Lagrangian properties of turbulent diffusion with passive chemical reaction in the framework of the premixed combustion theory

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In this paper, we analyze two effects caused by the Lagrangian nature of turbulent transfer which are usually ignored in the theory of turbulent premixed combustion. These effects are (i) the nonequilibrium behavior of the turbulent diffusion coefficient, which is important for modeling the initial stage of combustion (for example, in the spark ignition engine), and (ii) the existence of a traveling front of turbulent diffusion with finite speed, which controls the velocity of the steady state flame in strong turbulence. However, in order to derive simple and exact results, the hydrodynamical and the combustion subproblems are stated to be independent by assuming a constant density so that a passive chemical reaction is actually considered. First, we derive a parabolic diffusion equation with both diffusion and chemical source terms expressed by Lagrangian characteristics of turbulence. We show that, in general, the diffusivity of product particles is not zero in the moment of their generation by chemical transformation and this result is important for combustion theories that relate the formation of the initial flame with the development of the diffusion coefficient. Afterward, a hyperbolic diffusion equation based on hydrodynamics is derived with turbulent diffusion front velocity $\mathcal{U}_f = \langle u'^2 \rangle^{1/2}$, where $\langle u'^2 \rangle^{1/2}$ is the root mean square of turbulent velocity fluctuations, and we analyze the relationships between U_f and the speed of the steady state premixed flame \mathcal{U}_t^{ss} . In particular, for the flamelet combustion mechanism, we obtain $\mathcal{U}_t^{ss} = (\mathcal{U}_f^2 + S_L^2)^{1/2}$, where S_L is the normal laminar flame speed. This result shows that, in moderate turbulence $(\langle u'^2 \rangle^{1/2} \sim S_L)$, the usually assumed relation $\mathcal{U}_t^{ss} = \mathcal{U}_f + S_L$ is not consistent with an accurate statistical analysis and more when $\mathcal{U}_f \simeq S_L$ gives a percent error around 40%, which cannot be neglected in applications. In strong turbulence case $(\langle u'^2 \rangle^{1/2} \gg S_L)$, the value of the flame speed is very close to that of the diffusion front velocity. © 2011 American Institute of Physics. [doi:10.1063/1.3562842]

I. INTRODUCTION

The classical paradigm of diffusion transfer modeling is based on parabolic equations with turbulent diffusion coefficient D_t expressed in terms of Eulerian turbulent characteristics by the formula $D_t = \text{const} \langle u'^2 \rangle^{1/2} \mathcal{L}$, where $\langle u'^2 \rangle^{1/2}$ and \mathcal{L} are the root mean square of turbulent velocity fluctuation and the Eulerian integral lengthscale of turbulence, respectively. In the case of homogeneous and stationary turbulence, D_t is constant. However, such Eulerian approach misses qualitative effects that are caused by the Lagrangian nature of turbulent transfer. In this paper, we analyze two of these effects which are important for the theory of the turbulent premixed combustion: (i) the time-dependence of the diffusion coefficient due to finite value of the Lagrangian timescale of turbulence and (ii) the existence of the traveling turbulent diffusion front with finite velocity that is the geometrical locus of those Lagrangian particles that are the most advanced in space.

Although both these effects are discussed in the scientific literature devoted to turbulence mechanics,¹ they are usually ignored in the engineering handbooks,² and this neglect is justified in many practical applications because the relaxation time of the diffusion coefficient is small with respect to the reference time and, contemporaneously, the front speed is large with respect to the reference velocity of the process. Nevertheless, these effects play an important role in turbulent premixed combustion not only under the theoretical point of view but also in engineering applications. In fact, an accurate description of the nonequilibrium behavior of the turbulent diffusion coefficient, i.e., its growing in time, is important during the initial transient stage of the combustion process as, for example, in the spark ignition (SI) engine, the laboratory bombs with spark ignition, or the initial instants of the Bunsen flame, while the velocity of the diffusion front is important because it is one of the main parameters that control the speed of the steady state premixed flame at large elapsed times.

The problem of turbulent combustion actually consists on the coupling of the hydrodynamics subproblem with the combustion subproblem. The former describes the state and the dynamics of the hydrodynamic fields and the latter describes the transfer and the chemical processes that concern the species dispersed by the velocity field. These subproblems are connected through the common pressure and velocity fields. Combustion influences the average flow and the turbulent fluctuations by means of the variation of the density field. This means that in the case study of constant density, the combustion does not influence the turbulence. This approach is theoretically useful because it permits to split the two subproblems and then to consider the combustion pro-

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cess alone. So, if the parameters of turbulence are assumed known, it is possible to analyze separately the combustion subproblem in order to perform a theoretical analysis of combustion models to prove their theoretical foundation.

In this spirit, here we assume a constant density, so we actually consider turbulent diffusion with passive chemical reaction, with the aim to deduce exact kinematical results that can confirm some assumptions generally postulated in literature models for turbulent premixed combustion.^{3,4} The turbulent velocity field is assumed homogeneous, isotropic, and stationary, and, without loss of generality, the mean velocity is set equal to zero.

We tell here in advance that the choice to consider a constant density has repercussions on the so called countergradient turbulent flux, which is experimentally observed in premixed flames. In fact, since such countergradient transport phenomenon is generated by the fact that the pressure gradient accelerates the light products more than the heavy reactants,^{5,6} in the case of constant density the pressure gradient acts equally on both products and reactants and then there is no countergradient transport.

In the first part of the paper, we derive a parabolic equation with time-dependent diffusion coefficient and chemical source where both are expressed in terms of Lagrangian characteristics of turbulence. We show that, in this equation, the turbulent diffusion coefficient does not depend on the random appearance of product particles. This result is not trivial, as at first glance it appears, because, since new species result from chemical reaction, a zero diffusivity at the initial instant for products dispersion could be stated from a formal application of Taylor theory of turbulent dispersion, as it is sometimes discussed. However, our analysis proves that it is not so. In fact, since the product particles are randomly generated by the chemical reaction, the Taylor formula for particle dispersion variance cannot be straightforwardly applied. Then we discuss here the application of Taylor theory also to the general cases with random initial conditions. The derived result is important for combustion models where the formation of the developed turbulent flame during the initial stage is connected with the transient behavior of the Lagrangian turbulent diffusion coefficient.^{3,4} Furthermore, from the purely Lagrangian formulation of the process, we obtain that the spatial structure of the chemical source term is determined by the motion of mixture particles and that it results to be dependent on particle diffusion properties.

In the second part of the paper, following Refs. 4 and 8, we derive a hyperbolic equation with time-dependent coefficients on the basis of exact unclosed hydrodynamical equations. This hyperbolic transfer equation has the striking property that it does not contain parameters to be empirically determined. For an arbitrary Lagrangian velocity fluctuation autocorrelation function, this equation permits to have dispersion properties in agreement with the Taylor theory⁷ and gives a diffusion front velocity $U_f = \langle u'^2 \rangle^{1/2}$. In particular, when an exponential Lagrangian autocorrelation function is considered, the coefficients of the equation turn out to be constant.^{4,8} Moreover, when a chemical source is accounted for, the relationships between the diffusion front velocity U_f

and the speed of the steady state flame U_t^{ss} are analyzed. Considering the flamelet combustion mechanism, generally named BML model,⁹ it emerges that $U_t^{ss} = (U_f^2 + S_L^2)^{1/2}$, where S_L is the normal laminar flame speed. For strong turbulence $(\langle u'^2 \rangle^{1/2} \gg S_L)$, this result confirms and better specifies the classical estimation ¹⁰ $U_t^{ss} \sim \langle u'^2 \rangle^{1/2}$, which is based on intuitive considerations and from which it follows that the combustion front speed does not depend on chemistry. For moderate turbulence $(\langle u'^2 \rangle^{1/2} - S_L)$, this result shows that the usual estimation $U_t^{ss} \sim \langle u'^2 \rangle^{1/2} + S_L$ is not consistent with an accurate statistical analysis.

Depending on the investigation issue, we switch between three-dimensional and one-dimensional framework. Boldface notation indicates three-dimensional quantities, i.e., \mathbf{x} , otherwise one-dimensional, i.e., x.

The paper is organized as follows. To better highlight the aims of the later mathematical analysis, in Sec. II, the scientific problems that motivate the research are stated; in Sec. III, the Taylor theory is briefly reminded and its application in reactive mixture is discussed; in Sec. IV, a transport equation with finite front velocity is derived and the role of the front velocity in the premixed combustion process is analyzed; finally, in Sec. V, the conclusions are given.

II. DEFINITION OF THE PROBLEMS AND ASSUMPTIONS

The two Lagrangian characteristics of turbulent dispersion which are analyzed in the present paper are relevant, in the theory of turbulent premixed combustion, for the following two specific problems.

A. Product generation and turbulent diffusion coefficient

The classical paradigm of turbulent premixed combustion is based on a parabolic diffusion equation with chemical source $W(\mathbf{x}, t)$ for the average value *C* of the instantaneous progress variable *c* (*c*=0 in unburned mixture and *c*=1 in burned one). In the simplest case of homogeneous, isotropic and stationary turbulence, with constant density and zero mean velocity, the flame propagation is described by the following differential equation:

$$\frac{\partial C}{\partial t} = D_t \nabla^2 C + \mathcal{W}(\mathbf{x}, t).$$
(1)

As defined in Sec. I, the turbulent diffusion coefficient D_t is constant and expressed in terms of Eulerian characteristics of turbulence by the formula $D_t = \text{const} \langle u'^2 \rangle^{1/2} \mathcal{L}$. The study and prediction of the mean chemical source term \mathcal{W} is one of the main targets of combustion theory.

The developed analysis is intended for the flamelet combustion mechanism, when the transformation of reactants in products takes place in a strongly wrinkled surface that divides reactants and products. Every element of this surface (a flamelet sheet) travels in the unburned mixture with constant speed that is generally assumed to be equal to the velocity of the normal laminar flame S_L . This sheet is considered to be a laminar flame with constant speed S_L and zero laminar flame thickness δ_L . Mathematically, a process like this corresponds to assume the molecular diffusion coefficient D_m and the chemical time $\tau_{\rm ch}$ with the limit values $D_m \rightarrow 0$ and $\tau_{\rm ch} \rightarrow 0$ such that $D_m/\tau_{\rm ch} = {\rm const.}$ So we have that $\lim_{D_m \rightarrow 0} \delta_L \sim (D_m \tau_{\rm ch})^{1/2} = 0$ and $\lim_{D_m \rightarrow 0} S_L \sim (D_m/\tau_{\rm ch})^{1/2} = {\rm const.}$ Then, for this reason, in what follows, molecular diffusion is neglected and it is set $D_m = 0$.

We consider a plane initial dividing surface so that at $t \ge t_0$ it is described by the profile C(x,t) with stepwise initial condition $C(x < x_0, t_0) = 1$ and $C(x > x_0, t_0) = 0$. In the context of Taylor theory⁷ for nonreacting particles (W=0), the Lagrangian description of turbulent diffusion shows that in general, even in stationary turbulence, D_t is not constant but it is a function of the elapsed time and it grows from $D_t(0) = 0$ to the asymptotic constant value $D_t(\infty) = \langle u'^2 \rangle T_L$, where T_L is the Lagrangian integral timescale of turbulence. Assuming a Gaussian probability density function (PDF) for particle displacement, the diffusion coefficient grows following the law:

$$D_t(t-t_0) = \int_{t_0}^t B_L(\tau - t_0) d\tau,$$
(2)

where $B_L(t-t_0) = v(t)v(t_0)$, with $B_L(0) = \langle u'^2 \rangle$, is the autocorrelation function of Lagrangian velocity fluctuation v(t) that, from stationarity, depends solely on the elapsed time $t-t_0$. The Lagrangian integral timescale T_L is determined by

$$T_L = \int_0^\infty \frac{B_L(\tau)}{\langle u'^2 \rangle} d\tau.$$

However, during a flamelet combustion process, it is not clear how to statistically manage the random appearance of product particles at their generation instant t_0 . At first glance, with a Lagrangian approach, the initial moment of diffusion of these product volumes, which randomly appear in space and time in the flame, is their generation moment that is when reactant particles cross the random flamelet sheet. As a consequence, from application of Eq. (2) to these volumes, it emerges that combustion reduces the Lagrangian diffusion coefficient because each volume starts its spreading with a null diffusion coefficient. Moreover, in the context of such intuitive knowledge, at very large elapsed times the turbulent diffusion coefficient across the steady state flame is not constant because its initial value at the front edge would be zero and then growing across the flame driven by Eq. (2). Nevertheless, we show that such kinds of considerations, which sometimes may occur in discussions, are erroneous. We point out that Taylor theory is not applicable directly to this case with random initial data, and we deduce kinematical relation for this case in Sec. III. In particular, we show that the turbulent diffusion coefficient D_t does not depend on the random generation of product volumes.

B. Flame front velocity estimation

The second flaw of the classical paradigm expressed by Eq. (1) is connected to its parabolic character and it is again due to Lagrangian properties of fluid particles traveling. In fact, the consequence of this parabolic nature is that at $t > t_0$, the value of the mean progress variable is not zero everywhere, which means that the front edge of the flame has an infinitely large speed.

In this respect, we remind the classical estimation¹⁰ of the velocity of the steady state flame U_t^{ss} at strong turbulence that, based on limited fluctuation speed, it is of the order of magnitude of the root mean square of turbulent velocity fluctuation $U_t^{ss} \simeq \langle u'^2 \rangle^{1/2}$ and it follows that the flame speed does not depend on chemistry. At a first impression, this result can be considered implausible. In fact, since the flame speed is caused by combustion, one expects that chemical kinetics has a key role. Maybe for this reason, this classical estimation has been ignored in the most recent theories of turbulent combustion and just few attempts^{11,12} exist in literature to theoretically prove or disprove it.

The target of our analysis is not only to show that the mentioned and intuitive estimation $U_t^{ss} \simeq \langle u'^2 \rangle^{1/2}$ can be theoretically justified, but also quantitatively refined. In the analysis that follows, a hyperbolic transfer equation is derived, instead of parabolic equation (1), using a closure that does not require any empirical constant. This mass transfer description is consistent with the Lagrangian property of the turbulent field concerning the impossibility to have an infinitely large speed of the front edge.

III. PARABOLIC TRANSPORT EQUATION WITH LAGRANGIAN DIFFUSION AND SOURCE TERMS

A. Remind on Taylor theory and its applications

The Taylor theory of turbulent diffusion⁷ is an exact kinematical theory, unlike closure models for turbulence dynamics equations. Given a homogeneous and stationary turbulent velocity field with zero mean, the Taylor theory describes the random motion of a fluid particle in terms of the Lagrangian autocorrelation function. Since, in this section, we aim to apply the Taylor approach in the analysis of turbulent diffusion in a medium where passive chemical transformations randomly occur, we briefly remind the derivation of the main expression.

Consider the one-dimensional case. The position of a fluid particle x is kinematically described by

$$x(t) = x_0 + \int_{t_0}^t v(\tau) d\tau, \quad x_0 = x(t_0),$$
(3)

where v(t) is the component in the *x*-direction of the particle velocity. Taking a large number of realizations of the flow with the same initial condition (x_0, t_0) , the Lagrangian ensemble average is $\bar{x}=x_0$. The particle dispersion variance turns out to be

$$\overline{(x-x_0)^2} = \sigma^2 (t-t_0) = \int_{t_0}^t \int_{t_0}^t \overline{v(\tau_1)v(\tau_2)} d\tau_1 d\tau_2 = \int_{t_0}^t \int_{t_0}^t B_L(\tau_1 - \tau_2) d\tau_1 d\tau_2.$$
(4)

With the change of variables $\tau = \tau_2 - \tau_1$ and $s = (\tau_2 + \tau_1)/2$, the double integral in Eq. (4) reduces to

$$\sigma^{2}(t-t_{0}) = \int_{t_{0}-t}^{0} \left[(t-t_{0}) + \tau \right] B_{L}(\tau) d\tau + \int_{0}^{t-t_{0}} \left[(t-t_{0}) - \tau \right] B_{L}(\tau) d\tau,$$
(5)

which, using the symmetry property $B_L(\tau) = B_L(-\tau)$, gives the final result

$$\sigma^2(t-t_0) = 2 \int_{t_0}^t (t-\tau) B_L(\tau-t_0) d\tau.$$
 (6)

From Taylor formula (6), with a generic correlation function $B_L(t)$, it follows that¹

$$\begin{split} \sigma^2(t-t_0) &\simeq \langle u'^2 \rangle (t-t_0)^2, \quad t-t_0 \ll T_L, \\ \sigma^2(t-t_0) &\simeq 2 \langle u'^2 \rangle T_L(t-t_0), \quad t-t_0 \gg T_L. \end{split}$$

Let the Lagrangian PDF of particle displacement $P_L = P_L(x;t|x_0,t_0)$, with $P_L \rightarrow \delta(x-x_0)$ when $t \rightarrow t_0$, be Gaussian

$$P_L(x;t|x_0,t_0) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left\{-\frac{(x-x_0)^2}{2\sigma^2}\right\},$$
(7)

with $\sigma^2 = \sigma^2(t-t_0) = \int_{-\infty}^{+\infty} (x-x_0)^2 P_L(x;t|x_0,t_0) dx$, then it satisfies the following non-Markovian diffusion equation:

$$\frac{\partial P_L}{\partial t} = D_t (t - t_0) \frac{\partial^2 P_L}{\partial x^2},\tag{8}$$

which was originally derived in 1976 by Adelman, see Ref. 13, formula (2.19), on the basis of the linear Mori–Kubo generalized Langevin equation with a Gaussian but non-Markovian noise. Substituting Eq. (7) in Eq. (8) and subsequently using Eq. (6) gives

$$D_t(t-t_0) = \frac{1}{2} \frac{d\sigma^2}{dt} = \int_{t_0}^t B_L(\tau-t_0) d\tau,$$
(9)

from which it follows that D_t depends on time as a monotonically increasing function and formula (2) is recovered. From formula (9), we have that $D_t(0)=0$ and the asymptotic expressions for small and large elapsed times are

$$D_t(t-t_0) \cong \langle u'^2 \rangle(t-t_0), \quad (t-t_0) \ll T_L,$$
 (10)

$$D_t(t-t_0) \cong \langle u'^2 \rangle T_L = \langle u'^2 \rangle^{1/2} \ell_L, \quad (t-t_0) \gg T_L, \qquad (11)$$

where $\ell_L = \langle u'^2 \rangle^{1/2} T_L$ is the Lagrangian integral lengthscale of turbulence. Actually, as mentioned in Sec. I, the Eulerian approach to diffusion corresponds to assume the equilibrium of the turbulent diffusion coefficient and the proportionality between the Lagrangian and the Eulerian integral length scales: $\ell_L = \text{const} \cdot \mathcal{L}$.

It is worth to stress here that, unlike the equation for the PDF of scalar concentration in the Eulerian framework, which inevitably contains also the conditional dissipation rate of the random variable for which the PDF is considered, Eq. (8) is a simple unsteady diffusion equation because it is

stated in the Lagrangian framework and it is referred to the Lagrangian PDF of fluid particle displacements. The diffusion coefficient is positive because, in the same spirit of Taylor approach, a constant density is assumed. Moreover, we remark that Eq. (8) is a purely mathematical fact that univocally and straightforwardly follows from two classical statements: (i) the use of Taylor formula (6) for particle displacement variance, which is an exact result that follows from the kinematical formula dx/dt=v, and (ii) the assumption that the particle displacement probability density function is Gaussian (7).

Consider now the three-dimensional case. When each particle contains a unit amount of diffusing matter with an impulsive δ -source, in $\mathbf{x}=0$ at time $t=t_0$, the source is described by the function $S(\mathbf{x},t) = \delta(\mathbf{x}) \,\delta(t-t_0)$. Then the average concentration $Y(\mathbf{x},t)$ is given by the expression $Y(\mathbf{x},t) = \int P_L(\mathbf{x};t | \mathbf{x}_0, \tau) \,\delta(\mathbf{x}_0) \,\delta(\tau-t_0) d\mathbf{x}_0 d\tau$ and Eq. (7) is the Green function of the equation

$$\frac{\partial Y}{\partial t} = D_t (t - t_0) \nabla^2 Y.$$
(12)

It should be remarked that the initial data $S(\mathbf{x},t) = \delta(\mathbf{x}) \delta(t-t_0)$ are identical in all realizations. When, in a fixed initial moment $t=t_0$, the mean concentration is distributed in space according to a known function $S(\mathbf{x})$ and it is the same in all realizations, then the solution of Eq. (12) in terms of the Green function is

$$Y(\mathbf{x},t) = \int_{-\infty}^{+\infty} P_L(\mathbf{x};t|\mathbf{x}_0,t_0) S(\mathbf{x}_0) d\mathbf{x}_0.$$
 (13)

We stress that, however, for general initial conditions, the diffusion process cannot be in principle described by Eq. (12). We list below some simple examples that show this.

1. Double source

In the case when the initial source is composed by two impulsive δ -sources at two different times $t_{01} \neq t_{02}$, i.e., $S_1(\mathbf{x}_{01}, t_{01}) = \delta(\mathbf{x} - \mathbf{x}_{01}) \,\delta(t - t_{01})$ and $S_2(\mathbf{x}_{02}, t_{02}) = \delta(\mathbf{x} - \mathbf{x}_{02}) \,\delta(t - t_{02})$, Eq. (12) cannot be used because there is not a single diffusion coefficient D_t . In fact, in each point (\mathbf{x}, t) , the diffusivity of the species under consideration depends on both sources S_1 and S_2 and then this case obligatory requires to use two equations with two different diffusion coefficients $D_t(t - t_{01})$ and $D_t(t - t_{02})$. Such situation can occur in combustion when ignition takes place in two different points and times, i.e., $(\mathbf{x}_{01}, t_{01})$ and $(\mathbf{x}_{02}, t_{02})$, and after a while the flames cross so that a common process results.

2. Random initial data uncorrelated with the velocity field

In the case of statistically random initial data, Eq. (12) is valid only if the random initial position of each particle and the underlying velocity field are uncorrelated, because in this case the evolution of the process does not depend on the Lagrangian kinematical history before than $t=t_0$.

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3. Random initial data correlated with the velocity field

The case when the initial conditions are the solution to Eq. (12) at some intermediate instant, say $t=t^* > t_0$ so that $S(\mathbf{x}, t_*) = Y(\mathbf{x}, t_*)$, corresponds to the case when the random initial positions of particles are correlated with the underlying velocity field. Such kind of initial condition turns out to be controlled also by the Lagrangian kinematics in the temporal interval $t_0 < t < t^*$, then the diffusion coefficient in Eq. (12) must not be replaced by $D_t(t-t^*)$, so that $D_t(t=t^*)=0$, but the correct diffusion coefficient is still $D_t(t-t_0)$. The reason is that, given the instant $t=t^* > t_0$, the Lagrangian PDF $P_L(\mathbf{x};t^*|\mathbf{x}_0, t_0)$ contains kinematical information related to the temporal interval $t_0 < t < t^*$, while, obviously, it does not depend on Lagrangian kinematical history preceding t_0 .

4. Retarded source

Consider the initial condition $Y(\mathbf{x},t_0)=S_0\delta(\mathbf{x})\delta(t_0)$, where S_0 is the strength of the impulsive source. If the strength depends on time S=S(t), with $S(t_0)=S_0$, the mean concentration is given by the expression

$$Y(\mathbf{x},t) = \int_{t_0}^t \int_{-\infty}^{+\infty} P_L(\mathbf{x};t|\mathbf{x}_0,\tau) S(\mathbf{x}_0,\tau) d\mathbf{x}_0 d\tau.$$

Assume now that $S(t)=S_0+(1-S_0)H(t-t^*)$, where *H* is the Heaviside function so that $H(\xi)=0$ when $\xi < 0$ and $H(\xi)=1$ when $\xi > 0$, and t^* is the instant of the chemical transformation. In the limit $S_0 \rightarrow 0$, we have that $Y(\mathbf{x},t)=0$ when $t < t^*$ and $Y(\mathbf{x},t)>0$ when $t > t^*$, i.e., the initial condition of the mean concentration is nonzero only at $t=t^*$ when the diffused concentration appears in the turbulent medium and it is determined by $Y(\mathbf{x},t^*)=P_L(\mathbf{x},t^*)$. Nevertheless, we must use the diffusion coefficient $D_t(t-t_0)$, so that in the moment $t=t^*$ it is $D_t(t^*-t_0)>0$, quite similar to the example (c) when the intermediate solution $Y(\mathbf{x},t_*)$ has been adopted as initial data.

Finally, in the context of the Lagrangian formulation of the turbulent diffusion problem with known initial mean profile $Y(\mathbf{x}, t_0) = S(\mathbf{x}, t_0)$, we cannot, in contrast to the Eulerian approach, predict $Y(\mathbf{x}, t > t_0)$ for an arbitrary general case. In the framework of Taylor theory, such prediction can be performed (i) in the case of a deterministic initial condition, i.e., identical initial distributions in all realizations; (ii) in the case of random initial conditions, but with particles initial position and the underlying velocity field that are uncorrelated; (iii) in the case when an intermediate solution $Y(\mathbf{x}, t_* > t_0)$ is used as initial condition and the process is let to evolve according to the initial moment $t=t_0$, which means, with respect to Eq. (12), do not replace the diffusion coefficient with $D_t(t-t_*)$ but keep it in the form $D_t(t-t_0)$.

In premixed combustion, using the flamelet combustion mechanism, chemical transformation takes place on self traversing random surface (flamelet sheet) and the products of combustion randomly appear. This means that the correct application of Taylor theory for product particle diffusion corresponds to the example (c), where the initial positions are random and correlated with the velocity field. So attention must be paid to use the correct diffusion coefficient to include such temporal correlation.

B. Lagrangian form of chemical source term and diffusion coefficient in parabolic turbulent transport equation for passive reactive mixture

Let $y(\mathbf{x},t)$ be the *instantaneous* random concentration field. Following the probability theory, the mean concentration in the point \mathbf{x} at time t is determined by

$$Y(\mathbf{x},t) = \int y P_{y}(y;t|\mathbf{x}) dy,$$
(14)

where $P_y(y;t|\mathbf{x})$ is the PDF to have the concentration value y at the time t in the fixed point **x**. The quantity $Y(\mathbf{x},t)$ is the average of the amount of material brought in (\mathbf{x},t) by all particles of the mixture, without any selection on their initial conditions. Then, in other words, the mean concentration field $Y(\mathbf{x},t)$ is the Eulerian measure that results from the amount of material Lagrangianly transported by mixture particles.

Since the system considered can be described by the joint stochastic process (\mathbf{x}, y) , in analogy with the description of the joint stochastic process (\mathbf{x}, \mathbf{v}) , where \mathbf{v} is the Lagrangian velocity, in the case of constant average density, the following integral relation can be derived by Novikov theorem:¹⁴

$$P_{y}(y;t|\mathbf{x}) = \int P(y,\mathbf{x};t|\mathbf{x}_{0},t_{0})d\mathbf{x}_{0}.$$
(15)

Using Eq. (15), definition (14) becomes

$$Y(\mathbf{x},t) = \int \int y P(y,\mathbf{x};t|\mathbf{x}_0,t_0) d\mathbf{x}_0 dy.$$
(16)

In the most general case with statistical dependence between chemistry and particle kinematics, the joint (\mathbf{x}, y) Lagrangian PDF *P* turns out to be

$$P(\mathbf{y},\mathbf{x};t|\mathbf{x}_0,t_0) = f(\mathbf{y};t|\mathbf{x},\mathbf{x}_0,t_0)P_L(\mathbf{x};t|\mathbf{x}_0,t_0)$$

where $f(y;t|\mathbf{x},\mathbf{x}_0,t_0)$ is the PDF of concentration in **x** conditioned to those particles with initial condition (\mathbf{x}_0,t_0) and $P_L(\mathbf{x};t|\mathbf{x}_0,t_0)$ is the transitional PDF of particles from (\mathbf{x}_0,t_0) to (\mathbf{x},t) . In this case, formula (16) becomes

$$Y(\mathbf{x},t) = \int \left\{ \int yf(y;t|\mathbf{x},\mathbf{x}_0,t_0) dy \right\} P_L(\mathbf{x};t|\mathbf{x}_0,t_0) d\mathbf{x}_0,$$

where the term in brackets can be recognized as the fraction of average concentration in (\mathbf{x}, t) due to the particles started in (\mathbf{x}_0, t_0) , and, hereinafter, noted by

$$\overline{Y}(t|\mathbf{x},\mathbf{x}_0,t_0) = \int yf(y;t|\mathbf{x},\mathbf{x}_0,t_0)dy.$$
(17)

Since a chemically reactive mixture is considered, the action of chemical transformation on the concentration of the chemical species associated to the particles is given by the time-dependence of the function \overline{Y} . Moreover, \overline{Y} must be monotonically increasing with initial condition $\overline{Y}=0$ at $t=t_0$, and, due to the random chemical transformation, \overline{Y} is related to the probability for a particle started in (\mathbf{x}_0, t_0) to react in (\mathbf{x}, t) . Finally, the average concentration $Y(\mathbf{x}, t)$ can be rewritten in the form

$$Y(\mathbf{x},t) = \int \overline{Y}(t|\mathbf{x},\mathbf{x}_0,t_0) P_L(\mathbf{x};t|\mathbf{x}_0,t_0) d\mathbf{x}_0.$$
 (18)

When $P_L(\mathbf{x}; t | \mathbf{x}_0, t_0)$ is Gaussian, its evolution equation turns out to be Eq. (8), where the nonequilibrium turbulent diffusion coefficient D_t is expressed by formula (9) in terms of the Lagrangian autocorrelation function $B_L(t-t_0)$, in accordance with Taylor theory.^{7,1} We remember that Taylor theory can be used in three special cases of initial conditions: deterministic initial condition, random initial position that is uncorrelated by the velocity field, and a third initial condition that is an intermediate solution of the transport equation but taking care to keep the continuity and the growing-intime monotonicity of the turbulent diffusion coefficient. In one of the three previous cases, multiplying each side in Eq. (8) by $\overline{Y}(t | \mathbf{x}, \mathbf{x}_0)$ and integrating over $d\mathbf{x}_0$ yields the following transport equation for reactive mixture:

$$\frac{\partial Y}{\partial t} = D_t (t - t_0) \nabla^2 Y + \int \left\{ \left[\frac{\partial \overline{Y}}{\partial t} - D_t (t - t_0) \nabla^2 \overline{Y} \right] P_L - 2D_t (t - t_0) \nabla \overline{Y} \cdot \nabla P_L \right\} d\mathbf{x}_0,$$
(19)

where the integral term represents in general form the average chemical source $W(\mathbf{x},t)$ by means of Lagrangian functions (in contrast to usual representation with Eulerian functions¹⁵),

$$\mathcal{W}(\mathbf{x},t) = \int \left\{ \left[\frac{\partial \overline{Y}}{\partial t} - D_t (t - t_0) \nabla^2 \overline{Y} \right] P_L - 2D_t (t - t_0) \nabla \overline{Y} \cdot \nabla P_L \right\} d\mathbf{x}_0.$$
(20)

In order to physically interpret the chemical source term in Eq. (20), we observe that even if the chemical transformation takes place independently by the motion of particles and fully described by the function $\overline{Y}(t|\mathbf{x}, \mathbf{x}_0, t_0)$, the spatial structure of the chemical source term $W(\mathbf{x}, t)$ is indeed determined also by the motion of the mixture particles. In fact, the intensity of the chemical source W in a point \mathbf{x} is due to the number of particles that chemically react in \mathbf{x} , obviously the number of particles located in \mathbf{x} is determined also by the diffusion coefficient D_t and by the PDF of particle displacement $P_L(\mathbf{x};t|\mathbf{x}_0,t)$.

If the chemical transformation of reactant particles is statistically independent of particle positions, then the two random variables y and x are statistically independent. As a consequence of this statistical independence, the joint (x, y)Lagrangian PDF P is given by the product of the marginal PDF of y and the marginal PDF of x, where the first varies without functional dependence on x and the second without functional dependence on y. Then, with respect to the previous case, in the PDF of concentration f, the dependence on x is dropped. Finally, in the special case with statistical independence between chemistry and particle kinematics, the joint (\mathbf{x}, y) Lagrangian PDF *P* turns out to be

$$P(\mathbf{y},\mathbf{x};t|\mathbf{x}_0,t_0) = f(\mathbf{y};t|\mathbf{x}_0,t_0)P_L(\mathbf{x};t|\mathbf{x}_0,t_0),$$

and the mean concentration field can be rewritten as

$$Y(\mathbf{x},t) = \int \bar{Y}(t|\mathbf{x}_0,t_0) P_L(\mathbf{x};t|\mathbf{x}_0,t_0) d\mathbf{x}_0,$$
(21)

which is equal to a formula given by Corrsin, see Ref. 16, formula (3), and the transport equation reduces to

$$\frac{\partial Y(\mathbf{x},t)}{\partial t} = D_t(t-t_0)\nabla^2 Y + \int \frac{\partial \overline{Y}}{\partial t} P_L(\mathbf{x};t|\mathbf{x}_0) d\mathbf{x}_0.$$
(22)

C. Application to turbulent premixed combustion

Let us now apply the previous general formulation for reactive systems to turbulent premixed combustion, even if constant density is assumed. The concentration field y is replaced by the progress variable c. Assume that at the initial moment $t=t_0$, we have the stepwise distribution c=0 at x>0 (reactants) and c=1 at x<0 (products). At $t>t_0$, the average turbulent flame travels from left to right. In the case of the flamelet combustion mechanism, it is assumed that the chemical transformation takes place in a wrinkled selfmoving random surface. When a fluid particle crosses this surface, an instantaneous variation of the progress variable from c=0 to c=1 for such particle occurs. Let $t' \ge t_0$ be the random time when this transformation occurs, then the value of the progress variable is c=0 when t < t' and c=1 when $t \ge t'$.

In this case, the contribution to the progress variable in (x,t) by the particles that were in (x_0,t_0) can be determined as the sum of c=1 weighted by the PDF of the random reaction activation time $\Psi(t'|x,x_0,t_0)$,

$$Q(t|x,x_0,t_0) = \int_{t_0}^t 1 \cdot \Psi(t'|x,x_0,t_0) dt'.$$
(23)

In the framework of the flamelet combustion mechanism, the Lagrangian PDF of *c* is a bimodal function of the type $f(c;t|x,x_0,t_0)=Q\delta(1-c)+(1-Q)\delta(c)$, where *Q* is given in Eq. (23).

The average chemical source term W(x,t) is expressed by the integral in Eq. (20) and, noting that $\partial Q(t|x,x_0,t_0)/\partial t = \Psi(t|x,x_0,t_0)$, it turns out to be

$$\mathcal{W}(x,t) = \int \left\{ \left[\Psi(t|x,x_0,t_0) - D_t(t-t_0) \int_{t_0}^t \frac{\partial \Psi}{\partial x^2} dt' \right] P_L - 2D_t(t-t_0) \int_{t_0}^t \frac{\partial \Psi}{\partial x} dt' \cdot \frac{\partial P_L}{\partial x} \right\} dx_0.$$
(24)

Let $\bar{x}_f(t)$ be the mathematical expectation of the instantaneous location of the flame surface, then, in the onedimensional case and zero mean velocity reference system, the propagation of the plane turbulent flame is characterized by the burning consumption rate \mathcal{U}_t which is defined as

$$\frac{d\overline{x}_f}{dt} = \mathcal{U}_t(t) = \int_{-\infty}^{+\infty} \mathcal{W}(x, t) dx,$$
(25)

where W(x,t) is the average chemical source term. We stress here that, in the transient regime for small elapsed times, the consumption rate is not equal to the speed of the flame front edge, different from the stationary regime that we will consider in the following.

Since it is assumed that particle trajectories are not affected by chemical transformation, the diffusion coefficient of the combustion process is the same of the nonreacting passive scalar diffusion coefficient assumes the same equilibrium value assumed in nonreacting systems and, in agreement with Eq. (11), the turbulent diffusion coefficient becomes practically constant $D_t \approx \langle u'^2 \rangle T_L = \langle u'^2 \rangle^{1/2} \ell_L$. In other words, the instantaneous random appearance of products with c=1 in the flame does not influence the transfer process. This means that conclusions about a zero initial diffusivity of volume with c=1, which are based on a straightforward application of Taylor formula (2) to product particles following an intuitive knowledge, are not theoretically justified, see Sec. III.

The gradient form of Eq. (19) and the diffusion coefficient independence of the chemical source term (20) give the above result on the null influence of the flame on the transfer process. This result is obtained in spite of the random generation of product volumes by the chemical transformation, then it could be in contradiction with the countergradient transport that is experimentally established for premixed flames. But, since the countergradient transport phenomenon has a gas dynamical nature, because it is generated by the fact that the pressure gradient accelerates the light products more than the heavy reactants,⁵ when reactants and products have the same density, there is no countergradient transport. Hence, the present result, which is not in contradiction with the nongradient turbulent flux because a constant density is here assumed, is a remarkable result of the present analysis, mainly because it is not intuitively expected.

Remembering that Ψ is the PDF of the random time of chemical transformation t', as a consequence of the normalization condition $\int_{t_0}^{\infty} \Psi(t'|x, x_0, t_0) dt' = 1$, in the limit $t \to \infty$ it follows that

$$\frac{\partial}{\partial x}\int_{t_0}^{\infty}\Psi(t'|x,x_0,t_0)dt' = \frac{\partial^2}{\partial x^2}\int_{t_0}^{\infty}\Psi(t'|x,x_0,t_0)dt' = 0.$$

Hence, $\mathcal{W}(x,t)$ turns out to be

$$\mathcal{W}(x,t) = \int_{-\infty}^{+\infty} \Psi(t|x,x_0,t_0) P_L(x;t|x_0,t_0) dx_0.$$
(26)

For each particle, the burning instant is dependent on the particle initial position, because the more the particle is initially distant from the initial flame, the later it will burn. However, for large elapsed time, the motion of particles becomes independent of the initial position, and then also their probability Ψ to collide with the flame sheet becomes independent of the initial condition, so we have

$$\mathcal{W}(x,t) = \int_{-\infty}^{+\infty} \Psi(t|x,x_0,t_0) P_L(x;t|x_0,t_0) dx_0$$
$$= \Psi(t|x) \int_{-\infty}^{+\infty} P_L(x;t|x_0,t_0) dx_0 = \Psi(t|x).$$
(27)

In the steady state regime, applying the change of variable $\xi = x - U_t^{ss} t$ gives

$$\int_{-\infty}^{+\infty} \Psi(t|x) dx = \int_{-\infty}^{+\infty} \mathcal{W}(x,t) dx$$
$$= \int_{-\infty}^{+\infty} \mathcal{W}(\xi) d\xi = \mathcal{U}_t^{ss} = \text{const.}$$

where \mathcal{U}_t^{s} is the steady state flame speed. To conclude, selecting the coordinate system connected with the traveling turbulent front, the parabolic diffusion equation (19) becomes

$$-\mathcal{U}_t^{\rm ss}\frac{dC}{d\xi} = \langle u'^2 \rangle T_L \frac{d^2C}{d\xi^2} + \mathcal{W}(\xi), \quad \mathcal{W}(\xi) = \Psi(\xi).$$
(28)

We notice that the particular form of Ψ , which is the PDF of the random instants when the chemical transformation of reactants in products takes place, is controlled by turbulence and by the speed of the flamelet sheet. Here we do not consider the particular form of Ψ because our target is to show that in the steady state flame the turbulent diffusion coefficient is $D_i = \langle u'^2 \rangle T_L$. This means that the random appearance in the flame of products with c=1 does not influence the transfer process, and the formal application of Taylor formula (2) to this case is not physically justified. So speculations on the increasing of the turbulent diffusion coefficient across the steady state flame, motivated by the fact that from Eq. (2) it follows a null diffusion coefficient for product particles in the instant of their generation, are not theoretically supported.

However, since the main phenomenon that controls \mathcal{U}_t^s is the traveling of the front of turbulent diffusion, the description of the steady state premixed combustion is not conceptually correct in the context of a parabolic diffusion equation. In order to perform such analysis, a hyperbolic diffusion equation based on hydrodynamics is derived in the next section. We will see that, considering the flamelet combustion mechanism, the speed of the steady state flame \mathcal{U}_t^s is mainly controlled by the velocity of the passive diffusive turbulent front \mathcal{U}_f .

IV. TURBULENT DIFFUSION WITH FINITE FRONT VELOCITY AND PREMIXED COMBUSTION

A. A hyperbolic equation for turbulent transport

Diffusion with a finite front velocity is generally obtained in literature by the telegraph equation, which is usually derived on the basis of random walk arguments.¹ Here, following Refs. 4 and 8, a hyperbolic equation with timedependent coefficient is derived on the basis of the unclosed equations for the average concentration and the turbulent flux of concentration fluctuation.

Consider Navier–Stokes equation for the velocity field $\mathbf{u}(\mathbf{x},t)$ and the scalar conservation equation for the concentration field $y(\mathbf{x},t)$,

$$\frac{\partial u_i}{\partial t} + u_\alpha \frac{\partial u_i}{\partial x_\alpha} = -\frac{1}{\rho} \frac{\partial p}{\partial x_i} + \nu \frac{\partial^2 u_i}{\partial x_\alpha \partial x_\alpha} + f_i(\mathbf{x}, t),$$
(29)

$$\frac{\partial y}{\partial t} + u_{\alpha} \frac{\partial y}{\partial x_{\alpha}} = D_m \frac{\partial^2 y}{\partial x_i \partial x_i},$$
(30)

where $\mathbf{f}(\mathbf{x},t)$ is a random force for unit volume that provides a statistically steady state turbulence, ρ is the fluid density, $p(\mathbf{x},t)$ the pressure field, ν the kinematic viscosity, D_m the molecular diffusion coefficient, and the summation rule on the repeated Greek indices is used.

Multiplying Eq. (30) by u_i and Eq. (29) by y, using the standard composition rule, i.e., $u_i = U_i + u'_i$, $f_i = F_i + f'_i$, and y = Y + y', where capital letter and prime indicate average and fluctuation, respectively, stated $U_i = F_i = 0$, gives

$$\left\langle u_{i}^{\prime} \frac{\partial y^{\prime}}{\partial t} \right\rangle + \left\langle u_{i}^{\prime} u_{\alpha}^{\prime} \right\rangle \frac{\partial Y}{\partial x_{\alpha}} + \left\langle u_{i}^{\prime} u_{\alpha}^{\prime} \frac{\partial y^{\prime}}{\partial x_{\alpha}} \right\rangle$$

$$= D_{m} \left\langle u_{i}^{\prime} \frac{\partial y^{\prime}}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle,$$
(31)

$$\left\langle y' \frac{\partial u'_i}{\partial t} \right\rangle + \left\langle y' u'_{\alpha} \frac{\partial u'_i}{\partial x_{\alpha}} \right\rangle$$

= $-\frac{1}{\rho} \left\langle y' \frac{\partial p'}{\partial x_i} \right\rangle + \nu \left\langle y' \frac{\partial^2 u'_i}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle + \langle y' f'_i \rangle.$ (32)

Hence, averaging Eq. (30) and summing Eqs. (31) and (32), the following system of equations is obtained:

$$\frac{\partial Y}{\partial t} + \frac{\partial \langle u'_{\alpha} y' \rangle}{\partial x_{\alpha}} = D_m \frac{\partial^2 Y}{\partial x_i \, \partial x_i},\tag{33}$$

$$\frac{\partial \langle u_{i}' y' \rangle}{\partial t} + \langle u_{i}' u_{\alpha}' \rangle \frac{\partial Y}{\partial x_{\alpha}}
= -\frac{\partial \langle u_{\alpha}' u_{i}' y' \rangle}{\partial x_{\alpha}} - \frac{1}{\rho} \left\langle y' \frac{\partial p'}{\partial x_{i}} \right\rangle
+ D_{m} \left\langle u_{i}' \frac{\partial^{2} y'}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle + \nu \left\langle y' \frac{\partial^{2} u_{i}'}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle + \langle y' f_{i}' \rangle.$$
(34)

System (33) and (34) requires a closure.

The random force **f** can be selected such that it acts at low wave numbers, it is divergent free (i.e., it does not directly influence the pressure field), and it is independent of the velocity field.^{17,18} This choice means that the correlations concentration-pressure and concentration-velocity do not depend on $f_i(\mathbf{x}, t)$, but also that the velocity-force correlation vanishes¹⁷ as well as the concentration-force correlation and then $\langle y' f'_i \rangle = 0$.

In turbulent flows, the molecular diffusion effects embodied by D_m can be neglected with respect to turbulent diffusion effects. But the results derived with this neglect must be correct also for applications in premixed combustion. With respect to this, we remind that in Sec. II it has been highlighted that here it is considered a premixed combustion process with constant laminar flame speed S_L and zero laminar flame thickness δ_L , whose estimations are given by $S_L \sim (D_m/\tau_{ch})^{1/2}$ and $\delta_L \sim (D_m\tau_{ch})^{1/2}$, where τ_{ch} is the chemical time. This means that in the limit of infinite Damköhler number, i.e., $\tau_{ch} \rightarrow 0$, the molecular diffusion D_m is such that $\lim_{\tau_{ch} \rightarrow 0} D_m/\tau_{ch} = \text{const}$, hence for this kind of process, $D_m \rightarrow 0$. These arguments and assumptions motivate to disregard D_m and assure that there is no propagation of error when results obtained with this neglect are applied in turbulent premixed combustion. Then, hereinafter, we state $D_m = 0$.

Finally, the right-hand side (RHS) of Eq. (34) reduces to (1 + 1) = (1 + 1

$$\frac{\partial \langle u'_{\alpha} u'_{i} y' \rangle}{\partial x_{\alpha}} + \frac{1}{\rho} \left\langle y' \frac{\partial p'}{\partial x_{i}} \right\rangle - \nu \left\langle y' \frac{\partial^{2} u'_{i}}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle,$$

where, in order to have turbulent energy dissipation, the kinematic viscosity must be $\nu \neq 0$, and then, to have stationary turbulence, the turbulent energy generated by the force $\mathbf{f}(\mathbf{x},t)$ is equal to the constant mean dissipation rate.^{17,18} But, with the hypothesis that turbulent diffusion is controlled in general by the Lagrangian velocity fluctuation autocorrelation function $B_L(t)$, for infinite Reynolds number the RHS of Eq. (34) can be assumed to be independent of ν and to be dependent on $\langle u_i' y' \rangle$, which preserves the dependence on (\mathbf{x}, t) , and on a functional of $B_L(t)$ which includes the parameter T_L : $\varphi = \varphi[B_L(t), T_L] = \varphi(t, T_L)$. This assumption can be applied also to the addendum with the pressure fluctuation p' because, from dimensional arguments, it can be expressed in terms of the velocity fluctuation. To conclude, invoking the II-theorem,¹⁹ the closure is

$$\frac{\partial \langle u'_{\alpha} u'_{i} y' \rangle}{\partial x_{\alpha}} + \frac{1}{\rho} \left\langle y' \frac{\partial p'}{\partial x_{i}} \right\rangle - \nu \left\langle y' \frac{\partial^{2} u'_{i}}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle$$

$$= \frac{\varphi(t, T_{L})}{T_{L}} \langle u'_{i} y' \rangle.$$
(35)

Closure (35) is different from the previous one.^{8,4} In fact, the present includes the time-dependent function φ that is fundamental to describe all dispersion regimes. Finally, the transport equation turns out to be

$$\frac{\partial^2 Y}{\partial t^2} + \frac{\varphi(t, T_L)}{T_L} \frac{\partial Y}{\partial t} = \frac{\partial \langle u_i' u_\alpha' \rangle}{\partial x_i} \frac{\partial Y}{\partial x_\alpha} + \langle u_\alpha' u_i' \rangle \frac{\partial^2 Y}{\partial x_i \partial x_\alpha}.$$
 (36)

From homogeneity and isotropy, it follows that $\langle u'_i u'_{\alpha} \rangle = \langle u'^2 \rangle \delta_{i\alpha}$ and then Eq. (36) becomes

$$\frac{\partial^2 Y}{\partial t^2} + \frac{\varphi(t, T_L)}{T_L} \frac{\partial Y}{\partial t} = \langle u'^2 \rangle \frac{\partial^2 Y}{\partial x^2},\tag{37}$$

where x stands for each Cartesian component x_i .

The idea of lumping the transport term with the pressure term is based on the fact that the pressure term is proportional to the square of velocity fluctuations, i.e., $p' \sim \rho u'^2$, and then the terms $\partial \langle u'_{\alpha} u'_{i} y' \rangle / \partial x_{\alpha}$ and $\rho^{-1} \langle y' \partial p' / \partial x_{i} \rangle$ have the same dimensional representation. Since the model is complete, that is there are no empirical parameters, the factors of proportionality can be merged in the unique factor

 $\varphi(t, T_L) = \varphi_u + \varphi_p$, where φ_u and φ_p are the factors of proportionality for the transport and the pressure term, respectively. The lumping of the viscous term with the transport and the pressure terms follows from the fact that they have the same physical dimension. But, since an infinite high Reynolds number flow is considered, the resulting closure is independent of viscosity. The most important step, which makes the present closure strongly different from those generally introduced in literature, is that here the transport equation (35) is generated by the coupling of the Navier-Stokes and the mass conservation equations, see system (28) and (29). From this fact, it follows that the turbulent flux term does not need a closure, e.g., the classical gradient model, because it is determined by the evolution equation (33) and the requirement of a closure is moved to the RHS of Eq. (33). This result is mainly due to dimensional arguments that yield a linear proportionality of the RHS in Eq. (33) with the turbulent flux $\langle u'_{z} v' \rangle$.

The analysis of the characteristics of Eq. (37) shows that the front velocity U_f , for any arbitrary $\varphi(t, T_L)$, is

$$\mathcal{U}_f = \langle u'^2 \rangle^{1/2}.\tag{38}$$

The Lagrangian properties of transport process (37) can be derived. In fact, for a δ -function initial condition, the particle displacement $\overline{x^2}$ is $\overline{x^2} = \int x^2 Y(x, t) dx$, and then multiplying Eq. (37) by x^2 and integrating in dx, using definition (9), gives

$$\frac{d^2 \overline{x^2}}{dt^2} + \frac{\varphi(t, T_L)}{T_L} \frac{d \overline{x^2}}{dt} = 2\langle u'^2 \rangle.$$
(39)

Imposing the agreement with Taylor statements on dispersion, see formula (6), Eq. (39) yields the following determination of $\varphi(t, T_L)$:

$$\varphi(t,T_L) = T_L \frac{\langle u'^2 \rangle - B_L(t)}{\int_0^t B_L(\tau) d\tau}.$$
(40)

This means that, with $\varphi(t,T_L)$ defined as in Eq. (40), Eqs. (37) and (39) hold at all times and they meet Taylor statements for an arbitrary Lagrangian velocity fluctuation auto-correlation function $B_L(t)$.

When $t \rightarrow 0$, applying the Taylor expansion $B_L \approx \langle u'^2 \rangle + dB_L/dt(t=0)t+\cdots$ gives

$$\varphi(t,T_L) \simeq -T_L \frac{dB_L/dt(t=0)}{\langle u'^2 \rangle}, \quad t \to 0,$$
(41)

which is positive because $B_L(t)$ is a decreasing function at the origin and then $dB_L/dt(t=0) < 0$. Moreover, if $dB_L/dt(t=0)=0$, when $t \rightarrow 0$, function $\varphi(t,T_L)$ tends to 0. When $t \rightarrow \infty$, since $B_L \rightarrow 0$ and $\int_0^t B_L(\tau) d\tau \rightarrow \langle u'^2 \rangle T_L$, it follows from Eq. (40) that

$$\varphi(t,T_I) \to 1, \quad t \to \infty.$$
 (42)

When an exponential autocorrelation function is chosen, namely, $B_L(t) = \langle u'^2 \rangle \exp(-t/T_L)$, it follows from Eq. (40) that $\varphi(t, T_L) = 1$ for all times and Eq. (37) reduces to

$$T_L \frac{\partial^2 Y}{\partial t^2} + \frac{\partial Y}{\partial t} = \langle u'^2 \rangle T_L \frac{\partial^2 Y}{\partial x^2},$$
(43)

which is the telegraph equation. This means that previous formulation^{8,4} holds only for very large elapsed times, or it implicitly assumes an exponential correlation function $B_L(t)$, because φ is taken constant.

Equation (43) has a number of good properties for turbulent transport modeling: (i) it is a Eulerian-type equation with constant coefficient; (ii) it generates a particle diffusion process in agreement with Taylor statements; (iii) it is consistent with an exponential Lagrangian velocity fluctuation autocorrelation function; and (iv) it describes a diffusion process with finite velocity front.

B. Relationships between the velocity of turbulent diffusion front and that of turbulent premixed flame in the flamelet combustion mechanism

In order to analyze premixed combustion, the scalar conservation equation (30) for the instantaneous concentration field $y(\mathbf{x},t)$ is now referred to the progress variable $c(\mathbf{x},t)$ and it is modified to take into account a constant laminar flame speed S_L as

$$\frac{\partial c}{\partial t} + u_{\alpha} \frac{\partial c}{\partial x_{\alpha}} = S_L |\nabla c|, \qquad (44)$$

where also a constant density is assumed to use, in what follows, the results from previous section. Considering Navier–Stokes equations (29), a system of equations analog to Eqs. (33) and (34) can be derived and, after using closure (35), it turns out to be

$$\frac{\partial C}{\partial t} + \frac{\partial \langle u'_{\alpha} c' \rangle}{\partial x_{\alpha}} = S_L \langle |\nabla c| \rangle, \tag{45}$$

$$\frac{\partial \langle u_i'c' \rangle}{\partial t} + \langle u_i'u_{\alpha}' \rangle \frac{\partial C}{\partial x_{\alpha}} + \frac{\varphi(t, T_L)}{T_L} \langle u_i'c' \rangle = S_L \langle u_i' | \nabla c | \rangle.$$
(46)

System (45) and (46) requires equations for $\langle |\nabla c| \rangle$ and $\langle u'_i |\nabla c| \rangle$. Let $\hat{n}(\mathbf{x},t) = -\nabla c/|\nabla c|$ be the unit vector which generally depends on both space and time and it is directed normal to a local isosurface pointing toward reactants. From the property $\hat{n} \cdot \hat{n} = 1$, it emerges that $\hat{n} \cdot \partial \hat{n}/\partial t = \hat{n} \cdot \partial \hat{n}/\partial x_i = 0$ and the following identities can be derived:

$$\frac{\partial |\nabla c|}{\partial t} = -\hat{n} \cdot \nabla \frac{\partial c}{\partial t} \quad \text{and} \quad \frac{\partial |\nabla c|}{\partial x_i} = -\hat{n} \cdot \nabla \frac{\partial c}{\partial x_i}.$$

Then from Eq. (44), it follows that

$$\frac{\partial |\nabla c|}{\partial t} + \frac{\partial u'_{\alpha} |\nabla c|}{\partial x_{\alpha}} = - |\nabla c| (\hat{n}\hat{n}:\nabla \mathbf{u}') - S_L \hat{n} \cdot \nabla |\nabla c|, \qquad (47)$$

where $\hat{n}\hat{n}: \nabla \mathbf{u}' = n_i n_j \partial u'_j / \partial x_j$. Considering again Navier–Stokes equations (29) and applying previous formalism, equations for $\langle |\nabla c| \rangle$ and $\langle u'_i |\nabla c| \rangle$ constitute the new system

$$\frac{\partial \langle |\nabla c| \rangle}{\partial t} + \frac{\partial \langle u'_{\alpha} |\nabla c| \rangle}{\partial x_{\alpha}} = -\langle |\nabla c| (\hat{n}\hat{n}: \nabla \mathbf{u}') \rangle - S_L \langle \hat{n} \cdot \nabla |\nabla c| \rangle,$$
(48)

$$\frac{\partial \langle u_i' | \nabla c | \rangle}{\partial t} + \frac{\psi(t, T_{\nabla c})}{T_{\nabla c}} \langle u_i' | \nabla c | \rangle$$

$$= \langle | \nabla c | \rangle \frac{\partial \langle u_i' u_{\alpha}' \rangle}{\partial x_{\alpha}} - \langle u_i' | \nabla c | (\hat{n} \hat{n} : \nabla \mathbf{u}') \rangle$$

$$- S_L \langle u_i' \hat{n} \cdot \nabla | \nabla c | \rangle, \qquad (49)$$

where $\psi(t, T_{\nabla c})$ and $T_{\nabla c}$ are defined by the following closure:

$$\frac{\partial \langle u'_{\alpha} u'_{i} | \nabla c | \rangle}{\partial x_{\alpha}} + \frac{1}{\rho} \left\langle | \nabla c | \frac{\partial p'}{\partial x_{i}} \right\rangle - \nu \left\langle | \nabla c | \frac{\partial^{2} u'_{i}}{\partial x_{\alpha} \partial x_{\alpha}} \right\rangle$$

$$= \frac{\psi(t, T_{\nabla c})}{T_{\nabla c}} \langle u'_{i} | \nabla c | \rangle,$$
(50)

which is similar to Eq. (35).

Combining Eqs. (45) and (46) and using Eq. (48) gives

$$\frac{\partial^2 C}{\partial t^2} + \frac{\varphi(t, T_L)}{T_L} \frac{\partial C}{\partial t} = \langle u_i' u_\alpha' \rangle \frac{\partial^2 C}{\partial x_i \partial x_\alpha} + \frac{\partial \langle u_i' u_\alpha' \rangle}{\partial x_i} \frac{\partial C}{\partial x_\alpha} + S_L^2 \frac{\partial^2 C}{\partial x_\alpha \partial x_\alpha} + \frac{\mathcal{W}(\mathbf{x}, t)}{T_L},$$
(51)

where

$$\frac{\mathcal{W}(\mathbf{x},t)}{T_L} = S_L \left\{ \frac{\varphi(t,T_L)}{T_L} \langle |\nabla c| \rangle - 2 \frac{\partial \langle u'_{\alpha} |\nabla c| \rangle}{\partial x_{\alpha}} - \langle |\nabla c| (\hat{n}\hat{n}:\nabla \mathbf{u}') \rangle \right\}.$$
(52)

Finally, from homogeneity and isotropy, Eq. (51) reduces to

$$T_L \frac{\partial^2 C}{\partial t^2} + \varphi(t, T_L) \frac{\partial C}{\partial t} = \left[\langle u'^2 \rangle + S_L^2 \right] T_L \frac{\partial^2 C}{\partial x^2} + \mathcal{W}(x, t), \quad (53)$$

where x stands for each Cartesian component x_i . Since the average source function $\mathcal{W}(x,t)$ defined in Eq. (52) does not contain the terms $\partial^2 C / \partial t^2$ and $\partial^2 C / \partial x^2$, from the analysis of characteristics of Eq. (53), the front edge flame speed \mathcal{U}_{fe} turns out to be

$$\mathcal{U}_{\rm fe} = (\langle u'^2 \rangle + S_L^2)^{1/2} = (\mathcal{U}_f^2 + S_L^2)^{1/2}.$$
 (54)

Hence, taking into account that in the stationary traveling flame case all isosurfaces have the same velocity, in the steady state regime the value of the consumption rate U_t , which is defined in Eq. (25), turns out to be equal to the front edge flame speed U_{fe} and, hereinafter, we refer to this value as the flame speed U_{fs}^{s} ,

$$\mathcal{U}_t^{\rm ss} = \mathcal{U}_{\rm fe} = (\langle u'^2 \rangle + S_L^2)^{1/2}.$$
(55)

This means that the speed of the steady state flame \mathcal{U}_t^{ss} does not depend on the particular form of the chemical source \mathcal{W} . Furthermore, in the case of strong turbulence, this theoretical result justifies and refines the classical estimation¹⁰ $\mathcal{U}_t^{ss} \sim \langle u'^2 \rangle^{1/2}$ by the formula

$$\mathcal{U}_{t}^{ss} \simeq \langle u'^{2} \rangle^{1/2} \left(1 + \frac{1}{2} \frac{S_{L}^{2}}{\langle u'^{2} \rangle} \right).$$
(56)

Formula (55) is important also because it shows that the commonly accepted expression for the steady state speed

 $\mathcal{U}_t^{ss} \simeq \langle u'^2 \rangle^{1/2} + S_L$, which follows from intuitive knowledge, is not consistent with a more statistically accurate analysis as that one here developed.

Moreover, formula (55) turns out to be important even for quantitative reasons. In fact, when $U_f \simeq S_L$ the classical estimation of U_t^{ss} gives $2U_f$ while the present estimation gives $\sqrt{2}U_f$, then the percentage error is $[(2-\sqrt{2})/\sqrt{2}] \times 100$ $\simeq 40\%$, which is not negligible in applications.

Refined estimation (56) of the turbulent flame velocity in the steady state for strong turbulence can be compared with other formulas in literature which differ from classical Damköhler estimation. In particular, a previous formula derived by Yakhot^{11,20} for the density constant case in our notation reads

$$\mathcal{U}_t^{\mathrm{ss}} \simeq \langle u'^2 \rangle^2 \left[\ln \left(\frac{\langle u'^2 \rangle^{1/2}}{S_L} \right) \right]^{-1/p}, \quad \frac{\langle u'^2 \rangle^{1/2}}{S_L} \to \infty$$

so that $\mathcal{U}_t^{ss} \ll \langle u'^2 \rangle^{1/2}$. However, this estimation has been obtained as extrapolation abroad the validity domain of the considered assumptions.¹¹ Differently, formula (56) is obtained working into the validity domain of the assumptions here considered.

A further comparison can be performed with the recent formula derived by Kolla *et al.*,^{12,21} which looks to be different from classical estimation. It includes also the heat release effects, so it can be considered a refinement for the derivable density case. However, when density is assumed constant, it reduces to the Damköhler estimation. To conclude, the formula of Kolla *et al.* is a refinement of Damköhler estimation when derivable density is taken into account, while our formula (56) is an effective theoretical based quantitative refinement of the Damköhler estimation.

To conclude, it is also pertinent to compare formula (55) with the well-known Kolmogorov–Petrovskii–Piskunov (KPP) solution,²¹

$$\mathcal{U}_t^{\rm ss} = 2 \sqrt{\frac{D_t}{\rm Sc} \left(\frac{\partial \mathcal{W}}{\partial C}\right)_{C \to 0}},\tag{57}$$

where $D_t = \langle u'^2 \rangle T_L = \text{const}$ and Sc is the Schmidt number. Formula (57) was originally obtained for a reaction-diffusion system introduced for a biological problem,²² and now it is used in literature for the prediction of the speed of the turbulent premixed flame. Comparing Eqs. (55) and (57) in strong turbulence $(\langle u'^2 \rangle^{1/2} \gg S_L)$, it follows that

$$4\left(\frac{\partial \mathcal{W}}{\partial C}\right)_{C\to 0} \simeq \frac{\mathrm{Sc}}{D_t} \langle u'^2 \rangle.$$
(58)

At a first glance, it appears that formula (55), and so also Damköhler's estimation, is not consistent with KPP approach (57), because from Eq. (58) it follows that the derivative of the mean chemical source at the front edge $(\partial W / \partial C)_{C \to 0}$ turns out to be dependent on the combination of some turbulence characteristics. However, as it is discussed by Zeldovich *et al.*,²³ the KPP tool is applicable in combustion for the analysis of cold flame, where combustion is controlled by the kinetics of isothermal chain reactions, i.e., flames without visible increasing of the temperature as experimentally observed in some mixtures. In particular, in the same book²³ at

p. 129, one can read: "The propagation of a cool flame can be treated as the continuous introduction of seed particles into the fresh mixture by means of diffusion. Thus, the flame velocity (57) is determined by the local rate of production of active centers near the boundary of the cool flame with the fresh mixture (for $C \rightarrow 0$) and is independent of the form of this rate function inside the cool flame front. For thermal flame propagation, on the other hand, the velocity is determined by the integral of the rate of heat release over the entire chemical reaction zone." In our case, the seeds are particles with C=1 introduced into the fresh mixture, which has C=0, by the large turbulent eddies that drive also the velocity of the turbulent diffusion front \mathcal{U}_{f} , which is here theoretically estimated in Eq. (38). Then the processes near the front edge (similar to the case of catalytic combustion) control the flame speed. In the transient flame (including the analyzed initial stage), which precedes the steady state regime, the assumption of the KPP theory is not valid, so its application in this case seems to be questionable. The physical reason is that, different from the steady state regime, in the transient stage the consumption rate is not equal to the velocity of the front edge of the flame: the former is equal to the integral of the chemical source across the flame, while the latter is controlled mainly by the turbulent diffusion and at strong turbulence it does not depend on the chemical source.

C. Discussion on the steady state flame

The steady state flame is not observed in laboratory flames and then doubts can arise on its existence. However, this fact can be explained by the estimation of the necessary elapsed time for the steady state premixed flame to take place.

Consider the case $\langle u'^2 \rangle^{1/2} \gg S_L$. During a significant temporal interval, the increasing of the flamelet dispersion practically does not depend on the relatively small S_L , so in this regime the width of the turbulent flame grows. Remembering $U_t(t) = d\bar{x}_f/dt$ to be the consumption rate where \bar{x}_f is the mathematical expectation of the flame location, then the turbulent flame width $\delta_t(t)$ is given by $\delta_t(t) \propto \sigma_f(t) \approx (2\langle u'^2 \rangle T_L t)^{1/2}$, where $\sigma_f^2(t) = [x - \bar{x}_f(t)]^2$.

A strong influence of small flamelet speed on the flame width occurs when the transport due to the velocity fluctuations and that due to the flamelet travel are of the same order. This means that there exists a time Θ_{ss} such that $(2\langle u'^2 \rangle T_L \Theta_{ss})^{1/2} \sim S_L \Theta_{ss}$, and then the steady state regime is obtained when $t \ge \Theta_{ss} \sim T_L \langle u'^2 \rangle / S_L^2$, and from Eq. (56) the flame speed turns out to be $U_t^{ss} \simeq \langle u'^2 \rangle^{1/2}$.

This timescale estimation can be used to explain why the steady state flame is not observed in the laboratory flames. In fact, let us consider the Bunsen burner with the following characteristic flow parameters: $\langle u'^2 \rangle^{1/2} / w = 10^{-1}$, $\ell_L / d = 10^{-1}$, and $S_L / \langle u'^2 \rangle^{1/2} = 10^{-1}$, where *w* and *d* are the velocity and the diameter of the flow. Since the order of the magnitude of the flame speed is $U_t^{ss} \sim \langle u'^2 \rangle^{1/2}$, the length of the conical Bunsen flame (the height of the prolate cone) is equal to $\Lambda \simeq 0.5 dw / U_t^{ss}$. Using the characteristic values of the parameters gives $\Lambda \sim T_L w (\langle u'^2 \rangle^{1/2} / S_L)$. Hence, the residence time

in the Bunsen flame is of the order of $T_L(\langle u'^2 \rangle^{1/2}/S_L) \sim 10^1 T_L$, while the steady state flame takes place for times much more larger than $\Theta_{ss} \sim T_L \langle u'^2 \rangle/S_L^2 \sim 10^2 T_L$. Furthermore, even when $\langle u'^2 \rangle^{1/2} \sim S_L$ the residence time is comparable with T_L but $\Theta_{ss} \gg T_L$. This estimation explains why the width of the flame increases beginning from the edge of the Bunsen burner until the flame crosses and combustion ceases. Similar estimation can be done for the V-flame. However, direct numerical simulations show the existence of the steady state flame,^{24–26} which corresponds to the realization of an ideal experiment where the plane flame must travel for a long time in the reactant medium with uniform and stationary turbulence.

We remind here that a quasiconstant flame speed occurs also during the intermediate temporal interval $T_L < t < T_L \langle u'^2 \rangle / S_L^2$. In this interval, the dispersion of the flamelet, which is mainly controlled by the largescale wrinkles, is $\sigma_f^2(t) \simeq 2 \langle u'^2 \rangle T_L t$, i.e., the flame width is $\delta_t(t) \sim (D_t t)^{1/2}$ and it does not depend on S_L . But, at the same time, the consumption rate \mathcal{U}_t , which is also estimated by $\mathcal{U}_{t}(t) = S_{L}(\overline{A}/A_{0})$, where $\overline{A}(t)/A_{0}$ is the average dimensionless flame sheet area so that $\mathcal{U}_t(t_0) = S_L$, strongly depends on S_L , because the dimensionless flamelet surface area \overline{A} is mainly controlled by small-scale wrinkles, which are smoothed by the traveling flamelet. If it is assumed the statistical equilibrium between the generation by turbulence of the small-scale wrinkles of the flamelet surface and their consumption due to the flamelet surface traveling, the consumption rate \mathcal{U}_t turns out to be quasiconstant in time. Such turbulent premixed flames with nearly constant consumption rate and increasing width prevail in typical laboratory flames: the conical flame in the Bunsen burner and the reverse conical V-flame in uniform flow of combustible mixture behind a stabilizer. The same result is experimentally obtained for the Bunsen flame²⁷ and both experimentally and by direct numerical simulations for the V-flame.²⁸

As a concluding remark, we cite that a quantitative analysis of the problems mentioned above can be developed in the context of Kolmogorov approach,⁴ which is based on the statistical equilibrium of the small-scale and on the statistical nonequilibrium of the large-scale of the hydrodynamical structures in turbulent flows, by invoking some general statistical properties of random surfaces and the dimensional theory.

V. SUMMARY AND CONCLUSIONS

In this paper, we have analyzed two Lagrangian properties of turbulent diffusion that, even if they are ignored in the Eulerian approach used for the description of heat and mass transfer, are important in several applications, in particular in turbulent premixed combustion. The analyzed properties are connected (i) with the existence of a finite integral correlation time of Lagrangian particle velocity that yields a memory effect on particle motion and a turbulent diffusion coefficient which is not constant in time and (ii) with the forming of a front of turbulent diffusion constrained by the physical restrictions on the velocity of Lagrangian particles.

Here, the Lagrangian properties analyzed are important in the temporal interval $0 \le t \le T_L$, when the turbulent diffusion coefficient is not in equilibrium and the flame has an increasing consumption rate $U_t(t)$ and width $\delta_t(t)$, and for very large elapsed times $t \ge T_L \langle u'^2 \rangle / S_L^2$, when the constant flame speed U_t^{ss} is controlled by the speed of the turbulent front and practically it does not depend on S_L .

The analysis refers to turbulent premixed flame in the mathematical framework where combustion takes place in a strongly wrinkled flamelet surface that travels with constant speed S_L . In order to avoid the influence of combustion on turbulence, which is assumed homogeneous, isotropic, and stationary, the density is stated constant so that passive chemical reaction is actually considered. This choice deletes countergradient phenomenon, which is a consequence of density variability because the pressure gradient gives different acceleration to the light products and to the heavy reactants.

Here, the results derived are obtained on the basis of physical assumptions that yielded formulas which do not require any empirical parameters. This property follows from the fact that some of these results are kinematical and the Lagrangian correlation function is assumed known.

We have analyzed the following questions:

- The transient behavior of Lagrangian turbulent diffusion coefficient in premixed flame, which is the driving process in the raising of the developed flame, that is important, for example, to have an accurate modeling of the initial stage of combustion in the SI engine;
- (2) The relationship of the velocity of the steady state flame at large times with the theoretically predicted finite speed of the front of turbulent diffusion, which is connected with the physical restrictions on the velocities of the Lagrangian particles.

The main results are the following:

(1) The arbitrary chemical transformation of Lagrangian particles does not influence the turbulent diffusion coefficient that remains the same for nonreacting system, during both the nonequilibrium and the equilibrium regimes. This result follows from the expression of the arbitrary average chemical source term $W(\mathbf{x},t)$ that here, different from other representations, it is written in the Lagrangian framework (20). Then, the present analysis proves that the intuitive understanding about an initial zero diffusivity of product particles in the instant of their appearance is not correct in premixed combustion. Moreover, from formula (20), it follows also that the spatial structure of the chemical source term is dependent on the particle diffusive motion.

(2) We have shown that in the flamelet combustion mechanism, the speed of the front edge of the turbulent flame is equal to U_{fe}=(⟨u'²⟩+S_L²)^{1/2}, and hence at large time (t→∞), when the flame is in the steady state, we have U_t^{ss}=U_{fe}=(⟨u'²⟩+S_L²)^{1/2}. This result follows from the deduced hyperbolic equation (53) for the mean progress variable with the average chemical source term W(**x**,t)

written in general form. For purely diffusive case, i.e., $\mathcal{W}=0$, this hyperbolic equation with constant coefficients satisfies the Taylor theory with an exponential Lagrangian correlation function. However, we have also shown that for an arbitrary autocorrelation function, this hyperbolic equation exactly satisfies the Taylor theory with time-dependent coefficients (37)-(40). The result obtained for the flame speed gives, in the case of strong turbulence, the theoretically founded estimation $\mathcal{U}_t^{ss} \simeq \langle u'^2 \rangle^{1/2} [1 + (1/2)S_L^2 / \langle u'^2 \rangle]$, which is more accurate than the classical intuitive formula¹⁰ $\mathcal{U}_{t}^{ss} \sim \langle u'^{2} \rangle^{1/2}$, and, in the case of moderate turbulence, from formula (55) it emerges that the commonly used estimation $\mathcal{U}_t^{ss} \sim \langle u'^2 \rangle^{1/2} + S_L$ is not consistent with an accurate statistical analysis. The present formula (55) and the classical estimation of \mathcal{U}_t^{ss} give results that differ for a percentage error of 40%, which is not negligible for applications.

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- ¹A. S. Monin and A. M. Yaglom, *Statistical Fluid Mechanics* (MIT, Cambridge, MA, 1971), Vol. I.
- ²R. B. Bird, W. E. Stewart, and E. N. Lightfoot, *Transport Phenomena* (Wiley, New York, 2007).
- ³A. N. Lipatnikov and J. Chomiak, "A simple model of unsteady turbulent flame speed propagation," SAE Trans., J. Engines **106**, 2441 (1997); SAE Paper No. 972993.
- ⁴V. L. Zimont, "Kolmogorov's legacy and turbulent premixed combustion modeling," in *New Developments in Combustion Research*, edited by W. J. Carey (Nova Science, New York, 2006), pp. 1–93.
- ⁵S. Pope, "Turbulent premixed flames," Annu. Rev. Fluid Mech. **19**, 237 (1987).
- ⁶V. L. Zimont and F. Biagioli, "Gradient, counter-gradient transport and their transition in turbulent premixed flames," Combust. Theory Modell. 6, 79 (2002).
- ⁷G. I. Taylor, "Diffusion by continuous movements," Proc. London Math. Soc. s2-20, 196 (1922).
- ⁸V. L. Zimont and V. A. Sabel'nikov, "The equation of turbulent transport in the presence of molecular diffusion," Izv., Acad. Sci., USSR, Atmos. Oceanic Phys. **11**, 627 (1975).
- ⁹K. N. C. Bray and J. B. Moss, "A unified statistical model of the premixed turbulent flame," Acta Astronaut. **4**, 291 (1977).
- ¹⁰G. Damköhler, "Der einfluß der turbulenz auf die flammengeschwindigkeit in gasgemischen," Z. Elektrochem. **46**, 601 (1940); "The effect of turbulence on the flame velocity in gas mixtures," NACA TM No. 1112, 1947.
- ¹¹V. Yakhot, "Propagation velocity of premixed turbulent flames," Combust. Sci. Technol. **60**, 191 (1988).
- ¹²H. Kolla, J. W. Rogerson, N. Chakraborty, and N. Swaminathan, "Scalar dissipation rate modeling and its validation," Combust. Sci. Technol. **181**, 518 (2009).
- ¹³S. A. Adelman, "Fokker–Planck equations for simple non-Markovian systems," J. Chem. Phys. 64, 124 (1976).

¹⁴E. A. Novikov, "The Lagrangian-Eulerian probability relations and the random force method for nonhomogeneous turbulence," Phys. Fluids 29, 3907 (1986).

- ¹⁵D. Veynante and L. Vervisch, "Turbulent combustion modeling," Prog. Energy Combust. Sci. 28, 193 (2002).
- ¹⁶S. Corrsin, "Effect of passive chemical reaction on turbulent dispersion," AIAA J. 6, 1797 (1968).
- ¹⁷K. Alvelius, "Random forcing of three-dimensional homogeneous turbulence," Phys. Fluids 11, 1880 (1999).
- ¹⁸K. Alvelius and A. V. Johansson, "LES computations and comparison with Kolmogorov theory for two-point pressure-velocity correlations and structure functions for globally anisotropic turbulence," J. Fluid Mech. **403**, 23 (2000).
- ¹⁹G. I. Barenblatt, Similarity, Self-Similarity, and Intermediate Asymptotics (Consultants Bureau, New York, 1979).
- ²⁰A. R. Kerstein and Wm. T. Ashurst, "Propagation rate of growing interfaces in stirred fluids," Phys. Rev. Lett. **68**, 934 (1992).
- ²¹H. Kolla, J. W. Rogerson, and N. Swaminathan, "Validation of a turbulent flame speed model across combustion regimes," Combust. Sci. Technol. 182, 284 (2010).
- ²²A. N. Kolmogorov, I. G. Petrovskii, and N. S. Piskunov, "A study of the diffusion equation with increase in the quantity of matter, and its

application to a biological problem," Bjul. Moskovskogo Gos. Univ. 1, 1 (1937).

- ²³ Ya. B. Zeldovich, G. I. Barenblatt, V. B. Librovich, and G. M. Makhviladze, *The Mathematical Theory of Combustion and Explosions* (Consultants Bureau, New York, 1985).
- ²⁴T. Hasegawa and R. Himeno, "Transport properties in turbulent premixed flames," RIKEN Rev. **30**, 16 (2000).
- ²⁵S. Nishiki, T. Hasegawa, and R. Himeno, "Transport properties in fully developed turbulent premixed flames," RIKEN Rev. 40, 12 (2001).
- ²⁶D. Lee and K. Y. Huh, "Statistically steady incompressible DNS to validate a new correlation for turbulent burning velocity in turbulent premixed combustion," Flow, Turbul. Combust. **84**, 339 (2010).
- ²⁷H. Kobayashi, T. Tamura, K. Maruta, T. Niioka, and F. Williams, "Burning velocity of turbulent premixed flames in a high-pressure environment," in *Twenty-Sixth Symposium (International) on Combustion*, edited by The Combustion Institute (The Combustion Institute, Pittsburgh, 1996), pp. 389–396.
- ²⁸J. B. Bell, M. S. Day, I. G. Shepherd, M. R. Johnson, R. K. Cheng, J. F. Grcar, V. E. Beckner, and M. J. Lijewski, "Numerical simulation of a laboratory-scale turbulent V-flame," Proc. Natl. Acad. Sci. U.S.A. **102**, 10006 (2005).
- ²⁹F. Ghirelli, "Eulerian modeling of turbulent dispersion from a point source," Comput. Fluids 38, 1795 (2009).