is to reduce the complexity of the

closure by modelling the probability distributions in terms of their moments. Such a model has been proposed by Lockwood and Naguib [9] for arbitrarily fuelled flames. It rests on the assumptions that $\tilde{P}(\hat{f})$ and $\tilde{P}_A^*(\hat{f})$ have the same shape, namely, a "clipped-Gaussian" based on the values of \tilde{f} and \tilde{f}'^2 , and that the source of activated species is given by,

$$S_A = B_3 \langle \rho \rangle \frac{v}{l} (\tilde{A}(1 - \tilde{A}))^{1/2}. \quad (31)$$

In addition, unweighted and mass-weighted quantities are not distinguished explicitly.

The equality of $\tilde{P}(\hat{f})$ and $\tilde{P}_A^*(\hat{f})$ is suggested by the similar form of Eqs. (25) and (27) and indeed, were it not for the final term in Eq. (27), the two equations would yield identical results. Thus, this expedient assumption, which was not stated explicitly by the originators of the model, finds theoretical support. Experimental support for the contention that $\tilde{P}(\hat{f})$ can be expressed in terms of \tilde{f} and \tilde{f}'^2 is provided by Rhodes [27] who shows that probability distributions of passive scalars are well represented by incomplete beta functions. Not only do these functions appear to provide a better representation than do clipped-Gaussians, they are also easier to incorporate into a numerical solution procedure since the distribution can be determined explicitly from the moments \tilde{f} and \tilde{f}'^2 . The transport equations for these two scalars can be derived from Eq. (25);

$$\langle \rho \rangle \frac{D\tilde{f}}{Dt} = \frac{\partial}{\partial x_i} B_1 v l \langle \rho \rangle \frac{\partial \tilde{f}}{\partial x_i}, \quad (32)$$

$$\begin{aligned} \langle \rho \rangle \frac{D\tilde{f}'^2}{Dt} = & \frac{\partial}{\partial x_i} B_1 v l \langle \rho \rangle \frac{\partial \tilde{f}'^2}{\partial x_i} \\ & + 2B_1 v l \langle \rho \rangle \left(\frac{\partial \tilde{f}}{\partial x_i} \right)^2 - \int_0^1 G_f(\hat{f}) d\hat{f}. \end{aligned} \quad (33)$$

These equations are the same as those used by Lockwood and Naguib except that the viscous de-

struction term is modelled after Corrsin [28] in its more familiar form,

$$\int_0^1 G_f(\hat{f}) d\hat{f} = \Gamma \left\langle \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_i} \right\rangle = B_4 \langle \rho \rangle \frac{v}{l} \tilde{f}'^2. \quad (34)$$

While the modelling of $G_f(f)$ by assumed Gaussian behaviour or by Eq. (28) is not entirely satisfactory, the modelling of its integral by Eq. (34) is well established.

The transport equations for \tilde{f} and \tilde{f}'^2 together with an assumed probability distribution form a closure for diffusion flames for which all species are assumed active, i.e., $\tilde{A} = 1$. Successful predictions of diffusion flames [5-7], serve to support the modelling of $\tilde{P}(\hat{f})$ in this manner.

The final major assumption is in the modelling of S_A . Lockwood and Naguib suggested Eq. (31) by analogy with Spalding's eddy-break-up model, Eq. (29) results from modelling $G(\hat{c})$ by Eq. (28) while Dopazo's modelling of this term fails to produce a meaningful result. In the last section it was shown how S_A is related to $G(\hat{c})$ and consequently the practice of modelling S_A in terms of $\tilde{P}(\hat{c})$ is validated. However, in spite of successful predictions based on Eqs. (29) and (31), both formulae can be shown to break down in some circumstances producing implausible results. This failure, which stems from the fact that the predicted values of $\partial S_A / \partial \tilde{A}$ becomes infinite as \tilde{A} tends to zero, is best demonstrated by considering Eqs. (26) and (31) applied to a steady one-dimensional flow;

$$\begin{aligned} \langle \rho \rangle U_1 \frac{d\tilde{A}}{dx_1} = & \frac{d}{dx_1} B_1 v l \langle \rho \rangle \frac{d\tilde{A}}{dx_1} \\ & + B_3 \langle \rho \rangle \frac{v}{l} \{\tilde{A}(1 - \tilde{A})\}^{1/2}. \end{aligned} \quad (35)$$

Consider the case in which the convective term dominates the diffusion term and consequently the solution is only sensitive to the upstream boundary condition, at $x_1 = 0$, say. Then with the boundary condition $\tilde{A}(0) = 0$, a solution is given by $\tilde{A}(x_1) = 0$ which corresponds to inactive combustibles flowing downstream and the solution indicates that no combustion takes place. Clearly,

this solution can always exist and, for flow velocities above some limit (the turbulent flame speed) it is the only solution. However, in the vicinity of $x_1 = 0$, Eq. (35) displays the solution

$$\tilde{A} = (2B_3 v / U_1 x_1 / l)^2 \quad (36)$$

indicating a finite amount of combustion irrespective of the flow velocity. That is, the model predicts the incorrect result that combustion can propagate against any flow velocity. This finding condemns the specific modelling of S_A by Eqs. (29) and (31) but the principle of modelling S_A in terms of $\tilde{P}(\hat{c})$ or its moments remains sound: it does require, however, that $\partial S_A / \partial \tilde{A}$ be finite as \tilde{A} tends to zero.

In common with all the models mentioned in the Introduction (excepting [4]) Lockwood and Naguib ignored density fluctuation correlations which introduces unnecessary inaccuracy since density-weighted and unweighted scalars can be evaluated exactly through Eq. (13).

In spite of these criticisms Lockwood and Naguib's model, which was derived mainly by intuition, contains the essence of what may be considered the soundest closure at this level: namely, to solve equations for \tilde{f} , $\widetilde{f'^2}$ and \tilde{A} and hence to determine averaged quantities through Eq. (30) by assuming $\tilde{P}(\hat{f})$ and $\tilde{P}_A^*(\hat{f})$ to be incomplete beta-functions. The main uncertainty is in S_A which again stems from the unsatisfactory modelling of $G_c(\hat{c})$.

4.2 Diffusion Flames

For diffusion flames the assumption of fast reactions is interpreted to mean that fuel and oxidant are not copresent and, consequently, $\langle P(\hat{f}, \hat{c}) \rangle$ is only nonzero on $\hat{c} = c_A(\hat{f})$. Thus, inactive species can only occur at $\hat{f} = 0$ or 1 (corresponding to pure oxidant or pure fuel) and, as the properties of active and inactive species are the same in those regions, no error is caused by setting \tilde{A} to unity everywhere. As a consequence an equation for \tilde{A} need not be solved and the major uncertainty in the model for arbitrarily fuelled flames, S_A , is avoided.

The models used by Elgobashi and Pun [6] and

Lockwood and Naguib [7] for diffusion flames are equivalent to their model for arbitrarily fuelled flames with $A = 1$ and consequently the same assessment applies. On the other hand Khalil *et al.* [3] and Gosman and Lockwood [5] assumed a double delta function for $\langle P(\hat{f}) \rangle$ which, though computationally more economical, is clearly less realistic. Nevertheless, the agreement between measurements and the calculations of all these authors is good, confirming the principles of the modelling.

4.3 Premixed Flames

Homogeneously premixed flames are another special case of arbitrarily fuelled flames in that f is uniform and consequently $\langle P(\hat{f}) \rangle$ is known, i.e.,

$$\langle P(\hat{f}) \rangle = \delta(\hat{f} - f_0). \quad (37)$$

The degenerate form of the combustion model for this situation comprises the transport equation for \tilde{A} from which all scalars can be evaluated;

$$\begin{aligned} \langle \phi(f_0, c) \rangle = \langle \rho \rangle \{ & (1 - \tilde{A}) \phi(f_0, 0) / \rho(f_0, 0) \\ & + \tilde{A} \phi(f_0, c_A(f_0)) / \rho(f_0, c_A(f_0)) \}. \end{aligned} \quad (38)$$

Unfortunately, the most important constituent of the model, S_A , is also the least sound but even so it has been used to predict bluff-body-stabilised flames with a maximum error in flame width of 30% [4].

The traditionally used eddy-break-up model for premixed flames [1] differs significantly from that outlined above: it comprises transport equations for $\langle c \rangle$ and $\langle c'^2 \rangle$,

$$\begin{aligned} \langle \rho \rangle \frac{D\langle c \rangle}{Dt} &= \frac{\partial}{\partial x_i} B_1 v l \langle \rho \rangle \frac{\partial \langle c \rangle}{\partial x_i} + B_5 \langle \rho \rangle \frac{v}{l} \langle c'^2 \rangle^{1/2}, \quad (39) \\ \langle \rho \rangle \frac{D\langle c'^2 \rangle}{Dt} &= \frac{\partial}{\partial x_i} B_1 v l \langle \rho \rangle \frac{\partial \langle c'^2 \rangle}{\partial x_i} + 2B_1 v l \langle \rho \rangle \left(\frac{\partial \langle c \rangle}{\partial x_i} \right)^2 \\ &\quad - B_4 \langle \rho \rangle \frac{v}{l} \langle c'^2 \rangle, \quad (40) \end{aligned}$$

and relates averaged scalars such as density and

temperature by

$$\langle \phi(f_0, c) \rangle = \phi(f_0, \langle c \rangle). \tag{41}$$

In addition to the neglect of density fluctuation correlations, the eddy-break-up model suffers from three defects which indicate that it is far removed from the physical picture of flames at the fast reaction limit provided by the probability equations:

- (1) Equation (41) is incorrect and, in a typical flame, can be in error by 50% compared with Eq. (38) [4];
- (2) Equation (40) is only appropriate to passive scalars since a term, of order of magnitude $\langle \rho \rangle \langle c'^2 \rangle / l$, has been omitted;
- (3) Equations (39) and (40) do not necessarily provide unique solutions.

The first two points are easily seen since, in both respects, the model is contrary to the double delta function distribution of c appropriate to this situation.

The third point is best demonstrated by considering the hypothetical case in which the density is uniform and consequently the velocity and turbulence fields are independent of $\langle c \rangle$ and $\langle c'^2 \rangle$: then, the homogeneity of Eqs. (39) and (40) ensures that, if one solution is given by $\langle c \rangle = \langle c \rangle_1$ and $\langle c'^2 \rangle = \langle c'^2 \rangle_1$, an infinity of solutions are given by

$$\langle c \rangle = \langle c \rangle_{in} - a(\langle c \rangle_{in} - \langle c \rangle_1),$$

and

$$\langle c'^2 \rangle = a^2 \langle c'^2 \rangle_1 \tag{42}$$

where $\langle c \rangle_{in}$ is the inlet boundary condition on $\langle c \rangle$ and a is an arbitrary positive number. The reason that this defect has gone unnoticed is probably either that inhomogeneous boundary conditions were applied (e.g., [2]) or that $\langle c \rangle$ was artificially constrained to be bounded by zero and unity (e.g., [4]) so destroying the homogeneity of the equations and ensuring a unique solution.

Thus, while the model for premixed flames based on the equation for \tilde{A} contains the uncertain term S_A , it is to be preferred to the eddy-break-up model which has but modest foundations.

5. CONCLUSION

An arbitrarily fuelled flame has been considered which for a high Reynolds number, low Mach number, adiabatic flow can be characterised by the mass fraction of combustion products, c , and the total fuel mass fraction, f , provided that the reactants combine in fixed proportions to produce products of a given composition. The statistical properties of such a flame can be determined from the joint probability of f and c for which a transport equation has been derived. The joint probability is transported in position space by mean flow and turbulent convection and in composition space by chemical reaction and viscous mixing. In the limit of very rapid reaction, the joint probability distribution adopts a shape expressible in terms of the two single probability distributions, $\tilde{P}(f)$, $\tilde{P}_A^*(f)$, and the mass fraction of activated species \tilde{A} . A closure has been obtained by deriving and modelling transport equations for these quantities: turbulent convection is modelled by a mixing length hypothesis while the modelling of the term involving viscous mixing, $G_\phi(\hat{\phi})$, is only partially successful. These equations reflect the experimental observation that, in the limit of rapid reaction, the macroscopic properties of the flame are independent of the reaction rate.

This closure has been used to assess existing combustion models and to clarify the assumptions on which they are based. The models for arbitrarily fuelled flames [9] and for diffusion flames [6] fare well although there is evidence that incomplete beta functions provide better representations of the probability distribution of passive scalars than do the clipped-Gaussians used by these models. In addition, their neglect of density fluctuation correlations is unnecessary. On the other hand, the eddy-break-up model for premixed flames [1] is found to be deficient on both physical and mathematical grounds and an alternative is provided. The source of activated species, S_A , has been identified as the principal remaining weakness in the closures and its successful modelling has been facilitated by relating it exactly to the viscous mixing term, $G_c(\hat{c})$.

This work has been carried out with the support of Procurement Executive, Ministry of Defence.

NOMENCLATURE

A	mass fraction of activated species
a_1, a_2, a_3	constants of combination, Eq. (4)
B	constants in closure approximations
c	mass fraction of products
$c_A(f)$	maximum possible value of c , given f
f	total fuel mass fraction
$G_f(\hat{f})$, $G_{fc}(\hat{f}, \hat{c})$	one- and two-dimensional viscous mixing correlations, Eq. (20)
h	enthalpy
l	length scale of turbulence
m	mass fraction
$P(\hat{f}, \hat{c})$	joint probability distribution function
$P_A^*(\hat{f})$	normalised probability distribution of activated species
p	pressure
S	source
t	time
U_i	component of velocity vector
v	velocity scale of turbulence
x_i	component of position vector
y	constant of combination, Eq. (4)
Γ	diffusion coefficient
δ	Dirac delta function
ρ	density
ϕ	scalar
$\langle f \rangle$	ensemble average value of f
\tilde{f}	Favre average value of f
\hat{f}	independent variable corresponding to f
f'	fluctuating component of f
f''	Favre average fluctuating component of f

REFERENCES

- Spalding, D. B., *Thirteenth Symposium (International) on Combustion*, The Combustion Institute, 1970, p. 649.
- Mason, H. B., and Spalding, D. B., *Combustion Institute European Symposium*, Academic Press, 1973, p. 601.
- Khalil, E. E., Spalding, D. B., and Whitelaw, J. H., *Int. J. Heat Mass Transfer* 18, 775 (1975).
- Pope, S. B., The calculation of the flow behind bluff bodies with and without combustion, Ph.D. Thesis, Univ. London, 1976.
- Gosman, A. D., and Lockwood, F. C., *Proceedings of International Seminar on Heat Transfer from Flames*, Trogir, Yugoslavia, 1973.
- Elgobashi, S. E., and Pun, W. M., *Fifteenth Symposium (International) on Combustion*, The Combustion Institute, 1974, p. 1353.
- Lockwood, F. C., and Naguib, A. S., *Combust. Flame* 24, 109 (1975).
- Toor, H. L., *A.I.Ch.E.J.* 8, 70 (1962).
- Lockwood, F. C., and Naguib, A. S., *Second Combustion Institute European Symposium*, 1975, p. 502.
- Hopf, E., *J. Ration. Mech. Anal.* 1, 87 (1952).
- Lundgren, T. S., *Phys. Fluids* 12, 485 (1969).
- Lundgren, T. S., in *Statistical Models and Turbulence*, ed. M. Rosenblutt and C. Van Atta, Springer-Verlag, Berlin, 1972, p. 70.
- Fox, R. L., *Phys. Fluids* 14, 1806 (1971).
- Fox, R. L., *Phys. Fluids* 16, 977 (1973).
- Ribeiro, M. M., The turbulence structure of free jet flows with and without swirl, Ph.D. Thesis, Univ. London, 1976.
- Dopazo, C., and O'Brien, E. E., *Phys. Fluids* 16, 2075 (1973).
- Dopazo, C., *Phys. Fluids* 18, 397 (1975).
- Pope, S. B., The probability approach to the modelling of turbulent reacting flows, *Combust. Flame* 27, 299 (1976).
- Launder, B. E., and Spalding, D. B., *Mathematical Models of Turbulence*, Academic Press, London, 1972.
- Pope, S. B., and Whitelaw, J. H., *J. Fluid Mech.* 73, pt. 1, 9 (1976).
- Bray, K. N. C., Kinetic energy of turbulence in flames, University of Southampton, *AASU Report* 332 (1974).
- Howe, N. M., Shipman, C. W., and Vranos, A., *Ninth Symposium (International) on Combustion*, The Combustion Institute, 1962, p. 36.
- Lockwood, F. C., and Odidi, A. O. O., *Fifteenth Symposium (International) on Combustion*, The Combustion Institute, 1974, p. 561.
- Guenther, R., and Simon, H., *Twelfth Symposium (International) on Combustion*, The Combustion Institute, 1968, p. 1069.
- Bilger, R. W., *Comb. Sci. Tech.* 11, 215 (1975).
- Bray, K. N. C., and Moss, J. B., A unified statistical model of the premixed turbulent flame, University of Southampton, *AASU Report* 335 (1974).
- Rhodes, R. P., A probability distribution function for turbulent flows, in *Turbulent Mixing in Non-reactive Flows*, Purdue University, 1974, p. 235.
- Corrsin, S., *Phys. Fluids* 1, 42 (1958).

Received 22 June 1976; revised 10 November 1976