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Statistical Methods in Reacting Turbulent Flows

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Modern combustion theory often finds it necessary to take into account fluctuations in velocity, composition and thermodynamic state variables of a reacting flow system. Methods for doing this have been intensively explored, especially over the past decade. It has been found that reactive flows pose exceptionally difficult problems for moment methods of analysis. The alternative of a probability density function description is now developed sufficiently for its potential to be assessed. The possibilities of a mixed description, partially by moment formulation and partially by probability density functions, have also been explored.

I. Introduction

An escalating concern for the details of pollutant emissions and, more recently, a demand for higher performance of combustors have obliged researchers to incorporate as much relevant fluid mechanics as possible into their combustion analyses and models. Improved experimentation has also been a factor. Symptoms of this activity can be seen in the rapid increase in special meetings,[1] survey papers,[2] review articles[3-6] and texts[7,8] which explore the role of turbulence in reactive systems. It is not a one way street; turbulent reactive flow studies have found their way into specialized fluid mechanics meetings[9] because reactants pose special problems which demand a better understanding of the microscale processes in turbulence. In some instances reactants have been used experimentally to display small structure dynamics.[10] Furthermore, the reaction process is most naturally dealt with in physical space (as distinct from wave number space) where, on the continuum scale, only species which coexist at a point can react. During the last decade, much of the interest in non-reactive turbulent shear flows has also centered on events in physical space, usually dubbed "characteristic structures." It is probable that a better understanding of turbulent reactive shear flows will only come when both of these features have been exploited and joined to the more common spectral descriptions of turbulence and turbulent transport. Some current theoretical approaches to turbulent reactive flow seem now to have reached a plateau of development after a decade of intensive research activity. One of these, certainly, is the development of a probability density function (pdf) formalism based on the conservation laws for reactive systems. The method of obtaining the pdf equation is well established[11]; the proper technique for assigning boundary conditions is not.[12] Closure approximations have been proposed for unclosed terms in the pdf evolution equation but are not yet fully evaluated; nevertheless, they have begun to appear in reactive flow modeling with encouraging results.

A second theme to have seen significant development is the "fast chemistry/conserved scalar" approach to turbulent diffusion flames.[13] It now may be ripe for extension to systems in which reaction is not overwhelmingly rapid[14] compared to the turbulent mixing time, at least for some species. This technique also relies on the pdf description in a fundamental way and is probably the best explored avenue of application of the method. It is possible, however, that the application to diffusion flames does not represent the most stringent test of the method since only certain integral properties of the pdf may be important in these cases.[15]

On the other hand, the moment formalism, which includes correlation and spectral methods, seems not to have progressed as rapidly, despite having received the earliest attention and being the description which has succeeded best in describing turbulent shear flows. A major difficulty is the mean production rate term of the species balance equation[16] which is unclosed for all nonlinear reactions. For example, a global, second order, forward reaction with an Arrhenius rate expression has the rate of production of species i, $\dot{w}_i$ given by

$$\dot{w}_i = -\rho^2 k Y_i Y_j \exp\left(-T_a/T\right) \tag{1}$$

where ρ is the mixture density, k the rate constant, Y_i the mass fraction of species i, T_a the activation temperature, and T the temperature.

In a turbulent combustion situation ρ, Y_i, Y_j and T are random variables correlated through the mass conservation equation, the species balance equations, the energy equation, the Navier-Stokes equations, and some thermodynamic state relationships.

A closure at a low order moment level (say, second order) requires expressing the entire statistics of this conglomerate variable $\dot{w}_i$ in terms of the first and second moments of its constituent variables. See, for example, Ref. 18.

The term $\dot{w}_i$ occurs in a conservation equation of the type[16]

$$\rho\frac{\partial Y_i}{\partial t} + \rho\, \underset{\sim}{u}\cdot\nabla Y_i - \nabla\cdot\rho D_i \nabla Y_i = \dot{w}_i \tag{2}$$

where $\underset{\sim}{u}$ is the advecting velocity field and D_i the coefficient of molecular diffusion of species i. Since $T_a/T \gg 1$ in the typical combustion case, an expansion of the exponential in Eq. (1) is restricted to uninterestingly small temperature fluctuations. However, even in the case of constant density, isothermal, and statistically homogeneous flows ($\overline{\underset{\sim}{u}\cdot\nabla Y}=0$; the bar indicates an ensemble average), the average of Eq.(2) leads, on the right-hand side, to the unclosed expression $\dot{w}_i = C\overline{Y_i Y_j}$, where C is the (constant) reaction rate. For very rapid reactions of non-premixed species, the time scale of $\dot{w}_i$ has been shown to depend on the turbulent mixing time,[19] not on the chemical rate. Hence, $\overline{Y_i Y_j} = 0(C^{-1})$ and the fluctuations in concentration are then of the same order as the mean. Any attempt to relate third-order concentration moment by expansion about lower orders seems unwise.

For less severe reactive flow conditions than those posed by combustion, use can be made of the existence of bounds[20] on the mass fractions to suggest closure approximations. An important parameter to characterize the usefulness of this "realizability" approach is the relative intensity of the concentration $\overline{(Y_i')^2}/\overline{Y}_i^2$ where Y_i' is the fluctuation defined by $Y_i' = Y_i - \overline{Y}_i$. The simple recombination reaction $A + A \rightarrow 2A$, which obeys $dY/dt = -Y^2$ (the reaction rate has been incorporated into the time variable t) has been subjected to "realizable" closures and to exact stochastic solution.[21]

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The relative intensity obtained from different closures varied by factors of 6 or 7 before the mean had decayed 30% and, if the relative intensity is initially high, these results can affect profoundly the decay of the mean, since

$$\frac{\mathrm{d}\overline{Y}}{\mathrm{d}t} = -\overline{Y}^2\left(1+\frac{\overline{Y_1^2}}{\overline{Y}^2}\right)$$

Most recent modeling efforts using the moment formulation have also introduced the pdf to describe the production rate term, either explicitly[22] or implicitly through the construction of cells in composition space.[18]

II. Probability Density Function Method

Techniques for obtaining an equation for the pdf of any variables jointly obeying a set of partial differential equations were presented independently and almost simultaneously by Monin,[23] Novikov,[24] and Lundgren[11] in the context of the incompressible Navier-Stokes equations. The latter used the "fine-grained density" method which is the one we will discuss here since it is the simpler and more efficient route to the same result.

Consider, at any fixed point $\underset{\sim}{x}$ and time t, a scalar field $Y(\underset{\sim}{x},t)$ which takes random values, perhaps because of transport by a turbulent velocity field. It may also be reacting with other species and undergoing molecular diffusion. Moments can be represented in the usual way by taking ensemble averages of the set $Y(\underset{\sim}{x},t)$. For example, the mean would be represented by $\bar{Y}(\underset{\sim}{x},t)$. The role of the "fine-grained density" is to give a probabililty density representation to each realization of the random field. That is, it gives a pdf representation to any particular realization of $Y(\underset{\sim}{x},t)$, as we now show.

Suppose the function $Y^*(\underset{\sim}{x},t)$ describes one realization of the random field $Y(\underset{\sim}{x},t)$ and let us denote the "fine-grained density" of this realization by $\mathcal{P}(\hat{Y};\underset{\sim}{x},t)=\delta[Y^*(\underset{\sim}{x},t)-\hat{Y}]$ $\mathcal{P}(\hat{Y};\underset{\sim}{x},t)$ is clearly a function of $\hat{Y}$ and a functional of $Y^*(\underset{\sim}{x},t)$, since it depends on the entire set of values of Y^*. Consider a fixed value of $\hat{Y}$, say $\hat{Y}_F$, and fix also the space time point $(\underset{\sim}{x},t)$ of interest. Then $\mathcal{P}(\hat{Y}_F;\underset{\sim}{x},t)$ is zero if $Y^*(\underset{\sim}{x},t)\neq\hat{Y}_F$ and is infinite (the delta function) if $Y^*(\underset{\sim}{x},t)=\hat{Y}_F$. For any particular realization $Y^*(\underset{\sim}{x},t)$, it is also true that $\mathcal{P}$ has the required properties of a pdf including normalization, since $\int\mathcal{P}(\hat{Y};\underset{\sim}{x},t)\,\mathrm{d}\hat{Y}=1$ by definition of the δ function. Note also that for the $Y^*(\underset{\sim}{x},t)$ realization, the nth moment of Y is, by direct calculation, $\int\hat{Y}\mathcal{P}(\hat{Y};\underset{\sim}{x},t)\,\mathrm{d}\hat{Y}=Y^n(\underset{\sim}{x},t)$ for all n. Thus $\mathcal{P}(\hat{Y};\underset{\sim}{x},t)=\delta[Y^*(\underset{\sim}{x},t)-\hat{Y}]$ contains precisely the same information as the realization $Y^*(\underset{\sim}{x},t)$ on which it is based.

In other words, the "fine-grained density" $\mathcal{P}(\hat{Y};\underset{\sim}{x},t)$ can be thought of as a device by which each realization of the random field is written in a pdf format. In particular, the ensemble average of $\mathcal{P}(Y;\underset{\sim}{x},t)$ is the customary pdf of the random field, denoted by $P(\hat{Y};\underset{\sim}{x},t)=\overline{\mathcal{P}}(\hat{Y};x,t)$.

The justification for obtaining mostly smooth, continuous pdf's from such a spiky field of realizations as are represented by $\mathcal{P}(\hat{Y};\underset{\sim}{x},t)$ was given by Stratonovich.[25] The use of this device is quite common in statistical physics.[26]

As an example of the method, we now derive the pdf equation for a constant density, constant diffusivity, single-species reactive field. The technique can easily be generalized to apply to more complicated reactive systems.

From Eq. (2), the species conservation equation is

$$\frac{\partial Y}{\partial t}+\underset{\sim}{u}\cdot\nabla Y=D\nabla^2 Y+\dot{w}/\rho \tag{3}$$

Let

$$\mathcal{P}(\hat{Y};\underset{\sim}{x},t)=\delta[Y(\underset{\sim}{x},t)-\hat{Y}]$$

Therefore,

$$\frac{\partial\mathcal{P}}{\partial t}=\frac{\partial\delta}{\partial t}=\frac{\partial\delta}{\partial Y}\frac{\partial Y}{\partial t}=-\frac{\partial\mathcal{P}}{\partial\hat{Y}}\frac{\partial Y}{\partial t}$$

On using Eq. (3)

$$\frac{\partial\mathcal{P}}{\partial t}=-\frac{\partial\mathcal{P}}{\partial Y}\left(-\underset{\sim}{u}\cdot\nabla Y+D\nabla^2 Y+\dot{w}/\rho\right)$$

or

$$\frac{\partial\mathcal{P}}{\partial t}=-\underset{\sim}{u}\cdot\frac{\partial\mathcal{P}}{\partial Y}\nabla Y-D\frac{\partial}{\partial\hat{Y}}(\nabla^2 Y\mathcal{P})-\frac{\partial}{\partial\hat{Y}}\left(\frac{\dot{w}}{\rho}\mathcal{P}\right)$$

Therefore

$$\frac{\partial\mathcal{P}}{\partial t}+\underset{\sim}{u}\cdot\nabla\mathcal{P}+\frac{\partial}{\partial\hat{Y}}\left(\frac{\dot{w}}{\rho}\mathcal{P}\right)+D\frac{\partial}{\partial\hat{Y}}(\nabla^2 Y\mathcal{P}) \tag{4}$$

Equation (4) is a partial differential equation for the "fine-grained density" $\mathcal{P}$ in the sense that for any infinitely differentiable test function $\psi(\hat{Y})$(a "good" function in Lighthill's terminology)[27]

$$\int\psi(\hat{Y})\left[\frac{\partial\mathcal{P}}{\partial t}+\underset{\sim}{u}\cdot\nabla\mathcal{P}+\frac{\partial}{\partial\hat{Y}}\left(\frac{\dot{w}}{\rho}\right)\right.$$
$$\left.+D\frac{\partial}{\partial\hat{Y}}(\nabla^2 Y\mathcal{P})\right]\mathrm{d}\hat{Y}\equiv 0 \tag{5}$$

Integration of Eq. (5) by parts verifies this result, when Eq. (3) is used.

On taking the ensemble average of Eq. (4), one finds

$$\frac{\partial P}{\partial t}+\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}}+\frac{\partial}{\partial\hat{Y}}\left(\frac{\dot{w}}{\rho}P\right)=-D\frac{\partial}{\partial\hat{Y}}(\overline{\nabla^2 Y}) \tag{6}$$

Equation (6) is the evolution equation we seek. It demonstrates many of the advantages and disadvantages of the pdf formulation, and we shall use it as a basis for the subsequent discussion of the method, generalizing without proof to problems of interest in combustion, where not only species balance equations, but also mass, momentum, and energy conservation are to be satisfied. The Lundgren technique applied to these more general cases can be found in Refs. 28-30. The Russian literature, which follows Monin's functional method, reached the same general results somewhat earlier.

From Eq. (6) we can immediately see the major advantage of the pdf method. The third term on the left-hand side is the chemical production rate term which proved so troublesome in the moment formulation. Here $\dot{w}$ is a known coefficient and the production rate term needs no approximation in the pdf formalism.[31] This result follows from the fact that the $\dot{w}$ has no differential or integral space operators in its definition and it remains true for all such specifications of chemical kinetics no matter how complicated.

This is a spectacular simplification and one must be prepared to pay a high price for it. The first disadvantage to be noted from Eq. (6) is that P is a function of five variables, namely $P(\hat{Y};\underset{\sim}{x},t)$ whereas the original species conservation equation for $Y(\underset{\sim}{x},t)$ had only four independent variables. More generally, if one considers a system of N reactants, three velocity components, one density, one enthalpy, and no symmetries in space or time, it is easy to show that the one point pdf is a function of $N+9$ variables. Four or five variables are probably the most one can hope to deal with numerically at the present time. The three velocity components can be removed as independent variables by considering $\underset{\sim}{u}$ as an externally prescribed random coefficient, as we have done in deriving Eq. (6). This is clearly not permissible if there is interest in the effects of reaction on the turbulence. It is also sometimes possible to consider density as a function of only species concentration and enthalpy in which case it is no longer an independent random variable. This excludes random acoustic modes, for example. Finally, symmetry assumptions such as stationarity in time, homogeneity in space, or similarity in two or more spatial dimensions may reduce the number of independent variables further. In any case, at the present time, direct modeling of combustion with the pdf equation is probably limited to systems with no more than three reactants. Consequently, various modeling strategies have been devised which make only partial use of the pdf equation. A brief summary of this activity is included in Sec. III.

Closure of the Advective Term

Equation (6) has two terms which are unclosed: an advective term $\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}}$ on the left-hand side and a molecular diffusion term on the right. The advective term occurs in any pdf formulation in which the velocity is treated as an external random variable. It could be removed by including the Navier-Stokes equations with Eq. (3) and defining a more general "fine-grained density."

$$\mathcal{P}(\hat{Y},\hat{u}_1,\hat{u}_2,\hat{u}_3;\underset{\sim}{x},t)$$

$$=\delta[Y(\underset{\sim}{x},t)-\hat{Y}]\prod_{i=1}^{3}\delta[u_i(\underset{\sim}{x},t)-\hat{u}_i]$$

where u_1, u_2, and u_3, are the three components of $\underset{\sim}{u}$. As we have stated, the disadvantage is the addition of the components of $\underset{\sim}{u}$ as three more independent variables. Furthermore, the pressure terms and the molecular diffusion terms of the Navier-Stokes equations are unclosed in such a description, as was pointed out by Lundgren in his original paper.

The most common approach[32] to approximating $\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}}$ has been to use gradient transport theory. To the usual difficulties of justifying such an approximation one can add the mathematical problem that $\mathcal{P}$ is unbounded. Nevertheless, for an incompressible velocity field, it is easy to obtain formally by this approach:

$$\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}}=\bar{\underset{\sim}{u}}\cdot\nabla P-\nabla\cdot\underset{\approx}{K}\nabla P$$

where $\underset{\approx}{K}$ is an eddy diffusivity tensor which can be equated to the traditional one since

$$\overline{\underset{\sim}{u}\cdot\nabla Y}=\overline{\underset{\sim}{u}\cdot\nabla\int\hat{Y}\mathcal{P}(\hat{Y};\underset{\sim}{x},t)\mathrm{d}\hat{Y}}=\int(\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}})\,\hat{Y}\mathrm{d}\hat{Y}$$

An alternate approximation for $\overline{\underset{\sim}{u}\cdot\nabla\mathcal{P}}$ has also been proposed.[33] We will not discuss it here as it appears to have no experimental support at present and is difficult to use in modeling.

Closure of the Molecular Diffusion Term

The unclosed nature of the diffusion term on the right-hand side of Eq. (6) is characteristic of all pdf formulations. While the advective nonclosure will disappear in a statistically homogeneous field (a well-stirred reactor, for example) the molecular diffusion effect will always remain. It was shown by Fox[34] that it can be split into two phenomena, one of which is transport in physical space and the other is transport in composition space.

The physical space term in Eq. (6) becomes $D\nabla^2P$, which, at moderate Reynolds numbers, is usually negligible compared to the advective transport term $\nabla\cdot\underset{\approx}{K}\nabla P$. The composition space transport of pdf is the term which carries information about microscale mixing. It can be written for Eq. (6) in the form

$$-D\frac{\partial^2}{\partial\hat{Y}^2}\left(\overline{\frac{\partial Y}{\partial x_i}\frac{\partial Y}{\partial x_i}\mathcal{P}}\right) \tag{7}$$

The term $(\partial Y/\partial x_i)(\partial Y/\partial x_i)$ is easily recognized as the (unaveraged) dissipation function of the scalar field $Y(\underset{\sim}{x},t)$. It is always nonnegative, as is $\mathcal{P}$. Considerable effort and some controversy has been generated in attempts to model Eq. (7), or the full right-hand side of Eq. (6).

Simple stochastic assumptions such as the statistical independence[13] of $\mathcal{P}$ and $(\partial Y/\partial x_i)(\partial Y/\partial x_i)$ or linear mean square estimation techniques,[28] can yield unphysical results in some contexts. In particular, the details of the microscale mixing process seem to be badly misrepresented.

For example, one can show that a linear mean square estimation closure leads to the following unsatisfactory result. Suppose we consider molecular mixing of a binary temperature field which has the initial values T_1 and T_2, where T_1 is less than T_2, and suppose there are equal proportions of fluid carrying each temperature. The initial description in terms of pdf's is a pair of delta functions at T_1 and T_2, with coefficients of ½ to maintain normalization. It turns out[35] that the closure predicts that the delta functions will remain as such for all finite time. They simply move in at identical rates toward the line of symmetry at $T=½(T_1+T_2)$. This is clearly not microscale mixing; during the mixing process, one expects to generate a continuous distribution of temperatures rather than maintain a binary field for all time. There are two other difficulties with the simple closures; they cannot represent the physical boundary conditions correctly[28] and even in an infinite space, they are awkward to use in numerical modeling schemes.[36]

Recent proposals for closure of the molecular diffusion term suggest the use of an integral form in composition space. The most complete description of the method can be found in Refs. 29, 35, and 36. Vignon[37] reached a similar conclusion, independently.

For example, a mechanistic model of mixing[36] without length scales produces the following approximation to the right-hand side of Eq. (6)

$$-\frac{D_\tau}{\mathrm{T}_y}\left(\int_0^y\mathrm{d}\hat{Y}'P(\hat{Y}')\int_Y^1\mathrm{d}\hat{Y}''P(\hat{Y}'')P(\hat{Y}',\hat{Y}'',\hat{Y})-P(\hat{Y})\right) \tag{8}$$

where the integral term represents coalescence to produce $\hat{Y}$ and loss of $\hat{Y}$ is taken proportional to the existing $P(\hat{Y})$. $P(\hat{Y}',\hat{Y}'',\hat{Y})$ is a transition probability linking the production of $\hat{Y}$ to the existence jointly of $\hat{Y}'$ and $\hat{Y}''$ and it has clearly defined mathematical properties. The coefficients D_τ and T_y represent, respectively, a turbulent diffusivity, and a microscale mixing time. It is clear that a good deal of physics is lost in such modeling, although linear mean square estimation can be used to suggest the appropriate forms for D_τ and T_y in terms of moment statistics of the $Y(\underset{\sim}{x},t)$ field.[31] In such a way D_τ/T_y can be related to λ and ϵ and the Schmidt number, where λ and ϵ are the microscale and dissipation function of the turbulence, respectively. $P(\hat{Y}',\hat{Y}'',\hat{Y})$ can be chosen so as to produce a realistic small scale mixing of an initially binary field, but it is clearly not unique. Equation (8) is in a convenient form for numerical modeling and has been applied with some success to a simple diffusion flame and to advection of temperature fluctuation in a turbulent round jet. It is not quite satisfactory from a statistical mechanics point of view since it does not seem to be a member of a recognizable class of stochastic processes and it does not have the correct asymptotic behavior in simple cases. Molecular diffusion is fundamentally a multipoint process and it may be unreasonable to expect modeling by a single-point pdf to do more than characterize the phenomena qualitatively. It remains to be seen if reactive flows can be adequately treated by any single point model.

Favre Averaging[38]

Fluctuating density has not been considered in the derivation of Eq. (6). For most combustion problems, it is essential to account for such fluctuations and it has become common practice to use mass-weighted (Favre) averaging for this purpose. It is worthwhile to consider the role of Favre averaging in the pdf formulation.

In the case that ρ is a known function of the other random variables in the system, say $\underset{\sim}{\hat{Y}}$, (the composition variables and enthalpy) then the use of Favre averaging is not a problem since moments with ρ can still be computed using $P(\underset{\sim}{\hat{Y}})$. By the same token, Favre averaging is not particularly meaningful since there is then a direct conversion between a Favre average and a traditional unweighted average. Since Favre averages are essentially impossible to determine directly by experimentation they probably should be avoided where they are unnecessary.

In the general case ρ is not a known function of $\underset{\sim}{\hat{Y}}$ but, due perhaps to turbulent compressibility effects, it is random in its own right. In this situation, Favre averaging can be an important technique. However, unless the production rate term $\dot{w}$ is a linear function of $\hat{\rho}$ the Favre averaged production rate term in the pdf formalism is unclosed and the *raison d'être* for a pdf method may be lost.

We can demonstrate this simply by reconsidering Eq. (2) using the "fine-grained density" defined by

$$\zeta(\hat{Y},\rho;\underset{\sim}{x},t)=\delta[Y(\underset{\sim}{x},t)-\hat{Y}]\delta[\rho(\underset{\sim}{x},t)-\hat{\rho}]$$

The technique of obtaining a pdf equation, outlined following Eq. (3), then yields

$$\begin{aligned}\frac{\partial\hat{\rho}\zeta}{\partial t}+\nabla\cdot\underset{\sim}{u}\,\hat{\rho}\zeta=&\frac{\partial}{\partial\hat{\rho}}\left[\hat{\rho}^{2}(\nabla\cdot\underset{\sim}{u})\zeta\right]\\&-D\frac{\partial}{\partial\hat{Y}}\left[\nabla\cdot\rho\nabla Y\zeta\right]-\frac{\partial}{\partial\hat{Y}}\left[\dot{w}\zeta\right]\end{aligned}$$

We use Bilger's definition of the Favre averaged pdf as

$$\tilde{P}(\hat{Y};\underset{\sim}{x},t)=\frac{1}{\bar{\rho}(\underset{\sim}{x},t)}\int\hat{\rho}\langle\zeta\rangle\mathrm{d}\hat{\rho}\tag{9}$$

to obtain

$$\begin{aligned}&\frac{\partial}{\partial t}\bar{\rho}\tilde{P}+\nabla\cdot\bar{\rho}\langle\underset{\sim}{u}\,\zeta\rangle\\&=-D\nabla\cdot\int\langle\rho\nabla Y\frac{\partial\zeta}{\partial Y}\rangle\mathrm{d}\hat{\rho}-\frac{\partial}{\partial\hat{Y}}\int\dot{w}\langle\zeta\rangle\mathrm{d}\hat{\rho}\end{aligned}\tag{10}$$

Here, an ensemble average is represented by $\langle\ldots.\rangle$.

Not surprisingly, the molecular diffusion term has been somewhat complicated, as always, by Favre averaging. Of more damaging consequence is the form of the reaction rate term which, unless $\dot{w}$ is linearly dependent on ρ, cannot be written as a closed form function of $\tilde{P}$ [see Eq. (9)]. Such linear behavior has been assumed implicitly in most studies. Bilger[39] has shown it to be valid in a study of perturbation about equilibrium for the hydrogen/air reaction.

Conditioned Averaging

The existence of sharp boundaries for turbulent shear flows is still largely ignored in reactive flow modeling, although it seems to be an important phenomenon to account for if entrainment and mixing, and therefore reaction, are to be accurately described.

Experimentalists have explored both the vorticity-potential flow interface[40] and the interface between regions where a scalar field such as concentration exists and does not exist.[41] For many shear flows the vorticity and concentration interfaces appear to coincide, as, for example, in heated jets, boundary layers, and wakes. It is also clear that entrainment in such flows takes place predominately on the edge of the traveling interface which is leeward with respect to a frame in which the interface is held immobile.[42] For a two-dimensional jet discharging into a slow moving airstream, the dominant entrainment rate occurs on the upstream edge, as can be seen by measuring the mean temperature at fixed positions on a cut through bulges of a certain size. The mean temperature was found to increase from the upstream edge to the downstream edge, which can be interpreted as enhanced entrainment of cool air at the upstream edge.

It has been found useful experimentally to inspect selected regions of a flow by the technique of conditioned sampling. For example, one may look exclusively at the temperature inside the boundary of a heated jet discharging into a constant temperature ambient stream, by fixing the probe position and discarding all temperature signals below a chosen small threshold above ambient. Much more sophisticated conditioning has also been achieved.[42]

A corresponding conditioning of the conservation equations[43] and/or the pdf equation[44] has been developed for modeling purposes following Libby's introduction of the idea.[45] The advantage is that modeling assumptions can then be applied to each region separately with more chance of success. As a simple example, consider again the case in which a hot air jet is discharging into a constant temperature ambient stream. Molecular diffusion, at a point, is always modeled in such a way as to preserve the mean property and to decrease its variance. However, the uniform temperature of the external fluid is not diffusing at all. Modeling using the unconditioned mean misrepresents the physics entirely as far as the external fluid is concerned, as it does, less dramatically, for the heated fluid in the jet.

The improved modeling obtained by conditioned averaging also raises a problem. When the flowfield is separated into disparate regions, it is necessary to model the entrainment of mass, momentum, and energy from one section of the flow into another. In principle, this requires the resolution of difficult problems in geometric statistics associated with a random surface[46] as well as an understanding of the entrainment process which we do not yet possess. Numerical modeling has never been daunted by such obstacles; it may now be time to incorporate aspects of entrainment into the modeling of reactive flows. The pdf formulation seems well suited to this task and preliminary models of conditioned nonreactive scalar transport with entrainment and mixing are beginning to surface.[47]

III. Numerical Modeling of Diffusion Flames

We conclude with a brief description of the strategies used to model diffusion flames numerically, this being the combustion configuration which has seen the most extensive application of pdf modeling. The technique makes use of second-order moment modeling for the compressible turbulent shear flow, combined with a modeling of the pdf of transported scalars (mass fractions and enthalpy). The details are too complicated to reproduce here but a full description can be found in recent papers[29,36] along with extensive lists of references[28,29] and numerical predictions[36] for mean scalar quantities in a H_2-air flame.

There are several distinct approaches to the problem of incorporating the scalar pdf into the model. The first, and simplest, is to postulate the shape of the pdf and to represent it by a small number of parameters, usually the means and correlations, whose values are determined by solving the first and second order moment equations. The assumed pdf is used to represent the species production rate term in the equations. Shape assumptions for a single species have included the Gaussian[48] (sometimes clipped) and the beta function,[49] and two delta functions.[50] Intermittency, an important feature of the statistical behavior of rapidly reacting species, can not be incorporated by such shape assumptions. It can be introduced explicitly through an intermittency factor,[43] or via delta functions.[50]

Closely related studies using multispecies models are those of Donaldson and Varma[18] employing delta functions and Pope's[29] suggestion of defining a most likely distribution by maximizing the entropy of the distribution, subject to the constraints imposed by normalization, the mean scalar values and all second order scalar correlations. The technique appears to be an extension of one developed for information theory.[51] For the reasons stated at the beginning of this article, it is not clear that the low order moments will adequately define the pdf in a reacting flow of high relative scalar intensity. Nevertheless, these techniques make available the advantages of the pdf formulation to much more complicated combustion systems than the use of the pdf evolution equation allows. The second-order moment closure technique for turbulence is itself only well established for shear flows such as jets, boundary layers, and wakes which are neither too complicated nor too rapidly evolving.

Bilger's conserved scalar approach[13,14,39] is another method for treating multispecies reactions in equilibrium. It provides approximate solutions to practically important situations and can be used as a starting point for the analysis of trace species which follow finite kinetics embedded in a background mixture in equilibrium.

An alternate approach is to solve the scalar pdf evolution equation instead of the moment equations and combine it with second order moment methods applied to the turbulence.[52] This has only been carried out for a single nonreacting scalar field (temperature) in a heated round jet.[36] The results can be converted into a simple two reactant turbulent diffusion flame by using the conserved scalar approach (Lewis number of unity) combined with a flame sheet approximation.

Modeling of a H_2-air jet by this method uses a kinetic energy dissipation function (k,ϵ) model for the turbulence, which is decoupled from the scalar pdf by Favre averaging. The advection term in the pdf equation is closed by a gradient transport model using the accepted diffusivity coefficients for turbulent, scalar transport. The integral closure, Eq. (8), represents molecular transport in composition space and the coefficients are defined by the turbulent field solutions for k and ϵ. Qualitative agreement with measured data is obtained. Mean temperature and products are somewhat more accurately predicted than are the reactant species. The general assumptions of the model are too simplified to allow one to expect much better. For example, relatively rapid diffusion of H_2 makes the unity Lewis number a doubtful assumption.

IV. Conclusions

The use of pdf's in the description of reactive turbulent flows has led to improved modeling and better, although still inadequate, understanding of the microscale processes in such flows. Because of the complexity of combustion systems, it is virtually certain that the marginal single point pdf of the mass fractions and enthalpy will be the only practical dependent variable; the random velocity will be included through turbulence moment models and closures of the advective term in the pdf equation. It also appears likely that the evolution equation for the marginal scalar pdf will have to be augmented by second- (or higher) order modeling of the scalar field moment equations when more than two or three species participate importantly in the reaction.

It is necessary for both theoreticians and modelers to confront the problem of entrainment across random interfaces and some tools for doing that seem now to be available. Such studies may also lead to the ability to include more complex conditioned information into the description of classical turbulent shear flows such as jets, mixing layers, boundary layers and wakes, when they transport reactive species. In this way a closer collaboration between experimentation, theory and modeling may develop to the benefit of all research in chemically reactive flows.

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References

[1] Murthy, S. N. B., Ed., "Turbulent Mixing in Nonreactive and Reactive Flows," Plenum Press, New York, 1975.

[2] Edelman, R. B., and Harsha, P. T., "Laminar and Turbulent Gas Dynamics in Combustors—Current Status," *Progress in Energy Combustion Science,* Vol. 4, 1978, pp. 1-62.

[3] Bracco, F. V., Ed., "Special Issue: Turbulent Reactive Flows," *Combustion Science and Technology,* Vol. 13, 1976, pp. 1-275.

[4] Hill, J. C., "Homogeneous Turbulent Mixing With Chemical Reaction," *Annual Reviews of Fluid Mechanics,* Vol. 8, 1976, pp. 135-161.

[5] Libby, P. A. and Williams, F. A., "Turbulent Flows Involving Chemical Reactions," *Annual Reviews of Fluid Mechanics,* Vol. 8, 1976, pp. 351-376.

[6] Bilger, R. W., "Turbulent Jet Diffusion Flames," *Progress in Energy Combustion Science,* Vol. 1, 1976, pp. 87-109.

[7] Brodkey, R. S., Ed., *Turbulence in Mixing Operations,* Academic Press, Inc., New York, 1975.

[8] Libby, P. A., and Williams, F. A., Eds., *Turbulent Reacting Flows,* Springer-Verlag, New York, 1980.

[9] Janicka, J. and Kollmann, W., "Prediction Model for the pdf of Turbulent Fluctuations in a Heated Round Jet," *2nd International Symposium on Turbulent Shear Flows,* London, 1979, pp. 1.7-1.12.

[10] Breidenthal, R. E., and A. Roshko, "Reacting Flow Visualization of the Three-Dimensional Vorticity in a Perturbed Wake," Abstract DC 1, *Bulletin of the American Physical Society,* Vol. 24, Oct. 1979, p. 1141.

[11] Lundgren, T. S., "Distribution Functions in the Statistical Theory of Turbulence," *Physics of Fluids,* Vol. 10, 1967, pp. 969-975.

[12] Srinivasan, R., Giddens, D. P., Bangert, L. H., and Wu, J. C., "Turbulent Plane Couette Flow Using Probability Distribution Functions," *Physics of Fluids,* Vol. 20, 1977, pp. 557-567.

[13] Bilger, R. W., "The Structure of Diffusion Flames," *Combustion Science and Technology,* Vol. 13, 1976, pp. 155-170.

[14] Bilger, R. W., "Effects of Kinetics and Mixing in Turbulent Combustion," *Combustion Science and Technology,* Vol. 19, 1979, pp. 89-94.

[15] Bilger, R. W., "Turbulent Flows with Non-Premixed Reactants," *Turbulent Reacting Flows,* edited by P. A. Libby and F. A. Williams, Springer-Verlag, New York, 1980, Chap. V.

[16] O'Brien, E. E., "Theoretical Aspects of Turbulent Mixing of Reactants," *Turbulence in Mixing Operations,* edited by R. S. Brodkey, Academic Press, Inc., New York, 1975, Chap. I.

[17] Williams, F. A., *Combustion Theory,* Addison-Wesley, Reading, Mass., 1965, Chap. 1.

[18] Donaldson, C. du P., and Varma, A. K., "Remarks on the Construction of a Second-Order Closure Description of Turbulent Reacting Flow," *Combustion Science and Technology,* Vol. 13, 1976, pp. 55-78.

[19] Toor, H. L., "Mass Transfer in Dilute Turbulent and Nonturbulent Systems with Rapid Irreversible Reactions and Equal Diffusivities," *AIChE Journal,* Vol. 8, 1962, pp. 70-78.

[20] Lin, C. H., and O'Brien, E. E., "Two Species Isothermal Reactions in Homogeneous Turbulence," *Astronautica Acta,* Vol. 17, 1972, pp. 771-778.

[21] O'Brien, E. E., "Closure for Stochastically Distributed Second Order Reactants," *Physics of Fluids,* Vol. 11, 1968, pp. 1883-1888.

[22] Bonniot, C., and R. Borghi, "Probability Density of Fluctuations of Reactive Species in Turbulent Combustion," *6th*

Colloquium International on Dynamics of Gases in Explosions and Reactive Systems, Stockholm, 1977.

[23] Monin, A. S., "Equations for Finite Dimensional Turbulence Field Probability Distribution," *Prikladnaya Matematika i Mekhanika,* Vol. 31, 1967, pp. 1057-1062.

[24] Novikov, E. E., "Kinetic Equations for a Vortex Field," *Soviet Physics—Doklady,* Vol. 12, 1968, pp. 1006-1010.

[25] Stratonovich, R. L., *Topics in the Theory of Random Noise,* Gordon and Breach, New York, Vol. 1, 1963, p. 83.

[26] Brissaud, A., and Frisch, U., "Solving Linear Stochastic Differential Equations," *Journal of Mathematics and Physics,* Vol. 15, 1974, pp. 524-534.

[27] Lighthill, M. J., *Fourier Analysis and Generalized Functions,* The University Press, Cambridge, England, 1968, p. 15.

[28] O'Brien, E. E., "The Probability Density Function (pdf) Approach to Reacting Turbulent Flows," *Turbulent Reacting Flows,* edited by P. A. Libby and F. A. Williams, Springer-Verlag, New York, Heidelberg, 1980, Chap. III.

[29] Pope, S. B., "The Statistical Theory of Turbulent Flames," *Philosophical Transactions of Royal Society of London, Series A.,* Vol. 291, 1979, pp. 529-568.

[30] Janicka, J., Kolbe, W., and Kollman, W., "Closure of the Transport Equation for the Probability Density Function of Turbulent Shear Flows," *Journal of Non-Equilibrium Thermodynamics,* Vol. 4, 1979, pp. 47-66.

[31] Dopazo, C., "Non-Isothermal Turbulent Reactive Flows: Stochastic Approaches," Ph.D. Thesis, State Univ. of New York, Stony Brook, N. Y., 1973.

[32] Kuznetsov, V. R., "Probability of the Concentration of a Passive Impurity in Turbulent Flows with Lateral Shear," *Izvestia Akademii Nauk, SSSR, Mekhanika Zhidkosti i Gaza,* Vol. 2, 1972, pp. 58-63.

[33] Dopazo, C., "Probability Density Function Approach for a Turbulent Axisymmetric Heated Jet: Centerline Evaluation," *Physics of Fluids,* Vol. 18, 1975, pp. 397-404.

[34] Fox, R. L., "Solution for Turbulent Correlations Using Multipoint Distribution Functions," *Physics of Fluids,* Vol. 14, 1971, pp. 1806-1808.

[35] Dopazo, C., "Relaxation of Initial Probability Density Functions in the Turbulent Convection of Scalar Fields," *Physics of Fluids,* Vol. 22, 1979, pp. 20-30.

[36] Janicka, J., Kolbe, W., and Kollmann, W., "The Solution of a pdf-transport Equation for Turbulent Diffusion Flames," *Proceedings of the 1978 Heat Transfer and Fluid Mechanics Institute,* edited by C. T. Crowe, W. L. Grosshandler, Stanford Univ. Press, Stanford, 1978, pp. 296-312.

[37] Vignon, J-M., "Turbulence et Reaction Chimique," Thesis, L'Universite Claude Bernard de Lyon, July 1979.

[38] Favre, A., "Equation Des Gaz Turbulents Compressibles," *Journal de Mecanique,* Vol. 4, No. 3-4, 1965.

[39] Bilger, R. W., "Perturbation Analysis of Turbulent Nonpremixed Combustion," Charles Kolling Res. Lab., Tech. Note F-88, Aug. 1979.

[40] Corrsin, S., and Kistler, A. L., "Free Stream Boundaries of Turbulent Flows," NACA Rept. 1244, 1955.

[41] Tutu, N. K., "An Experimental Investigation of a Heated Turbulent Round Jet," Ph.D. Thesis, State Univ. of New York, Stony Brook, N. Y., 1973.

[42] Anderson, P., La Rue, J. C., and Libby, P. A., "Preferential Entrainment in a Two Dimensional Turbulent Jet in a Moving Stream," *Physics of Fluids,* Vol. 22, 1979, pp. 1857-1861.

[43] Dopazo, C., and O'Brien, E., "Intermittency in Free Turbulent Shear Flows," *Turbulent Shear Flows 1,* edited by Durst, Launder, Schmidt and Whitelaw, Springer-Verlag, New York, 1979, pp. 6-18.

[44] O'Brien, E. E., and Dopazo, C., "Behavior of Conditioned Variables in Free Turbulent Shear Flows," *Structure and Mechanisms of Turbulence,* Vol. 2, Springer-Verlag, New York, 1978, pp. 124-133.

[45] Libby, P. A., "On the Prediction of Intermittent Turbulent Flows," *Journal of Fluid Mechanics,* Vol. 68, 1975, pp. 273-295.

[46] Swerling, P., "Statistical Properties of the Contours of Random Surfaces," *IRE Transactions on Information Theory,* Vol. IT-S, 1962, pp. 315-321.

[47] Dopazo, C., O'Brien, E., and Wu, M-Z., "Modeling of Advection, Entrainment and Microscale Mixing in Free Turbulent Shear Flows," Paper EA 4, *Bulletin of the American Physical Society,* Vol. 24, Oct. 1979, p. 1146.

[48] Hawthorne, W. R., Weddell, D. S., and Hottel, H. C., "Mixing and Combustion in Turbulent Gas Jets," *3rd Symposium International on Combustion, Flame and Explosive Phenomena,* Williams and Wilkens, 1949, pp. 266-288.

[49] Rhodes, R. P., "A Probability Distribution Function for Turbulent Flows," *Turbulent Mixing in Non-Reactive and Reactive Flows,* Plenum Press, New York, 1975, pp. 235-241.

[50] Bush, W. B., and Fendell, F. E., "On Diffusion Flames in Turbulent Shear Flows," *Acta Astronautica,* Vol. 1, 1974, pp. 645-666.

[51] Lumley, J. L., *Stochastic Tools in Turbulence,* Academic Press, N. Y., 1970, pp. 33-37.

[52] Launder, B. E., "Heat and Mass Transport," *Turbulence,* edited by P. Bradshaw, Springer-Verlag, Berlin, 1976, Chap. 6.