

The Relationship Between the Probability Approach and Particle Models for Reaction in Homogeneous Turbulence

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Chemical reaction in homogeneous turbulence has been studied theoretically using both the probability approach [9-15] and particle models [3-5]. The relationship between these two approaches is determined, thus enabling the advantages of each to be exploited. A sound physical interpretation of the particle model is obtained and in some cases the pdf (probability density function) equations are easier to solve in particle-model form.

INTRODUCTION

An analysis of chemical reaction in statistically homogeneous turbulence should take account of the spatial and temporal variations in the instantaneous chemical compositions. Two ways of accounting for these variations have been proposed. The first—the pdf approach—is based on the statistical description provided by the joint probability density function of the species concentrations. In the second approach—the particle model—the reacting turbulent mixture is viewed as an ensemble of representative particles or lumps of fluid. In this paper it is shown how these two approaches are related, thus enabling the advantages of each to be exploited. The particle model is provided with an alternative and more sound physical interpretation, whereas the pdf equations can in many cases be solved more readily when expressed in particle-model form.

BACKGROUND

Both the pdf and particle-model approaches originate in the coalescence-dispersal model proposed by Curl [1] in 1963. Curl's model was proposed for mixing and reaction in clouds of liquid droplets, but its application to reaction in homogeneous

turbulence is obvious. The model supposes that there is a large number of equal-sized particles, within each of which the composition is uniform. At a given rate, the particles coalesce (two at a time), mix completely, and then redisperse instantly to form two identical particles. In the time between particle interactions, chemical reaction proceeds within each particle.

Spielman and Levenspiel [2] noted that this mixing process could be simulated by the Monte Carlo technique and applied the resulting stochastic model to droplets in a stirred-tank reactor. The same stochastic model has been used subsequently for homogeneous turbulent reaction by Kattan and Adler [3], Flagan and Appleton [4], and Pratt [5].

Curl [1], rather than using this technique, deduced from his model an integrodifferential equation for the evolution of the pdf of the particle compositions. This he, and later Shain [6], solved by a finite-difference method. The same integrodifferential equation was solved (by an approximate method) by Evangelista et al. [7, 8] for homogeneous turbulent reaction. However, the equation for the evolution of the joint pdf of the species compositions can be derived directly from conservation principles, without recourse to the artificial notion of particles [e.g., 9]. In this

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equation the mixing appears as an unknown correlation and so a model is required in order to close the equation. Such models have been proposed by Dopazo and O'Brien [10], Pope [9], Dopazo [11], and Kollmann [12]. Again, the resulting equations are nonlinear integrodifferential equations.¹

The pdf approach has the advantage of being well founded—the pdf equation can be obtained from the species-conservation equation without assumption. The disadvantage is that the equation is difficult to solve. Not only is it a nonlinear integrodifferential equation, but its dimensionality is equal to the number of independent reactants. A simple analysis indicates that the finite-difference solution of the pdf equation with more than two independent reactants is beyond the capability of present computers.

For the particle model, the advantages and disadvantages are reversed. It cannot be claimed that the model is well founded since the notion of a turbulent particle is contrary to the nature of turbulence. However, the equations are comparatively easy to solve. In the stochastic model, between particle interactions, the species concentrations change according to a set of simultaneous ordinary differential equations: the number of equations is equal to the number of independent reactants. The task of integrating such equations is trivial compared with that of solving the pdf equations.

ANALYSIS

For simplicity of exposition, the density is taken to be constant (unity), the system to be adiabatic, and simple gradient diffusion is assumed. Then the turbulent reactive system can be characterized by the velocity field $\mathbf{U}(\mathbf{x}, t)$ and the set of species mass fractions $\phi(\mathbf{x}, t) \equiv \phi_1, \phi_2, \dots, \phi_\sigma$ where σ is the number of independent reactants. The conservation equation for each mass fraction is

$$\frac{\partial \phi_\alpha}{\partial t} + U_i \frac{\partial \phi_\alpha}{\partial x_i} = \Gamma \frac{\partial^2 \phi_\alpha}{\partial x_i^2} + S_\alpha(\phi), \quad (1)$$

¹ In fact, if the Dopazo–O'Brien model [10] is used, the equation contains no integral. However, it appears that only models containing integrals can be physically realistic, [9, 13].

where $S_\alpha(\phi)$ is the rate of production of species α due to chemical reaction and Γ is the molecular diffusivity.

Particle Model

In the particle model there is a large number, N , of particles, the n th of which has composition $\phi^{(n)}(t)$. Each of these particles evolves according to the equation

$$\frac{d\phi_\alpha^{(n)}(t)}{dt} = S_\alpha(\phi^{(n)}) + \text{“mixing,”} \\ \alpha = 1, 2, \dots, \sigma. \quad (2)$$

If the stochastic model is used, the “mixing” is zero nearly all the time: mixing occurs by particle interactions that cause a discontinuous change in $\phi^{(n)}(t)$.

The pdf Approach

The joint pdf, p , of the set of species mass fractions ϕ is defined such that $p(\psi, t)d\psi$ is the probability of $\phi(t)$ being in the range

$$\psi < \phi(t) < \psi + d\psi.$$

Then $\psi \equiv \psi_1, \psi_2, \dots, \psi_\sigma$ is a set of independent variables corresponding to the set of dependent variables ϕ ; and $d\psi = d\psi_1 d\psi_2 \dots d\psi_\sigma$ represents an infinitesimal volume in ψ -space. It follows from equation (1) and the definition of $p(\psi)$ [9] that, in homogeneous turbulence, $p(\psi)$ satisfies the equation

$$\frac{\partial p(\psi)}{\partial t} + \frac{\partial}{\partial \psi_\alpha} \{p(\psi)S_\alpha(\psi) + F_\alpha(\psi)\} = 0. \quad (3)$$

$F(\psi)$ represents the flux of $p(\psi)$ due to mixing and is given by

$$F_\alpha(\psi) = \Gamma \left\langle \delta(\psi - \phi) \frac{\partial^2 \phi_\alpha}{\partial x_i^2} \right\rangle, \quad (4)$$

where angled brackets indicate a statistical mean and δ is the Dirac δ function. It is this term, F_α ,

that needs to be modeled and usually leads to integrals over ψ -space.

Relationship Between Approaches

Since the two approaches are formulated differently they cannot be compared directly. However, both formulations allow the mean of an arbitrary function $Q(\phi)$ to be evaluated, and a comparison of these means can be made. According to the pdf approach, the mean is

$$\langle Q(\phi) \rangle = \int Q(\psi) p(\psi) d\psi, \quad (5)$$

and according to the particle model, it is

$$\langle Q(\phi) \rangle = \frac{1}{N} \sum_{n=1}^N Q(\phi^{(n)}). \quad (6)$$

If the means given by (5) and (6) are to be equal for any Q , it is necessary that as N tends to infinity the particle density distribution tends to the pdf p . This is another way of saying that the particles must be representative of the distribution, which is an obvious condition.

Two evolution equations for $\langle Q(\phi) \rangle$ can be obtained by differentiating equations (5) and (6) with respect to time. This is performed in the Appendix, and the condition that the two equations should produce the same value of $\langle Q(\phi) \rangle$ leads to the following equation for $\phi^{(n)}(t)$:

$$\frac{d\phi_\alpha^{(n)}}{dt} = S_\alpha(\phi^{(n)}) + \frac{F_\alpha(\phi^{(n)})}{p(\phi^{(n)})}. \quad (7)$$

The probability distribution $p(\phi^{(n)})$ is not known explicitly in the particle-model formulation but is, simply, the local particle density.

Thus the evolution of the system given by the pdf approach, equation (3), is equivalent to that given by the particle-model formulation, equation (7).

DISCUSSION

From the viewpoint of the pdf approach, the significance of this result is that the pdf equation is a good deal easier to solve in particle-model form,

equation (7). For example, if the conditionally-Gaussian model [10] is used for $F_\alpha(\psi)$, the pdf equation becomes

$$\frac{\partial p(\psi)}{\partial t} + \frac{\partial}{\partial \psi_\alpha} \{p(\psi) S_\alpha(\psi) + (\langle \phi_\alpha \rangle - \psi_\alpha) p(\psi) / \tau\} = 0, \quad (8)$$

where τ is a turbulent time scale. It may be recalled that $p(\psi, t)$ is a $(\sigma + 1)$ -dimensional quantity. Consequently, the solution of equation (8) by a finite-difference technique can only be contemplated for a few independent reactants—that is, for small σ . Such calculations (with $\sigma = 2$) have been performed (on a rather coarse grid) by Bonniot and Borghi [14]. In particle-model form the same equation becomes

$$\frac{d\phi_\alpha^{(n)}}{dt} = S_\alpha(\phi^{(n)}) + (\langle \phi_\alpha \rangle - \phi_\alpha^{(n)}) / \tau. \quad (9)$$

In contrast, this is a set of σ ordinary differential equations that is to be solved for each of N particles. Clearly, this formulation is more amenable to numerical solution, and solutions for significantly more than two reactants can be envisaged. The use of more realistic (integral) models of $F_\alpha(\psi)$ becomes progressively more expensive computationally as σ increases. Nevertheless, the advantages of the particle-model formulation remain.

From the viewpoint of the particle-model approach, the analysis provides a sound foundation for the formulation, defines the “mixing” term, and provides an alternative to the usual Monte Carlo treatment. In the Appendix it is shown that $\phi^{(n)}$ can be interpreted not as a particle, but as a somewhat abstract though well-defined quantity. This verifies the propriety of regarding the continuous turbulent field as an ensemble of “particles.” Also, comparing equations (2), (4), and (7), it may be seen that a definition of particle mixing has been obtained in terms of the turbulent field. That is,

$$\begin{aligned} \text{“Mixing”} &= \frac{F_\alpha(\phi^{(n)})}{p(\phi^{(n)})} \\ &= E \left\{ \Gamma \frac{\partial^2 \phi_\alpha}{\partial x_i^2}; \phi^{(n)} \right\}. \end{aligned} \quad (10)$$

The last expression is the conditional expectation of $\Gamma \partial^2 \phi_\alpha / \partial x_i^2$. The Monte Carlo method provides a simple method of simulating Curl's model but has the disadvantage of causing discontinuous changes in $\phi^{(n)}(t)$, which can result in unrealistic compositions. On the other hand, the equation for $\phi^{(n)}(t)$ obtained here, equation (7), has smooth solutions and so has no such difficulty.

The results presented here are for reaction in homogeneous turbulence. The particle representation can be expected to be useful in the inhomogeneous case also, but the straightforward extension of the present work meets with some difficulty. This is because an ensemble of particles is not convected along a mean streamline, but each particle is converted along a conditional streamline. Hence the ensemble disperses and so cannot be held to be representative of the pdf at a point. (This observation has no bearing on the homogeneous case.)

APPENDIX

Before deriving an evolution equation for $\phi^{(n)}$, it is necessary to obtain a mathematical definition of $\phi^{(n)}$ in terms of ψ and $p(\psi)$. To this end, consider the σ -dimensional transform space η that has a one-to-one correspondence to ψ wherever $p(\psi)$ is nonzero. In particular, there is an inverse transform ψ^* ,

$$\psi = \psi^*(\eta, t). \quad (\text{A.1})$$

Transforming equation (5) yields

$$\langle Q(\phi) \rangle = \int Q(\psi^*) p(\psi^*) \left| \frac{\partial \psi^*}{\partial \eta} \right| d\eta, \quad (\text{A.2})$$

where $|\partial \psi^* / \partial \eta|$ is the determinant of the Jacobian of the transformation. The product of $p(\psi^*)$ with the Jacobian can be identified as the pdf of η , $p(\eta)$. Now, since $p(\psi)$ is positive semidefinite, η can be chosen such that

$$\left| \frac{\partial \eta}{\partial \psi^*} \right| = \left| \frac{\partial \psi^*}{\partial \eta} \right|^{-1} = p(\psi^*) \quad (\text{A.3})$$

In other words, η can be chosen to be a space of uniform probability with $p(\eta)$ equal to unity

everywhere. Then equation (A.2) becomes

$$\langle Q(\phi) \rangle = \int Q(\psi^*) d\eta. \quad (\text{A.4})$$

The integral in equation (A.4) can be expressed as the limit of a sum

$$\langle Q(\phi) \rangle = \lim_{N \rightarrow \infty} \sum_{n=1}^N Q[\psi^*(\eta^{(n)})], \quad (\text{A.5})$$

where $\eta^{(n)}$ are N equally spaced points in η -space. By comparing this with the particle-model equation for $\langle Q(\phi) \rangle$, equation (6), it may be seen that the two relations are the same in the limit of large N if the particles are chosen by

$$\phi^{(n)} = \psi^*(\eta^{(n)}). \quad (\text{A.6})$$

An implication of this equation is that the particles should be uniformly distributed in η -space, which is a space of uniform probability. In other words, in ψ -space, the particle density distribution should be equal to the probability density function—which is an obvious result. The importance of equation (A.6) is that the ill-defined particle $\phi^{(n)}$ can be identified with the well-defined, if more abstract, inverse transform ψ^* .

Having identified $\phi^{(n)}$ with $\psi^*(\eta)$ through equation (A.6), an evolution equation for $\phi^{(n)}(t)$ can be obtained as follows. Differentiating equation (5) with respect to t gives

$$\frac{d\langle Q \rangle}{dt} = \int Q(\psi) \frac{\partial p(\psi)}{\partial t} d\psi. \quad (\text{A.7})$$

Substituting for $\partial p(\psi) / \partial t$ from equation (3) and integrating by parts produces

$$\frac{d\langle Q \rangle}{dt} = \int \frac{\partial Q(\psi)}{\partial \psi_\alpha} \{p(\psi) S_\alpha(\psi) + F_\alpha(\psi)\} d\psi, \quad (\text{A.8})$$

and transforming to η space leads to

$$\frac{d\langle Q \rangle}{dt} = \int \frac{\partial Q(\psi^*)}{\partial \psi_\alpha^*} \left\{ S_\alpha(\psi^*) + \frac{F_\alpha(\psi^*)}{p(\psi^*)} \right\} d\eta. \quad (\text{A.9})$$

Alternatively, differentiating equation (A.4) with

respect to t yields

$$\frac{d\langle Q \rangle}{dt} = \int \frac{\partial Q(\psi^*)}{\partial \psi_\alpha^*} \left\{ \frac{\partial \psi_\alpha^*}{\partial t} \right\} d\eta \quad (\text{A.10})$$

Clearly, these last two equations are compatible if the terms in braces are equated, which results in the required evolution equation

$$\frac{\partial \psi_\alpha^*}{\partial t} = S_\alpha(\psi^*) + \frac{F_\alpha(\psi^*)}{p(\psi^*)}. \quad (\text{A.11})$$

Or, in terms of particles, we have

$$\frac{\partial \phi_\alpha^{(n)}}{\partial t} = S_\alpha(\phi^{(n)}) + \frac{F_\alpha(\phi^{(n)})}{p(\phi^{(n)})}, \quad (\text{A.12})$$

where $p(\phi^{(n)})$, it may be recalled, is the local particle density.

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