

# Combustion Modeling Using Probability Density Function Methods

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## I. Introduction

**P**ROBABILITY density function (pdf) methods have been applied to a variety of turbulent flows both with and without combustion. In general, single-phase, low-Mach-number flows have been considered, in which radiation is not a major factor. For such flows, the fundamental dependent variables are the velocities  $U(x,t)$  and the compositions  $\Phi(x,t)$  (e.g., the species mass fractions and enthalpy). Different probabilistic approaches to modeling turbulent flows can be categorized according to the statistics of  $U(x,t)$  and  $\Phi(x,t)$  that are considered. For example, in a mean-flow closure,  $\langle U(x,t) \rangle$  and  $\langle \Phi(x,t) \rangle$  are the primary dependent variables; in second-order closures, the variances  $\langle u_i u_i \rangle$ ,  $\langle \phi'_\alpha \phi'_\beta \rangle$  and covariances  $\langle u_i \phi'_\alpha \rangle$  are also included. Angled brackets denote means (i.e., mathematical expectations), and  $u_i(x,t) = U_i(x,t) - \langle U_i(x,t) \rangle$  and  $\phi'_\alpha(x,t) = \phi_\alpha(x,t) - \langle \phi_\alpha(x,t) \rangle$  are the fluctuating components of  $U_i$  and  $\phi_\alpha$ , respectively.

In pdf methods, the dependent variable is a pdf, or joint pdf of  $U(x,t)$  and  $\Phi(x,t)$ . The pdf contains information equivalent to all the moments; hence, in this sense, pdf methods are more comprehensive than moment closures (e.g., second-order closures). The methods that have proved most successful are based on one-point, one-time pdf's, which contain information at each point in the flow separately, but no joint information at two or more distinct points.

In the last 15 years, pdf methods have advanced from being only of theoretical interest to a small group of specialists, to being a practical approach for calculating the properties of turbulent reactive flows. In addition to having been applied to idealized flames and simple laboratory flames, as the subsequent review indicates, the methods have been applied to flames requiring multistep chemical kinetics (e.g., Refs. 1 and 2) and to computationally difficult flows (e.g., that in the cylinder of a spark-ignition engine<sup>3,4</sup>).

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This chapter reviews the work on pdf methods with some emphasis on the numerical issues and on the applications to turbulent combustion. In the next section, the different pdf methods are described, along with the modeling they entail. Monte Carlo methods have proved to be the most successful means of solving pdf transport equations. The essence of these solution techniques is described in Sec. III. Sections II and III briefly describe the principal features with no attempt at rigor. The theoretical foundations of pdf methods (including the modeling and Monte Carlo solution algorithms) are described comprehensively by Pope.<sup>5</sup> Section IV reviews the applications of pdf methods to turbulent diffusion flames and premixed flames. Recent applications to constant-density inert flows have been reviewed in Ref. 6. In Sec. V, some of the outstanding problems and future directions are assessed.

## II. PDF Methods

### A. Definitions and Properties

Let  $\phi$  denote the value of a composition variable (the mass fraction of oxygen, for example) at a particular location  $\mathbf{x}_0$  and time  $t_0$  in a turbulent reactive flow. For ease of exposition, we suppose that the flow can be realized any number of times, and the time  $t$  is measured from the initiation of the flow. Thus, from each realization we obtain a value of  $\phi$ ; given the nature of turbulence, these values are, in all probability, different. In other words,  $\phi$  is a random variable. It is not possible to predetermine the value of  $\phi$  that will be obtained in a given realization. However, it is possible to ascribe probabilities to its value being in a given interval: this can be done through the pdf.

For every random variable, we introduce an independent (sample-space) variable: in particular,  $\psi$  is the sample-space variable corresponding to  $\phi$ . The cumulative distribution function (cdf),  $F_\phi(\psi)$ , is then defined as the probability that  $\phi$  is less than  $\psi$ :

$$F_\phi(\psi) \equiv \text{Prob}\{\phi < \psi\} \quad (1)$$

And the pdf of  $\phi$ ,  $f_\phi(\psi)$ , is defined to be

$$f_\phi(\psi) \equiv \frac{d}{d\psi} F_\phi(\psi) \quad (2)$$

Whereas  $F_\phi(\psi)$  is a probability function,  $f_\phi(\psi)$  is a probability density function. That is,  $f_\phi(\psi)$  is the probability *per unit*  $\psi$  of the event  $\phi = \psi$ . Equivalently,  $f_\phi(\psi) d\psi$  is the probability of the  $\phi$  being in the range  $\psi < \phi \leq \psi + d\psi$ .

The three fundamental properties of the pdf [in addition to Eq. (2)] follow:

$$f_\phi(\psi) \geq 0 \quad (3)$$

since probabilities are nonnegative;

$$\int_{-\infty}^{\infty} f_{\phi}(\Psi) d\Psi = 1 \quad (4)$$

since  $\text{Prob}\{\phi < \infty\} = 1$  and  $\text{Prob}\{\phi < -\infty\} = 0$ ; and, for any (nonpathological) function  $Q(\phi)$ ,

$$\langle Q \rangle = \int_{-\infty}^{\infty} f_{\phi}(\Psi) Q(\Psi) d\Psi \quad (5)$$

Equation (5) shows that, if the pdf is known, the mean (or mathematical expectation) of any function of the random variable can be calculated. In particular, the mean  $\langle \phi \rangle$  and the  $m$ th central moment  $\langle \phi'^m \rangle$  ( $m > 1$ ) can be determined (if it exists).

For a general turbulent reactive flow, we need to consider a set of  $\sigma \geq 1$  composition variables  $\Phi \equiv \{\phi_1, \phi_2, \dots, \phi_{\sigma}\}$ . Accordingly, the  $\sigma$  sample-space variables  $\Psi \equiv \{\psi_1, \psi_2, \dots, \psi_{\sigma}\}$  are introduced, and the joint pdf of  $\Phi$ ,  $f_{\Phi}(\Psi)$  is defined to be the probability density of the compound event  $\Phi = \Psi$  (i.e.,  $\phi_1 = \psi_1, \phi_2 = \psi_2, \dots, \phi_{\sigma} = \psi_{\sigma}$ ).

Clearly, the joint pdf defined at the particular location  $x_0$  and time  $t_0$  can be defined at any  $(x, t)$ . We denote by  $f_{\Phi}(\Psi; x, t)$  the joint pdf of  $\Phi(x, t)$ . It is important to realize that this is a one-point, one-time joint pdf: it contains no joint information between  $\Phi$  at two or more positions or times. The pdf method described in the next subsection is based on  $f_{\Phi}(\Psi; x, t)$ , which is called the *composition joint pdf*.

Another pdf method described (in Sec. II.C) is based on the *velocity-composition joint pdf*,  $f(\mathbf{V}, \Psi; x, t)$ . Here  $\mathbf{V} = \{V_1, V_2, V_3\}$  are the three independent velocity variables, and  $f$  is the probability density of the compound event  $\{U(x, t) = \mathbf{V}, \Phi(x, t) = \Psi\}$ .

In the treatment of variable-density flows, two other probability functions prove useful, and are now defined. By assumption (see Ref. 5), the set of composition variables is sufficient to determine the fluid density. Thus, if the composition is  $\Phi$ , the density is given by the function  $\rho_{\sigma}(\Phi)$ , which can be determined from a thermodynamic calculation. Consequently, at  $(x, t)$  the fluid density is

$$\rho(x, t) = \rho_{\sigma}(\Phi[x, t]) \quad (6)$$

and the mean density [evaluated using Eq. (5)] is

$$\langle \rho(x, t) \rangle = \int f_{\Phi}(\Psi; x, t) \rho_{\sigma}(\Psi) d\Psi$$

where integration is over the whole of the composition space. Having made the distinction between the different functions  $\rho(x, t)$  and  $\rho_{\sigma}(\Phi)$ , we now follow conventional (if imprecise) notation and denote both by  $\rho$ .

Favre, or density-weighted, pdf's are defined by, for example,

$$\tilde{f}_{\Phi}(\Psi) \equiv \rho(\Psi) f_{\Phi}(\Psi) / \langle \rho \rangle \quad (7)$$

It then follows that density-weighted means are given by

$$\bar{Q} \equiv \frac{\langle \rho Q \rangle}{\langle \rho \rangle} = \int_{-\infty}^{\infty} Q(\boldsymbol{\psi}) \tilde{f}(\boldsymbol{\psi}) \, d\boldsymbol{\psi} \quad (8)$$

[cf. Eq. (5)]. The *mass density function*  $\mathcal{F}$  is defined by

$$\mathcal{F}(\mathbf{V}, \boldsymbol{\psi}, \mathbf{x}; t) = \rho(\boldsymbol{\psi}) f(\mathbf{V}, \boldsymbol{\psi}; \mathbf{x}, t) \quad (9)$$

The use of these functions is made apparent in the next two subsections.

### B. Composition Joint PDF Equation

Dopazo and O'Brien<sup>7</sup> were the first to consider the transport equation for  $f_{\Phi}(\boldsymbol{\psi}; \mathbf{x}, t)$ . Since then, a number of derivations have been given.<sup>5,8-11</sup> Here we state the result, and refer the reader to Ref. 5 for a detailed derivation.

The composition  $\phi_{\alpha}(\mathbf{x}, t)$  evolves according to the conservation equation

$$\frac{D\phi_{\alpha}}{Dt} = - \frac{1}{\rho} \frac{\partial J_i^{\alpha}}{\partial x_i} + S_{\alpha} \quad (10)$$

where  $J^{\alpha}$  is the (molecular) diffusive flux of  $\phi_{\alpha}$ , and  $S_{\alpha}$ —a known function of  $\boldsymbol{\phi}$ —is the rate of creation of  $\phi_{\alpha}$  due to chemical reaction. The pdf transport equation corresponding to Eq. (10) is

$$\begin{aligned} & \frac{\partial \tilde{f}_{\Phi}}{\partial t} + \bar{U}_i \frac{\partial \tilde{f}_{\Phi}}{\partial x_i} + \frac{\partial}{\partial \psi_{\alpha}} [S_{\alpha}(\boldsymbol{\psi}) \tilde{f}_{\Phi}] \\ &= - \frac{1}{\langle \rho \rangle} \frac{\partial}{\partial x_i} [\langle \rho \rangle \langle u_i'' | \boldsymbol{\psi} \rangle \tilde{f}_{\Phi}] + \frac{\partial}{\partial \psi_{\alpha}} \left[ \left\langle \frac{1}{\rho} \frac{\partial J_i^{\alpha}}{\partial x_i} \middle| \boldsymbol{\psi} \right\rangle \tilde{f}_{\Phi} \right] \end{aligned} \quad (11)$$

On the left-hand side, the first two terms represent the rate of change following the Favre-averaged mean flow. The third term is—in composition space—the divergence of the flux of probability due to reaction. The form of this term gives this pdf method its advantage over other statistical approaches. Since  $S(\boldsymbol{\psi})$  is known,  $\tilde{f}_{\Phi}$  is the subject of the equation, and  $\psi_{\alpha}$  is an independent variable, the term contains no unknowns. Thus, however complicated and nonlinear the reaction scheme, in the composition joint pdf equation the effect of chemical reaction is in closed form, requiring no modeling.

In contrast, the terms on the right-hand side require modeling. The quantity  $\langle u_i'' | \boldsymbol{\psi} \rangle$  is the conditional mean of the Favre velocity fluctuation ( $\mathbf{u}' \equiv \mathbf{U} - \bar{\mathbf{U}}$ )—conditional, that is, upon the event  $\boldsymbol{\phi} = \boldsymbol{\psi}$ . The term in  $\langle u_i'' | \boldsymbol{\psi} \rangle$  represents the transport of  $\tilde{f}_{\Phi}$  in physical space by the fluctuating velocity. Although there have been other suggestions, this term generally is modeled by gradient diffusion:

$$\langle \rho \rangle \langle u_i'' | \boldsymbol{\psi} \rangle \tilde{f}_{\Phi} = - \Gamma_T \frac{\partial}{\partial x_i} \tilde{f}_{\Phi} \quad (12)$$

where  $\Gamma_T$  is a turbulent diffusivity. Such gradient transport models are, of course, subject to many objections, especially when applied to variable-density reactive flows. The final term in Eq. (11) represents the effect of molecular mixing. It is generally treated by a stochastic mixing model (see, for example, Refs. 5, 12, and 13). Although some aspects of this modeling are discussed later, the cited references should be consulted for a full account.

The composition joint pdf equation [Eq. (11)] is not a self-contained model. Mean momentum equations must be solved for  $\bar{U}$ , and a turbulence model ( $k$ - $\epsilon$ , say) is needed to determine both  $\Gamma_T$  ( $\sim k^2/\epsilon$ ) and the mixing rate ( $\sim \epsilon/k$ ) used in the stochastic mixing model.

### C. Velocity-Composition Joint PDF Equation

Two shortcomings of the composition pdf approach are that turbulent transport ( $\langle u_i'' | \Psi \rangle$ ) has to be modeled, and that the velocity and turbulence fields have to be treated separately. Both of these shortcomings are overcome in the velocity-composition joint pdf approach.

The instantaneous momentum equation is

$$\rho \frac{DU_j}{Dt} = \frac{\partial \tau_{ij}}{\partial x_i} - \frac{\partial p}{\partial x_j} + \rho g_j \quad (13)$$

where  $\tau_{ij}$  is the stress tensor,  $p$  the pressure, and  $g$  the gravitational acceleration.

From this equation [and that for  $\Phi$ , Eq. (10)], the following equation can be derived<sup>5</sup> for the mass density function  $\mathcal{F}(V, \Psi, x; t)$  [Eq. (9)]:

$$\begin{aligned} \frac{\partial \mathcal{F}}{\partial t} + V_j \frac{\partial \mathcal{F}}{\partial x_j} + \left[ g_j - \frac{1}{\rho(\Psi)} \frac{\partial \langle p \rangle}{\partial x_j} \right] \frac{\partial \mathcal{F}}{\partial V_j} + \frac{\partial}{\partial \Psi_\alpha} [\mathcal{F} S_\alpha(\Psi)] \\ = \frac{\partial}{\partial V_j} \left[ \left\langle -\frac{\partial \tau_{ij}}{\partial x_i} + \frac{\partial p'}{\partial x_j} \middle| V, \Psi \right\rangle \mathcal{F} / \rho(\Psi) \right] \\ + \frac{\partial}{\partial \Psi_\alpha} \left[ \left\langle \frac{\partial J_j^\alpha}{\partial x_j} \middle| V, \Psi \right\rangle \mathcal{F} / \rho(\Psi) \right] \end{aligned} \quad (14)$$

None of the terms on the left-hand side requires modeling. In order, the terms represent the following: rate of change with time; transport in position space (by both mean and fluctuating components of velocity); transport in velocity space (by gravity and the mean pressure gradient); and, as before, transport in composition space due to reaction.

The terms requiring modeling [on the right-hand side of Eq. (14)] are means conditional on the compound event  $\{U(x, t) = V, \Phi(x, t) = \Psi\}$ . The term involving  $J^\alpha$ —as in the composition pdf equation—represents molecular mixing. The remaining term involving  $\tau_{ij}$  and  $p'$  represents transport in velocity space due to molecular stresses and the fluctuating pressure gradient. A discussion of how the term can be modeled is deferred to the next subsection.

It may be seen, then, that the velocity-composition joint pdf method retains the advantage of treating reaction without approximation, and, in addition, treats transport in physical space (turbulent convection) exactly, thus avoiding gradient-diffusion assumptions. It also provides a more complete closure: the mean velocity  $\bar{U}(\mathbf{x}, t)$ , the Reynolds stresses, and indeed all one-point velocity-composition statistics can be calculated from  $\mathcal{F}$ . The model equation for  $\mathcal{F}$  is not quite self-contained because the modeled terms require a knowledge of the turbulent time scale ( $k/\epsilon$ ) that cannot be deduced from  $\mathcal{F}$ .

#### D. Lagrangian Viewpoint

Thus far, the Eulerian view has been adopted: we have considered functions [e.g.,  $\mathcal{F}(\mathbf{V}, \boldsymbol{\psi}, \mathbf{x}; t)$ ] at a fixed position  $\mathbf{x}$ . It proves extremely helpful, both to the modeling and to the numerical solution technique, to take the alternative Lagrangian viewpoint also.

Let  $\mathbf{x}^+(t)$ ,  $\mathbf{U}^+(t)$ , and  $\boldsymbol{\Phi}^+(t)$  denote the position, velocity, and composition of the fluid particle that was at a reference point  $\mathbf{x}_0$  at a reference time  $t_0$ . These particle properties evolve according to

$$\frac{d\mathbf{x}^+(t)}{dt} = \mathbf{U}^+(t) = \mathbf{U}(\mathbf{x}^+[t], t) \quad (15)$$

since, by definition, a fluid particle moves with the local fluid velocity;

$$\frac{dU_j^+}{dt} = A_j \equiv g_j - \frac{1}{\rho(\boldsymbol{\Phi}^+)} \left[ \frac{\partial \langle p \rangle}{\partial x_j} \right]_{\mathbf{x}^+} + \left\{ \frac{\partial \tau_{ij}}{\partial x_i} - \frac{1}{\rho(\boldsymbol{\Phi}^+)} \frac{\partial p'}{\partial x_j} \right\}_{\mathbf{x}^+} \quad (16)$$

[from Eq. (13)]; and

$$\frac{d\phi_\alpha^+}{dt} = \Theta_\alpha \equiv S_\alpha(\boldsymbol{\Phi}^+) - \left\{ \frac{\partial J_i^\alpha}{\partial x_i} \right\}_{\mathbf{x}^+} \quad (17)$$

[from Eq. (10)].

The connection between these equations for the properties of a fluid particle and the equation for the mass density function  $\mathcal{F}$  [Eq. (14)] is immediately apparent. Equation (14) can be written

$$\frac{\partial \mathcal{F}}{\partial t} + \frac{\partial}{\partial x_j} [\mathcal{F} \langle U_j^+ | \rangle] + \frac{\partial}{\partial x_j} [\mathcal{F} \langle A_j | \rangle] + \frac{\partial}{\partial \psi_\alpha} [\mathcal{F} \langle \Theta_\alpha | \rangle] = 0 \quad (18)$$

where the expectations are conditional on the compound event  $\{\mathbf{x}^+(t) = \mathbf{x}, \mathbf{U}^+(t) = \mathbf{V}, \boldsymbol{\Phi}^+(t) = \boldsymbol{\psi}\}$ . Furthermore, it may be noticed that the terms in braces in Eqs. (16) and (17) appear on the right-hand side of Eq. (14)—that is, they need to be modeled—whereas all other terms appear on the left-hand side and are treated exactly.

#### E. Stochastic Models

The standard approach to turbulence modeling is to construct constitutive relations for the unknown correlations (see, for example, Ref. 14).

In the context of the mass density function, this approach is to model the unknown conditional expectations on the right-hand side of Eq. (14) in terms of known quantities, i.e., functions or functionals of  $\mathcal{F}(\mathbf{V}, \boldsymbol{\psi}, \mathbf{x}; t)$ . However, the Lagrangian viewpoint offers a different approach to modeling, namely, to use stochastic processes to simulate unknown contributions to  $U^+(t)$  and  $\Phi^+(t)$  [i.e., the terms in braces in Eqs. (16) and (17)].

To illustrate this approach, we consider  $U^*(t)$ —a stochastic model for  $U^+(t)$ . If the model is accurate, then  $U^*(t)$  is (statistically) an accurate approximation to  $U^+(t)$ . In general, the time series  $U^*$  is not differentiable. Consequently, we express the models in terms of the infinitesimal increment

$$dU^*(t) \equiv U^*(t + dt) - U^*(t) \quad (19)$$

rather than in terms of the derivative  $dU^*/dt$ . Note that for a deterministic, differentiable process [e.g.,  $U^+(t)$ ], the infinitesimal increment is nonrandom (i.e., zero variance) and is of order  $dt$ .

In view of the equation for  $U^+(t)$  [Eq. (16)], the increment  $dU^*$  can be written

$$dU_j^* = \left( g_j - \frac{1}{\rho(\Phi^*)} \left[ \frac{\partial \langle p \rangle}{\partial x_j} \right]_{\mathbf{x}^*} \right) dt + dU_j^S \quad (20)$$

where (similar to  $U^*$ )  $\mathbf{x}^*$  and  $\Phi^*$  are models of  $\mathbf{x}^+$  and  $\Phi^+$ . The stochastic increment  $dU^S$  models the effects of the fluctuating pressure gradient and viscous stresses, whereas the term in  $dt$  is an exact expression for the effect of gravity and the mean pressure gradient.

Two types of models for stochastic increments such as  $dU^S$  have been used. The first type—of which the stochastic mixing model is an example—processes, these are *point processes*. According to these models, the infinitesimal increment  $dU^S$  is nearly always zero. But with probability of order  $dt$ , the increment is of order unity. Thus, the time series is a piecewise constant, with a finite number of jumps per unit time.

The second type of model uses *diffusion processes* in which  $dU^S$  is a random variable with (conditional) mean and variance both of order  $dt$ . Note that this implies that the rms is of order  $dt^{1/2}$ , and hence the process—though continuous—is not differentiable. The different variants of the Langevin model are diffusion processes (see, for example, Refs. 5, 15–17).

For more information on this general modeling approach, the reader is referred to Ref. 5, whereas the current status of the Langevin model is described in Ref. 17.

### III. Numerical Solution Algorithms

The velocity-composition joint pdf  $f(\mathbf{V}, \boldsymbol{\psi}; \mathbf{x}, t)$  is a single function defined in a multidimensional space. In general,  $f$  depends on the three velocity variables,  $\sigma$  composition variables, three spatial variables, and time—(7 +  $\sigma$ ) independent variables in all. In many cases, the dimensionality may

be less, but still large. For example, in a statistically stationary and two-dimensional flow with a single composition variable,  $f(\mathbf{V}, \psi_1; x_1, x_2)$  depends on six independent variables. The composition joint pdf  $f_\phi(\boldsymbol{\psi}; \mathbf{x}, t)$  in general depends on  $(4 + \sigma)$  variables; however, for the simpler flow cited earlier,  $f_\phi(\psi_1; x_1, x_2)$  is a function of just three variables.

Given the large dimensionality of joint pdf's, it is clear that conventional grid-based numerical methods (e.g., finite differences) are impractical for all but the simplest cases. Just to provide an accurate representation of a function of six independent variables is a major task. Consequently, although one or two finite-difference solutions have been obtained for  $f_\phi(\psi_1; x_1, x_2)$  (e.g., Refs. 9 and 10), all investigators currently use Monte Carlo methods instead.

In the next subsection, the general Monte Carlo method devised by Pope<sup>5</sup> to solve for the velocity-composition joint pdf is outlined. Then, in Sec. III.B, Monte Carlo solution algorithms for the composition joint pdf are reviewed.

#### A. Monte Carlo Method for the Velocity-Composition Joint PDF

The Monte Carlo method to solve the modeled equation for the velocity-composition joint pdf is conceptually simple and natural. Rather than discretizing the space, we discretize the mass of fluid into a large number  $N$  of representative or *stochastic particles*. At a given time  $t$ , let  $M$  be the total mass of fluid within the solution domain. Then each stochastic particle represents a mass  $\Delta m \equiv M/N$  of fluid. The  $n$ th particle has position  $\mathbf{x}^{(n)}(t)$ , velocity  $\mathbf{U}^{(n)}(t)$ , and composition  $\boldsymbol{\phi}^{(n)}(t)$ .

Starting from appropriate initial conditions, the particle properties are advanced in time by the increments

$$d\mathbf{x}^{(n)}(t) = \mathbf{U}^{(n)}(t) dt \quad (21)$$

$$d\mathbf{U}^{(n)}(t) = [\mathbf{g} - \rho(\boldsymbol{\phi}^{(n)})^{-1} \nabla \langle p \rangle] dt + d\mathbf{U}^S \quad (22)$$

$$d\boldsymbol{\phi}^{(n)}(t) = \mathbf{S}(\boldsymbol{\phi}^{(n)}) dt + d\boldsymbol{\phi}^S \quad (23)$$

where  $d\mathbf{U}^S$  and  $d\boldsymbol{\phi}^S$  are the stochastic increments that simulate molecular processes and the fluctuating pressure gradient. At symmetry boundaries, particles are reflected; at inflow boundaries, particles are added with appropriate properties; and, at outflow boundaries, particles are discarded. Although wall boundaries have been treated,<sup>18</sup> a comprehensive account of this treatment is not available in the literature.

The correspondence between the ensemble of stochastic particles and the joint pdf has been established by Pope.<sup>5</sup> The main results follow:

- 1) The expected density of the stochastic particles in physical space

$$\left[ \Delta m \sum_{n=1}^N \langle \delta[\mathbf{x} - \mathbf{x}^{(n)}(t)] \rangle \right]$$

is equal to the fluid density  $\langle \rho(\mathbf{x}, t) \rangle$ .

- 2) The joint pdf of the stochastic particle properties  $\mathbf{U}^{(n)}(t)$ ,  $\boldsymbol{\phi}^{(n)}(t)$  is the density-weighted joint pdf  $\tilde{f}(\mathbf{V}, \boldsymbol{\psi}; \mathbf{x}^{(n)}[t], t)$ .



3) From particle properties, expectations [e.g.,  $\bar{U}(\mathbf{x}, t)$ ] can be approximated as ensemble averages, with a statistical error of order  $N^{-1/2}$ .

The accurate numerical determination of means [such as  $\bar{U}(\mathbf{x}, t)$ ] from the particle properties [ $\mathbf{x}^{(n)}(t)$ ,  $\mathbf{U}^{(n)}(t)$ ] is far from trivial. The straightforward method is to bin the particles in physical space, and then to approximate  $\bar{U}$  (at bin centers) as the ensemble average formed from the particles within the bin. Although correct in principle, this method produces large statistical errors. An accurate alternative, based on statistical techniques, is to estimate means in terms of cubic splines.<sup>5,19</sup>

Several implementations of the algorithm based on Eqs. (21–23), and variants of it, have been reported. For example, the turbulent jet diffusion flame calculations reported in Sec. IV are performed using a “boundary-layer” variant.<sup>5</sup> Haworth and Pope<sup>20</sup> report a variant of the algorithm designed specifically for self-similar shear flows. From a numerical standpoint, this work is of particular interest, because the convergence of the method (as  $N^{-1/2}$ ) is demonstrated. The basic algorithm has been implemented and demonstrated for statistically two-dimensional recirculating flows by Anand et al.<sup>18</sup> Haworth and El Tahry<sup>3,4</sup> report calculations of the three-dimensional time-dependent flow in the cylinder of a spark-ignition engine. In these calculations, the pdf algorithm is coupled to a conventional finite-volume algorithm that is used to calculate the mean pressure field and the turbulent time-scale field.

## B. Monte Carlo Algorithms for the Composition Joint PDF

Two different algorithms have been proposed to solve the modeled transport equation for the composition joint pdf.

The algorithm proposed by Pope<sup>5</sup> is similar to that described earlier for the velocity-composition joint pdf. Again, it is a grid-free algorithm in which the mass of fluid is discretized into  $N$  stochastic particles, the  $n$ th of these having position  $\mathbf{x}^{(n)}(t)$  and composition  $\Phi^{(n)}(t)$ . In each time step, the composition is incremented according to Eq. (23), while the position is incremented by

$$d\mathbf{x}^{(n)}(t) = \bar{U}(\mathbf{x}^{(n)}[t], t) dt + d\mathbf{x}^S \quad (24)$$

where the stochastic component  $d\mathbf{x}^S$  causes a random walk to simulate gradient diffusion. No implementations of this algorithm have been reported in the literature.

A different Monte Carlo algorithm for the composition joint pdf that has been used extensively was devised by Pope.<sup>21</sup> In this method, there is a finite-difference grid in physical space. At each grid node, the composition joint pdf is represented by  $N$  particles, the  $n$ th having composition  $\Phi^{(n)}(t)$ . Reaction and mixing are performed according to Eq. (23), while particles are moved from node to node to simulate convection and turbulent diffusion. This algorithm is used in the premixed flame calculation of Refs. 21 and 22, and in the diffusion flame calculations of Refs. 2, 23, and 24.

#### IV. Turbulent Flame Calculations

##### A. Turbulent Diffusion Flames

Some of the first pdf calculations are of turbulent diffusion flames.<sup>9,10,23,25,26</sup> The calculations reported by Nguyen and Pope<sup>23</sup> are the first use of the Monte Carlo method for jet flames. The results include demonstrations of convergence of the solutions as  $N^{-1/2}$  tends to zero.

In the calculations cited earlier, the thermochemistry is handled in a simple manner—by assuming chemical equilibrium, for example. This reduces the number of composition variables to one, namely, the mixture fraction  $\xi$ . Finite-rate, multistep kinetics have been used by Pope and Correa<sup>27</sup> (see also Ref. 28), Jones and Kollmann,<sup>24</sup> and Chen and Kollmann.<sup>2,29</sup> A computational challenge is to implement the integration of the rate equation, i.e.,

$$\frac{d\Phi}{dt} = S(\Phi) \quad (25)$$

in an efficient manner. This equation has to be integrated for every particle on every time step to determine the change in composition due to chemical reaction. All investigators have used table look-up algorithms. In the conceptually simplest algorithm, the change in composition  $\Delta\Phi$  over the time step  $\Delta t$  is tabulated as a function of the composition  $\Phi$  at the start of the step.

Considerable attention has been paid to the CO/H<sub>2</sub>-air turbulent diffusion flame studied experimentally by Drake et al.<sup>30</sup> Using the velocity-composition joint pdf approach, Pope and Correa<sup>27</sup> and Correa et al.<sup>28</sup> report calculations based on a partial equilibrium assumption. This reduces the number of composition variables to two: the mixture fraction  $\phi_1 = \xi$  and a reaction progress variable  $\phi_2 = \eta$  (for the radical recombination reactions). Consequently, the general set of coupled ordinary differential equations [Eq. (25)] reduces to the single equation

$$\frac{d\phi_2}{dt} = S_2(\phi_1, \phi_2) \quad (26)$$

since the mixture fraction is conserved (i.e.,  $S_1 = 0$ ). The numerical integration of Eq. (26) is a simple task. As an example of the joint pdf calculations made for this CO/H<sub>2</sub>-air turbulent diffusion flame, Fig. 1 shows the profiles of the mean mole fractions of the major species at an axial location of 25 nozzle diameters. It may be seen that there is good agreement between the measurements and the calculations. Further comparisons can be found in the original works.<sup>27,28</sup>

Again using the velocity-composition joint pdf approach, Haworth et al.<sup>31,32</sup> have made calculations of the CO/H<sub>2</sub>-air flame using a flamelet model. In this approach, the instantaneous composition of the fluid is assumed to be a unique function of the mixture fraction  $\xi$  and of its dissipation rate  $\chi$ . Consequently, the determination of statistics of composition—the mean density, for example—requires a knowledge of the joint pdf of  $\xi$  and  $\chi$ . The pdf of  $\xi$  is determined from the velocity-mixture fraction

pdf equation, whereas different assumptions are made about the distribution of  $\chi$ .

### B. Turbulent Premixed Flames

The composition joint pdf approach using the Monte Carlo method has been applied to premixed flames by Pope<sup>1</sup> and McNutt.<sup>22</sup> The former calculation demonstrated the ability of the pdf method to handle nonlinear reaction kinetics. A three-variable kinetics scheme was used to calculate the oxidation of CO and the formation of NO in a propane-air flame stabilized behind a perforated plate.

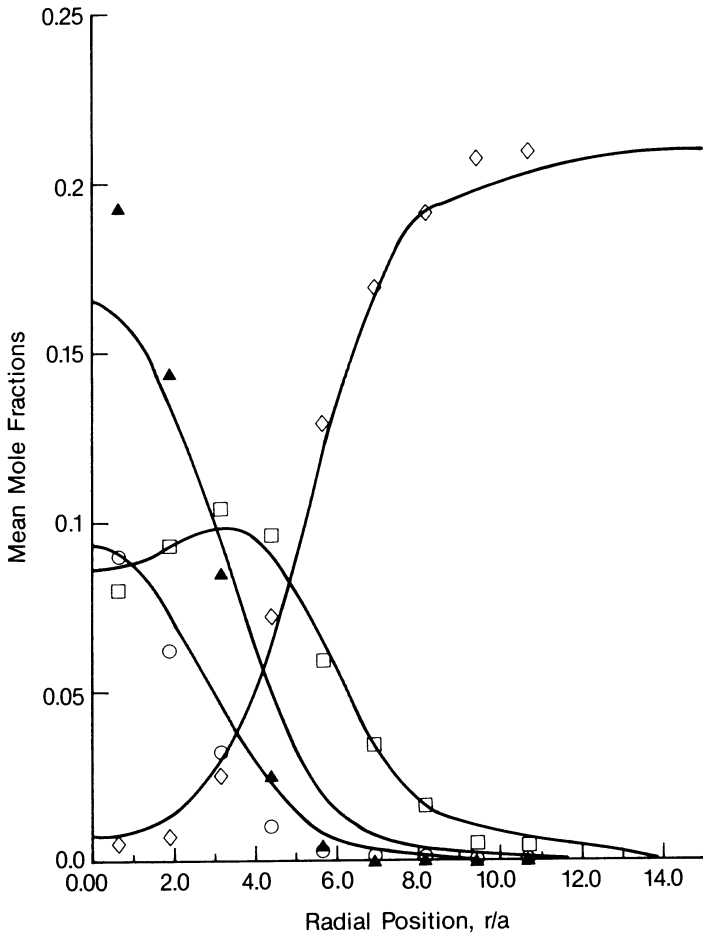


Fig. 1 Comparison of velocity-composition joint pdf calculations (lines) with experimental data (symbols) for a turbulent syngas diffusion flame.<sup>27</sup> Mean mole fractions against radial distance (normalized by the nozzle radius  $a$ ) at an axial distance of 25 nozzle diameters. Symbols:  $\circ$ ,  $H_2$ ;  $\blacktriangle$ ,  $CO$ ;  $\square$ ,  $CO_2$ ;  $\diamond$ ,  $O_2$ .

The works of McNutt,<sup>22</sup> Pope and Anand,<sup>33</sup> and Anand and Pope<sup>34</sup> are concerned with the idealized case of a statistically steady, one-dimensional turbulent premixed flame. In Ref. 34, the velocity-composition joint pdf method is used, and the effects of variable density are studied. It is shown that, similar to the Bray–Moss–Libby model,<sup>35</sup> the pdf method is capable of accounting for countergradient transport and large turbulence energy production due to heat release. The application of the method to a spark-ignited turbulent flame ball is described by Pope and Cheng.<sup>36</sup>

Turbulent premixed combustion usually occurs in the flamelet regime.<sup>37</sup> This fact presents a challenge to any statistical approach, since the small scales of the composition fields are no longer governed by the turbulent straining motions, rather they are determined by reaction and diffusion occurring in thin flame sheets. Pope and Anand<sup>33</sup> present and demonstrate a model applicable to the flamelet regime. However, as discussed by Pope,<sup>5,37</sup> this model is not entirely satisfactory. An alternative approach to treating flamelet combustion is the stochastic flamelet model of Pope and Cheng.<sup>38</sup> This can be viewed as a pdf approach in which a modeled pdf equation is solved by a Monte Carlo method. In this case, however, the pdf is not that of fluid properties (i.e., velocity and composition), but is rather the pdf of flamelet properties (i.e., position, area, and orientation of flamelets).

## V. Discussion and Conclusion

The works reviewed briefly in the previous section demonstrate that pdf methods provide a practicable means of calculating the properties of turbulent reactive flows. Calculations have been made with thermochemical schemes involving up to three composition variables with finite-rate kinetics.<sup>1,2</sup> The Monte Carlo method used to solve the pdf equations has been implemented for a variety of flows, including two-dimensional recirculating flows<sup>18</sup> and the three-dimensional transient flow in a spark-ignition engine.<sup>34</sup>

The most advanced method considered here is the velocity-composition joint pdf approach. This approach has the advantage, compared with moment closures, that chemical reaction can be treated exactly, without approximation. Compared with the composition joint pdf approach, it has the advantages that turbulent transport is treated exactly, and that a separate turbulence model is not needed to determine the Reynolds stresses.

A shortcoming of the velocity-composition joint pdf approach is that it does not provide a completely self-contained model, in that the turbulence frequency  $\langle \omega \rangle \equiv \langle \epsilon \rangle / k$  must be determined by separate means. For example, in some calculations of simple free shear layers, it has been assumed that  $\langle \omega \rangle$  is constant across the flow, and scales with the mean-flow velocity and length scales.<sup>17,27</sup> In more complex flows, another approach is to solve the standard model equation for  $\langle \epsilon \rangle$  (e.g., Ref. 3), or, similarly, to solve a modeled equation for  $\langle \omega \rangle$  deduced from those for  $k$  and  $\langle \epsilon \rangle$ .<sup>39</sup>

A natural extension is to consider  $f(V, \psi, \zeta, x, t)$ —the joint pdf of velocity, composition, and dissipation. This is the probability density function of the compound event  $\{U(x, t) = V, \phi(x, t) = \psi, \epsilon(x, t) = \zeta\}$ , where  $\epsilon(x, t)$  is the instantaneous mechanical dissipation. Following some preliminary

investigations,<sup>2,40,43</sup> a satisfactory model equation for  $f(V, \psi, \zeta; x, t)$  has been developed.<sup>41,42</sup> The incorporation of dissipation within the pdf allows more realistic and accurate modeling. More important, however, the single equation for  $f(V, \psi, \zeta; x, t)$  provides a completely self-contained model for turbulent reactive flows.

There are three major areas in which progress can be expected in the next five years. The first area concerns turbulent mixing models. As discussed in Sec. II, the stochastic mixing models used in pdf methods lead to discontinuous composition time series—this is clearly contrary to the physics of the problem. Nevertheless, the stochastic models have many advantages over alternative suggestions in spite of their lack of physical appeal. For inert mixing, their performance is generally acceptable; for reactive flows, especially in the flamelet regime, their performance is highly suspect. We expect that stochastic models will be improved and refined to account better for the microstructure of the composition fields, and also to allow mixing and reaction to proceed simultaneously at finite rates.

The second area of expected progress is in the computational implementation of complex kinetics. When the Monte Carlo method is used to solve the joint pdf equation for an inert flow involving  $\sigma$  compositions, the computer time and storage increase nearly linearly with  $\sigma$ . In a naive implementation with complex reaction kinetics, it is necessary to solve the coupled set of  $\sigma$  ordinary differential equations

$$\frac{d\phi_\alpha}{dt} = S_\alpha(\phi_1, \phi_2, \dots, \phi_\sigma), \quad \alpha = 1, 2, \dots, \sigma \quad (27)$$

for each particle, on each time step. The right-hand side (which is a combination of reaction rates) is computationally expensive to evaluate scaling roughly as  $\sigma^2$ , and, as is well known, the set of equations is likely to be stiff. Hence, such a naive implementation is impracticable for all but the lowest values of  $\sigma$ .

As mentioned in Sec. IV.A, the more efficient alternative approach followed by all investigators is to implement Eq. (27) through a table look-up scheme.<sup>24,27</sup> To date, this has been done on an ad hoc basis, although progress toward a general methodology can be expected.

The third area of expected progress is in the determination of the mean pressure field  $\langle p(x, t) \rangle$  using the Monte Carlo algorithm. For thin shear flows, the mean pressure is determined readily by invoking the boundary-layer approximations. For statistically stationary, constant-density, two-dimensional recirculating flow, an algorithm to determine  $\langle p \rangle$  has been developed and demonstrated.<sup>17</sup> However, for the general case, a computationally efficient and robust algorithm needs to be developed. (In the three-dimensional transient calculations of Refs. 3 and 4, the Monte Carlo method is coupled to a finite-volume code that determines  $\langle p \rangle$ .)

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