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Observation, analysis and modelling in complex fluid media

# Raison d'être and general formulation of two-point statistical description of turbulent premixed combustion

Alexey A. Burluka

*School of Mechanical Engineering, The University of Leeds, Leeds, LS2 9JT, UK*

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**Abstract**

This work undertakes an analysis of parameters defining the rate of chemical reactions in turbulent premixed combustion with particular emphasis on the strong turbulence case. Experimental data show that not only the laminar flame speed and rms turbulent velocity but also both the details of chemical kinetics and the molecular transport coefficients are important in this case. Determination of the molecular transport rates inside a turbulent flame requires a two-point statistical description. In order to construct such a description the transport equation for the two-point joint velocity and scalars probability density function is considered. It proved possible to achieve a closure of the unclosed terms in this equation, including the micromixing terms, following Ievlev's method. ***To cite this article: A.A. Burluka, C. R. Mecanique 334 (2006).***


**Résumé**

**Raison d'être et formulation générale d'une description statistique en deux-point pour une combustion turbulente pré-mélangée.** Ce travail considère les paramètres définissant les taux de réactions chimiques dans une combustion turbulente pré-mélangée en insistant sur le cas d'une turbulence forte. Les données expérimentales montrent que non seulement la vitesse de flamme laminaire et l'écart type de la vitesse turbulente mais aussi à la fois les détails de la cinétique chimique et les coefficients de transport moléculaire sont impotants dans ce cas. La détermination des taux de transport moléculaire dans une flamme turbulente nécessite une description statistique en deux points. Afin de construire une telle approche on considère l'équation de transport de la fonction densité de probalité conjointe en deux points de la vitesse et des scalaires. Il s'avère qu'il est possible d'obtenir une fermeture de cette équation, incluant les termes de micro-mélange, à partir de la méthode de Ievlev. ***Pour citer cet article : A.A. Burluka, C. R. Mecanique 334 (2006).***


*Keywords:* Turbulence; Turbulent premixed combustion; Molecular transport effects; Small-scale mixing; Two-point probability density functions

*Mots-clés :* Turbulence ; Combustion turbulente prémélangée ; Effet du transport moléculaire ; Mélange à petite échelle ; Fonctions densité de probabilité en deux points

*E-mail address:* a.a.burluka@leeds.ac.uk (A.A. Burluka).

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## 1. Introduction

Conversion of fuel chemical bonds energy into thermal energy of the medium is made by means of turbulent premixed flames in a number of circumstances. Most important, from an applied point of view, instances of premixed combustion occur in spark-ignition engines, certain gas turbine combustion chambers and explosions of concern for fire and hazard control. Generally speaking, flame in a turbulent medium composed of initially mixed reactants is a propagating exothermic wave, however, the structure of this wave and its response to variation of turbulence or reacting medium properties are not yet perfectly understood despite a large number of research papers devoted to the subject. However, one very important step to understanding turbulent premixed combustion has been accomplished with the classification [1,2] of different regimes of turbulent premixed combustion in terms of ratios of turbulence rms velocity $u'$ to the laminar flame speed $u_n$ and turbulent integral length scale $l_t$ to the laminar flame thickness $\delta_n$. Graphical representation of these dependencies has become known as a "Borghi diagram" and its extensive recent discussion can be found in, e.g., [3, Chapter 7]. At the same time, while providing a useful insight into the nature of turbulent combustion, the Borghi diagram does not give an immediate answer about the rate of combustion which is the primary quantity of applied interest.

Two types of approaches have so far been developed for determination of the average heat release rate or chemical reaction rates in premixed turbulent combustion. The first type is based on solution of transport equations for several first moments of the fluctuating velocity and concentration fields, and the reaction rates are found from some a priori assumptions, more or less according to the Borghi diagram, about the structure of the reaction fronts. Among these one can cite the Eddy-Break-Up type models, e.g., [4], which assume that the reactions occur in infinitesimally thin flame fronts, laminar flamelet models, e.g., [5,6] presuming that the local reactions rates equal those in (strained and curved) laminar flames, as well as various models based on the concept of average flame surface, e.g., [7] or related to it quantity of the average scalar dissipation, e.g., [8,9]. This type of model is usually related to only one of combustion regimes identified in the Borghi diagram.

The possibility of describing several combustion regimes within the same model is offered, at least in principle, by the second type of models, commonly referred to as 'statistical' or 'probabilistic'. A model of this type aims at the calculation of statistical properties, such as probability density functions (pdf's), of reacting scalar, such as a species concentration or temperature, fields and the reactions rates are calculated from their 'exact' instantaneous rates (if those are known) taking into account the fluctuation statistics. This type of model was pioneered in 1960 by a then second-year PhD student V.A. Frost [10] who suggested a transport equation for a progress variable pdf. While the original proposal [10] did consider both one and two-point statistics, most subsequent work has been limited to one-point pdf's only. Following the work [11], the Monte Carlo method is commonly employed for solution of the pdf transport equation which is reformulated in terms of equations for properties of a discrete ensemble of statistical particles. Alternatives to pdf's concepts in this class include the Linear-Eddy model [12], reference scalar field model [13,14] and recent attempts [15] to adapt the Conditional Moment Closure for the premixed combustion. As compared with models of the first type which employ presumed formula(ae) for the mean reaction rate(s), the models of the second class are invariably more complicated, heavier in required computational resources and the question then arises as to whether this additional complexity is justified by their wider generality and applicability.

Very recently, two papers of monumental proportions [16,17] (featuring 350 and 571 referenced papers, respectively) presented a comprehensive review of turbulent premixed flame properties and physical agents defining these properties. These reviews also considered to what extent the models of the first group are capable of describing the experimental data on turbulent flame speed and thickness. A number of critical factors, e.g., molecular transport rates or unsteady character of a flame, have been highlighted in the analysis of [16,17] where it was found that the turbulent burning rate strongly correlates with an extremum of the chemical time scale for curved developing laminar flames. This chemical time scale depends not only upon the elementary reaction rates but also on the molecular heat and mass diffusivities.

The probabilistic models capable of incorporating instantaneous rates of elementary chemical reactions have not been considered in [16,17] and this provides the rationale for the present work. Departing from the conclusion of [16, 17] that the molecular transport coefficients are important, this work attempts to clarify what requirements have to be satisfied by a probabilistic model in order to compete successfully against simpler models of the first group. Some experimental data on burning rates are first discussed with a special emphasis on stronger turbulence where, according to the Borghi diagram [2] one can expect a change in the combustion regime. Following this review an attempt is

made to construct a model free of any presumptions about turbulent reactive zones structure and capable of taking into account the molecular diffusivities explicitly. With this aim, the two-point joint velocity-scalars pdf is necessary and a novel closure for its transport equation is developed here following the technique envisaged in [18].

## 2. Main factors determining the rate of turbulent premixed combustion

There exists a large number of variables which may affect the average rate of heat release in a turbulent premixed flame. Some of these variables, such as the laminar flame speed $u_n$ or molecular heat diffusivity $\kappa$ are determined by the mixture composition, pressure and temperature; some, such as turbulence properties, are dependent on flow and flame geometry. A brief summary of how a particular variable can affect the average reaction rates is presented below; the emphasis is put here on changing combustion regime which is usually associated with strong turbulence.

### *2.1. Turbulence parameters*

In any 'practical' combustion chamber, e.g., an internal combustion spark-ignition (IC SI) engine, the turbulence is neither homogeneous nor isotropic, so different velocity components $u_i$ have unequal rms values and nine different integral length scales can be defined at any given point $\underrightarrow{x}$ from correlations $\langle u_i'(\underrightarrow{x}, t)u_j'(\underrightarrow{x} + \underrightarrow{r}, t)\rangle, i, j = 1, 2, 3$. While this may be true in theory, in reality measurements of turbulence inhomogeneity are scarce and most often the turbulence description is given in terms of a single rms velocity $u'$ and either longitudinal or lateral integral length-scale $l_t$ thus presuming local homogeneity and isotropy. Moreover, very often, the spectral properties of turbulence are assumed to be given by Kolmogorov's laws [19], thus assuming a steady velocity fluctuation spectrum. This may not necessarily be true in non-steady flames, e.g., in explosions, however, while there is an experimental evidence [20] that combustion modifies the turbulence spectrum, no assessment of applicability of Kolmogorov's laws to combustion flows is known. As far as this can affect the burning rate, there exists an early experimental evidence [21] that the rate of combustion depends upon the spectral characteristics of the turbulence upstream of the flame *paribus cæteris*. While this observation must not be discarded because of the lack of published follow-up research, a further study with modern techniques is needed to clarify the influence of turbulence spectrum on combustion rate. In the absence of measurements of the effects of turbulence inhomogeneity and spectral characteristics on combustion, the data analysis has to be confined solely to the effects of $u'$ and $l_t$.

The review [16] concluded that dependency of the burning rate $U_t$ upon the integral length-scale $l_t$ cannot be established without ambiguity because of controversy and paucity of available experimental data. It is also appropriate to mention that the remarkable success of the Zimont–Lipatnikov model applied for very diverse flame configurations [16], indicates that the effects of the integral scale are likely to be weak, because this model assumes that $U_t \sim l_t^{1/4}$. Herein the burning rate $U_t$ is defined as the mass rate of the fresh mixture consumption, sometimes referred to as 'the consumption speed'. Use of the mass burning rate rather than the visible flame speed is chosen because the latter is affected by the thermal expansion of the burnt gases differently, for different flame configurations.

For moderate turbulence the same model assumes that $U_t \sim u'^{3/4}$, and this dependency is well supported by the data shown in Fig. 1 which were obtained in high pressure methane-air explosions. It can be seen that the power-law dependency $U_t = Const \cdot u'^{3/4}$ used in the Zimont–Lipatnikov model provides quite a good approximation to these data over the entire range of $u'$ achievable in the experiment, clearly demonstrating applicability of the model advocated in the [16] for these conditions. However, this simple dependency of $U_t$ upon $u'$ is no longer valid when $u'$ is increased beyond a certain value $u'_m$ [24,25]. When $u'$ is increased beyond $u'_m$ the corresponding increase of $U_t$ with $u'$ slows down and, under certain conditions, it may even be negative, see Fig. 2. Finally, the combustion is extinguished when $u'$ exceeds another threshold value $u'_q$ [24,25]. This situation is illustrated on Fig. 2 presenting the burning rates in explosions in the same mixtures as used in Fig. 1 but performed at the atmospheric pressure [23]. One can see that for the mixtures with $\phi = 0.7,\ 0.8$ and $1.0$ $u'_m \approx 1.8,\ 2.5$ and $3.0$ m/s, respectively, while $u'_q \approx 3.0, 5.7$ for the former two mixtures while no quenching was observed for the stoichiometric mixtures [23]. It must be stressed that the assessment of various models in [16,17] has been performed for $u' \leqslant u'_m$ while it has been remarked that the stronger turbulence case with $u' \geqslant u'_m$ cannot, as a general rule, be adequately described with the models analysed therein without invoking additional assumptions.

It should be noted that a general rule for calculating $u'_m$ has not so far been proposed, perhaps partly because all the changes in flame behaviour are gradual rather than sudden. Similarly, much uncertainty exists about the rms velocity

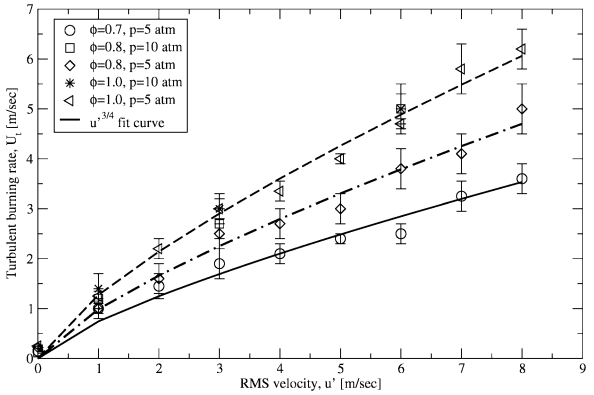


Fig. 1. Mass burning rate in $CH_4$-air spherical explosions for different rms velocities measured at flame radius $r_{fl} = 6$ cm at initial temperature and pressures $T_0 = 358$ K and $p_0 = 5$ and 10 bar. Lines show fitted dependency of the form $U_t = Const \cdot u'^{3/4}$ for explosions at $p_0 = 5$ bar. The experimental points are taken from [22].

Fig. 1. Taux de consommation de masse dans des explosions sphèriques de $CH_4$-air pour plusieurs écarts types de vitesses mesurées au rayon de flamme $r_{fl} = 6$ cm aux température et pressions initiales $T_0 = 358$ K et $p_0 = 5$ et 10 bar. Les courbes montrent une dépendance de la forme $U_t = Const \cdot u'^{3/4}$ pour des explosions à $p_0 = 5$ bar. Les points expérimentaux proviennent de [22].

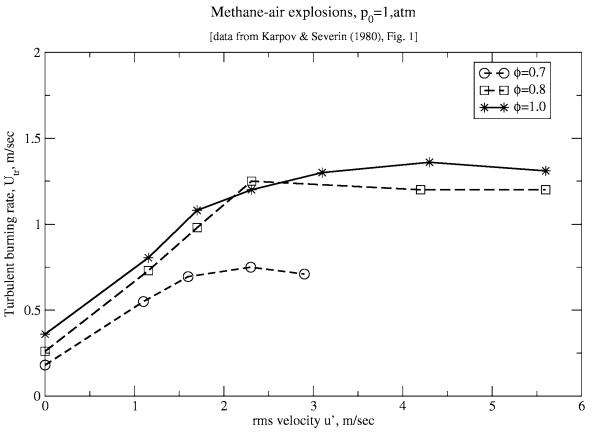


Fig. 2. Mass burning rate in $CH_4$-air spherical explosions for the different rms velocities averaged over the flame radii $r_{fl} = 1.5$–4.5 cm at the initial temperature of $T_0 = 20\,^{\circ}$C and atmospheric pressure. The data are re-plotted from [23].

Fig. 2. Taux de consommation de masse dans des explosions sphèriques de $CH_4$-air écarts types de vitesses moyennées sur les rayons de flamme $r_{fl} = 1.5$–4.5 cm à la température initiale $T_0 = 20\,^{\circ}$C et à la pression atmosphèrique. Les données proviennent de [23].

$u'_q$ at which the flame is extinguished, and proposed expressions for $u'_q$ [26] predicts that $u'_q$ should decrease with the pressure and this trend is contradicted by all available experimental evidence, as can be seen from comparison of the curves for $\phi = 0.7$ in Figs. 1 and 2, see also [20]. The pressure rise decreases the laminar burning velocity $u_n$ and greatly increases $u'_q$ and the flame resistance to quenching by turbulence; the decrease in the mixture equivalence ratio decreases both $u_n$ and $u'_q$, therefore $u'_q$ cannot be expressed solely in terms of $u_n$. Lack of general rules for predicting $u'_m$ and $u'_q$ translates into an uncertainty about the validity domain of the simpler models such as those developed in [16,17], and this provides a justification for further research with the probabilistic models in which both $u'_m$ and $u'_q$ are outcomes rather that external input. In other words, use of more complicated models is justified provided any such model *must*, firstly, describe adequately reaction rates at $u' \leqslant u'_m$ and reproduce the trends analysed in [16,17] and, secondly, yield burning rate $U_t$ dependency on $u'$ up to and including the extinction.

### 2.2. Mixture properties

The combustion regime in the Borghi diagram is characterised in terms of two variables defined by the mixture, namely the laminar flame mass burning rate $u_n$ and the laminar flame thickness $\delta_n$. The latter is usually calculated as $\delta_n = \kappa/u_n$ where $\kappa$ is the molecular heat diffusivity in the cold reactants. Hence, the question about the influence of the mixture properties is whether the burning rate in turbulent flow can be expressed in terms of only these two parameters, especially for strong turbulence, $u'_m \leqslant u' \leqslant u'_q$. An answer to this question, among other things, requires a study of dependency of $u'_m$ and $u'_q$ on mixture properties.

Fig. 3 presents the burning rates measured [27] for diluted lean hydrogen–oxygen–nitrogen mixtures having equal laminar flame speed of $u_n = 17$ cm/s and very close values of the Lewis number (defined as $Le = \kappa/D_{H_2}$ where $D_{H_2}$ is the hydrogen mass diffusivity), but different adiabatic flame temperature. The values of $\kappa$ in the cold reactants is approximately the same for these three mixtures, and so is the laminar flame thickness. While for moderately strong turbulence, $u' \leqslant 3$ m/sec, the burning rates do not differ appreciably, the flame behaviour is very different for stronger turbulence. The $2H_2 + 4.2(O_2 + 3.76N_2)$ mixture burns faster than that of $2H_2 + 4.5(O_2 + 3.76N_2)$ at $u' = 3.5$ m/s but the quenching of these mixtures occurs at approximately the same conditions. Decrease of the oxygen content in the mixture increases the flame adiabatic temperature; while this does not seem to affect the burning rates, the

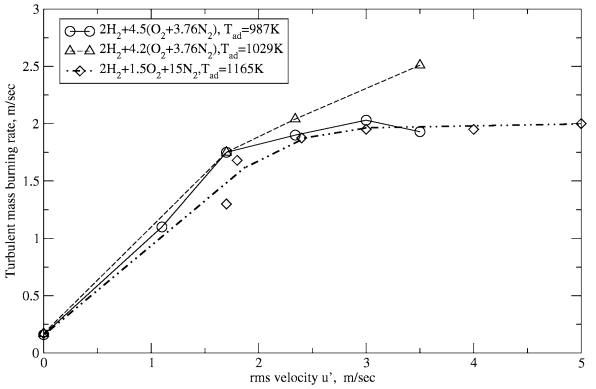


Fig. 3. Mass burning rate for atmospheric explosions of diluted lean hydrogen–oxygen–nitrogen mixtures. The data are taken from [27].

Fig. 3. Taux de consommation de masse pour des explosions atmosphèriques de mélanges pauvres d'hydrogène–oxygène–nitrogène. Les données proviennent de [27].

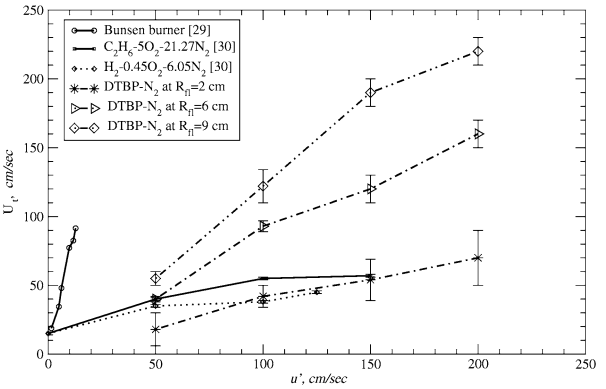


Fig. 4. Comparison of the mass burning rate versus rms turbulence in the DTBP decomposition flame with the literature values reported for the flames with the same $U_n$ and $Le$ (taken from [28]).

Fig. 4. Comparison des taux de consommation de masse en fonction de l'écart type dans la flamme de decomposition thérmique de DTBP avec les données de la literature reportées pour des flammes de mêmes $U_n$ et $Le$ (prises dans [28]).

quenching limit is greatly enhanced. Therefore, the data presented in Fig. 3 indicate that, in addition to $u_n$ and $\delta_n$, or $\kappa$, some additional factors must be considered. For the case of Fig. 3, because different degrees of dilution were used, the rates of reactions involving the nitrogen as a third body may be different, however, it is most likely that the observed differences are caused by variation in the adiabatic flame temperature.

The fact that the influence of mixture composition, temperature and pressure cannot be reduced to effects of the laminar flame speed alone can as well be inferred from a comparison of any curve in Fig. 3 with the data for the methane-air flame at the equivalence ratio $\phi = 0.8$ shown in Fig. 1. This flame has approximately the same laminar flame speed $u_n \approx 16$ cm/s as the mixtures in Fig. 3 but its turbulent burning rates for $u' \geqslant 3$ m/s and resistance to extinction are distinctly different from theirs.

An attempt to explore to which extent the details of the flame chemical kinetics mechanism affect the burning rates was undertaken in [28]. In contrast to commonly employed hydrocarbon and hydrogen oxidation flames which are characterised with multi-step chain-branching chemical kinetics, this work reports a study of di-tert-butyl-peroxide (DTBP) thermal decomposition flame. This DTBP thermal decomposition flame in an inert gas atmosphere is described [28] with a single first-order reaction of which the rate constant is:

$$W = 2 \times 10^{15} \exp\left(-\frac{18282}{T}\right) \left[\frac{1}{\mathrm{s}}\right] \tag{1}$$

Fig. 4 presents comparison of the flame mass burning rates measured in the fan-stirred 'bomb' [28] for this simple kinetics flame with the literature [29,30] values reported for the mixtures having the same values of $u_n$ and $Le$. While the inspection of Fig. 4 shows the effects of unsteady flame development, especially important in comparison between the freely propagating flames [28,30] and Bunsen burner ones [29], it also indicates that the $U_t(u')$ curves for DTBP decomposition flames barely have any saturation effects, that is for this mixture the definition of an exact value for $u'_m$ is rather ambiguous. At the same time, for $u' \leqslant 1$ m/s there is no noticeable difference in the burning rates for all freely propagating flames regardless of their chemistry. This observation indicates that the chemical kinetics mechanism details should be accounted for in determination of the average reaction rates for 'strong', i.e., $u' \geqslant u'_m$, turbulence, while their effects for 'moderate' $u' \leqslant u'_m$ are most likely to be encapsulated in terms of $u_n$, $\kappa$ and $Le$, in full agreement with the analysis of [17].

As a summary of the experimental observations presented above, as well as from the extensive analysis of data performed in [16,17], one can state that the average reaction rates in turbulent premixed combustion depend, in addition to commonly considered turbulence rms velocity $u'$, length-scale $l_t$ and laminar flame speed $u_n$, upon the molecular transport coefficients, such as heat diffusivity $\kappa$ and the Lewis number $Le$. The arguments presented above shows that when $u'$ exceeds a certain value $u'_m$ then, in addition to these factors, the adiabatic flame temperature and the details of chemical kinetics mechanism become important. Because $u_n$ itself is a function of elementary reactions rates and the

molecular transport properties, a model of turbulent premixed combustion should take these into account in order to be applicable across entire range of $u'$ from weak to strong turbulence. For strong turbulence the available experimental data indicate that the dependency of the turbulent burning rate upon the molecular transport coefficients cannot be expressed solely in terms of $u_n$, $\kappa$ and $Le$.

## 3. Statistical model with the explicit account of molecular transport and details of the chemical kinetics

A turbulent combustion model valid across several combustion regimes must be capable of taking into account both the molecular transport and the chemical kinetics in the flame. Molecular transport rate of a variable is defined by the instantaneous values of its gradient, and its description therefore requires knowledge of a two-point statistics of this variable. An additional advantage of the two-point statistical model is that, unlike the majority of existing turbulent combustion models, it is self-contained, in a sense that it does not require any external information about the turbulence length-scales. Below follows an attempt to develop such a model, taking as a departure the method proposed by Ievlev in [18,32].

Let $\hat{A}_n = (a^j,\ j = 1, \ldots n)$ be a set of values of variables describing the reacting flow field: $(\underset{\rightarrow}{u}, \rho, Y_\alpha,\ \alpha = 1, \ldots l)$ at points $\underset{\rightarrow}{x}^k,\ k = 1, 2, \ldots n$. Here $\underset{\rightarrow}{u}$ is the velocity field, $\rho$ is the density and $\underset{\rightarrow}{Y}$ is the concentration vector. Let $P_n(\hat{A}_n)$ be a joint $n$-point probability density function for the set $\hat{A}_n$. Following [18,32], the 'balance', or transport equation for $P_n(\hat{A}_n)$ can be written as:

$$\frac{\partial P_n(\hat{A}_n)}{\partial t} + \hat{u}_i^k \frac{\partial P_n(\hat{A}_n)}{\partial x_i^k} = -\frac{\partial}{\partial \hat{a}_i^k}\langle F_i^k \rangle_{\hat{A}_n} P_n\big(\hat{A}_n\big) \tag{2}$$

where conditional rate of change (acceleration for a velocity component) $\langle F_i^k \rangle_{\hat{A}_n} = \langle \frac{\mathrm{d}a_i}{\mathrm{d}t} \rangle_{\hat{A}_n}$ of variable $a_i^k$ is expressed in terms of pdf $P_{n+1}(\hat{A}_{n+1})$ taken at $n+1$ points, because they are defined by conditions at $n$ points and derivatives at the $k$th point. To obtain $\langle F_i^k \rangle_{\hat{A}_n}$, the usual set of equations of motion (Navier–Stokes), continuity and scalar transport should be used:

$$\big\langle F_i^k \big\rangle_{\hat{A}_n} = \left\langle -\frac{1}{\rho^k} \cdot \frac{\partial p}{\partial x_i^k} + \frac{\partial}{\rho^k \partial x_j^k} \frac{\rho^k \nu \partial u_i^k}{\partial x_j^k} \right\rangle_{\hat{A}_n} \quad \text{for } \hat{a}_i^k = \hat{u}_i^k \tag{3}$$

$$\big\langle F_i^k \big\rangle_{\hat{A}_n} = \left\langle -\frac{\partial u_j^k}{\partial x_j^k} \right\rangle_{\hat{A}_n} \quad \text{for } \hat{a}_i^k = \hat{\rho}^k \tag{4}$$

$$\big\langle F_i^k \big\rangle_{\hat{A}_n} = \left\langle -\frac{\partial}{\rho^k \partial x_l^k} \frac{\rho^k D_j \partial Y_j^k}{\partial x_l^k} \right\rangle_{\hat{A}_n} + W_j\big(\underset{\rightarrow}{Y}^k\big) \quad \text{for } \hat{a}_i^k = \hat{Y}_j^k \tag{5}$$

where $\nu$ is the viscosity, $D_j$ is the mass diffusivity of the $j$th species and $W_j$ is its rate of conversion in chemical reactions. Because $\langle F_i^k \rangle_{\hat{A}_n}$ depend upon the $(n+1)$-point pdf Eqs. (3)–(5) require the so-called closure hypothesis expressing these conditional rates of change in terms of $n$-point pdf's and $\langle F_i^k \rangle_{\hat{A}_{n-1}}$. For the scalar values pdf such closure hypothesis for Eq. (5) are known as the micro- (or small-) scale mixing models. A considerable number of small-scale mixing models have been proposed [31], but they are restricted to one-point pdf's and the effects of molecular diffusivities cannot be, as a matter of principle, described in such models [33].

### 3.1. Closed two-point joint velocity-scalars pdf equation

In 1970, V.M. Ievlev [18,32] extended the derivation of Eq. (2) to the case of joint velocity-scalars pdf and proposed a general method of expressing the $\langle F_i^k \rangle_{\hat{A}_n}$ terms through $n$-point pdf's using exclusively the formal mathematical constraints such as normalisation and reduction properties. This methods allows one to achieve a closed description of a turbulent flow for $n \geqslant 2$ without resorting to any a priori ideas about structure of the turbulent vortices, spectra, etc. However, the proposed method was sufficiently complicated and the only application of Ievlev's closure had so far been made for studying the mixing of one non-reacting scalar in a homogeneous and isotropic turbulence [33], where it was shown that the Ievlev's method predicts correct physical trends in the evolution of an initially bi-modal pdf.

The method [18] is based on an observation that the following properties of reduction, separation and coincidence which should be satisfied by any $P_n(\hat{A}_n)$ can be expressed as a following set of *integral* equations for $\langle F_i^k \rangle_{\hat{A}_n}$:

– Separation:

$$\int \langle F_i^j \rangle_{\hat{A}_n} P_n(\hat{A}_n)\, \mathrm{d}\, \underset{\rightarrow}{a}^k = \langle F_i^j \rangle_{\hat{A}_{n-1}^{(k)}} P_{n-1}\big(\hat{A}_{n-1}^{(k)}\big), \quad j \neq k$$

– Coincidence and reduction:

$$\lim_{\underset{\rightarrow}{x}^k \to \infty,\, \underset{\rightarrow}{x}^j} \langle F_i^m \rangle_{\hat{A}_n} \to \langle F_i^m \rangle_{\hat{A}_{n-1}^{(k)}}$$

Here $\hat{A}_{n-1}^{(k)}$ is the set of $\underset{\rightarrow}{a}$ values in all points except the $k$th one. Henceforth, one may attempt construction of expression for $\langle F_i^k \rangle_{\hat{A}_n}$ as:

$$\langle F_i^k \rangle_{\hat{A}_n} = \sum_j g_i^{k,j}\big(\hat{A}_{n-1}^{(k)}\big) \langle F_i^k \rangle_{\hat{A}_{n-1}^{(j)}} + u_i^j \cdot \frac{\partial \xi_j^k}{\partial x_i^j} \tag{6}$$

where $\xi_j^k$ are arbitrary functions of $A_{n-1}^{(k)}$ and the weight coefficients $g_j$ depend as well on the coordinates of all $n$ points. Eq. (6) is the original Ievlev's formulation, and unfortunately it does not provide an explicit expression for $g_j$ functions and one has to resort to numerical quadratures as was done in [33].

At the same time, for the two-point statistics a remarkable simplification can be obtained if one considers an expression slightly more general than the original Ievlev approximation equation (6), namely:

$$\langle F_i^k \rangle_{\hat{A}_{1,2}} = g_i^{k,1}\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2, \hat{A}_1\big) \langle F_i^k \rangle_{\hat{A}_2} + g_i^{k,2}\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2, \hat{A}_2\big) \langle F_i^k \rangle_{\hat{A}_1} \tag{7}$$

It is easy to see that the generalisation is simply the postulated dependency of $g_i^{k,1}$ on $\hat{A}_1$. In writing Eq. (7) the condition $\xi_j^k = 0$ has been used, and it is trivial to show that this condition corresponds to a constant density case.

The next step is to multiply Eq. (7) by $P_2(\hat{A}_1, \hat{A}_2)$ and integrate with respect to $\mathrm{d}\hat{A}_1$:

$$\langle F_i^k \rangle_{\hat{A}_2} = \big\langle g_i^{k,1} \big\rangle\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2\big) \langle F_i^k \rangle_{\hat{A}_2} + g_i^{k,2}\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2, \hat{A}_2\big) \langle F_i^k \rangle$$

Similarly, integration with respect to $\mathrm{d}\hat{A}_2$ yields:

$$\langle F_i^k \rangle_{\hat{A}_1} = g_i^{k,1}\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2, \hat{A}_1\big) \langle F_i^k \rangle + \big\langle g_i^{k,2} \big\rangle\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2\big) \langle F_i^k \rangle_{\hat{A}_1}$$

Multiplying either of the above equations by $P_2$ and integrating leads to

$$\big\langle g_i^{k,1} \big\rangle\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2\big) + \big\langle g_i^{k,2} \big\rangle\big(\underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2\big) = 1$$

Expressing $g_i^{k,1}$ and $g_i^{k,2}$ from the above three equations and substituting them back into Eq. (7) results in the sought closure model:

$$\langle F_i^k \rangle_{\hat{A}_{1,2}} = \frac{\langle F_i^k \rangle_{\hat{A}_1} \cdot \langle F_i^k \rangle_{\hat{A}_2}}{\langle F_i^k \rangle} \tag{8}$$

It is straightforward to see that this closure indeed satisfies re-numbering, reduction, coincidence and separation properties. Its physical meaning is that the accelerations, or rates of change, averaged with simultaneous conditions at two points can be expressed in terms of product of accelerations averaged with conditions at only one point.

Resulting from this closure Eq. (8) equations are best illustrated on the example of velocity pdf for an incompressible flow. In this case, the pressure gradient can be expressed from solution of a Poisson equation as, e.g., for an infinite unbounded flow, the averaging of which results in a clearly non-local term:

$$\left\langle -\frac{1}{\rho} \frac{\partial p(\underset{\rightarrow}{x})}{\partial x_i} \right\rangle_{\hat{A}_m} = \frac{1}{4\pi} \int \mathrm{d}^3 \underset{\rightarrow}{x}' \cdot \frac{x_i' - x_i}{\| \underset{\rightarrow}{x} - \underset{\rightarrow}{x}' \|^3} \int \mathrm{d}^3\, \underset{\rightarrow}{\hat{u}}'\, \hat{u}_j' \hat{u}_k' \frac{\partial^2 P_2(\underset{\rightarrow}{x}', \underset{\rightarrow}{x}^m, \hat{A}', \hat{A}_m)}{\partial x_j' \partial x_k'}$$

At the same time the small-scale mixing term is local and contains explicitly the molecular viscosity:

$$\left\langle \nu \frac{\partial^2 u_i^k}{\partial x_j^k \partial x_j^k} \right\rangle_{\hat{A}_m} = \nu \int \mathrm{d}^3 \underset{\rightarrow}{\hat{u}}' \hat{u}_i' \lim_{\underset{\rightarrow}{x}' \to \underset{\rightarrow}{x}^k} \frac{\partial^2 P_2(\underset{\rightarrow}{x}', \underset{\rightarrow}{x}^m, \hat{A}', \hat{A}_m)}{\partial x_j' \partial x_j'}$$

Using the reduction property for a two-point pdf $f_2$:

$$\lim_{\underset{\rightarrow}{x}^2 \to \underset{\rightarrow}{x}^1} P_2\left(\underset{\rightarrow}{u}^1, \underset{\rightarrow}{u}^2, \underset{\rightarrow}{x}^1, \underset{\rightarrow}{x}^2, t\right) = \delta\left(\underset{\rightarrow}{u}^1 - \underset{\rightarrow}{u}^2\right) P_1\left(\underset{\rightarrow}{u}^1, \underset{\rightarrow}{x}^1, t\right)$$

the small-scale mixing terms can be further simplified to the sum:

$$-\nu \sum_{k,j} \left( \int \mathrm{d}\hat{u}_j^k \cdot \hat{u}_j^k \int \mathrm{d}A^{(k)} \frac{\partial^2 P_2}{\partial x_l^k \partial x_l^k} \right)^{-1} \left( \int \mathrm{d}\hat{u}_j^k \cdot \hat{u}_j^k \frac{\partial^2 P_2}{\partial x_l^k \partial x_l^k} \right) \frac{\partial}{\partial \hat{u}_j^k} \left( \left( \int \mathrm{d}\hat{u}_j^k \cdot \hat{u}_j^k \frac{\partial^2 P_1(\underset{\rightarrow}{\hat{u}}^k, \underset{\rightarrow}{x}^k, t)}{\partial x_l^k \partial x_l^k} \right) P_2 \right)$$

Similar terms proportional to the molecular diffusivities will arise in the micro-scale mixing description for the scalars within the same framework.

The resulting set of Eqs. (2), (3), (8) form a closed set of non-linear equations, solution of which is, as was shown in [18], the best possible approximation for the two-point statistical properties. At the same time, it is formulated in terms of $15 + 2 \times$ (number of scalars) dimensional functions and because of its high dimensionality the numerical solution may be challenging. For a single reacting scalar problem, its numerical solution with grid discretisation technique would require approximately $10^{35}$ bytes which is way beyond any foreseeable computer capacity. Problematic with alternatives to grid discretisation methods, such as Monte Carlo or discrete particles methods, is the representation of the non-linear terms involving various derivatives in Eq. (2). Therefore an obvious direction of study consists in exploring whether a solution to Eqs. (2), (3), (8) can be found as a truncated series expansion in some suitable function set.

## 4. Conclusions

Reviews [16,17] have demonstrated that the account of molecular transport properties is essential for an accurate calculation of the rate of combustion. Those arguments are supplemented here with the observation that the details of chemical kinetics also become important for stronger turbulence, in addition to all the factors reviewed in [16,17]. The present work explores a possible framework for including both molecular transport properties and details of chemical kinetics into a mathematical model of a reactive turbulent flow. This required the consideration of the two-point fluctuation statistics and this work presents a closure for the two-point joint velocity-scalars pdf transport equations.

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