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TURBULENCE COMBUSTION MODELING: FLUCTUATIONS AND CHEMISTRY

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1 Introduction

Aerodynamic CFD calculations are routinely conducted that calculate the performance of an airfoil or wing with remarkable accuracy. Turbulent combustion is, of course, a much more complex phenomenon than the flow of air around an aircraft. Nevertheless, a major aim of research on turbulent combustion should be to develop models that have the necessary quantitative accuracy when applied to reacting flows of importance in engineering.

The fundamental problem of turbulent combustion modeling — as it has been for the last 25 years — is to accurately represent three interconnected phenomena: inhomogeneous turbulent flow; the chemistry of combustion; and turbulent fluctuations of temperature, species and density. Of course, in applications there are other phenomena — such as sprays and soot — that may be important; but the core of the problem is the interconnection of these three phenomena. In gas-phase laminar flows, great strides have been made so it is now possible to calculate laminar flames to good accuracy. It remains a challenge to calculate turbulent flames to the same degree of accuracy.

The most common CFD approach to turbulent reactive flows is based on the mean -flow closure. Model transport equations are solved for the mean velocity, for the mean compositions, and for turbulence model quantities such as k and ϵ. Models of this type provide little or no account of the composition fluctuations. Turbulent combustion models, such as the assumed PDF of mixture fraction, can

provide reasonable treatments for some simple situations. But in general, the mean-flow approach lacks the level of description needed to account accurately for finite-rate chemical kinetics.

In contrast to mean-flow closures, PDF methods provide a full statistical description of the composition fluctuations. There are different levels of PDF closure. The current work at Cornell is focused on the joint PDF of velocity, composition and turbulence frequency. The single modelled equation for this joint PDF provides a self-contained model for turbulent reactive flows. The principle virtues of this approach are that the two dominant processes in turbulent combustion are treated in closed form: namely, reactions of arbitrary complexity, and convection by both the mean and fluctuating components of velocity.

In the next section, the basic ideas of this PDF method are briefly described. Section 3 provides a summary of recent developments. An important advance is the method of *in situ* adaptive tabulation (ISAT) that enables the computationally efficient implementation of combustion chemistry. Recent work on this method is described in more detail in Section 4.

The purpose of this paper is solely to provide a sketch of recent work in the author's research group at Cornell. Consequently, the references given are strongly biased towards this work. A different kind of paper would be needed to give appropriate credit to all who have contributed to the development of the PDF methodology.

2 PDF Methodology

The PDF methodology is most easily understood in the setting of its numerical implementation, which is a particle-mesh method. As in finite-volume methods, there is a grid that divides the solution domain into cells. But the primary representation of the turbulent reactive flow is in terms of particles. There is a large number N of particles, each of which is ascribed the following properties: mass m^*, position $\mathbf{X}^*(t)$, velocity $\mathbf{U}^*(t)$, composition $\boldsymbol{\phi}^*(t)$, and turbulence frequency $\omega^*(t)$. The particle mass remains fixed while the other properties evolve in time. These particles model fluid particles, and so the position evolves simply as

$$\frac{d\mathbf{X}^*(t)}{dt} = \mathbf{U}^*(t)\,.$$

At any given time, mean fields are estimated from the particle properties. Typically, at grid nodes (*i.e.*, cell vertices) the method of local least squares is used to estimate means, and then linear interpolation is used within each cell. The model evolution equations for the remaining particle properties involve these mean fields. For example, the simplest model equation for composition is

$$\frac{d\boldsymbol{\phi}^*(t)}{dt} = -C_\phi\langle\omega\rangle(\boldsymbol{\phi}^*(t) - \langle\boldsymbol{\phi}\rangle) + \mathbf{S}(\boldsymbol{\phi}^*)\,.$$

In this equation, the final term is the rate of change due to chemical reaction, which is treated exactly, without modeling assumptions. The first term on the right-hand side is the interaction by exchange with the mean (IEM) model for molecular mixing. This causes $\phi^*(t)$ to relax to the local mean value $\langle\phi\rangle$ at a rate proportional to the local mean frequency $\langle\omega\rangle$.

The simplest model for velocity is the simplified Langevin model (SLM) given by the stochastic differential equation

$$\begin{aligned} d\mathbf{U}^*(t) = -\frac{1}{\langle\rho\rangle}\nabla\langle p\rangle dt \quad &- \quad \left(\frac{1}{2}+\frac{3}{4}C_0\right)\langle\omega\rangle(\mathbf{U}^*-\langle\mathbf{U}\rangle)dt \\ &+ \quad (C_0\langle\omega\rangle k)^{\frac{1}{2}}\, d\mathbf{W}. \end{aligned}$$

The first term on the right-hand side involves the mean pressure gradient, and this term alone guarantees that the mean momentum equation is satisfied. The next term is similar to the IEM model and makes the particle velocity relaxed to the local mean. The final term involves the Wiener process which causes a diffusion in velocity space. The quantity k is the turbulent kinetic energy, and so the product $\langle\omega\rangle k$ is the mean dissipation rate ϵ. It may be recognized, then, that the diffusion coefficient is $C_0\epsilon$ as given by Kolmogorov theory.

The model for turbulence frequency also takes the form of a diffusion process [1]. A valuable feature of this model is the definition of a conditional — or "above average" — mean of ω:

$$\Omega = C_\Omega\langle\omega|\omega \geq \langle\omega\rangle\rangle.$$

The constant C_Ω is defined so that in homogeneous turbulence the conditional and unconditional means are equal. At the edge of free shear flows, Ω goes to zero much more slowly than $\langle\omega\rangle$, so that the ratio $\langle\omega\rangle/\Omega$ behaves like the intermittency factor γ. The behavior of the model equations in the intermittent region is greatly improved by the selective use of Ω in place of ω.

3 Recent Advances

3.1 Velocity Models

A discussion of the basis of the Langevin model is provided [2, 3]. Haworth & Pope [4] developed the generalized Langevin model (GLM) which introduces a dependence on the mean velocity gradients, in accord with the modeling of the rapid pressure-rate-of-strain in Reynolds stress closures. In fact, there is a close correspondence between Reynolds stress closures and velocity models of the form of the generalized Langevin model [5]. Based on this, velocity models have been developed corresponding to the isotropization of production model (IPM) and to the Sarkar Speziale and Gatski (SSG) model.

3.2 Near Wall Flows

As in standard turbulence modeling, there are two ways to treat flows near walls. One is to use wall functions, the other is to characterize the flow through the viscous sublayer to the wall and these approaches have been studied recently [6]–[8]. The wall function approach amounts to reflecting particles off the wall with appropriate reflection conditions, to yield a specified wall shear stress.

In order to make calculations through the viscous sublayer to the wall, it is of course necessary to introduce the viscous effects that are neglected in the standard high Reynolds number modeling. This is done by replacing the position equation by

$$d\mathbf{X}^*(t) = \mathbf{U}^*(t)dt + \sqrt{2\nu}d\mathbf{W}'.$$

This equation describes the statistics of the motion of a molecule, and so the particle now models the continuum fluid properties following the motion of a molecule. Using this model, calculations have been made of turbulent channel flow and good agreement is obtained with the Reynolds stress budgets obtained from DNS.

3.3 Wave Vector Model

In turbulence modeling, a useful limit to consider is that of rapid distortion. When homogeneous turbulence is rapidly distorted, the resulting change in the turbulence spectrum can be determined from rapid distortion theory (RDT). It has been shown [9] that Reynolds–stress closures are incapable of accounting for the evolution of the Reynolds stresses when the turbulence is subjected to rapid distortions that include mean rotation. These authors have gone on to augment Reynolds-stress models with additional dependent variables in order to describe all rapid distortions.

Drawing heavily on this work, a PDF closure that is exact for rapid distortions of homogeneous turbulence, has been developed [10]. A unit vector — called the wave vector — is added to the PDF formulation. This model has been applied to homogeneous turbulence and to the plane mixing layer with excellent results. It is expected that this model will prove advantageous compared to Reynolds-stress closures in flows with complex mean velocity gradients, such as swirling jets.

3.4 High Speed Flows

Most of the PDF studies performed at Cornell are for low Mach number flows. The fluid density may vary appreciably due to composition and temperature variations but, at low Mach number, pressure variations have a negligible effect on the density and on the energy. In high speed flows, on the other hand, the pressure variations significantly affect both the density and the energy. PDF methods have been extended to high Mach number flows [11]. This involved adding pressure as a particle property, and in specifying coupled stochastic differential equations for pressure and energy.

The method has been applied to several flows with good results, including the supersonic reacting mixing layer studied experimentally.

3.5 Turbulent Mixing Models

In the evolution equation for the particle composition $\phi^*(t)$, a turbulent mixing model — the simplest being IEM — is used to model the effects of molecular diffusion. Since PDF methods (as described here) provide a one-point statistical representation of the flow, and yet molecular diffusion depends on the microstructure of the composition fields, there are significant difficulties in formulating a general and accurate mixing model. It is simpler to point out deficiencies in existing models than it is to devise new models that remedy these deficiencies!

Over the years, there has developed a growing list of constraining conditions and desiderata for turbulent mixing models. In the context of combustion, an important condition is *localness* [12]. The evolution of the composition $\phi^*(t)$ should be determined by particles whose composition is close (in composition space) to $\phi^*(t)$. The IEM model does not satisfy this localness condition, and as a consequence its predictions are qualitatively incorrect in some important circumstances, notably the flamelet regime of nonpremixed combustion.

To remedy this problem, Subramaniam and Pope [13] have developed the EMST mixing model, based on Euclidean minimum spanning trees. Demonstrations of the success of the model are provided [14, 15], a subtler consideration is the dependence of mixing on the particle velocity [16].

3.6 PDF Methodology in Large Eddy Simulations

In some quarters, statistical approaches are viewed as being incapable of accurately describing turbulent reactive flows, and more deterministic approaches such as DNS and LES are advocated instead. Part of the reason for this view is that (in some minds) the large scale organized turbulent motions observed in turbulent flows are at odds with statistical notions. This viewpoint prompts several comments.

1. In spite of this view, that has been heard since the 1970's, statistical approaches remain (with very few exceptions) the design tool used in engineering industries.

2. For engineering flows, DNS requires many orders of magnitude more computer power than will be available for several generations, and therefore should not be considered as a possible engineering design tool.

3. Large eddy simulations (LES) require models for the subgridscale motions. For reactive flows, the straightforward LES approach of solving equations for the filtered composition fields leads to the same formidable closure problem that is encountered in mean flow closures.

4. Several calculations have been performed [17] of flows with shedding. They show that time means statistical methods are indeed inaccurate, but that the same statistical models yield accurate results if only the very largest motions are treated in a time-dependent way.

For LES of reactive flows, the subgrid closure problem associated with nonlinear reaction rates can be circumvented by using the PDF methodology. The appropriate quantity to consider is the filtered density function (FDF) that was introduced by Pope [18] and developed by Gao & O'Brien [19]. At any point and time, the FDF gives the density in composition space of fluid around the point, weighted by the filter. Just as reaction is in closed form in the PDF equations, so also reaction is in closed form in the FDF equations.

The first implementation of an LES/FDF methodology is reported by Colucci *et al.* [20]. For two test flows they compare results from DNS, from LES/FDF and from LES with the neglect of composition fluctuations. The results confirm that the neglect of composition fluctuations can lead to huge errors, and that the LES/PDF approach is both tractable and accurate. Further work is needed to apply the LES/FDF approach to turbulent flames so that direct comparison with experimental data can be made.

4 Efficient Implementation of Combustion Chemistry — ISAT

For reactive flows, the great advantage of the PDF approach is that reaction is in closed form. This advantage can be realized only if the reaction mechanisms of interest can be implemented in a computationally tractable way.

As a definite example, a skeletal mechanism for methane consisting of 16 species and 41 reactions is considered [21]. To integrate the reaction equation

$$\frac{d\boldsymbol{\phi}}{dt} = \mathbf{S}(\boldsymbol{\phi})$$

once on an SGI workstation required about 0.2 s of CPU time. For a full-scale PDF computation this reaction calculation must be made for each of (maybe) 500,000 particles on each of (maybe) 2,000 time steps, *i.e.*, or order 10^9 times. Simple arithmetic shows that this requires 6 years of CPU time, which is generally deemed to be prohibitive.

To avoid such huge computational requirements, in the past, simple reaction mechanisms or representations of the chemistry have been used: for example, global mechanisms, systematically reduced mechanisms, and manifold methods. These simpler descriptions of the chemistry have often been implemented via table look-up schemes. That is, in a preprocessing stage, a table is constructed of the composition after reaction for a time interval Δt as a function of the composition $\boldsymbol{\phi}$ before reaction. We denote this tabulated function by $\mathbf{R}(\boldsymbol{\phi})$. Then, in the turbulent reactive

flow calculation, rather than solving the reaction equation for each particle on each time step, instead the result $\mathbf{R}(\phi)$ is obtained from the table by linear interpolation.

It has generally been supposed that table look-up methods are restricted by computational considerations to simple schemes. For, the table must be constructed in the composition space whose dimension is the number of degrees of freedom in the chemistry — which is roughly equal to the number of species. But in fact, for a number of reasons, the region of the composition space that is accessed in a turbulent reactive flow calculation is but a small fraction of the whole composition space. As a consequence, with an appropriately designed algorithm, a tabulation is possible.

The *in situ* adaptive tabulation algorithm (ISAT) has been demonstrated for a partially stirred reactor using the 16-species skeletal mechanism for methane [21]. Because the accessed region of the composition space is not known *a priori*, the tabulation is performed *in situ* as the flow calculation is in progress, rather than in a pre-processing stage. The table is unstructured and adaptive so that the interpolation errors are controlled relative to a specified tolerance.

4.1 Demonstration of ISAT

New results are now reported of the use of ISAT for a one-dimensional unsteady laminar flame test case. A motivation for these tests was to examine the performance of ISAT used in conjunction with finite-difference schemes as opposed to particle methods.

The flow is one-dimensional and time dependent. At the left boundary ($x = 0$) there is an in-flow of premixed stoichiometric hydrogen and air at atmospheric conditions. The fluid velocity is uniform in x, but varies sinusoidally in time as:

$$u(t) = 1 + \sin\left(\frac{t}{T}\right) \text{ m/s},$$

where $T = 10^{-5}$ s. The compositions evolve in time according to the following simple convection-diffusion-reaction equation

$$\frac{\partial \phi}{\partial t} + u(t)\frac{\partial \phi}{\partial x} = D\frac{\partial^2 \phi}{\partial x^2} + \mathbf{S}(\phi),$$

where the diffusivity is $D = 10^{-4}$ m/s^2. The initial condition is that the fluid is in chemical equilibrium everywhere (except at the inlet boundary). Typical profiles of the major species are shown in Fig. 1 and the minor species are shown in Fig. 2. The reaction-convection-diffusion equation is solved by a simple finite-difference method. Strang's splitting algorithm is used so that convection and diffusion are treated (using Crank-Nicolson) on one fractional step, and reaction is treated using ISAT on another.

The tests show that the method is second-order accurate in time; and that the ISAT algorithm is convergent in the sense that the global error in the calculation

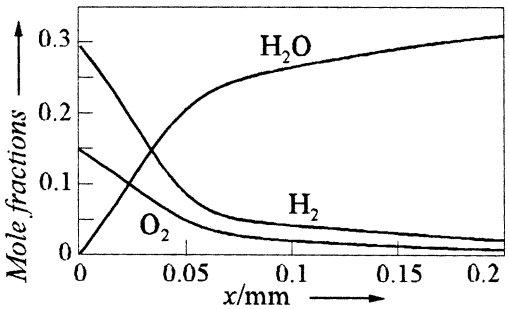

Figure 1. Major species in the one-dimensional unsteady laminar flame test

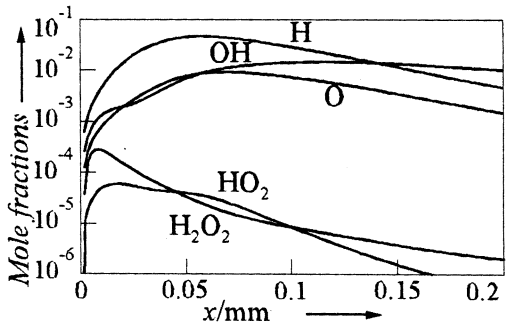

Figure 2. Minor species in the one-dimensional unsteady laminar flame test

(resulting from tabulation errors) decreases to zero as the specified tolerance is decreased. Figure 3 shows the global error in the calculation as a function of the time step. In this log-log plot the slope of two confirms the second order accuracy of the splitting scheme. Figure 4 shows the same global error this time plotted against the specified error tolerance in the ISAT algorithm. As may be seen the global error may be reduced to the level of round-off error by specifying a suitably small value of the error tolerance.

The next two figures illustrate the computation performance of the ISAT algorithm. For the early stages of the calculation, Fig. 5 shows the CPU time plotted against the number of queries — that is, the number of times the reaction calculation is performed. If direct integration of the reaction equation is used (rather than ISAT) then the CPU time is simply linearly proportional to the number of queries. It may be seen from the figure that initially ISAT consumes slightly more CPU time than direct integration (DI) but after about 20,000 queries the CPU time required by ISAT levels off and becomes dramatically less than that required by direct integration. The initial steep slope of this ISAT curve corresponds to the building of the table whereas after 20,000 queries nearly every queries is satisfied by a retrieve from the existing table rather than by the addition of a new table entry.

Figure 6 shows the speed-up factor of ISAT relative to direct integration as a function of the number of queries. It may by seen that little speed-up is achieved until 10^4 queries. After 10^8 queries a speed-up of over a factor of 100 is achieved.

For the partially-stirred reactor test case [21], the speed-up factor is even larger — a factor of about 1,000. Full scale PDF calculations are now under way for the piloted jet diffusion flame with the 16-species skeletal mechanism for methane, and it is already clear that such calculations are quite tractable on a workstation.

It is likely that the ISAT algorithm will greatly increase the application of skeletal and detailed chemical mechanisms to a variety of combustion problems not only in the context of PDF methods for turbulence combustion.

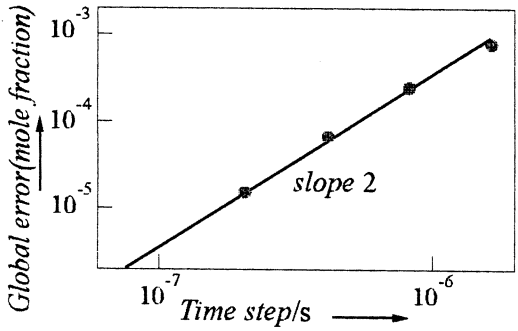

Figure 3. Time-stepping error as a function of the time step in the one-dimensional unsteady laminar flame test. The global error is the average (over grid nodes) of the two-norm of the difference between the numerical solution with the specified time step and that with a very small time step

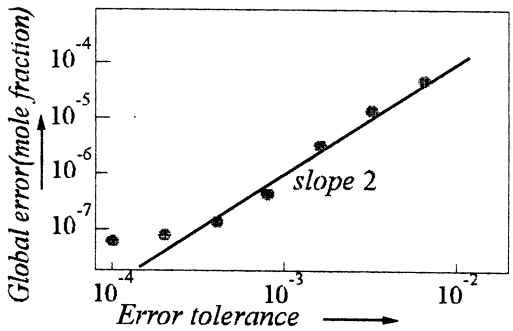

Figure 4. Global error (average two-norm of mole fraction errors) as a function of the specified ISAT error tolerance in the one-dimensional unsteady laminar flame test

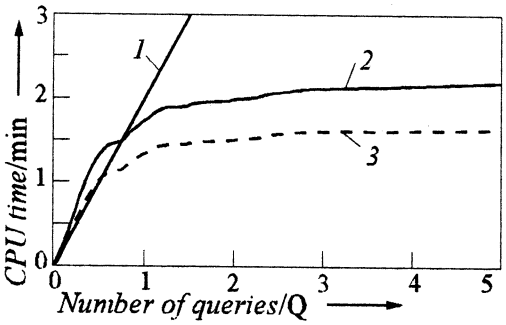

Figure 5. CPU time (on an SGI Indigo workstation) and number of table entries as functions of the number of queries (*i.e.*, reaction calculations). *1* — direct integration, *2* — *in situ* adaptive tabulation, *3* — $N_{records}/1000$

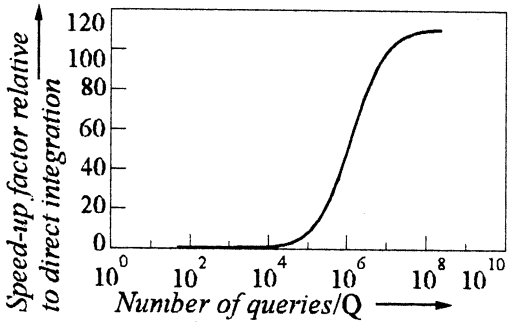

Figure 6. Speed-up factor (total time for Q queries using direct integration / total time for Q queries using ISAT) as a function of the number of queries

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