

# Lagrangian properties of diffusion in the theory of turbulent combustion

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**Abstract** — In this paper we analyze two effects of the Lagrangian nature of turbulent transfer that are usually ignored in the theory of turbulent combustion. They are (i) nonequilibrium behavior of the turbulent diffusion coefficient, which is important for modeling of initial stage of combustion in the SI 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. First we derive the parabolic diffusion equation with both the diffusion and chemical source terms expressed by Lagrangian characteristics of turbulence. We show that chemical transformation does not influence the turbulent diffusion coefficient and this result is important for combustion theories that relate the formation of the initial flame with the development of the diffusion coefficient. Second, an hyperbolic diffusion equation based on hydrodynamics is derived and we analyze relationship between velocities of the turbulent diffusion front and the speed of the steady state premixed flame. In particular, in the case of the flamelet combustion mechanism and strong turbulence, we state that the flame speed is very closed to the theoretical value of the diffusion front velocity, which is equal to the root mean square of turbulent velocity fluctuations.

## 1. Introduction

The classical paradigm of diffusion transfer modeling is based on parabolic equations with the turbulent diffusion coefficient  $D_t$  that is expressed in terms of Eulerian turbulent characteristics  $D_t = \text{const} \langle u'^2 \rangle^{1/2} \mathcal{L}$ , 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 here 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: *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 due to limited values of the instantaneous fluctuations of the velocity, which requires to use an hyperbolic equation instead of a parabolic one.

Though both these effects are discussed in the scientific literature (see, for example, [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 and, at the same time, the front speed is large with respect to the reference time and velocity of the process, respectively. But these effects play an important role in the theory of turbulent premixed combustion. In fact, an accurate description of nonequilibrium behavior of the turbulent diffusion coefficient, i.e. its growing in time, is important during the initial transient stage of the combustion process in the SI engine and laboratory bombs with spark ignition (as well as in initial instants of the Bunsen flame),

and the velocity of the diffusion front is one of the main parameters that controls the speed of the steady state premixed flame. Mathematical analysis of these effects, using statistical tools, is the subject of this paper.

In the first part of the paper we derive the parabolic equation with time-dependent diffusion coefficient and chemical source expressed in terms of the Lagrangian probability density function (PDF) and other Lagrangian characteristics of turbulence. We show that the turbulent diffusion coefficient in this equation does not depend on chemical transformation. 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, as it is discussed sometimes in the literature [3]. However, our exact mathematical analysis proves that it is not so and this 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 turbulent diffusion coefficient [4, 5].

In the second part of the paper, following [6, 7], on the basis of exact unclosed hydrodynamical equations we derive an hyperbolic equation with time dependent coefficients which does not contain parameters to be empirically determined and this is a striking property of the obtained equation. This equation gives a diffusion front velocity  $\mathcal{U}_f$  that equals  $\langle u'^2 \rangle^{1/2}$  and permits to have dispersion properties in agreement with the Taylor theory [8] for an arbitrary Lagrangian velocity fluctuation autocorrelation function. In particular, when constant coefficients are considered, the Taylor statement for the dispersion is satisfied by an exponential Lagrangian autocorrelation function. Moreover, when a chemical source is accounted for, the relationships between the front velocity and the speed of the steady state flame  $\mathcal{U}_t^{ss}$  are analyzed. This result is discussed especially with respect to the classical prediction in strong turbulence  $\mathcal{U}_t^{ss} \simeq \langle u'^2 \rangle^{1/2}$  [9, 10], when the combustion front speed does not depend on chemistry.

Since homogeneous, isotropic and stationary turbulence is considered, without loss of generality the one-dimensional case with zero mean velocity is analyzed in the paper, which is organized as follows. In Section 2 the Taylor theory is briefly reminded and a spread misleading application in reactive mixture is highlighted and discussed. In Section 3 the finite front velocity transport equation is derived and the role of the front velocity in the combustion process is analyzed. Finally in Section 4 the conclusions are given.

## 2. Parabolic transport equation with Lagrangian diffusion and source terms

### 2.1. Remind on the Taylor theory

The Taylor theory of turbulent diffusion [8] is an exact kinematical theory, unlike closure models of the equations of fluid dynamics. Given an 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 velocity fluctuation autocorrelation function, which is one of the statistical characteristics of turbulence. Since in this section we aim to discuss some misunderstandings on the application of the Taylor results, especially in combustion processes, in what follows we briefly remind the derivation of the main result.

The position of a fluid particle  $x$  is kinematically described by

$$x(t) = x_0 + \int_{t_0}^t v(\tau) d\tau, \quad (1)$$

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

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

where  $B_L(t - t_0) = \overline{v(t)v(t_0)}$ , with  $B_L(0) = \langle u'^2 \rangle$ , is the Lagrangian velocity fluctuation autocorrelation function and, from stationarity, it depends solely on the elapsed time. With the change of variables  $\tau = \tau_2 - \tau_1$  and  $s = (\tau_2 + \tau_1)/2$  the double integral in (2) reduces to

$$\overline{x^2}(t - t_0) = \int_{t_0-t}^0 [(t - t_0) + \tau] B_L(\tau) d\tau + \int_0^{t-t_0} [(t - t_0) - \tau] B_L(\tau) d\tau, \quad (3)$$

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

$$\overline{x^2}(t - t_0) = 2 \int_0^{t-t_0} [(t - t_0) - \tau] B_L(\tau) d\tau. \quad (4)$$

Let us consider the problem of turbulent transfer in the context of the parabolic diffusion equation. 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 the Gaussian function

$$P_L(x; t|x_0, t_0) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left\{ -\frac{(x - x_0)^2}{2\sigma^2} \right\}, \quad \sigma^2 = \sigma^2(t - t_0), \quad (5)$$

which satisfies the parabolic diffusion equation

$$\frac{\partial P_L}{\partial t} = D_t \frac{\partial^2 P_L}{\partial x^2}. \quad (6)$$

Substituting (5) in (6) and subsequently using (4) give

$$D_t(t - t_0) = \frac{1}{2} \frac{d\overline{x^2}}{dt} \quad (a), \quad D_t(t - t_0) = \int_0^{t-t_0} B_L(\tau) d\tau \quad (b), \quad (7)$$

from which it follows that  $D_t$  depends on time and it is a monotonically increasing function. From formula (7b) we have that  $D(0) = 0$  and the asymptotic expressions for small and large elapsed times are

$$D(t - t_0) = \langle u'^2 \rangle (t - t_0), \quad \text{when } (t - t_0) \ll T_L, \quad (8)$$

$$D(t - t_0) = \langle u'^2 \rangle T_L = \langle u'^2 \rangle^{1/2} \ell_L, \quad \text{when } (t - t_0) \gg T_L, \quad (9)$$

where

$$T_L = \int_0^\infty \frac{B_L(\tau)}{\langle u'^2 \rangle} d\tau \quad \text{and} \quad \ell_L = \langle u'^2 \rangle^{1/2} T_L$$

are the Lagrangian integral time and space scales of turbulence, respectively. Actually, as mentioned in the Introduction, the Eulerian approach to diffusion corresponds to the

assumption of the equilibrium of the turbulent diffusion coefficient and to the proportionality of the Lagrangian and Eulerian integral lengthscales  $\ell_L = \text{const } \mathcal{L}$ .

It is worth noting that Taylor formula (4) is valid only if the initial coordinate of particles, at the initial moment, is the same in all the realizations, then also (8-9) are valid only in this case. Below we discuss a misunderstanding connected with this constraint. In fact, any attempt to apply (9) together with (6) to situations where the initial position of particles is random (as for product particles in premixed turbulent flame) yields erroneous conclusions.

Let  $c(x, t)$  be the concentration field, in the case when each particle contains a unit amount of diffusing matter with an impulsive source  $c(x_0, t_0) = \delta(x - x_0)$ , the average concentration is described by the expression  $C(x, t) = \int P_L(x; t|x_0, t_0)\delta(x - x_0)dx_0$ , i.e. formula (5) is the Green function of the equation

$$\frac{\partial C}{\partial t} = D_t(t - t_0) \frac{\partial^2 C}{\partial x^2}. \quad (10)$$

When the mean concentration, in a fixed initial moment  $t = t_0$ , is distributed and known  $C(x, t_0) = S(x_0)$ , then the solution of Eq. (10) in terms of the Green function is

$$C(x, t) = \int_{-\infty}^{+\infty} P_L(x; t|x_0, t_0)S(x_0) dx_0. \quad (11)$$

We remind that, given an instant  $t = t^*$ , the Lagrangian PDF  $P_L(x; t^*|x_0, t_0)$  contains kinematical prehistory of turbulence during the temporal interval  $t_0 < t < t^*$  and, obviously, it does not depend on kinematical prehistory due to moments previous than  $t_0$ . So, for the validity of Eqs. (10) and (11), the initial condition  $C(x, t_0) = S(x_0)$  obligatory must not be yielded by kinematics in temporal interval previous than  $t_0$ .

The main misunderstanding is connected with ignoring this fact. A mistake is that if the solution to (10) in some intermediate instant, say  $t = t^* > t_0$ , is considered as an initial condition then the diffusion coefficient in (10) is changed in  $D_t(t - t^*)$ , i.e.  $D_t(t = t^*) = 0$ . But it is not like this, if this intermediate solution  $C(x, t^*) = S(x^*)$  is used as initial condition it is controlled also by the turbulence kinematics in  $t_0 < t < t^*$ . For this reason the correct diffusion coefficient in Eq. (10) is still  $D_t(t - t_0)$  rather than  $D_t(t - t^*)$ . Only in this case the descriptions of the diffusion with initial data  $C(x, t_0) = S(x_0)$  and  $C(x, t^*) = S(x^*)$  are identical for  $t > t^*$ . A simple example illustrates this statement.

Let us consider the initial condition  $C(x, t_0) = S_0\delta(x - x_0)$ , 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

$$C(x, t) = \int_0^t \int_{-\infty}^{+\infty} P_L(x; t|x_0, t_0)S(x_0, t_0) dx_0 dt_0.$$

Assume that  $S(t) = S_0 + (1 - S_0)H(t - t^*)$ , where  $H$  is the Heaviside function so that  $H(\alpha) = 0$  when  $\alpha < 0$  and  $H(\alpha) = 1$  when  $\alpha > 0$ . In the limit  $S_0 \rightarrow 0$  we have that  $C(x, t) = 0$  when  $t < t^*$  and  $C(x, t) > 0$  when  $t > t^*$ , i.e. the initial condition of the mean concentration is nonzero only at  $t = t^*$ , namely when the diffusive concentration appears in the turbulent medium, and it is  $C(x, t^*) = P_L(x, t^*)$ . Nevertheless the diffusion coefficient for fluid particles in the moment  $t = t^*$  is  $D_t(t^* - t_0) > 0$ .

In the context of the Lagrangian formulation of the turbulent diffusion problem with known initial data  $C(x, t_0) = S(x_0)$ , in the general case we cannot predict  $C(x, t > t_0)$ .

Only in the case of a deterministic initial condition, i.e. the identity of initial distributions in all realizations  $C_n(x, t_0) = S(x)$ , where  $n$  stands for the number of the realization, (definitely, in this case there is no prehistory with respect to  $t_0$  in the Lagrangian process of diffusion) and  $C(x, t)$  is described by (10) with  $D_t(t - t_0)$ . In the case of non deterministic initial data (with known PDF  $P(x, t_0)$  that yields initial mean distribution  $C(x, t_0) = S(x_0)$ ) we must additionally define if this initial condition is connected with previous prehistory or no: If “no” then the diffusion coefficient is  $D_t(t - t_0)$ ; if “yes” , i.e. the initial profile is formed by the prehistory, in simple cases we can resolve the problem using the idea of the *image source*. If, for example, the PDF is unimodal and  $C(x, t_0) = S(x_0)$  is the Gaussian function with known dispersion  $\sigma^2(t_0)$ , this initial data is the result of diffusion from the *image source* in previous moment  $t_{00} < t_0$  with variance  $\sigma^2(t_{00})$ . We can find the moment  $t_{00}$  from the Taylor equation ( $B_L(t)$  being supposed to be known) and hence the diffusion coefficient is equal to  $D_t(t - t_{00})$  so that  $D_t(t_0 - t_{00}) > 0$ . In the most general case, the number of such image source could be large or even infinite so, strictly speaking, large or infinite number of diffusion equations with different diffusion coefficients describe turbulent diffusion.

In chemically reacting system the concentration of liquid particles changes. In premixed combustion with the flamelet combustion mechanism, chemical transformation takes place on self traversing random surface (flamelet sheet). So in the flame it takes place a random stepwise instantaneous (described by the Heaviside function) change of the progress variable  $c$  (a generalized concentration) from  $c = 0$  in reactants to  $c = 1$  in products. The application of the Taylor theory for description of turbulent diffusion of the progress variable to this situation would be erroneous because the Taylor theory assumes, as we pointed out above, that the particle coordinate in the initial moment must be the same in all realizations. The problem of turbulent diffusion in the premixed flame with random generation of products, both in space and time, is mathematically addressed in the next section.

## 2.2. Turbulent transport equation for reactive mixture

Let  $c(x, t)$  be the *instantaneous* random concentration field. Following the probability theory, the “Eulerian” mean concentration, in the point  $x$  at time  $t$ , is determined by

$$C(x, t) = \int c P_c(c; t|x) dc, \quad (12)$$

where  $P_c(c; t|x)$  is the PDF to have the concentration value  $c$  at the time  $t$ , in the fixed point  $x$ . The quantity  $C(x, t)$  is the average of the amount of material brought in  $(x, t)$  by all particles of the mixture, without any selection on their initial conditions. Then, in other words, the mean concentration field  $C(x, t)$  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  $(x, c)$ , in analogy with the description of the joint stochastic process  $(x, v)$ , where  $v$  is the Lagrangian velocity, in the case of constant average density, the following integral relation can be derived by the Novikov theorem [11, 12]

$$P_c(c; t|x) = \int P(c, x; t|x_0, t_0) dx_0. \quad (13)$$

Using (13), definition (12) becomes

$$C(x, t) = \int \int c P(c, x; t|x_0, t_0) dx_0 dc. \quad (14)$$

In the most general case with statistical dependence between chemistry and particle kinematics, the joint  $(x, c)$  Lagrangian PDF  $P$  turns out to be

$$P(c, x; t|x_0, t_0) = f(c; t|x, x_0, t_0)P_L(x; t|x_0, t_0),$$

where  $f(c; t|x, x_0, t_0)$  is the Lagrangian PDF of concentration given by particles with initial condition  $(x_0, t_0)$  and  $P_L(x; t|x_0, t_0)$  is the transitional PDF of particles from  $(x_0, t_0)$  to  $(x, t)$ . In this case, formula (14) becomes

$$C(x, t) = \int \left\{ \int c f(c; t|x, x_0, t_0) dc \right\} P_L(x; t|x_0, t_0) dx_0,$$

where the term in brackets can be recognized as the Lagrangian average concentration due to the particles started in  $(x_0, t_0)$ , and, hereinafter, noted by

$$\bar{c}(t|x, x_0, t_0) = \int c f(c; t|x, x_0, t_0) dc. \quad (15)$$

Finally, the ‘‘Eulerian’’ ensemble averaged concentration  $C(x, t)$  can be re-written in the form

$$C(x, t) = \int \bar{c}(t|x, x_0, t_0) P_L(x; t|x_0, t_0) dx_0. \quad (16)$$

When  $P_L(x; t|x_0, t_0)$  is Gaussian, the evolution equation turns out to be Eq. (6) where the nonequilibrium turbulent diffusion coefficient  $D_t$ , as in (7b), is expressed in terms of the Lagrangian autocorrelation function  $B_L(t - t_0)$ , in accordance with the Taylor theory [8, 1].

Multiplying each side in (6) by  $\bar{c}(t|x, x_0)$  and integrating over  $dx_0$ , the following transport equation for reactive mixture is obtained

$$\frac{\partial C}{\partial t} = D_t(t - t_0) \frac{\partial^2 C}{\partial x^2} + \int \left\{ \left[ \frac{\partial \bar{c}}{\partial t} - D_t(t - t_0) \frac{\partial^2 \bar{c}}{\partial x^2} \right] P_L - 2 D_t(t - t_0) \frac{\partial \bar{c}}{\partial x} \frac{\partial P_L}{\partial x} \right\} dx_0, \quad (17)$$

where the integral term represents in general form the source expressed by Lagrangian functions (in contrast to usual representation with Eulerian functions, see e.g. [13]).

In the special case with statistical independence between chemistry and particle kinematics, the joint  $(x, c)$  Lagrangian PDF  $P$  turns out to be

$$P(c, x; t|x_0, t_0) = f(c; t|x_0, t_0)P_L(x; t|\mathbf{x}_0, t_0),$$

and the mean concentration field can be re-written as

$$C(\mathbf{x}, t) = \int \bar{c}(t|x_0, t_0) P_L(x; t|x_0, t_0) dx_0, \quad (18)$$

hence the transport equation is

$$\frac{\partial C(x, t)}{\partial t} = D_t(t - t_0) \frac{\partial^2 C(x, t)}{\partial x^2} + \int \frac{\partial \bar{c}}{\partial t} P_L(x; t|x_0) dx_0. \quad (19)$$

We here emphasize again that Eq. (7b) is valid only if the initial coordinates of particles, at the initial moment  $t = t_0$ , are the same in all realizations. In fact, any attempt to apply Eqs. (7b) to situations where the initial position of particles is random yields erroneous conclusions, as illustrated by the following example.

Assume that  $C(x, t_0) = 0$  at  $t \leq t^*$  and chemical transformation takes place only when  $t \geq t^*$ . In this case we can shift the initial condition to the moment  $t = t^*$  ( $C(x_0, t) = 0$ ), but it does not mean that the turbulent diffusion coefficient in Eq. (17) becomes  $D_t(t - t^*)$ . It remains  $D_t(t - t_0)$ , i.e. at the initial moment of the chemical transformation  $t = t^*$  the diffusion coefficient for products is  $D_t > 0$ .

### 2.3. Application to the turbulent premixed combustion

Let  $c$  be the progress variable ( $c = 0$  in reactants and  $c = 1$  in combustion products). 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 self-moving random surface with speed  $S_L$ , that is equal to the velocity of the normal laminar flame. When this surface crosses a fluid particle, an instantaneous variation of the progress variable from  $c = 0$  to  $c = 1$  for such particle occurs. Let  $t'$  be the random time when this transformation occurs, than  $c = 0$ , when  $t < t'$ , and  $c = 1$ , when  $t \geq t'$ .

In this case the Lagrangian average concentration  $\bar{c}$  can be determined as the sum of concentrations  $c = 1$  weighted by the PDF of the random reaction activation time

$$\bar{c}(t|x, x_0, t_0) = \sum 1 \cdot \Psi(t'|x, x_0, t_0), \quad (20)$$

and in integral form

$$\bar{c}(t|x, x_0, t_0) = \int_{t_0}^t \Psi(t'|x, x_0, t_0) dt', \quad (21)$$

where  $\Psi$  is the PDF of  $t'$ . In this framework, the Lagrangian PDF of  $c$ , i.e.  $f(c; t|x, x_0, t_0)$ , is a bimodal function of the type  $f(c; t|x, x_0, t_0) = \bar{c}\delta(1-c) + (1-\bar{c})\delta(c)$ , where  $\bar{c}$  is given in (21).

The average chemical source term  $W(x, t)$  is expressed by the integral in (17) and, noting that  $\partial\bar{c}(t|x, x_0, t_0)/\partial t = \Psi(t|x, x_0, t_0)$ , it is determined as

$$W(x, t) = \int \left\{ \left[ \Psi(t|x, x_0, t_0) - D_t(t-t_0) \frac{\partial^2}{\partial x^2} \int_{t_0}^t \Psi(t'|x, x_0, t_0) dt' \right] P_L - 2D_t(t-t_0) \frac{\partial}{\partial x} \int_{t_0}^t \Psi(t'|x, x_0, t_0) dt' \frac{\partial P_L}{\partial x} \right\} dx_0, \quad (22)$$

and the speed of the flame  $\mathcal{U}_t$  is determined by  $\mathcal{U}_t(t) = \int_{-\infty}^{+\infty} W(x, t) dx$ .

The consequence of this process is that, at sufficiently large time  $t \gg T_L$ , the turbulent diffusion coefficient, in agreement with (9), is practically constant  $D_t \simeq \langle u'^2 \rangle T_L = \langle u'^2 \rangle^{1/2} \ell_L$ , i.e. it is the same as in nonreacting systems at large elapsed time of diffusion. With other words, the instantaneous random appearing of products with  $c = 1$  in the flame does not influence the transfer process, i.e. conclusions about a zero initial diffusivity of volume with  $c = 1$ , which is based on erroneous application of the Taylor theory to product particles, are not theoretically justified.

In the case of strong turbulence with  $\langle u'^2 \rangle^{1/2} \gg S_L$ , the PDF of the flamelet sheet and hence the chemical source can be assumed Gaussian  $W = -\mathcal{U}_t(t) \exp(-0.5(x - x_0)^2/\sigma_f^2)/(2\pi\sigma_f^2)^{1/2}$ , where  $\sigma_f^2 = (\bar{x}^2 - x_0^2(t))^{1/2}$  and  $x_0(t) = \int_0^t \mathcal{U}_t(\tau) d\tau$  are the dispersion and the coordinate of the mathematical expectation of the sheet. These expressions are valid for times  $t \ll T$ , where  $T$  is a time-scale to be determined, when the influence of flamelet traveling with small speed  $S_L$  on the large-scale sheet wrinkles is negligible in comparison with the effect of the large velocity fluctuations connected with the energy eddies. In order to estimate  $T$ , we assume that the influence of  $S_L$  on the PDF takes place when the transfer due to the velocity fluctuations and the flamelet traveling with speed  $S_L$  are of the same order,  $(2\langle u'^2 \rangle/T_L T)^{1/2} \simeq S_L T$ , i.e.  $T \simeq 2T_L(\langle u'^2 \rangle^{1/2}/S_L)^2 \gg T_L$ .

Now we show that for times  $t \ll T$  the chemical transformation does not influence the turbulent flame width  $\delta_t \simeq \sigma_f$ .

The moment analysis gives

$$\frac{d\sigma_f^2}{dt} = 2D_t(t - t_0) + \int_{-\infty}^{+\infty} x^2 \frac{\partial W}{\partial x} dx - 2\mathcal{U}_t^2(t) t, \quad (23)$$

and, since  $\int x^2 \partial W / \partial x dx = 2\mathcal{U}_t^2(t) t$ , it follows that the flame width is independent of the chemical source and driven only by the passive dispersion. At the same time the flame speed  $\mathcal{U}_t(t)$  depends on both turbulence and chemistry because of the interaction of the traveling flamelet with the small-scale wrinkles, which are controlled by small eddies with  $\langle u_{se}^2 \rangle^{1/2} \leq S_L$ . We notice that at times  $T_L \ll t \ll 2T_L(\langle u^2 \rangle^{1/2}/S_L)^2$ , when small-scale wrinkles are already in statistical equilibrium and large-scale ones are still in nonequilibrium, flames have quasi constant speed and increasing width. A theoretical analysis of this situation is given in [7], and a comprehensive review of experimental data referring to such flames is presented in [14].

At  $t \gg 2T(\langle u^2 \rangle^{1/2}/S_L)^2$  the turbulent flame reaches the steady state regime. We will see in the next section that, in the flamelet combustion mechanism, the speed of the steady state flame  $\mathcal{U}_t^{ss}$  is mainly controlled by the velocity of the diffusive front  $\mathcal{U}_f$ . In order to perform the steady state analysis, an hyperbolic diffusion equation based on hydrodynamics is derived.

### 3. Turbulent Diffusion with Finite Front Velocity and Premixed Combustion

#### 3.1. An hyperbolic equation for turbulent transport

Diffusion with a finite front velocity is generally obtained in literature by the telegraph equation, which usually is derived on the basis of random walk arguments [1]. Here, following [6, 7], an hyperbolic equation with time dependent coefficient is derived on the basis of the unclosed equations for the average concentration and the turbulent flux of concentration fluctuation.

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

$$\frac{\partial u_i}{\partial t} + u_\alpha \frac{\partial u_i}{\partial x_\alpha} = -\frac{1}{\rho} \frac{\partial p}{\partial x_i} + \nu \frac{\partial^2 u_i}{\partial x_\alpha \partial x_\alpha}, \quad (24)$$

$$\frac{\partial c}{\partial t} + u_\alpha \frac{\partial c}{\partial x_\alpha} = 0, \quad (25)$$

where  $\rho$  is the fluid density,  $p(\mathbf{x}, t)$  the pressure field and  $\nu$  the kinematic viscosity, the molecular viscosity is neglected and the summation rule on the repeated Greek indexes is used.

Then, multiplying (25) by  $u_i$  and (24) by  $c$ , using the standard compositions rule, *instantaneous = average + fluctuation*, and stated  $U_i = 0$ , give respectively

$$\left\langle u'_i \frac{\partial c'}{\partial t} \right\rangle + \langle u'_i u_\alpha \rangle \frac{\partial C}{\partial x_\alpha} + \left\langle u'_i u_\alpha \frac{\partial c'}{\partial x_\alpha} \right\rangle = 0, \quad (26)$$

$$\left\langle c' \frac{\partial u'_i}{\partial t} \right\rangle + \left\langle c' u'_\alpha \frac{\partial u'_i}{\partial x_\alpha} \right\rangle = -\frac{1}{\rho} \left\langle c' \frac{\partial p'}{\partial x_i} \right\rangle + \nu \left\langle c' \frac{\partial^2 u'_i}{\partial x_\alpha \partial x_\alpha} \right\rangle. \quad (27)$$



Hence, averaging (25) and summing (26) and (27), the following system of equations is obtained

$$\frac{\partial C}{\partial t} + \frac{\partial \langle u'_\alpha c' \rangle}{\partial x_\alpha} = 0, \quad (28)$$

$$\frac{\partial \langle u'_i c' \rangle}{\partial t} + \langle u'_i u'_\alpha \rangle \frac{\partial C}{\partial x_\alpha} = - \frac{\partial \langle u'_\alpha u'_i c' \rangle}{\partial x_\alpha} - \frac{1}{\rho} \left\langle c' \frac{\partial p'}{\partial x_i} \right\rangle + \nu \left\langle c' \frac{\partial^2 u'_i}{\partial x_\alpha \partial x_\alpha} \right\rangle. \quad (29)$$

System (28-29) requires a closure. 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 (29) can be assumed to be independent of  $\nu$  and to be dependent on  $\langle u'_i c' \rangle$ , which includes the dependence on  $(\mathbf{x}, t)$ , and on a functional of  $B_L(t)$  which includes the parameter  $T_L$ , i.e.  $\varphi = \varphi(B_L(t), T_L) = \varphi(t, T_L)$ . 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. Finally, invoking the  $\Pi$ -Theorem [15]

$$\frac{\partial \langle u'_\alpha u'_i c' \rangle}{\partial x_\alpha} + \frac{1}{\rho} \left\langle c' \frac{\partial p'}{\partial x_i} \right\rangle - \nu \left\langle c' \frac{\partial^2 u'_i}{\partial x_\alpha \partial x_\alpha} \right\rangle = \frac{\varphi(t, T_L)}{T_L} \langle u'_i c' \rangle. \quad (30)$$

Closure (30) is different from that in [6, 7]. In fact, the present includes the time dependence that is fundamental to describe all dispersion regimes. Finally, the transport equation turns out to be

$$\frac{\partial^2 C}{\partial t^2} + \frac{\varphi(t, T_L)}{T_L} \frac{\partial C}{\partial t} = \frac{\partial \langle u'_i u'_\alpha \rangle}{\partial x_i} \frac{\partial C}{\partial x_\alpha} + \langle u'_\alpha u'_i \rangle \frac{\partial^2 C}{\partial x_i \partial x_\alpha}. \quad (31)$$

From homogeneity and isotropy  $\langle u'_i u'_\alpha \rangle = \langle u'^2 \rangle \delta_{i\alpha}$  and equation (32) becomes

$$\frac{\partial^2 C}{\partial t^2} + \frac{\varphi(t, T_L)}{T_L} \frac{\partial C}{\partial t} = \langle u'^2 \rangle \frac{\partial^2 C}{\partial x^2}, \quad (32)$$

where  $x$  stands for each Cartesian component  $x_i$ .

Analysis of the characteristics of (32) shows that the front velocity  $\mathcal{U}_f$ , for an arbitrary  $\varphi(t, T_L)$ , is

$$|\mathcal{U}_f| = \left| \frac{dx}{dt} \right| = \langle u'^2 \rangle^{1/2}. \quad (33)$$

The Lagrangian characteristics of the transport process (32) can be derived. In fact, for a  $\delta$ -function initial condition the particle displacement  $\overline{x^2}$  is  $\overline{x^2} = \int x^2 C(x, t) dx$ , and then multiplying (32) by  $x^2$  and integrating in  $dx$ , using definitions (7), gives

$$\frac{d^2 \overline{x^2}}{dt^2} + \frac{\varphi(t, T_L)}{T_L} \frac{d \overline{x^2}}{dt} = 2 \langle u'^2 \rangle. \quad (34)$$

Imposing the agreement with the Taylor statements on dispersion (7), equation (34) yields the following determination of  $\varphi(t, T_L)$

$$\varphi(t, T_L) = T_L \frac{\langle u'^2 \rangle - B_L(t)}{\int_0^t B_L(\tau) d\tau}. \quad (35)$$

This means that, with  $\varphi(t, T_L)$  defined as in (35), Eqs. (32) and (34) hold at all times and they meet the Taylor statements on turbulent dispersion for an arbitrary Lagrangian velocity fluctuation autocorrelation function  $B_L(t)$ .

Unlike the present analysis, the one developed in [6, 7] holds only for very large elapsed times because  $\varphi$  is taken constant. In fact, when  $t \rightarrow \infty$ ,  $B_L \rightarrow 0$  and  $\int_0^t B_L(\tau) d\tau \rightarrow \langle u'^2 \rangle T_L$ , from (35) follows that  $\varphi(t, T_L) \rightarrow 1$ . However, when an exponential autocorrelation function is chosen, i.e.  $B_L(t) = \langle u'^2 \rangle \exp(-t/T_L)$ , from (35) it follows that  $\varphi(t, T_L) = 1$  for all times and equation (32) reduces to

$$T_L \frac{\partial^2 C}{\partial t^2} + \frac{\partial C}{\partial t} = \langle u'^2 \rangle T_L \frac{\partial^2 C}{\partial x^2}, \quad (36)$$

that is the telegraph equation, mathematically an hyperbolic equation with constant coefficient.

Equation (36) has a number of good properties for turbulent transport modelling: *i*) it is an 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, *iv*) it describes a diffusion process with finite velocity front.

### 3.2. A relationship between the velocities of turbulent diffusion front and turbulent premixed flame in the flamelet combustion mechanism

In order to analyze premixed combustion, we equip the hyperbolic transport equation (36) with an average chemical source term  $W(x, t)$  as it follows

$$T_L \frac{\partial^2 C}{\partial t^2} + \frac{\partial C}{\partial t} = \langle u'^2 \rangle T_L \frac{\partial^2 C}{\partial x^2} + W(x, t). \quad (37)$$

Assume that at the initial moment  $t = t_0$  we have the stepwise distribution:  $c = 0$  at  $x > 0$  and  $c = 1$  at  $x < 0$ .

In the case of dispersion without combustion ( $W = 0$ ), the leading points of the mixing boundary layer are passive and they move with the front velocity  $\mathcal{U}_f = \langle u'^2 \rangle^{1/2}$ , while when a flame exists they travel with constant speed  $S_L$  in respect of the reactants, then, if the flamelet combustion mechanism with constant flamelet speed  $S_L$  is considered, the velocity of the front edge of the turbulent flame turns out to be  $\mathcal{U}_{fe} = \mathcal{U}_f + S_L = \langle u'^2 \rangle^{1/2} + S_L = \text{const}$ .

Moreover, the turbulent flame velocity (volume consumption speed)  $\mathcal{U}_t$  depends on time as  $\mathcal{U}_t(t) = S_L A(t)/A_0$ , where  $A(t)/A_0$  is the average dimensionless flame sheet area. At the initial time the dimensionless flame sheet is  $A(t_0)/A_0 = 1$ , so that  $\mathcal{U}_t(t_0) = S_L$ , but the wrinkling of the sheet and the increasing of flame width causes the growing in time of the ratio  $A(t)/A_0$  and the corresponding increasing of the  $\mathcal{U}_t(t)$  up to reach the asymptotic steady state value  $\mathcal{U}_t^{ss}$ . Since in the stationary traveling flame all the iso-surfaces has the same speed, the value of the steady state flame velocity is  $U_t^{ss} = \mathcal{U}_{fe} = \langle u'^2 \rangle^{1/2} + S_L$ .

In order to describe this qualitative behavior of the flame speed, we introduce the source in equation (37) as the sum of two contributions:  $W(x, t) = W_1(x, t) + W_2(x, t)$ , where the first term controls the consumption rate, i.e.  $\int_{-\infty}^{+\infty} W_1(x, t) dx = \mathcal{U}_t(t)$ , while the second term yields the front edge speed  $U_{fe} = \langle u'^2 \rangle^{1/2} + S_L = \text{const}$  but it does not give contribution to the consumption rate, i.e.  $\int_{-\infty}^{+\infty} W_2(x, t) dx = 0$ . In order to have from

characteristics analysis the front edge speed of the flame equal to  $\langle u'^2 \rangle^{1/2} + S_L$ , equation (37) becomes

$$T_L \frac{\partial^2 C}{\partial t^2} + \frac{\partial C}{\partial t} = \left( \langle u'^2 \rangle^{1/2} + S_L \right)^2 T_L \frac{\partial^2 C}{\partial x^2} + W_1(x, t), \quad (38)$$

and  $W_2$  is uniquely determined as  $W_2 = (2\langle u'^2 \rangle^{1/2} + S_L) S_L T_L \partial^2 C / \partial x^2$ .

In the case of strong turbulence ( $\langle u'^2 \rangle^{1/2} \gg S_L$ ), the speed of the front edge of the flame is practically equal to the velocity of the diffusion front, i.e.  $U_t^{ss} \cong U_f = \langle u'^2 \rangle^{1/2}$ , that corresponds and refines the classical intuitive estimation  $U_t^{ss} \sim \langle u'^2 \rangle^{1/2}$  [9, 10], and the flame speed does not depend on chemistry. At the same time, the width of the flame, adapted to this speed, obviously depends on  $S_L$  (the smaller  $S_L$  the larger  $\delta_t$  and vice versa). In the case  $\langle u'^2 \rangle^{1/2} \simeq S_L$ , the flame speed is  $U_t^{ss} = \langle u'^2 \rangle^{1/2} + S_L$  and then  $U_t^{ss} > U_f$ , i.e. the flame velocity is larger than the speed of the diffusion front. The opposite situation, when  $U_t^{ss} < U_f = \langle u'^2 \rangle^{1/2}$ , is possible in the case of distributed combustion where coupled transfer and chemical processes yield a turbulent flame with a speed that is insufficient to reach the faster traveling diffusion front, so the diffusion front does not influence the combustion. In this case both the flame speed and width depend on turbulence and chemistry.

## 4. Summary and Conclusions

1. In this paper we have analyzed Lagrangian properties of turbulent diffusion that, even if they are ignored in many engineering works devoted to heat and mass transfer, are important in several fields, in particular, in the theory of turbulent combustion. Our analysis refers to the turbulent premixed flame where (i) the transient behaviour of Lagrangian turbulent diffusion coefficient is relevant for initial forming of developed flame (this is important, in particular, for accurate modelling of initial stage of combustion in the SI engine) and (ii) the existence of the traveling front of turbulent diffusion (connected with finite turbulent velocity of Lagrangian fluid particles) that plays decisive role on the speed of the steady state flame in the flamelet combustion mechanism.

2. We have considered one-dimensional situations in homogeneous, isotropic and stationary turbulence with constant density. After a short remind of the derivation of the main result of the Taylor theory of Lagrangian diffusion, to point out the conditions when it is valid, we have stressed the fact that the classical Taylor expression describing the variance of diffusing particles  $\sigma^2$  in terms of the Lagrangian autocorrelation function  $B_L(t)$ , and corresponding to a turbulent diffusion coefficient that increases in time from zero to a constant value, is valid only when the initial position of the particle is the same in all realizations. Then, the estimation of diffusivity, using the Taylor theory, of the products of combustion (or of the progress variable), that randomly appear in the turbulent premixed flame, is not theoretically justified and any claim on zero diffusivity of products in the moment of their appearance is erroneous.

3. In the context of the parabolic balance equation in terms of the progress variable, we have proved that the Lagrangian turbulent diffusion coefficient does not depend on chemical transformation and it is identical to the case of nonreacting systems. Though this result is valid for arbitrary chemical source term, that here differently from other papers is expressed in terms of Lagrangian functions, we have attracted special attention to a traveling premixed flame with flamelet combustion mechanism and stepwise initial condition. We have showed that instantaneous transformation of reactants in products, which takes place in strongly wrinkled flamelet sheet, does not influence the turbulent

diffusion coefficient. In the case of strong turbulence  $\langle u'^2 \rangle^{1/2} \gg S_L$  we have additionally proved that at times  $t \ll T_L(\langle u'^2 \rangle^{1/2}/S_L)^2$  the increasing widths of the premixed flame and mixing layer (for initial stepwise profile) are practically the same, while at  $t > T_L(\langle u'^2 \rangle^{1/2}/S_L)^2$  the flame width is smaller and becomes asymptotically constant. The velocity of this asymptotic premixed flame strongly depends on the speed of the front of turbulent diffusion, which is a Lagrangian characteristic of turbulent transfer.

5. An hyperbolic equation of turbulent diffusion that follows from unclosed moment equation, where solely the Lagrangian autocorrelation function  $B_L(t)$  is assumed to be known, has been derived. More, we have showed that (i) in the case of an exponential  $B_L(t)$  the hyperbolic equation has known constant coefficients, which are expressed in terms of  $\langle u'^2 \rangle^{1/2}$  and  $T_L$  and it satisfies exactly the Taylor theory, and (ii) for an arbitrary  $B_L(t)$  the hyperbolic equation exactly satisfies the Taylor theory with time-dependent coefficients that are expressed in terms of the autocorrelation function  $B_L(t)$ . In all cases, the speed of the diffusion front is  $U_f = \langle u'^2 \rangle^{1/2}$ .

6. We have shown that, in the premixed flame asymptotic steady state regime, the flame speed is  $U_t^{ss} \cong U_f = \langle u'^2 \rangle^{1/2}$ , which at strong turbulence  $\langle u'^2 \rangle^{1/2} \gg S_L$  corresponds and refines the classical intuitive estimation  $U_t^{ss} \sim \langle u'^2 \rangle^{1/2}$  [9, 10]. At the same time it is possible, in the case of distributed combustion mechanism, that  $U_t^{ss} < U_f$  and the more fast traveling diffusion front does not influence, at large times, the speed of the turbulent premixed flame.

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