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Paradigms in turbulent combustion research

R.W. Bilger[a,*], S.B. Pope[b], K.N.C. Bray[c], J.F. Driscoll[d]

[a] School of Aerospace Mechanical and Mechatronic Engineering, University of Sydney, NSW 2006, Australia
[b] Sibley School of Mechanical and Aerospace Engineering, Cornell University, Ithaca, NY 14853, USA
[c] 23, De Freville Ave, Cambridge CB4 1HW, UK
[d] Department of Aerospace Engineering, University of Michigan, Ann Arbor, MI 48109, USA

Abstract

The development of the basic conceptual viewpoints, or paradigms, for turbulent combustion in gases over the last 50 years is reviewed. Significant progress has been made. Recent successes in the prediction of pollutant species and extinction/re-ignition phenomena in non-premixed flames are seen as the result of close interaction between experimentalists, theoreticians, and modellers. Premixed turbulent flames seem to be dependent on a much wider range of factors, and predictive capabilities are not so advanced. Implications for large eddy simulation (LES) and partially premixed combustion are outlined.

1. Introduction

This week we celebrate a half-century of what has been a remarkable, sustained endeavour to surmount the challenges of combustion science and application. The Combustion Institute was formed to provide a permanent home for the multi-disciplinary community of scientists and engineers that had grown together, after the Second World War, with the common interest of grappling at a fundamental level with the problems of combustion processes in engines (spark-ignition, diesel, gas-turbine, and rocket), furnaces, explosions, and fires, and the aero-thermochemistry of rocket nozzles and re-entry of objects from earth orbit. This community had already produced the fabulous archive of its three post-war symposia. It knew that it needed to sustain and grow itself if it were to surmount the great challenges that lay ahead. How else to assimilate many still-emerging sciences—thermochemistry, chemical kinetics, molecular transport phenomena, heat, and mass transfer in single and multi-phase systems, and laminar and turbulent fluid flow—into a coherent science capable of application to engineering problems?

Fifty years is a long time in science. Just look at the biological sciences! Why have we not solved the problem in this time? Surely, back then, our founders projected a far shorter time frame than half a century when seeking support for the fledgling Institute and for their own individual research projects. The opening speeches at the Royal Institution in London for the 1958 Symposium make interesting reading. Sir Cyril Hinshelwood, then President of the Royal Society and winner, with Semenov, of the 1956 Nobel Prize in chemistry for his work on the chemical kinetics of combustion, gives a sobering assessment [1] of the difficulties of achieving a satisfactory theory for laminar flames. It includes, by the way, a complete dismissal of any future for Zel'dovich's asymptotics! Lewis [2] bemoans the Edisonian approach, still

* Corresponding author. Fax: +61 2 9351 7060.
E-mail address: bilger@aeromech.usyd.edu.au (R.W. Bilger).

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universally used then in combustion development. He cites the discovery of lead tetraethyl as an anti-knock additive after an exhaustive and expensive testing program. And further, in spite of much continued effort at a fundamental level, the science behind this technology was still not understood when Lewis was speaking 35 years later. Surely, combustion science could eventually outreach such blind empiricism! Both Lewis and Hinshelwood knew that practical applications would need the mastering of turbulence—a formidable task in itself—and would need to be coupled to all the intricacies of hydrocarbon chemical kinetics that were then emerging. They did not explicitly mention this, however! Experts advising the Australian government in the 1960s listed Combustion as one of a few areas of research that would not be cost-effective!

Have we solved the problem? Laminar flames? On the whole, for simple fuels: yes—but we still have many papers in this area! Hydrocarbon kinetics? Well along the road, it would seem. Turbulent combustion and its science-based application to engineering problems? Not yet, but significant progress has been made.

At the 27th Symposium, Correa [3] reviewed the remarkable progress that has been achieved in gas turbine technology over the past 50 years. To illustrate the value of combustion science, he shows that CFD using PDF methods [4] can produce accurate calculations of the NO emissions from combustors. This methodology is now gradually being incorporated into commercial CFD codes [5], and hence available for broader application. We can also take heart that Michael Schumacher and the Ferrari Formula 1 team put their trust in one of our combustion science community [6]—Thierry Baritaud—for the most crucial aspects of their engine design! Over the last five, Ferrari-winning years, Thierry and the Ferrari team have been able to get a 2–3% increase in peak power every year. He makes use of CFD codes for fuel mixing and evaporation, and advanced turbulent combustion codes for flame travel to guide the empirical development of the combustion design [7,8]. He looks forward to combustion codes that can be even better predictors of flame travel and of the post-flame reactions that also affect performance.

Increasingly, advanced combustion modelling is finding application across the broad range of industry. Increasingly, such computational modelling is becoming more sound in its scientific base. There is still a long way to go until it enjoys the status of being an accurate predictive tool as is finite element analysis in elasticity and conductive heat transfer.

Turbulent combustion has been the subject of regular invited lectures and topical reviews over the last 20 years [9–19] of the biennial symposia of The Combustion Institute. The Hottel Lecture given by Ken Bray at the 26th Symposium [15] is particularly comprehensive. It pre-empts any attempt at such a comprehensive review for this 50th anniversary lecture.

Our approach is prompted, instead, by the outstanding advances that have been made in recent years in turbulent non-premixed combustion. These advances have been closely associated with a series of international workshops on turbulent non-premixed flames [20]. The aim of these workshops has been to foster close interaction between experimentalists, modellers, and theoreticians, with a focus on elucidating the physics and computational modelling of the chemistry–turbulence interactions that occur, and on improving measurement techniques. It is generally agreed that advances here have been more rapid than in premixed turbulent flames, and that this is partly because there exists a well-developed and widely accepted hierarchy of experiments that is well aligned with the capabilities of the modelling and simulation. In contrast, there is no generally accepted set of experiments for the study of turbulent premixed flames.

It is also widely held that premixed turbulent combustion is inherently more complex than non-premixed [21], and that this is because there is a much stronger coupling between the chemistry and the turbulence. Strong coupling also occurs in piloted jet diffusion flames at high turbulent mixing rates such as in the Sandia [20,22] flames E and F where local extinction and re-ignition occur. Modelling of these flames has, however, been quite successful [23–27].

Our objective here was to explore possible reasons for this disparity in achievement. We review the historical development of the conceptual viewpoints or paradigms associated with these two branches of the subject. Thomas Kuhn argues [28] that the most interesting aspects of scientific enquiry are the paradigm shifts and the events causing them, which may be experimental, theoretical or sociological. We hope to gain insight into new approaches for the study of turbulent premixed combustion and for the emerging subject of partially premixed turbulent combustion. Much that has been learned in the past must play a central role in addressing today's problems of perfecting large eddy simulation (LES) for combustion.

We limit our scope to the crucial issue of turbulence-chemistry interactions in gases. We leave questions of buoyancy, radiant loss, multi-phase transport, etc., aside. The views presented here are not intended to reflect the wide range of views held within the turbulent combustion research community as a whole. Rather they are intended to reflect the views of the authors, who perhaps represent a mainstream of the research community. Furthermore, we have not tried to reach a consensus, but present instead the spread of views that we have among us.

2. Non-premixed turbulent combustion

The major, well-founded paradigms in turbulent non-premixed combustion that are considered here are given in Table 1. These are categorised in terms of their modelling of the chemistry and mixing. These are discussed in more detail in the following sub-sections together with the innovative experiments that have arisen from them or that they suggest for future investigation.

2.1. The mixing-controlled paradigm

The original paradigm for turbulent non-premixed combustion has the viewpoint that such combustion is basically mixing controlled. The classical paper by Hawthorne et al. [29] appears in the first of the post-war combustion symposia. In it, the flame lengths and mean structure of turbulent jet diffusion flames are studied and are found to correlate well with the mixing laws of turbulent jets. The paradigm is closely related to that for laminar diffusion flames as studied by Burke and Schumann [30] in the first combustion symposium held in 1928. In that early work, the mixing is by molecular diffusion in laminar flow, and the flame is situated at the surface where the fuel and oxidant are in stoichiometric proportions—the reaction zone, itself, being very thin compared with the diameter of the flame. The main idea was that chemical rates were much faster than mixing by molecular diffusion. This concept was carried across into the paradigm for turbulent diffusion flames—if fuel is in excess, the mixture consists of products and excess fuel; if oxygen is in excess, the mixture consists of products and excess oxidant. A variable, essentially the same as today's mixture fraction [31], can be defined that describes the stoichiometry of the mixture, and the instantaneous temperature and composition of the mixture would be uniquely related to the mixture-fraction variable. In turbulent flow, the mixture fraction at a particular point has random fluctuations, but these have statistics similar to those in non-reacting flow. Mean values of composition and temperature can be obtained by weighting their instantaneous relationship to the mixture fraction by the probability density function (PDF) of the mixture fraction. Such mean values seem to show co-existence of fuel and oxidant. This is not, however, due to the slowness of chemical reaction, but due to the fluctuating stoichiometry: significant concentrations of both fuel and oxidant may exist at the same place, but not at the same time.

Such a *mixing-controlled* paradigm immediately raises the question as to its range of validity. If the mixing can be made fast enough, then surely the reaction rates would be controlled by the chemistry and not by the mixing. The well-stirred reactor of Longwell and Weiss [32] used small sonic jets to produce what was thought to be a homogeneous mixture of reactants and products within the reactor. Would the length of jet diffusion flames be affected by limits on the reaction rate? In a little-known study, Hottel reports [33] on experiments at MIT [34] where this was investigated by studying jet diffusion flames at a range of pressures from 0.3 to 6 atm. The original idea was that chemical rates would go approximately as the square of the pressure whereas the Reynolds number would go linearly with pressure. The findings were that the flame lengths were still controlled by mixing but that buoyancy was more important in determining rates of mixing in such flames than was the Reynolds number. A modified Froude number was needed to incorporate the effects of buoyancy on mixing.

In the early 1970s, interest was renewed in the effects of chemical kinetics, particularly for pollutants such as nitric oxide. This interest came as a result of sociological pressures for reduction in air pollution emissions arising from combustion processes. Nitric oxide is formed by relatively slow reactions, and so could not come directly under the mixing-controlled paradigm. Since the formation of nitric oxide was not important for the major species and overall heat release, efforts were made to correlate its production with the convective and mixing time scales in the flow. Early results on hydrogen jet diffusion flames, with Froude number held constant [35], suggested that the reactive time scale had a minus half power dependence on the Reynolds number—and so proportional to the Kolmogoroff, or fine-scale time scale of the turbulence. Later work [36] has indicated that radiation and differential diffusion effects are also important, particularly in hydro-

Table 1
Paradigms in non-premixed turbulent combustion

Paradigm	Chemistry	Mixing
Mixing controlled	Fast for major species	Scaling laws/second-order closures
Laminar flamelet	Precalculated tables/'Lagrangian' Pdes	Laminar counter-flow/second-order closure
PDF approach	Already closed/reduced mechanisms	Mixing models
Conditional moment closure	Closure in terms of conditional moments	PDF integrals
Multiple mapping conditioning	Closure in terms of conditional means	Mapping closure

carbon flames, and such attempts at simple correlation are not sustainable.

For a while in the 1960s and 1970s, the primary emphasis was on how the rate of mixing was affected, not only by buoyancy, but by density variations and/or fluctuations, and by the postulated phenomenon of 'flame-generated turbulence' [37]. Kent and Bilger [38] studied a horizontal round-jet diffusion flame of hydrogen into a co-flowing airstream, the main idea being that minimal deflection of the flame in the vertical direction and, preservation, essentially, of axi-symmetry would gainsay any effects of buoyancy—the main interest was in density effects and 'flame generated turbulence'. On the down side was the introduction of new non-dimensional parameters such as the ratio of the jet to co-flow velocity and the probable increase in the influence of the boundary layer characteristics at the jet nozzle exit—something that could involve a host of new non-dimensional parameters. This development reflected the new paradigm for the study of non-reacting turbulence. The then classical work on self-preserving flows and equilibrium flows was being superseded by attempts to describe the evolution of turbulence by partial differential equations involving several significant terms such as production, advection, turbulent diffusion, and dissipation [39]. These equations were derived from Reynolds averaging of the Navier–Stokes equations. As Bray pointed out [40], many more terms would be involved in flows with strong density fluctuations. Density-weighted or Favre averaging of the Navier–Stokes and species conservation equations [41] appeared to be a way around this problem and has enjoyed some success [42,43].

The modelling of chemistry was extended to include the concept of chemical equilibrium being the fast chemistry limit so that the effects of high temperature dissociation could give estimates for the concentrations of intermediate species and radicals such as H, OH, and O. This was particularly helpful for modelling nitric oxide. Early work in hydrogen jet diffusion flames [44] assumed that the kinetics were essentially those of the Zel'dovich reduced mechanism [45] and that the temperature and oxygen concentrations were given by the fast chemistry paradigm. Consequently, the average production rate, needed in the Reynolds/Favre-averaged equation for this species, could be obtained by weighting instantaneous reaction rates, derived from fast-chemistry theory for the temperature and reactant species, by the PDF of the mixture fraction [44].

A significant result arising from this paradigm was the discovery by analysis [46] that reaction rates in flames with fast chemistry are proportional to the rate of scalar dissipation or the rate of molecular mixing (of the mixture fraction). Scalar dissipation rates in turbulent jets into still air scale with the minus fourth power of the distance from the nozzle exit—so finite-rate chemistry effects would be most apparent near the nozzle exit. The bulk of the carbon monoxide burnout occurs much further downstream where scalar dissipation rates are very low. No wonder that Hottel [33] and Homsy [34] could find no effects of kinetic rates on flame lengths defined in terms of CO burnout!

An immediate upshot of this discovery was the new viewpoint that the reactive species and temperature should depend not just on mixture fraction, as under the 'fast chemistry' paradigm, but also on local values of scalar dissipation. High values of local scalar dissipation would need high values of chemical reaction rates to balance the high rates of reactant arrival in the reaction zone from turbulent mixing. Tsuji and Yamaoka [47] had shown, in a beautiful series of investigations on laminar counter-flow diffusion flames, that the composition and temperature were dependent on the strain-rate in the flow—a quantity linearly related to the scalar dissipation of the mixture fraction. At high strain rates (scalar dissipation rates), temperatures are lowered, and there is increased overlap in the profiles of fuel and oxidant concentrations. At very high strain rates, extinction occurs.

Turbulent diffusion flames of hydrocarbons posed a great problem in these times. The 'fast chemistry' paradigm implied that chemical equilibrium would be the limit applicable at low rates of scalar dissipation. Experiments showed that levels of CO concentration were far below those predicted by full chemical equilibrium [48]. Analysis of experimental data on laminar diffusion flames [49] indicated that such equilibrium structure was not approached at the low strain rates of interest—some sort of metastable state was involved. It was proposed [49] that such experimental data could be used for turbulent flame calculations. Lieuw et al. [50] showed that CO levels in turbulent diffusion flames could be well predicted by assuming that moderately low strain-rate data from laminar flame calculations could be used to predict CO levels in turbulent (methane) flames. These concepts [49,50] were the forerunners of what has become known as laminar flamelet modelling (which is the subject of the next sub-section).

The general idea of the chemistry being dependent on the effects of scalar dissipation rates in regions where the mixture fraction is close to stoichiometric led, eventually, to the now-much-heralded Sandia experiments [20,22] on piloted-jet diffusion flames. Crucial to this experimental concept was the notion that high scalar dissipation rates should not be at the nozzle exit where the flow was subjected to the detailed minutiae of the 'initial' conditions arising from the upstream boundary layers in and outside of the jet tube or subjected to transitional 'coherent'

structures. It would be better to have such high scalar dissipation delayed to 10–20 diameters downstream where the turbulence was more 'fully developed' and amenable to the then, currently available, techniques for turbulence modelling. A large pilot flow with burned products at close to stoichiometric mixture could provide such a streamwise holiday for stoichiometric values of the scalar dissipation that would, eventually, be overcome by the dynamics of the turbulence, and eventually give rise to rates high enough to cause local extinction. An unenclosed jet flow was also amenable to the latest advances in laser-diagnostic measurement. This experimental configuration was initially realised at the University of Sydney by Stårner and Bilger [51]. The paper in the Munich 1986 Symposium [52] involving probe measurements was accorded the 1988 Silver Medal of the Combustion Institute: the judges were perhaps influenced by the oral presentation by Assaad Masri that included later measurements by Rayleigh/ Raman scattering made at Sandia with Bob Dibble, and eventually published in Combustion and Flame [53,54]. These laser-diagnostic measurements were, indeed, the first of a long series of collaborations between Sydney University and Sandia [20,55–57], initially with Bob Dibble and then with Rob Barlow, that has led to the superb experimental databases that are the current benchmark for turbulent non-premixed combustion modelling [20]. Recognition is needed for the vision of Sandia administrators, Dan Hartley, Peter Mattern, and Bill McLean, who have been strong supporters of this international collaboration and the value of this experimental endeavour. The TNF collaboration now includes many groups around the world. The database now comprises excellent measurements for all the major species, temperature, and for CO, OH, and NO for a wide range of jet and bluff-body flames [20]. The high quality of the data has put the onus onto the modellers to refine their models and onto theoreticians to develop new paradigms.

2.2. The laminar flamelet paradigm

A new paradigm appeared in the early 1980s when Peters showed [58] that the species conservation equation can, locally and instantaneously, be transformed into the so-called stationary laminar flamelet (SLFM) equation by assuming terms involving transients and those involving gradients parallel to the instantaneous surface of the mixture fraction to be small. The underlying concept is that flame reaction zones are thin, and their structure is essentially the same as in laminar flames subjected to the same scalar dissipation. Pre-calculated libraries of quantities such as composition, temperature, and reaction rates, as functions of mixture fraction and scalar dissipation, are used to obtain closure for the chemistry–turbulence interactions. Detailed chemical reaction mechanisms and molecular diffusion processes can be included in these laminar flame calculations. A presumed joint PDF of mixture fraction and scalar dissipation allows mean values to be predicted [50], in a manner analogous to that introduced for mixing-controlled combustion. However, the flamelets are relatively insensitive to variations in scalar dissipation, and adequate predictions are sometimes obtained by assuming a constant mean value.

This innovation has been taken up widely by modellers because it provides a simple and easy-to-implement physical picture of the turbulent flame structure. Within its range of validity, it is an attractively simple way to include effects of complex chemistry in turbulent flame calculations. An early success [50] was the improvement in matching experimental CO concentrations, in comparison with calculations assuming chemical equilibrium. However, strong arguments have also been put forward [59] against flamelet models. These include the effects of variations in scalar dissipation through flamelets, and the influence of neglected advection terms in the transport equations. It is clear that the SLFM paradigm cannot remain valid in the presence of local extinction and re-ignition when unsteady and flame edge effects [60] must become important. Nevertheless, the whole question of the range of validity of flamelet models remains controversial. It cannot be denied that the laminar flamelet paradigm can provide an accurate description for sufficiently large turbulence scales and low turbulence intensities for combustion chemistry that is close to irreversible. It is equally obvious that there is also a regime of intense turbulence, whose length and time scales are small relative to those of a laminar flame, for which laminar flamelet models will not be appropriate. But the *range of validity* of these models is not yet agreed.

More recently, the SLFM has been modified to include effects of transients [61,62] and to incorporate a Lagrangian viewpoint [63,64], with associated treatment of the strong fluctuations that can occur in scalar dissipation, and to accommodate the effects of the advection terms parallel to the surfaces of constant mixture fraction. These changes can be helpful, particularly for extinction and re-ignition processes, and for kinetically slow species such as nitric oxide, carbon monoxide or soot. Large eddy simulation incorporating such a viewpoint has been successful [26]. New interpretations of the flamelet paradigm continue to appear. For example, Vervisch et al. [65] combines a flamelet-generated chemistry tabulation with a simplified Conditional Moment Closure model (see Section 2.5) in which some conditional moments are related to the flame-tabulated chemistry.

Another factor that affects the flamelet paradigm is the competition between the broadening of flamelets and their extinction, both of which are governed by the local Damköhler number and the level of mixture fraction fluctuations [11]. For the particular conditions of very high turbulence levels reported by Ratner et al. [66], the CH PLIF images indicate that the flamelets "extinguish before they broaden." When the velocity fluctuations exceeded 10 m/s, "shredded" flamelets were observed, but each CH reaction layer remained about as thin as a layer in a laminar flame (0.5–1 mm). This finding implies that, for these specific conditions, the Damköhler number required to extinguish the flamelet exceeds that required to enter a distributed reaction regime. However, most experiments to date have been conducted using room temperature reactants and relatively low gas velocities, which favor flamelet formation. Additional work is needed at elevated inlet temperatures and large mixing rates to measure the boundaries between flamelet and non-flamelet behavior.

Despite these developments the applicability of flamelet models to specific circumstances remains controversial. The TNF Workshop has adopted bluff-body experiments to test Lagrangian flamelet modelling, as it is not clear how widely applicable this will be, for example, in a recirculation zone, unless an LES approach is used.

2.3. The PDF approach

The mixing-controlled and stationary laminar flamelet models lead to the prediction that the instantaneous composition at a point in a turbulent flame is uniquely determined by just one or two variables, mixture fraction (for fast-chemistry), and mixture fraction and scalar dissipation (for the steady flamelet model). In the 1980s, single-shot Rayleigh/Raman data capable of testing these predictions became available and were presented in the form of scatter plots [53]. For hydrocarbon flames, the degree of scatter observed, and especially the occurrence of compositions outside those realised in steady laminar flames, indicated the need for a more general modelling approach [67]. This was provided by PDF methods, whose development for turbulent reactive flows started in the mid-1970s [68–70].

The primary advantages of the PDF approach are that independent turbulent fluctuations of all species can be represented, and that the direct effects of reaction appear in closed form in the PDF equations [4,12,71–73]. While no modelling is required of the reaction term in the PDF equations, a mixing model is needed to account for mixing by molecular diffusion.

The development of Eulerian [74] and Lagrangian [4] Monte Carlo particle methods enabled PDF calculations to be performed of piloted-jet flames using 4-step reduced mechanisms [67]. The resulting calculated scatter plots were, in many respects, in qualitative agreement with the measurements; but improvements in both calculations and measurements were required to draw firmer quantitative conclusions.

The improvements in laser diagnostics led to more accurate measurements of the Sandia flames D, E, and F [22]; and the in situ adaptive tabulation (ISAT) algorithm [75,76] facilitated the use of more detailed chemistry [77]. At the 28th Symposium, quantitatively accurate calculations of these flames were reported [24,25] based on PDF computations involving around 20 species. The PDF calculations of Xu and Pope [23] accurately represent the level of local extinction in these flames as a function of jet velocity and axial distance—a feat as yet unparalleled by other approaches. Predictions in methanol flames [78] are also good, even though the reaction zone is thinner.

In spite of the success of these PDF calculations, there remain significant questions about the physical realism of the mixing models. The primary models used are: IEM [79] or, equivalently, LMSE [68]; the modified Curl model (MC) [80,81]; and the EMST model [82]. These are found to yield substantially different distributions (as revealed by scatter plots) [83], and also to predict extinction at substantially different Damköhler numbers [83,84].

The three major issues with mixing models in PDF methods are as follows. First, there is no explicit coupling between reaction and mixing; although (especially in the EMST model) such coupling is implicit through the shape of the PDF. Second, the IEM and MC models are non-local in composition space: even at high Damköhler number, according to the models, rich and lean mixtures can mix to form unreacted stoichiometric mixtures [85]. The EMST model was developed to overcome this shortcoming. Third, none of the models includes a physically realistic representation of the fluctuations in scalar dissipation. Since our notions of local extinction are based on extreme values of the scalar dissipation—many times the mean value—it remains a puzzle that PDF models (which do not explicitly represent the distribution of scalar dissipation) are capable of calculating local extinction and re-ignition in piloted jet flames. An important goal of research in this area is to delineate the regimes of applicability of the different models, and of course to develop improved models. Some progress in this direction is discussed below in Section 2.5.

PDF methods have also been applied to the more challenging bluff-body stabilised jet flames [86] (including calculations with detailed chemistry [87,88]) and to the swirling bluff-body flames [89]. For these complex recirculating flows, the primary issue is the extent to which the turbulence

modelling in PDF methods is adequate, and when it is necessary to account explicitly for the large-scale turbulent motions (e.g., via unsteady RANS or LES).

While still an active and fruitful area of research, PDF methods have reached the level of maturity that they are available in commercial CFD codes for use both in research (e.g., [90,91]) and in industrial applications. There are still significant areas requiring careful research, however.

2.4. Conditional moment closure

Conditional moment closure (CMC) [92–94] is a modelling viewpoint that has been inspired by the laser-diagnostic measurements in jet diffusion flames [20] and by experiments in reactive mixing layers [95]. It was apparent that flamelet methods would not be applicable in the latter case as the reaction between nitric oxide and ozone was slow and was occurring throughout the mixed fluid. The basic concept is that the fluctuations in temperature and composition that occur in turbulent combustion can be closely cross-linked to the fluctuations in one or two key variables. In non-premixed combustion, mixture fraction is the key variable of interest. In many such flows, the temperature and species mass fractions that exist are found to have values that vary little from others that have the same value of the mixture fraction. The conditional average is defined as the average of all those members of the subset having the same value of the mixture fraction. The known conservation equations for species and enthalpy can be reformulated in terms of such conditional averages. They involve important terms for the conditional average reaction rates.

In CMC, it is necessary to close these conditional average rates of reaction [94]. Fluctuations about the conditional averages are often small, and the conditional average reaction rates are well approximated using the conditional average temperature and species mass fractions. So-called first-order CMC achieves conditional reaction rate closure by neglecting the contributions of conditional variances and co-variances. Many non-premixed problems, including flows with recirculation [96,97] and auto-ignition of sprays [98], have been successfully modelled using first-order CMC.

In flames with significant local extinction and re-ignition, it has been found [99] that such first-order closure is not sufficiently accurate. Second-order closure [94] incorporates the effects of conditional variances and co-variances in the conditional reaction rates. Equations for these conditional variances and co-variances can be formulated from the conservation equations. To keep the number of such equations small, a reduced mechanism can be used for second-order corrections to the reaction rates as done for hydrogen by Kronenburg et al. [100] for improving prediction of the temperature sensitive species NO. For hydrocarbons, this approach has not, so far, been successful. Kim and Huh have, instead, applied second-order corrections to the forward rates of only a few key reactions and achieved excellent results for piloted-jet diffusion flames [27,101].

An alternative to second-order closure is to use double conditioning. Kronenburg [102,103] uses averaging conditional on both mixture fraction and sensible enthalpy. Closure for conditional reaction rates is found to be in excellent agreement with DNS in homogeneous turbulence with multi-step chemistry under conditions having strong local extinction and re-ignition. Full closure will require models for other unclosed terms such as the double conditioned dissipation terms. Provided these challenges can be surmounted, double-conditioning CMC has considerable promise for application to flames and practical combustors.

Recently, it has been found that, to satisfy boundary conditions correctly, it is necessary to use the fully conservative form of the CMC equation [104]. In this form [94], it is the conditional average multiplied by the probability density that appears as the dependent variable for which the solution is being sought. Earlier work used the finite-difference form of the CMC equation that is closely similar to that of the unsteady laminar flamelet equation [64]. In the finite-difference form, problems arise in parts of the flow where the probability density of the mixture fraction is essentially zero for part of its range. The difficulty with boundary conditions is most obvious in such flows as the auto-ignition of a jet in a hot co-flow. Although partially blocked by the turbulent diffusion term including the PDF, computations in regions with unmixed hot co-flow may result in significant advection of heated mixture containing radicals into regions where there should be little progress in the reaction. There is then the problem of deciding where to establish the upstream region of the CMC computation. These problems are readily resolved when using the fully conservative form of the CMC equation [104].

A big advantage with CMC is that it uses conventional turbulence models to predict the flow and mixing fields. For the CMC calculation to be fully consistent with this CFD it is important for the models for conditional velocity and conditional scalar dissipation to be properly modelled. In earlier work, a linear model has been used for the conditional velocity [94], but this is inconsistent with the gradient model commonly used for closure of the turbulent flux of the variance of the mixture fraction. A gradient model for the conditional velocity is fully consistent [105] with conventional CFD modelling. It also leads to improved modelling of the conditional scalar dissi-

pation [105] by integration of the PDF transport equation using a presumed form of the PDF of the mixture fraction [106].

In second-order closure and in double conditioning, it is necessary to model the scalar dissipation fluctuations. The root mean square (rms) of the scalar dissipation divided by the unconditional mean is known to increase with the Reynolds number. Measurement of scalar dissipation requires the spatial resolution to be finer than the Kolmogoroff scale [107]. Measurements being made in flames [108,109] have yet to achieve such fine resolution and may be under-estimating the mean and rms of the scalar dissipation. Double-conditioning closure [102,103] also requires models for the double-conditioned scalar dissipation of mixture fraction, sensible enthalpy, and their cross dissipation [102]. The laser measurements will be of great assistance in developing models for these.

CMC is showing some success in the prediction of lifted diffusion flames [110,111], the classical model problem for partially premixed combustion. A gradient model is being used for the conditional turbulent scalar flux, and this allows stabilisation to occur. Modelling so far uses first-order closure and is not fully coupled to the flow and mixing field, so the results so far are very preliminary but are, nevertheless, encouraging.

In summary, CMC has a very sound basis and is capable of continued refinement. It has already achieved considerable success for a wide range of problems and is providing stimulus for advanced measurements in flames.

2.5. Multiple mapping conditioning

While in the early stages of its development, the approach of multiple mapping conditioning (MMC) [112] holds the promise of unifying the CMC and PDF approaches, and of providing a more soundly based mixing model, stemming from a generalisation of the mapping closure [113,114]. In the first-order CMC closure, all species are assumed to take their mean values, conditioned on mixture fraction. In MMC, in place of mixture fractions, one considers a set of m "reference variables," with m being specified between 1 and the number of species. The reference variables may be of different types, roughly related to mixture fraction, the velocity, the scalar dissipation, or other quantities. The distribution of compositions is then represented by two quantities: the joint PDF of the reference variables (which, by construction, is a known joint normal); and the mean of the species conditioned on the reference variables, for which a consistent modelled transport equation is obtained.

The initial applications of MMC are very encouraging. An analytic solution is obtained [112] for the three-stream mixing problem in isotropic turbulence that is in excellent agreement with DNS data [115]; whereas the existing models (IEM, MC, and EMST) yield a qualitatively incorrect behaviour for this test problem. The effects of fluctuations in the scalar dissipation can be incorporated [116], and the resulting model is consistent with CMC and SLFM in the appropriate limits.

Related to the concepts in MMC is the following fundamental question [103]: in the multi-dimensional composition space, what is the shape and dimensionality of the region accessed in turbulent combustion? Is it one- or two-dimensional as implied by CMC and SLFM? Or does it have the full dimension of the space as allowed by PDF methods? Theoretical arguments [117] suggest that the dimensionality is indeed large, but that compositions lie close to a low-dimensional manifold, as assumed in MMC. Very accurate simultaneous measurements of all species are necessary to determine the dimensionality and geometry of the accessed region, and hence DNS is better suited to addressing these questions than is experimental measurement.

Further development and application of the MMC approach to simple flows is needed to explore its potential and to gain experience of its performance. A key issue is the development of an accurate and efficient particle method (or other numerical method) so that MMC can be applied in the computation of turbulent flames.

2.6. Implications for LES

In the modelling approaches discussed above, the primary focus is on the coupling between reaction and molecular mixing, and the associated closure problem faced by statistical approaches. Until recently, these approaches to turbulent combustion have been applied in conjunction with conventional turbulence models (i.e., at the Reynolds-averaged Navier–Stokes [RANS] level). Over the last decade, we have seen the beginning of the application of large-eddy simulation (LES) to turbulent combustion. Does LES represent a new paradigm? Our answer is: yes and no.

With respect to the treatment of the turbulent velocity field, LES emerged in the 1960s as a new paradigm [118]. In place of a purely statistical approach in terms of turbulent viscosity or Reynolds stresses, in LES the large-scale unsteady motions are explicitly represented. This seems to be particularly beneficial for the bluff-body flame and swirling flames in which such large-scale unsteady motions are evident. Furthermore, the inherently unsteady framework of LES makes it more suitable for the study of combustion dynamics and acoustics [119,120].

With respect to the coupling between reaction and molecular mixing, LES is not a new paradigm. This is because these two coupled processes occur, predominantly, at the smallest, unresolved scales. Hence, to a large extent, LES faces the same closure problem as RANS-based approaches, and indeed similar models have been used, e.g., flamelet models [121,122]. The demands on the models are often somewhat reduced, as the amplitudes of the sub-grid fluctuations are less than those of the Reynolds-averaged fluctuations. Also, the resolved fields provide additional information that can be incorporated in the modelling [26]. Because of the substantial computational requirements of LES, there has been a tendency to use quite simple turbulent-combustion submodels; but we are beginning to see the combination of LES with more comprehensive submodels, e.g., CMC [123], PDF [124], and linear eddy modelling (LEM) [125,126].

While impressive LES calculations have been performed of practical combustors [127–130] and results validated against flow-field and temperature data, quantitative testing of the sub-grid models for the chemistry against detailed Rayleigh/Raman data is at an early stage. Hopefully, within this decade a clearer picture will emerge of the relative merits and ranges of applicability of RANS and LES methodologies. Crucial questions are the abilities of the models to predict local extinction and re-ignition, and the need and ability to represent the distribution of mixing rates (e.g., fluctuations in the scalar dissipation rate).

3. Premixed turbulent combustion

The major paradigms considered here are that of Damköhler [131] and what are, essentially, extensions of the Damköhler paradigm. Damköhler's paradigm is here considered to embody the hypothesis that the turbulent flame speed and flame structure can be related to correlations and classifications dependent on a small number of parameters. This is detailed in Section 3.1 together with experimental and DNS evidence for the validity of such a hypothesis under wide ranges of effects of flow geometry, Lewis number, and other factors on turbulent flame speed and flame-front structure. It seems that such simple correlation and classification may not be possible. As with non-premixed turbulent combustion, the way ahead seems to involve modelling of unclosed terms in suitable forms of the known conservation equations. In Sections 3.2, 3.3, and 3.5, we consider such paradigms that involve thin flame-front concepts, including Bray–Moss–Libby modelling, coherent flame and flame surface density modelling, and the level set approach. These paradigms consider the chemistry to be fast and often to be the same as that in stretched laminar flames. Means for directly calculating the effects of turbulence on the chemistry are not yet evident. This is becoming an important shortcoming as significant effects can be expected on prompt NO formation and CO burnout in lean premixed combustors such as those used in modern stationary gas turbines. Addressing such questions seems to be more tractable when turbulent mixing rates at small scales become high compared with chemical rates and the reaction zone becomes thickened—Lewis number effects should then be less significant. Such thickened flame-front paradigms are considered in Section 3.5. Conditional moment closure appears to be a feasible approach at this limit. Such issues, and new ideas on the design of 'watershed' experiments for turbulent premixed combustion, together with new ideas on using 'marker fields' to link the CMC to the mixing are outlined in this section. Finally, some implications for LES are considered in Section 3.6.

3.1. Damköhler's paradigm

The basic paradigm for turbulent premixed combustion is that of Damköhler [131]. It has its origins in the paradigm for laminar premixed flames. In laminar premixed flows, the flame front is thin and propagates normal to itself at a speed relative to that of the unburned mixture that is primarily dependent on the state of the unburned mixture (composition, temperature, and pressure). In turbulent flow, the large scales of the turbulence wrinkle the flame front, so increasing its effective area, and hence the rate of reactant consumption and propagation of the mean front. If the smallest scales of the turbulence are larger than the flame-front thickness, it is hypothesised that the instantaneous front will propagate locally at the laminar burning velocity. The turbulent flame will then propagate at a speed equal to the laminar burning velocity multiplied by the ratio of the wrinkled instantaneous front area to the projected area. Small-scale turbulence, smaller than the laminar flame-front thickness, is hypothesised to increase the propagation speed of the instantaneous flame–front by a factor equal to the ratio of the effective turbulent diffusivity of the small scales to the molecular diffusivity raised to the one-half power. Here, the paradigm is considered to embody the hypothesis that the turbulent flame speed and flame structure can be related to correlations and classifications dependent on a small number of parameters.

These concepts have been applied, mainly, to flow situations, such as flames propagating in spark-ignition (SI) engines and flames stabilised on Bunsen-type burners, where the turbulence present in the unburned mixture is significant. (This is in contrast to jet diffusion flames in non-premixed combustion where the turbulence is generated by shear in the flow. We will later return to

premixed turbulent flames where the turbulence is dominated by generation from gradients.) Of major interest is the prediction of turbulent flame speed. This determines flame travel times in spark-ignition (SI) engines and the length of flames on Bunsen-type burners. The Damköhler paradigm suggested that, in problems of practical interest, the degree of flame wrinkling would be the dominant effect and that this should be largely a kinematic effect dependent on the ratio of a turbulent velocity scale to the laminar burning velocity. The paradigm became focused on an ideal turbulent flame that was one-dimensional in the mean and was statistically stationary in a reference frame moving with the turbulent flame 'brush.' As in laminar flows, it was assumed that angled flames could be treated by consideration of propagation in the direction normal to the mean front.

Efforts have been made over 60 years (and still continue) to correlate experimental data on turbulent flame speed in terms of this paradigm and extensions of it. Extensions include dependence on other parameters such as the turbulence length scale and Reynolds number, the laminar flame thickness and volumetric expansion ratio, and the effects of non-unity Lewis number. Many authors have claimed success. Their correlations are limited, however, to a very limited number of flow configurations and their correlation coefficients are, even then, unsatisfactorily low. Results seem to be particularly sensitive to the flow configuration.

Some advances have evolved from a series of workshops organised by R.K. Cheng and F.C. Gouldin in association with the International Combustion Symposia. The workshops have identified several approaches to overcome the difficulties that are specific to turbulent premixed flames, which include the determination of "canonical" flame geometries to use for model validation, and an unambiguous definition of the turbulent burning velocity. The workshop has led to agreement that premixed turbulent flames should be separated into the "Oblique" category (i.e., rod stabilised V-flames), the "Envelope" category (i.e., Bunsen flames that form an envelope about all of the reactants), and the "Unattached" category, which includes low-swirl flat flames and counter-flow flames that do not attach to burner hardware. Each category is associated with different boundary conditions and has a different flame wrinkling process. For example, extensive merging of flamelets occurs near the tip of a Bunsen flame, but this degree of merging does not occur in a counter-flow geometry. Therefore, it is recommended that any scaling relation that is determined (from computations or experiments) for one category should be identified only with that category and not applied to all categories.

It is noted that none of these flow configurations meet the ideal paradigm of one-dimensionality in the mean and some are also not statistically stationary in a framework moving with the flame brush. Insight into the limitations on application of the ideal paradigm in turbulent flows can be gained from considering its mother paradigm—that for premixed laminar flames. Ideal behaviour there is not exhibited where a characteristic length scale of the flow—flame-ball diameter or distance from a wall—is of the same order of magnitude as the laminar flame thickness or strain rate in the flow is comparable with the inverse of the convective time scale of passage through the laminar flame. Furthermore, initially planar flames will often become highly distorted due to thermo-diffusive instabilities. Accordingly, we should expect that turbulent flame speeds should strongly depend on similar considerations. Flames stabilised at fixed locations (such as Bunsen-burner types, V-flames, etc.) should be sensitive to convective time scales comparable to those for passage through a normal turbulent flame brush. Low swirl flames, counter-flow flames, and stagnation flames have mean strain rates that are often comparable with the mean convective time through the turbulent flame brush. There has been much evidence that the external pressure gradients that occur in such flows affect the turbulent burning velocity to a significant extent. This is probably because the conditional momentum balance for burned and unburned gases is crucial in determining the conditional velocities, which are closely related with the turbulent burning velocity.

To properly define the turbulent burning velocity, it has been proposed [132] that one should use the consumption speed rather than the displacement speed that has been reported in the past. Consumption speeds are determined by defining a control volume (which can be a local control volume [132] or a global control volume [133]) and computing or measuring the mass flowrate of reactant that crosses all boundaries, including the mass flowrate associated with density–velocity correlations. Displacement speed is defined as the velocity of the wave (which is the velocity of an iso-surface of the mean reactedness) with respect to the gas velocity ahead of the wave, in the direction normal to the wave. It is difficult to exactly define where the gas velocity ahead of the wave should be determined, and this leads to large uncertainties. Thus, the use of a displacement speed is not recommended.

If turbulent flame speeds are not easily correlated by such a simple paradigm as that of Damköhler, perhaps the *structure* of the turbulent flame could be so captured. So-called 'Borghi diagrams' have proliferated [134,64] in which the flame-front structure is mapped onto a two-dimensional plane with axes of relative velocity scales versus relative length scales, turbulent to

laminar (or of Damköhler number versus Reynolds number). Such diagrams could, perhaps, also be applicable in flows with gradient-generated turbulent mixing where the concept of a turbulent flame speed is less meaningful. Regions identified on these diagrams are given names such as "wrinkled laminar flamelets," "corrugated flamelets," "distributed reaction zone," "thin reaction zone," "quenched reaction zone," etc. This is often done on the basis of scaling arguments, and on experimental observations [135–137] and DNS [138] for two-dimensional laminar flame–vortex interactions. Two-dimensional turbulence lacks the vortex stretching phenomena present in three-dimensional turbulence. Vortex stretching is likely to be important as it is a means by which the turbulence can be kept at high intensity in the presence of large dilatation and an order of magnitude increase in kinematic viscosity. DNS for three-dimensional turbulence with realistic chemistry and turbulence Reynolds numbers is still beyond present capabilities although some results for special geometries are becoming available [139]. Experimental measurements in fully turbulent flames have been limited by spatial resolution requirements that are more severe than in non-premixed turbulent flames. Credible experimental results that have been obtained so far do not present clear confirmation that flame structure can be correlated on the basis of a few parameters such as envisaged by Borghi diagrams.

Several experimental investigations using planar Rayleigh scattering have been made with the object of determining the limit of validity of the "wrinkled laminar flamelet" regime. Recent work by Shepherd, et al. [140] using the low-swirl stabilised burner finds no effect of high turbulence intensity on the thickness of the preheat zone of the instantaneous flame front. This contradicts the findings of O'Young and Bilger [141] made with a two-sheet technique in a very similar flame, and Chen and Bilger in Bunsen-type flames [142,143] and stagnation-plate stabilised flames [144] where progress variable gradients are found to be decreased by high turbulence levels. Soika et al. [145] find flame-front thinning at high turbulence levels for very lean flames. All of these results discussed so far are for hydrocarbon flames, mainly methane. For lean hydrogen flames, Chen and Bilger [146] find that flame fronts remain thin up to high turbulence levels, but local extinction occurs at the trailing cusps. Lewis number effects are clearly important. Planar laser-induced fluorescence of OH [142,143] and CH [133,147] radicals shows little effect of turbulence intensity on gradients of these species in measurements made to date. This has been taken as evidence for the "thin reaction zones" regime of the flame-front structure proposed by Peters [64]. Peters' proposal seems to be founded on an estimate of the fuel-consumption layer thickness in a premixed laminar flame being around 0.018 mm [64] for a stoichiometric methane flame at 1 atm and 300 K. This estimate is an order of magnitude smaller than the thickness of the flame derived from its maximum temperature gradient. It is also, however, an order of magnitude smaller than the fuel reaction layer thickness obtained in full flame calculations, and so the Peters' structure is clearly questionable. Further investigation of the effect of high turbulence on flame structure is needed to clarify these issues.

There also appears to be close coupling between the overall flow field and the detailed structure of the instantaneous flame front. This is exemplified by the findings of Swaminathan et al. [148]. They show, analytically, that the scalar turbulent flux (a macro-scale quantity) in statistically one-dimensional flames is inherently linked to the structure of the instantaneous flame front—very much important at the micro-scale level. This has been confirmed by reference to DNS [148] that are one–dimensional in the mean. Flow divergence present in Bunsen and stagnation plate flows introduces other terms in the analysis, and these flames do not show such behaviour [149]. Whatever the explanation for these effects, it is apparent that the overall flow field can have marked effects on the instantaneous flame-front structure.

It appears that the Damköhler paradigm may not lead to simple correlations for turbulent flame speeds or for classification of the structure of the instantaneous flame front. In the much more tractable case of turbulent non-premixed combustion, we have seen that efforts to correlate the coupled turbulence–chemistry interaction effects on minor species, such as nitric oxide, using appropriate non-dimensional parameters have not been successful. Too many phenomena are involved. In that case, a strategy that has had some success has been to adopt paradigms that are not based on flow-geometric and dimensional reasoning but rather on concepts that involve modelling-closures for terms that appear in suitable forms (flamelet, PDF, and CMC) of the known conservation equations. Such paradigms have also been explored in turbulent premixed combustion, particularly for flows in which the chemistry is thought to be relatively fast. These approaches have embodied closure assumptions that assume that the instantaneous flame front is thin, and its fine-scale structure is like that in a (stretched) laminar flame. Thickened flame-front paradigms, on the other hand, are for flows in which mixing rates associated with the small scales of the turbulence are high compared with chemical reaction rates. They have received little attention, probably because they have been thought to be of little practical importance. They may, however, be more tractable theoretically.

Laminar flamelet and other thin flame-front models of turbulent premixed combustion are an extension of the Damköhler paradigm for the wrinkled laminar flame mode. The instantaneous flame front is assumed to be thin and is often assumed to behave as a stretched laminar flame. Various means are used to incorporate this concept into appropriate closures of the known conservation equations. In the Bray–Moss–Libby (BML) (Section 3.2) approach, the focus is on making closure of the unconditional moment equations for the progress variable. In coherent flame models and flame surface density models (Section 3.3), equations are formulated for the area of flame front per unit volume. In the level set approach (Section 3.4), moment equations are formulated for a conserved scalar, G, that is zero at the flame surface and propagates at this surface at the laminar flame speed modified for stretch effects.

3.2. BML modelling

Bray–Moss–Libby (BML) models [150–155] invoke simplifying assumptions enabling the thermochemical state of the mixture to be expressed in terms of a single quantity, the progress variable $c(\boldsymbol{x},t)$. This is defined either as a normalised temperature or as a normalised product concentration. Mean quantities can then be written in terms of a PDF $P(c;\boldsymbol{x})$, which is expressed in three parts, representing pockets of unburned reactants and fully burned products, with probabilities $\alpha(\boldsymbol{x})$ and $\beta(\boldsymbol{x})$, respectively, and a partially burned mixture with probability $\gamma(\boldsymbol{x})$. In the fast-chemistry or *thin-flame* combustion regime, in which a turbulent flame brush consists of unburned and fully burned gases separated by thin burning zones, it is found that $\gamma = O(1/D_a) \ll 1$, where D_a is a Damköhler number representing a ratio of turbulence and chemical times. The novel conceptual viewpoint following from this formulation sees all mean quantities as being the sum of three contributions. These are the *conditional means* in reactants and products, weighted by their probabilities, α and β, respectively, together with a burning zone contribution with weight γ. To the leading order in an expansion in powers of γ [151], the mean values of quantities such as velocity or progress variable are simply weighted averages of their conditional means in reactants and products, and are independent of γ. On the other hand, reaction rates and reactant concentration gradients are zero everywhere, except in the thin-flame reaction zones, so their means are proportional to γ.

This perspective has proved valuable in developing new closures for the second-order Favre-mean turbulence equations that form the basis of BML models. These closures identify and describe consequences of heat release in the turbulent flow. In the fast-chemistry limit, with $\gamma \ll 1$, turbulent transport is represented in terms of conditional means and variances in reactants and products, allowing processes of flame-generated turbulence [153] and counter-gradient scalar transport [152] to be described. Mean dissipation terms are partitioned into their three conditional contributions, proportional to α, β, and γ, respectively [156]. Thus, for example, the viscous dissipation can be described in terms of conditional dissipations in reactants and products together with a contribution from the thin reaction zones (dilatation dissipation), which is proportional to the mean reaction rate. The mean scalar dissipation for fluctuations in progress variable, which is proportional to the mean square gradient of $c(\boldsymbol{x},t)$, has no contributions from reactants and products, and so is proportional to γ. Both analysis [157] and physical arguments [158] then show that the mean reaction rate in the fast-chemistry limit is proportional to the mean scalar dissipation, a finding analogous to Bilger's result [46] for non-premixed combustion. As such, the BML PDF decomposition does not provide an expression for the mean reaction rate nor does it involve any necessary assumption that thin-flame reaction zones consist of laminar flamelets. Both algebraic models [155] and transported flame surface density models [159] have been used.

The BML PDF decomposition paradigm has been experimentally explored and in DNS. Early experiments by Moss [160] in 1980 show good agreement with predictions of flame-generated turbulence [153] and counter-gradient transport [152]. DNS [161] confirms the occurrence of counter-gradient transport at low turbulence intensities and large values of heat release, and shows that a transition to gradient transport takes place at higher turbulence intensities. This transition has also been experimentally observed [162,163]. Experiments in flames stabilised in counter-flows [164] and flows impinging on plates [165–167] provide a large database for flames at relatively low turbulence intensities. These flames have been successfully simulated in a series of papers [168–170] in which systematic use is made of a small parameter characterising the turbulence intensity. It is interesting to note that the turbulence and scalar flux within the flame are brought into excellent agreement with the experiments *only* after new reacting flow models for the pressure gradient covariance terms in the transport equations are introduced [169]. These models [171], which are well supported by DNS [65,171], are based on BML PDF decomposition, and the pressure gradient within the flame brush is found to have an important role. Lindstedt and Vaos [172] include effects that may be important in flows featuring combustion oscillations. The final paper in this series [170] tests various previously published models for the mean heat release rate,

and none are found to provide good agreement with the available experimental data.

It may be seen that BML models concentrate attention on the *fluid mechanics* of premixed turbulent combustion, and considerable progress has been made in this area. A satisfactory description of the mean rate of heat release, applicable in a wide range of circumstances, remains elusive.

3.3. Coherent flamelet and flame surface density models

The coherent flamelet model (CFM) [173,138] is based on the concept that the mean chemical reaction rate per unit volume is the product of two quantities: the reaction rate per unit area of the flamelets and the average flamelet area per unit volume. The first quantity is assumed to be proportional to the laminar burning velocity; the second quantity is the flame surface density. It has been used to simulate turbulent premixed spherical flames [175], V-flames [176], Bunsen flames [177], and flat flames [178,179]. It has also been combined with the KIVA code to design complex internal combustion engine geometries [180]. If conditions favour the existence of flamelets, the reaction rate can be expressed in terms of the flame surface density (FSD); therefore, a balance equation for the flame surface density is solved. The source terms in the FSD equation are based on a model of the average stretch rate, and the destruction terms are based on a model of flamelet merging and extinction. The approach is a physically based one because the exact conservation equation for the FSD can be derived from first principles [174,179,138], and each of the terms consists of quantities that can be directly measured or simulated with DNS. Therefore, a number of fundamental modelling assumptions can be assessed, and not just the final results. It is interesting to note that there are similarities between the FSD and the PDF approaches [174,181], and the mathematical analysis that relates the FSD evolution equation and the PDF transport equation is described in Vervisch et al. [181]. The source term in the FSD equation has been modelled by Cant et al. [179], and by Meneveau and Poinsot [138]; one term accounts for the stretching of the flame front due to the mean flow velocity gradients, and a second term accounts for small-scale stretch rates. A third term accounts for the destruction of flame surface area due to merging of flamelets and extinction. This term is assumed to be proportional to the square of the flame surface density.

An appealing aspect of the coherent flamelet paradigm is the way it rigorously incorporates information from direct numerical simulations as well as from experiments into submodels for stretch rate and extinction. The submodel for the mean stretch rate is based on the Stretch Efficiency Function, which has been determined from many previous measurements and simulations of stretch rates. These values were determined for laminar counterflow conditions, for unsteady flame–vortex interactions [136–138], and for some fully turbulent conditions [133,182]. The Stretch Efficiency submodel is based on the fundamental concept that the flame–vortex interaction is the basic building block of the structure of turbulence–chemistry interactions. It accounts for the way that each eddy stretches the flamelet, increases the area of the reaction zone, and changes the local scalar gradients.

However, this approach requires that some improvements be made to properly model differential diffusion. It has been experimentally shown that the importance of differential diffusion is not limited to laminar or transitional conditions; for high-Reynolds number turbulent premixed flames, it has been shown that the turbulent burning velocities differ by a factor of 2 when all conditions are held constant, except for the Markstein number [183]. Also of importance is the unsteady nature of the stretch rate. It has been shown that premixed [133] and non-premixed [147] turbulent flames experience rapid-changing stretch rates and do not respond to the high-frequency components of the stretch. For this reason, they do not extinguish, even when the average stretch rate exceeds that required for extinction for the steady-state case. The Coherent Flamelet approach attempts to account for the unsteadiness by using unsteady flame-vortex interaction information that is integrated over all eddy sizes in the turbulence energy spectrum [138].

The Coherent Flamelet Model also simulates the loss of flamelet area due to merging and extinction, which can be significant near walls or the tip of a Bunsen flame. An interesting finding is that the flamelet merging submodel successfully explains the well-known non-linear relation between the turbulent burning velocity and the turbulence intensity, which has been extensively studied in the past and has been called the "bending" phenomenon. For sufficiently large turbulence intensities, the rate of destruction of flame surface density is found to exceed the rate of creation, and this tends to reduce the turbulent burning velocity [178].

A subgrid model for Large Eddy Simulations [184,185] has included many of the concepts in the Coherent Flamelet Model, including the Stretch Efficiency Function. However, it becomes increasingly difficult to assess the validity of this approach at the subgrid scale, since measurements of subgrid surface density are not yet available. Furthermore, premixed flames normally do not form subgrid scale wrinkles that are smaller than approximately ten times the laminar flamelet thickness [186]. This is because of the large dilata-

tion velocities of the products that move away from the flame front as the product gases expand. So, it is not expected that the Stretch Efficiency Function that is determined at large scales will be valid at small scales.

3.4. The level set approach

In the level set approach, which has been described in detail by Peters [64], the flame surface is identified by defining a function $G(\mathbf{x},t)$ such that $G(\mathbf{x},t) = G_0$ on the flame, which is usually, but not necessarily, assumed to have negligible thickness. As it is, only the surface $G(\mathbf{x},t) = G_0$ that is of interest, the variation of $G(\mathbf{x},t)$ at other locations is assumed to be able to be chosen for convenience. The question of how such field variation of G is matched to the surface variation at $G = G_0$ seems to be unresolved. Values of $G > G_0$ are taken to represent fully burned gas while $G < G_0$ describes unburned mixture. Kinematic considerations lead to a transport equation for G. When turbulent flow is to be considered, this equation is averaged, and closures are required for the resulting transport equations for the mean and variance of G. This approach is in many ways similar to the flame surface density formulation, which provides the basis for the Coherent Flame Model [138]. The flame surface density can be shown [187] to be proportional to the mean of the modulus of the gradient of $G(\mathbf{x},t)$. Furthermore, in both cases, the reaction zone is considered to have negligible thickness, and the chemical closure problem is replaced by the need to specify the propagation speed of this reacting surface. If the propagation speed is not constant, then its covariances with other variables must also be specified.

Peters [64] describes how these closures are developed and interpreted for the level set approach using a combination of analysis and data from DNS. He considers two sets of circumstances, in the first of which the turbulence cannot penetrate the interior of the laminar flamelet, and the propagation speed is then that of a stretched laminar flame, while the second case is one where turbulence disrupts the preheat zone but the reaction zone remains thin and intact. Recent experimental data [188] illustrate the second of these cases. Finally, a single set of closed mean and variance equations are proposed [64] for application in both these regimes. It is argued [64] that, because the effects of gas expansion through the thin flame are explicitly considered, complications associated with countergradient transport are avoided. The analytical relationship between level set and BML models is set out in the review by Bray and Peters [187].

In his book [64], Peters describes applications of his level set model to the prediction of turbulent burning velocities and to the simulation of a turbulent Bunsen flame stabilised on a slot burner and, in both cases, agreement with available experimental data may be judged as being satisfactory. If we confine our attention to circumstances where laminar flamelet models are applicable, then level set models and flame surface density models (such as the Coherent Flame Model) make the same basic assumptions, and should give similar predictions for RANS closures. In LES, the broad field characteristics of the level set approach may have advantages, even though there may be fundamental problems with the basic assumptions of the model.

3.5. Thickened flame-front paradigms

Although thin flame-front paradigms may be the closest to what is of practical importance, they may not be the most tractable, theoretically. Lewis number effects are likely to be very important in thin flame fronts, but much less so in thickened flame fronts where turbulence within the front decreases these effects. Even when the Lewis numbers of reactant and product species are close to unity, differential-diffusion effects may be important due to the crucial role that molecular hydrogen has in determining radical levels. The work of Chen and Bilger [146] in lean premixed hydrogen flames indicates that chemical time scales can be greatly affected. Similar effects seem likely to be present in hydrocarbon flames. In non-premixed combustion, it seems that differential diffusion effects are reduced at high Reynolds number [189]. There, the appropriate length scale is clearly that of the large eddies. In premixed combustion, the important length scale is probably that of the instantaneous flame front. Accordingly, theoretical development in premixed combustion may be simplified under conditions where the effects of small-scale turbulence thicken the instantaneous flame front, and Lewis number effects on transport within the flame front are reduced. In non-stoichiometric flames, the laminar flame thickness can be greatly increased, particularly for rich mixtures, and thick flame paradigms may be of direct importance.

With Damköhler number, Da, defined as a large-scale turbulence time scale divided by a chemical time scale associated with overall heat release, it is seen that thickened flame fronts are favoured by low Da, but also by high Reynolds number of the turbulence, Re. At high Re, the reaction zone is thickened by the small-scale turbulence, even though Da is not small. The thickness may still be small compared with the integral length scales of the turbulence, and the PDF of the progress variable may still be dominated by spikes at zero and unity, so that BML-type models are still applicable to the fluid mechanics.

In non-premixed turbulent combustion, it is found that there is a fast-chemistry limit under which rates of chemical reaction are determined by fluid mechanics alone [46]. The fast-chemistry limit in premixed turbulent combustion appears, however, to be dependent on chemical time scales. In the previous section on thin flame-front paradigms, candidate closures for the chemical source term involve chemical time scales. Perhaps, this arises from the low turbulence *Re* of the experimental database being addressed.

There appears to have been little experimental work on premixed combustion under high turbulence *Re* conditions. Spalding [190] considers flame spread in a duct for a flame stabilised behind a V-gutter. The data of Wright and Zukoski [191] show little influence of approach velocity and temperature, or of stoichiometry, on the flame spread rate. Spalding finds that an eddy break-up model gives good predictions. This model does not involve chemical time scales. Reaction rates are largely governed by the rate of entrainment arising from the turbulence generated by the strong shear developed in the flow due to heat release. In non-premixed combustion, the Spalding eddy break-up model for mean reaction rate is clearly linked to the fast chemistry paradigm [192]. Bray's result [157] linking the mean reaction rate to the mean scalar dissipation rate is an important step in this regard. There is yet no evidence, however, that the mean rate of dissipation of the progress variable will become independent of chemical time scales at high *Re* and *Da*. The mixing-controlled limit for the mean reaction rate may apply at high Da, but only when the instantaneous reaction zone is thickened by the small-scale turbulence. It may be that flow geometry and flow-induced mean pressure gradients are also important.

Pope and Anand [193] use Monte-Carlo simulation of the PDF transport equation to investigate premixed flames at both the flamelet and distributed reaction zone limits. They use one-step kinetics with unity Lewis numbers so that reaction rates are an immutable function of the progress variable. At the distributed reaction limit, the mixing model is expressed in terms of turbulence scales, alone. They present results for the width of the turbulent flame brush and for the variance of the progress variable fluctuations as a function of the Damköhler number. Presumably, the distributed reaction model could be used to address questions arising for multi-step kinetics including prompt NO formation in lean flames and post-flame reactions in systems with rich mixtures. Recent work by Vicente et al. [194] makes predictions for CO and NO in a turbulent premixed flame behind a bluff-body stabiliser using such methods. Interestingly, they incorporate a distribution of mixing time scales into their mixing model.

The experiments of Magre et al. [195] address premixed combustion under conditions that are not those of fast chemistry. Their Coherent Anti-Stokes Raman (CARS) measurements in a planar mixing layer between hot products and cold premixture show high levels of carbon monoxide. These appear to be much higher than can be explained by flamelet models. Modelling of these data has been addressed by few investigators and remains a challenge to theoreticians and modellers.

Conditional moment closure has been formulated for turbulent premixed combustion [196,94] but little further development has occurred. Martin et al. [197] have made a start with application to an idealised lean premixed gas turbine combustor. This general lack of development for premixed is largely because of efforts being directed to the more tractable application in non-premixed combustion. CMC can predict only the effects of turbulence on species relative to the progress variable. Closure for the progress variable statistics is still required. This could be achieved by coupling CMC with moment closure methods such as PDF methods. At low Damköhler numbers, closure may also be possible using the new concept of a marker field [199].

Marker fields, denoted as *S*, can be generated by two different methods [199]. In modelling and simulation, the marker *S* obeys the usual transport and diffusion equation with a simple source term. In experiments, an inert species such as NO is seeded into the flow in a time-varying manner. Unlike Peters *G* field [64], the *S* field is not locked (by assumption) to the progress variable, *C*, field at the centre of the flame reaction zone. How the *C* field maps onto the *S* field is a matter of considerable interest. The characteristics of the marker field are expected to be close to Gaussian with a monotonically varying mean gradient in a simple, statistically stationary one-dimensional flow. At low Damköhler numbers, a first-order conditional moment closure seems possible leading to plausible results for the turbulent flame speed and for the structure of the turbulent flame. DNS of turbulent premixed flames with the marker field is under study in current research. Experimental implementation will be attempted first on the stagnation plate burner used in earlier studies of turbulent premixed flame structure [144,163], and the extension of this approach to more complex flow geometries and to flames with complex chemistry will be attempted.

The CMC formulation is also leading to new experimental endeavours in premixed turbulent combustion. As in the non-premixed case, a term appears in the premixed CMC equations that involves the product of the conditional scalar dissipation rate for the progress variable times the second derivative of the conditional average mass

fraction of individual species with respect to the progress variable. It seems that this term can give us insight into the effects of flow geometry and of Da on turbulent flame brush width and extinction phenomena:

- For the classical one-dimensional turbulent flame (one-dimensional in the mean), the turbulent flame brush becomes wider than the integral length scale as *Da* is lowered at low *Da*, as has been found by Pope and Anand [193]. Within the flame brush, the PDF of the progress variable is no longer U-shaped and trends to an increasingly narrow distribution around the local mean. Assuming, for the moment, that the chemical structure of the flame as a function of the progress variable is not greatly affected, the decrease in *Da* is largely due to decrease in the turbulence time scale. If the unconditional scalar dissipation is now controlled by turbulent mixing laws, it will remain approximately constant, due to reduction in both the progress variable variance and in the turbulence time scale. The conditional scalar dissipation rate for the progress variable will also remain approximately constant and profiles of conditional average species mass fractions against progress variable (and their second derivatives) will also remain approximately constant, as has been assumed. Decreasing *Da* just broadens the turbulent flame brush, and rates of molecular mixing remain at the level at which the chemistry can cope. No extinction occurs.
- For flames in shear layers, such as the Magre experiment [195] or a high velocity jet of premixture into a co-flow of hot products, and in counterflow and stagnation plate flames, the width of the flame brush is limited by the laws of turbulent mixing. At low *Da*, further decrease in *Da* by increase in velocity leads to no increase in the flame brush width or reduction of the progress variable variance. Unconditional and conditional scalar dissipation rates will increase to a level where the chemical reactions will no longer be able to cope with such high rates of molecular mixing. Local extinction will eventually occur. Full extinction will manifest itself as all species mass fractions are linear functions of the progress variable, including H_2 and other intermediates, and the chain carrying radicals H, OH, and O. The mass fractions of these intermediates and radicals will be limited by their equilibrium values. Partial extinction will involve high levels of intermediates such as H_2 and CO, as found in the Magre experiment [195]. Downstream in these flows the flame brush will broaden, and scalar dissipation rates will drop allowing re-ignition of the flame.

Clearly, flame structure at low Da will strongly depend on the flow geometry.

These ideas have given rise to attempts at the University of Sydney to develop a premixed jet burner that is the analogue of the piloted-jet diffusion flame burner that has proved to be such an important asset in the development of non-premixed combustion modelling. The configuration is, once again, of a small central high-velocity jet, this time of combustible premixture, an annulus of slowly moving radical-rich combustion products and an outer high temperature co-flow. Near the central jet exit, scalar dissipation rates of the progress variable are low in the annulus that is rich in radicals and intermediates. This ensures that combustion of ad-mixed jet fluid occurs. Further downstream, the influence of the annulus flow has gone, mean gradients in progress variable become steep, and scalar dissipation rates soar. Local extinction or partial extinction becomes apparent. Further downstream, mean gradients of the progress variable drop, scalar dissipation rates fall, and robust combustion is resumed. This experiment is still in early stages of development. Further details may be found on the web [198]. It is expected that advanced laser diagnostic line measurements and imaging in such flames will guide development of advanced paradigms and models for finite-rate chemistry effects in turbulent premixed flames.

3.6. Implications for LES

LES appears to be especially promising for the simulation of premixed combustion because it explicitly resolves the large-scale unsteady motions that are known to play a significant role when the reactants are premixed. Premixed flames are sensitive to the burner geometry and to large-scale, unsteady phenomenon such as the existence of an anchoring point, the passage of vortices, flame-wall interactions, and the merging of adjacent flame brush regions, such as those near the tip of a Bunsen flame. These factors are important because they control the large-scale wrinkling of the reaction surface.

LES for premixed has the problem that the flame usually lies within one grid cell, and this creates numerical problems. One approach to sub-grid scale modelling has been to artificially thicken the flame front [184,185], as has, been mentioned already in the sub-section on flame surface density modelling. Other flame surface density approaches include the work of Cant and co-workers [200–202]. Sub-grid models have been proposed, which assume that a sub-grid turbulent burning velocity exists and that this velocity depends on certain fractal properties of the subgrid flame surface area [189]. However, it has not yet been experimentally determined if the sub-grid

flame surface area is fractal, since the flame wrinkling process may not be scale-independent. For example, premixed flames, in general, do not form wrinkles smaller than several times the laminar flame thickness because there is rapid gas expansion at the flame front that prevents the reaction zone from rolling up. Another sub-grid model [203] requires that a differential equation for the sub-grid flame surface density be solved.

DNS is beginning to play a role in the simulation turbulent premixed combustion: recently, a turbulent flat flame was simulated on a 3-D grid (8 mm × 8 mm × 16 mm) with a spatial resolution of 31 μm, which is less than half of the laminar flame thickness [204]. The complex methane-air chemistry mechanism included 20 species and 84 reactions. It is immediately apparent from these results that the effects of Lewis number and flame stretch rates are significant. Although such resolution is as yet insufficient, it can be expected that DNS will play an even more important role in the future, because many practical premixed combustion devices do not operate at excessively large Reynolds numbers. Laboratory experiments and automotive IC engines typically have small dimensions and only a limited time for the flame to wrinkle, so future application of DNS appears promising. For other practical devices, such as the premixed gas turbine engine, the Reynolds numbers are large, so LES should continue to have an inherent advantage.

4. Partially premixed turbulent combustion

In many practical combustion systems, including some types of reciprocating and gas turbine engines, it cannot be assumed that combustion meets the most basic assumption of either classical premixed or non-premixed combustion theories. The reason is that the combustible gases entering localised reaction zones are not fully premixed to a uniform composition, and nor do pure fuel and pure oxidiser enter reaction zones from opposite sides. Combustion is then sometimes described as *partially premixed*. It is helpful to distinguish between different categories of partial premixing: the most important distinction is between non-uniform combustible mixtures in which the range of compositions excludes stoichiometric mixtures and those in which stoichiometric mixtures do occur. (It is noted that some partially premixed flames, such as the Sandia A–F series [22], have a fuel stream that is so fuel rich that they may be treated as non-premixed [20–26]: we will not be concerned further with such flames in this section.)

Situations where the non-uniform combustible mixture is either all fuel-rich or all fuel-lean, and stoichiometric mixtures do not occur, will be referred to as *stratified premixed* flames. In the absence of stoichiometric mixture, local combustion zone structure may be expected to resemble premixed burning because fuel and oxidiser will enter from the same side. However, composition variations will lead to local fluctuations in burning rate, and thus provide an additional mechanism for flame wrinkling. If the composition of the combustible mixture includes stoichiometric as well as fuel-lean and fuel-rich mixtures, then the possibility exists that local burning zone structures may resemble both premixed and non-premixed flames. This more challenging situation will be called *premixed/non-premixed* combustion.

Pollution control strategies often lead designers to favour stratified premixed combustion. However, the requirement to limit combustion instabilities in gas turbine engines, together with other factors, such as the need for rapid mixing rates to eliminate localised fuel-rich and stoichiometric pockets left by the evaporation of liquid fuel droplets, implies that premixed/non-premixed burning is hard to avoid. What laboratory experiments and DNS geometries allow these two different types of partial premixing to be studied? The standard test case for premixed/non-premixed turbulent combustion is the lifted jet flame [205]; here, triple flames and edge flames provide the paradigm shift that appears able [206] to reconcile competing claims of earlier theories to predict the lift-off height, based on fully premixed or strictly non-premixed burning models. Triple flames are found to play a key role in DNS of turbulent autoignition in mixtures containing fuel-rich and fuel-lean pockets [207] but the results may be an artefact of the one-step Arrhenius kinetics that is used. DNS of lifted hydrogen jet flames [208] indicates a far more complex structure involving edge flames, turbulent premixed flames, and floating diffusion flames. Bunsen flames and V flames will involve mixture fraction variations if, as often happens, the combustion zone overlaps with the mixing layer between the combustible gases and the surrounding co-flow or ambient air. The partially premixed burning will be of the stratified premixed type if the premixed flow is fuel-lean, and the surrounding gas is air, whereas a fuel-rich premixed flow will lead to premixed/non-premixed combustion. Other convenient geometries include counterflows [209] and a dump combustor [210] fed with two parallel streams with different compositions. The fuel jet in a vitiated co-flow as studied by Cabra et al. [211] is interesting as it appears to be a lifted flame in the classical flame-propagation sense under low jet velocity and high co-flow temperature conditions, and as a non-premixed auto-ignition flow under high jet velocity, lower co-flow temperature conditions: modelling and experimental investigation [91,198] are continuing.

Current modelling strategies are related to those developed for fully premixed and non-pre-

mixed combustion. For example, flamelet approaches for premixed/non-premixed situations, using both RANS [64,212] and LES [206], involve a combination of premixed and non-premixed flamelets. It is not yet clear to what extent it is necessary to take the local reaction zone structure explicitly into account in, for example, models for the various scalar dissipations or equivalent terms that appear in most models. As we have seen, PDF methods yield excellent predictions of the Sandia non-premixed jet flames, even in cases where significant local extinction is known to occur. Local extinction allows reactants to partially mix before they burn, and the success of the current generation of PDF models implies that their mixing closures provide an adequate description of the local reaction zone structure at least for these particular experiments. For practical combustors, PDF methods have the advantage that they are able to handle many different inflow streams of unrelated temperature and composition. CMC, which also contains no representation of triple flames or edge flames, nevertheless makes adequate predictions of the lift-off height of a lifted jet flame [110,111]. It must be concluded that we do not yet know how much detail of local flame structure is required, and we cannot yet determine the range of applicability of the various modelling approaches for partially premixed turbulent combustion. Recent advanced work on DNS of lifted diffusion flames involving more than 200 million mesh points and multi-step kinetics for the hydrogen fuel [213] promises to yield more insight than can be obtained from advanced laser measurements. It is apparent that such DNS will play an important role in enhancing our capability in modelling turbulent partially premixed combustion.

5. Concluding remarks

Over the 50 years of the Combustion Institute, great progress has been made in our understanding of turbulent combustion, although this understanding remains far from complete. Only partially do we have answers to the following, fundamental questions (applicable to both premixed and non-premixed combustion) which have been asked since the earliest investigations.

1. What different regimes of turbulent combustion can be identified? And where, in a multi-parameter space, are the boundaries between different regimes?
2. In a given regime, what is the instantaneous spatial structure of the flame?
3. Can the local properties of the instantaneous flame be characterised by a few variables?
4. How can a statistical model (applicable in RANS or LES) be constructed based on a characterisation of the instantaneous flame structure (or otherwise)?

For non-premixed combustion, the sequence of modelling paradigms—mixing-controlled, laminar flamelets, CMC, and PDF—seeks to describe successively more challenging regimes, from the flame-sheet regime to flames with substantial local extinction. In the simpler regimes, mixture fraction and scalar dissipation arise naturally in all approaches as the primary quantities determining the local properties. The understanding gained of the connection between scalar dissipation and local extinction led to the piloted jet flames (and other configurations) for the study of more challenging regimes. Here, experiments and DNS are beginning to address questions 2 and 3, above.

For premixed combustion, controversy remains on the existence and nature of some combustion regimes, such as a thin-reaction zone regime and a thickened flame-front regime. It appears that a large number of parameters may be needed to delineate the space for many regimes including parameters describing the flow geometry and method of stabilisation. In the most-studied thin flame-front regime, the behaviour of the instantaneous flame is characterised by the progress variable and the flame stretch but the limitations on the range of validity of such modelling remains controversial. The flame surface density (FSD) provides a statistical framework for describing turbulent premixed flames. But, because the flame surface is not passive (it propagates and is subjected to instabilities), model equations for the FSD involve more uncertainty than its non-premixed counterpart (i.e., the joint PDF of mixture fraction and scalar dissipation). This approach, and the other statistical frameworks described above, may eventually incorporate enough of the complexity in the physics to throw light on the nature and boundary of the various combustion regimes.

Well before the centennial of the Combustion Institute we will, surely, have conquered turbulent combustion in gases and also in multi-phase flows. The increasing power of laser diagnostics and DNS can be applied to thoughtfully designed configurations to refine our understanding of the fundamental questions posed above. The increasing power of computers and algorithms will allow sufficiently comprehensive statistical approaches to accurately describe turbulent combustion processes in the different regimes. We shall then have fulfilled the vision of the Combustion Institute's founders, to have predictive methods based on fundamental principles, which can be applied to the development of improved combustion equipment for the benefit of society.

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