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To cite this version:

A.N. Lipatnikov, A. Sabelnikov. Scalar flux at the leading edge of premixed turbulent flame brush.. 8th Mediterranean Combustion Symposium (MCS 2013), Sep 2013, IZMIR, Turkey. hal-01061349

HAL Id: hal-01061349

https://onera.hal.science/hal-01061349

Submitted on 5 Sep 2014

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SCALAR FLUX AT THE LEADING EDGE OF PREMIXED TURBULENT FLAME BRUSH

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Abstract
It is widely accepted that turbulent scalar flux can show the countergradient behavior almost everywhere within a premixed flame brush with the exception of a narrow zone at the leading edge (LE) of the flame where the flux always shows the gradient behavior. Moreover, many experts consider the existence of such a zone to be of crucial importance in order for the flame to be able to propagate into unburned mixture. The goal of the present work is to dispute this widely-recognized belief by studying an asymptotic case of density variations localized to infinitely thin, wrinkled flamelets that separate unburned and burned mixture and self-propagate at a finite speed into the former mixture. First, simple mathematical and physical examples are discussed in order to argue that a premixed flame can propagate into unburned mixture even if averaged scalar flux does not show the gradient behaviour at the LE. This phenomenon is associated with the straightforward influence of large-scale velocity oscillations (i.e. turbulent diffusion as far as a premixed turbulent flame is concerned) on the mean rate of product creation at the LE. Second, by considering a fully-developed, statistically stationary, planar, one-dimensional turbulent premixed flame, the following criterion is obtained. Turbulent scalar flux shows the countergradient behavior at the LE if turbulent burning velocity is less than the laminar flame speed multiplied by the density ratio and by a factor that (i) is equal to unity if perturbations of the local burning rate in flamelets are disregarded, but (ii) can substantially depend on the Lewis number and preferential diffusion effects if such perturbations are taken into account. Third, development of a premixed turbulent flame and straining of the flame brush by non-uniform mean flow are argued to suppress countergradient scalar flux at the LE.

Introduction
Since the pioneering work by Clavin and Williams [1], Moss [2], Tanaka and Yanagi [3], and Libby and Bray [4], turbulent transport of a scalar quantity q in a premixed flame is well known to occur not only in the direction of a decrease in the mean value $\bar{q}$ of this quantity, as happens in many non-reacting flows, but also in the opposite direction, i.e. $\overline{\rho \mathbf{u}'' q''} \cdot \nabla \bar{q} > 0$. Here, ρ is the density, $\mathbf{u}$ is the flow velocity vector, overlines and overbars designate the Reynolds average, e.g. $\bar{q}$ with $q' = q - \bar{q}$, and $\tilde{q} = \overline{\rho q}/\bar{\rho}$ is the Favre-averaged value of the scalar q with $q'' = q - \tilde{q}$.

The latter phenomenon, called often countergradient diffusion, is commonly associated with a higher magnitude $|\bar{\mathbf{u}}_b|$ of velocity conditioned on burned mixture than the magnitude $|\bar{\mathbf{u}}_u|$ of velocity conditioned on unburned mixture. Indeed, because the probability γ of finding intermediate (between unburned and burned) states of a reacting mixture is much lower than unity in many weakly and moderately turbulent premixed flames [5, 6], the well-known BML approach [4, 7, 8, 9] is commonly considered to be a reasonable approximation. Within its framework, (i) the combustion progress variable c is introduced

($0 \leq c \leq 1$ with $c = 0$ and 1 in unburned and burned mixture, respectively) so that the probability of finding the burned mixture is equal to the Reynolds-averaged $\bar{c}$ and (ii)

$$\overline{\rho \mathbf{u}'' c''} = \bar{\rho}\tilde{c}(1-\tilde{c})(\bar{\mathbf{u}}_b - \bar{\mathbf{u}}_u) + O(\gamma) \tag{1}$$

i.e. the direction of the flux $\overline{\rho \mathbf{u}'' c''}$ is controlled by the direction of the slip velocity vector $\Delta \mathbf{u} \equiv \bar{\mathbf{u}}_b - \bar{\mathbf{u}}_u$. For a statistically planar one-dimensional flame that propagates from right to left (or from left to right), $\partial \tilde{c}/\partial x > 0$ (< 0) and the conditioned velocities $\bar{u}_u$ and $\bar{u}_b$ are positive (negative), i.e. $\overline{\rho \mathbf{u}'' c''} \cdot \nabla \tilde{c} > 0$ if $|\bar{u}_b| > |\bar{u}_u|$. DNS data [10, 11, 12] indicate that the scalar flux and the slip velocity vector have the same direction even if the probability γ is not negligible.

As reviewed elsewhere [6], countergradient scalar flux (or $|\bar{\mathbf{u}}_b| > |\bar{\mathbf{u}}_u|$) was documented in many premixed turbulent flames. Nevertheless, it is widely assumed that countergradient scalar flux cannot occur at the leading edge (LE) of a turbulent flame brush. Moreover, many experts consider the existence of the region of gradient diffusion, i.e. $\overline{\rho \mathbf{u}'' q''} \cdot \nabla \bar{q} < 0$ at $\tilde{c} \to 0$, to be the necessary condition in order for the flame to be able to propagate into unburned mixture. The goal of the present work is to dispute this widely-recognized belief.

A Key Simplification

It is worth stressing that, if the opposite is not specified, the following discussion will address an asymptotic case of an **infinitely thin** flame front (flamelet) that separates unburned and burned mixtures in a turbulent flow and self-propagates at a finite speed S_L with respect to the unburned gas. In other words, flamelet thickness δ_L asymptotically vanishes. This key assumption has two important consequences.

First, it allows us to skip the molecular diffusion in the c-balance equation [13], i.e. the contribution of the molecular diffusion to the total flux of c is beyond the scope of the present work.

Second, if $\delta_L \to 0$, the BML equations are asymptotically exact, i.e. the last term $O(\gamma)$ on the Right Hand Side (RHS) of Eq. (1) asymptotically vanishes everywhere and the direction of the flux $\overline{\rho \mathbf{u}'' c''}$ is solely controlled by the direction of the slip velocity. In the case of a finite thickness δ_L, the term $O(\gamma)$ could play a role at the leading and trailing edges of the flame brush, because the first term on the RHS of Eq. (1) vanishes as $\tilde{c} \to 0$ or $\tilde{c} \to 1$.

The discussed assumption is equivalent neither to an assumption of a high Reynolds number nor to an assumption of $D = 0$. If the molecular diffusivity D is equal to zero, then, a flamelet cannot propagate, as the laminar flame speed S_L vanishes. To the contrary, the considered asymptotic case of $\delta_L \to 0$ and a finite S_L can be realized if (i) a chemical time scale and D that characterize a typical fuel-air mixture are multiplied with ε and (ii) $\varepsilon \to 0$. It is worth stressing that the case of $\delta_L \to 0$ and a finite S_L is widely recognized to be a valuable scientific problem. For instance, the seminal theory of hydrodynamical instability was developed by Darrieus [14] and Landau [15] (see also textbooks [16, 17]) by studying the asymptotic case of $\delta_L \to 0$ and a finite S_L.

The invoked assumption of $\delta_L \to 0$ will allow us to significantly simplify discussion, but it seems to be of a minor importance for the main goal of the following dispute, because, to the best of the present authors' knowledge, the constraint of gradient turbulent transport at the LE is commonly associated with neither the contribution of molecular diffusion to the total flux of c nor with the term $O(\gamma)$. To the contrary, this constraint and constraint of $\Delta \mathbf{u} \cdot \nabla \tilde{c} > 0$ were considered to be equivalent in many papers.

Flame Can Propagate in the Case of Countergradient Scalar Flux at the LE

Two mathematical examples

Let us consider the following continuity

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x}(\bar{\rho}\tilde{u}) = 0 \tag{2}$$

and combustion progress variable balance equations

$$\bar{\rho}\frac{\partial \tilde{c}}{\partial t} + \bar{\rho}\tilde{u}\frac{\partial \tilde{c}}{\partial x} = -\frac{\partial}{\partial x}\overline{\rho u'' c''} + \overline{W}, \tag{3}$$

which model a statistically planar 1D premixed turbulent flame, and (ii) invoke the following widely-used closure relations for the mean mass rate of product creation

$$\overline{W} = \left(\frac{\bar{\rho}}{\rho_u}\right)^n \frac{\rho_u \tilde{c}(1-\tilde{c})}{\tau_f} \tag{4}$$

and the turbulent scalar flux

$$\overline{\rho u'' c''} = -\bar{\rho} D_t \frac{\partial \tilde{c}}{\partial x} + \bar{\rho} N_B \left(\frac{D_t}{\tau_f}\right)^{1/2} \tilde{c}(1-\tilde{c}). \tag{5}$$

Here, t is time, x is spatial distance, $D_t > 0$ is turbulent diffusivity, τ_f is a flame time scale, $n = 0$ or 2, and N_B is a non-dimensional input parameter that (i) characterizes the magnitude of the countergradient contribution to the turbulent scalar flux $\overline{\rho u'' c''}$ and (ii) is associated with the well-known Bray number [9, 10]. The mean density is equal to [7]

$$\bar{\rho} = \frac{\rho_u}{1 + (\sigma - 1)\tilde{c}}, \tag{6}$$

where $\sigma = \rho_u/\rho_b$ is the density ratio.

For a flame that propagates from right to left, the boundary conditions read

$$\tilde{c}(-\infty, t) = 0, \qquad \tilde{c}(\infty, t) = 1. \tag{7}$$

Equation (4) subsumes closure relations invoked in many numerical studies of turbulent combustion, e.g. see Ref. [18] or Table 1 in Ref. [19]. Following Chomiak and Nisbet [20] and Veynante et al. [10], Eq. (5) divides the flux $\overline{\rho u'' c''}$ into turbulent diffusion and pressure-driven countergradient transport and invokes the gradient closure of the turbulent diffusion, while the last term on the RHS is basically similar to the model of the countergradient transport, developed by Veynante et al. [10]. Therefore, the problem given by Eqs. (2)-(7) is highly relevant to modeling of premixed turbulent combustion.

This problem is studied analytically and numerically elsewhere [21]. Here, we restrict ourselves to a brief summary of results relevant to the subject of the present dispute.

First, if D_t and τ_f are constant, there is the following explicit traveling wave solution

$$\tilde{c} = \frac{1}{1 + \mathrm{e}^{-4\xi}}, \qquad \xi = \frac{x + U_t t}{\Delta_t} \tag{8}$$

to Eqs. (2)-(7), where the turbulent burning velocity and the mean flame brush thickness are equal to

$$\frac{U_t}{\sqrt{D_t/\tau_f}} = \begin{cases} 1/N_B & \text{if } n = 0 \\ 2(1-\alpha)/(N_B + \sqrt{N_B^2 - 4\alpha}) & \text{if } n = 2 \text{ and } N_B \geq 2\sqrt{\alpha} \end{cases} \tag{9}$$

and

$$\frac{\Delta_t}{\sqrt{D_t\tau_f}} = \begin{cases} 4/N_B & \text{if } n=0 \\ 8/(N_B+\sqrt{N_B^2-4\alpha}) & \text{if } n=2 \text{ and } N_B \geq 2\sqrt{\alpha} \end{cases}, \tag{10}$$

respectively. Here, $0 \leq \alpha = (\sigma-1)/\sigma < 1$. Note that the profile of $\tilde{c}(\xi)$ given by Eq. (8) is widely accepted in order to parametrize experimental data obtained from various turbulent flames, as reviewed elsewhere [5].

The use of constant (independent of x and t) time τ_f, length $\sqrt{D_t\tau_f}$, and velocity $\sqrt{D_t/\tau_f}$ scales when obtaining the above explicit solution implies that the turbulence is stationary and homogeneous, i.e. combustion does not affect it. For the goals of the present study, such a simplification is fully justified, because the hypothesis that $\overline{\rho\mathbf{u}''c''}\cdot\nabla\tilde{c} < 0$ at the LE of a premixed flame brush is not based on an analysis of the influence of combustion on turbulent time, length, and velocity scales.

Second, if the initial conditions are sufficiently steep (e.g. a step function) and N_B is larger than a certain critical value N_B^{cr}, i.e. $N_B^{cr}(n=0)=1$ and $N_B^{cr}(n=2)=1+\alpha$, then, a solution to the initial value problem tends to the above explicit solution as $t/\tau_f \to \infty$.

Substitution of Eqs. (8)-(10) into Eq. (5) shows that, if $N_B \geq N_B^{cr}(n)$, then,

$$\frac{\overline{\rho u''c''}}{\rho_u\sqrt{D_t/\tau_f}} = \begin{cases} 0 & \text{if } n=0 \text{ and } N_B \geq 1 \\ 0.5\bar{\rho}\tilde{c}(1-\tilde{c})(N_B-\sqrt{N_B^2-4\alpha}) > 0 & \text{if } n=2 \text{ and } N_B \geq 1+\alpha \end{cases}. \tag{11}$$

The former ($n=0$) example proves that a combustion wave modeled by Eqs. (2)-(7) with $N_B \geq 1$ can propagate even if the transport term on the RHS of Eq. (24) vanishes in the entire flame brush. The latter ($n=2$) example proves that a combustion wave modeled by Eqs. (2)-(7) with $N_B \geq 1+\alpha$ can propagate even if the flux $\overline{\rho u''c''}$ shows the countergradient behavior everywhere within the flame brush, including its LE. Thus, the two examples do prove that, from the purely mathematical viewpoint, the constraint of $\overline{\rho\mathbf{u}''c''}\cdot\nabla\tilde{c} < 0$ at $0 < \tilde{c} < \varepsilon \ll 1$ is not a necessary condition for a premixed turbulent flame to propagate into unburned gas. The constraint could be the necessary condition only for particular models of the mean rate $\overline{W}$, e.g. for models that yield $\overline{W}=0$ if $\tilde{c} < c_0$.

In the subsequent subsections, the above analytical results will be supported by simple physical models and reasoning.

Darrieus-Landau Instability

Although the well-known problem of Darrieus-Landau (DL) instability of a premixed flame addresses a laminar flow, the classical DL solution offers also an opportunity to demonstrate that the flux $\overline{\rho u''c''}$ on the RHS of Eq. (24) can show the countergradient behaviour at any $\tilde{c}$.

Indeed, Darrieus [14] and Landau [15] theoretically investigated the following problem; A laminar flame is reduced to an infinitely thin surface that separates unburned and burned mixtures and propagates at a constant speed S_L with respect to the unburned mixture. The flow outside the flame is governed by the non-reacting Euler equations, with the density being equal to either ρ_u or ρ_b, ahead or behind the flame, respectively. Jump conditions on the flame surface are used to close the model. A stability analysis of such a planar flame, reproduced in many textbooks [16, 17], shows that the flame is unconditionally unstable to infinitesimal ($kl \ll 1$) harmonic perturbations $l\sin(ky)$ of the x-coordinate of the flame surface, with the growth rate β of the amplitude of the perturbation being linearly increased by the wavenumber k, i.e. $\beta = S_L k f(\sigma)$ and

$l = l_0 e^{\beta t}$, with $f(\sigma > 1) > 0$ and $f(1) = 0$. Here, the x-axis is normal to the position of the unperturbed flame.

To draw an analogy with turbulent combustion, let us decompose the well-known DL solution, e.g. see Eq. (40) in [6], into (i) a mean part $\bar{q}(x,t) = \int_y^{y+2\pi/k} q(x,\eta,t)d\eta$ by integrating along the y-axis, which is parallel to the unperturbed flame surface, and (ii) an oscillating part $q'(x,y,t) = q(x,y,t) - \bar{q}(x,t)$. Then, the evolution of $\tilde{c}(x,t)$ is modeled by Eq. (24). Moreover, the BML equations hold in this case, because the laminar flame front is assumed to be infinitely thin in the DL analysis. Furthermore, the amplitude u' of perturbations in the x-component of the flow velocity is much less than S_L within the framework of the DL theory, i.e. $u'/S_L = O(kl) \ll 1$, see Eq. (40) in [6]. Therefore, the conditioned velocities $\bar{u}_u$ and $\bar{u}_b$ are approximately equal to S_L and σS_L, respectively, i.e. $\bar{u}_b > \bar{u}_u$. Accordingly, turbulent scalar flux evaluated using Eq. (1) shows the countergradient behavior at any $\tilde{c}$.

Thus, the analysis by Darrieus [14] and Landau [15] provides us with an example of a propagating transient flame that can be modeled by Eq. 24 with the flux $\overline{\rho u'' c''}$ being countergradient everywhere within the flame brush including its LE. This flame is unstable. An example of a stable flame associated with $\overline{\rho \mathbf{u}'' c''} \cdot \nabla \tilde{c} > 0$ for any $\tilde{c}$ is given below.

A Planar Oscillating Laminar Flame

Let us consider an infinitely thin planar laminar flame that oscillates in one-dimensional oncoming flow $u = S_L + s(t)$. Here, $s(t)$ is a random function such that (i) $\overline{s(t)} = 0$, (ii) $\int_0^\infty \overline{s(t)s(t+\theta)}d\theta = u'^2 T$ and $\overline{s(t)s(t)} = u'^2$ do not depend on time, (iii) the length scale $L = u'T$ of velocity oscillations is much larger than the laminar flame thickness. In this simple case, (i) the spatial profile of $\tilde{c}(x)$ is modeled by Eq. (24) and (ii) $\bar{u}_u = S_L$ and $\bar{u}_b = \sigma S_L$. Therefore, the turbulent scalar flux evaluated using Eq. (1) shows the countergradient behaviour in the entire flame brush.

In the considered case, the flame front retains its planar shape, $\bar{c}(x) = \int_{-\infty}^x \gamma d\zeta$, the probability γ of finding flame front is equal to $\Delta_t d\bar{c}/dx$, and, therefore, the rate $\overline{W} = \rho_u S_L d\bar{c}/dx$ is controlled by the gradient of $\bar{c}$, rather than by $\bar{c}$. Such a dependence of $\overline{W} \propto |\nabla \bar{c}|$ substantially changes the general properties of Eq. (24) as compared with the same balance equation closed by invoking an algebraic expression $\overline{W} = \overline{W}(\bar{c})$. In particular, in the former case, the flame speed does not depend on the transport term, which controls the mean flame thickness and structure.

Discussion

The above mathematical and physical examples show that the sign of $\overline{\rho \mathbf{u}'' c''} \cdot \nabla \tilde{c}$ at low $\tilde{c}$ is not a criterion of the ability of a premixed turbulent flame to propagate as far as an arbitrary source term $\overline{W} \geq 0$ on the RHS of Eq. (24) is concerned. To the contrary, in the theory of a premixed laminar flame of a finite thickness, the gradient behaviour of scalar flux at the cold boundary is necessary for the flame to propagate. This difference between laminar and turbulent combustion is associated with the following differences between the RHS of the c-balance equation

$$\rho \frac{\partial c}{\partial t} + \rho u \frac{\partial c}{\partial x} = \frac{\partial}{\partial x}\left(\rho D \frac{\partial c}{\partial x}\right) + W, \tag{12}$$

which describes a laminar premixed flame of a finite thickness, and the RHS of Eq. (24) which addresses a turbulent premixed flame.

First, from the mathematical viewpoint, the rate W in Eq. (12) is exponentially small at $c \ll 1$, whereas the mean rate $\overline{W}$ in Eq. (24) can be finite at $\tilde{c} \ll 1$, e.g. Eq. (4) yields $\overline{W}(\tilde{c} \to 0) \propto \tilde{c}$.

Second, from the physical viewpoint, the transport and source terms on the RHS of Eq. (12) model two totally different physical mechanisms, molecular transport and chemical reactions, respectively. Balance between these two mechanisms controls the c-flux from a thin reaction zone to a thicker preheat zone associated with vanishing reaction rate W [17]. The c-flux, in turn, controls both the laminar flame speed S_L and thickness Δ_L. Therefore, the balance between the molecular transport and chemical reaction terms controls both S_L and Δ_L.

To the contrary, the transport and source terms on the RHS of Eq. (24) are associated with basically the same physical mechanism, i.e. random convection of flamelets by turbulence, even if there are two different manifestations of this mechanism; (i) an increase in flame brush, controlled by large-scale eddies (turbulent diffusion) and (ii) an increase in the flamelet surface area due to wrinkling of the surface by turbulence. Accordingly, at least in the asymptotic case of infinitely thin flamelets, the underlying physics of the propagation of a premixed turbulent flame can basically differ from the underlying physics of the propagation of a premixed laminar flame.

To show this basic difference in a clear manner, let us consider a simple case of an interface that self-propagates at a speed S_L in constant-density Kolmogorov turbulence. The speed S_t of the leading edge of the "flame brush" formed due to random motion of the interface is mainly (if $u' \gg S_L$) controlled by throwing the interface far into unburned mixture by large-scale turbulent eddies. Because exactly the same physical mechanism controls the growth rate of a turbulent mixing layer, this mechanism is associated with turbulent diffusion.[1] As soon as the interface arrives at a point x_1 associated with $0 < \tilde{c}(x = x_1) \ll 1$, the mean reaction rate $\overline{W}(x = x_1)$ becomes positive, because the local reaction rate $\rho_u S_L |\nabla c|$ is infinitely high at the infinitely thin interface. Therefore, in the considered constant-density case, turbulent diffusion affects not only the propagation of the LE of the flame brush, but also the mean flame surface density $\overline{\Sigma} = \overline{|\nabla c|}$ and, hence, $\overline{W} = \rho_u S_L \overline{\Sigma}$ at the leading edge (in the middle of the flame brush, the surface density is mainly controlled by the balance between an increase in Σ due to wrinkling of the interface by small-scale turbulent eddies and a decrease in Σ due to collisions of different segments of the highly-wrinkled self-propagating interface). Consequently, because, at the LE of a turbulent flame brush, a correct closure of the mean rate $\rho_u S_L \overline{|\nabla c|}$ should allow for the influence of turbulent diffusion on it, the flame can propagate even if the transport term on the RHS of Eq. (24) vanishes or shows the countergradient behavior, as proved in the beginning of this section.

Moreover, if the thickness Δ_t of the mean flame brush is constant, the speed S_t of its leading edge should be equal to the turbulent burning velocity $U_t = \rho_u^{-1} \int_{-\infty}^{\infty} \overline{W} dx$. Therefore, if (i) $u' \gg S_L$, (ii) effects of turbulence on S_L, including local combustion quenching, are neglected (such a simplification is justified for infinitely thin flamelets, because response of a laminar flame to a stretch rate $\dot{s}$ scales as $\delta_L \dot{s}/S_L$ [23]), and (iii) collisions of flamelets are disregarded at $\tilde{c} \to 0$ for geometrical reasoning; then, it is tempting to assume that solely turbulent diffusion controls burning velocity. Accordingly, a balance between an increase in the interface surface area by turbulent wrinkling and

[1] Even if $u' \gg S_L$, the large-scale motion of a self-propagating interface differs from the large-scale motion of a material surface, because, due to merging of the interface elements, its surface area is much less than the area of the material surface, with all other things being equal [22].

turbulent transport appear to control Δ_t, but not U_t, at least in the case of a fully-developed flame brush formed due to random motion of a self-propagating, infinitely thin interface in constant-density Kolmogorov turbulence.[2] Consequently, the well-known equality $\rho_u U_t = \int \overline{W} dx \propto \max\{\overline{W}\}\Delta_t$ does not necessitate that the burning velocity is controlled by the balance between the rate $\overline{W}$ and turbulent transport that controls the thickness Δ_t. The equality can also mean that both Δ_t and, maybe, $\overline{W}$ adjust themselves in order for $U_t = S_t$ in a fully-developed flame.

A randomly oscillating planar laminar flame addressed in the previous subsection gives a very simple example of an averaged combustion wave that moves at a mean speed S_L that is not affected by the transport term on the RHS of Eq. (24). This example also implies that even large-scale random motion of a locally planar flamelet can make the spatial profile of $\overline{W} = \rho_u S_L \overline{|\nabla c|}$ very different from the spatial profile of W in the counterpart unperturbed laminar flame. Accordingly, in a turbulent flow, the local value $\overline{W}(\mathbf{x}, t)$ of the mean rate of product creation can be affected not only by S_L and by wrinkling of flamelet surface by small-scale turbulent eddies, but also by large-scale random motion of flamelets, which is commonly associated with turbulent diffusion.

The above fundamental differences between laminar and turbulent premixed flames and, in particular, the straightforward dependence of the mean rate $\overline{W}$ on turbulent diffusion allow a premixed turbulent flame to propagate even if turbulent scalar flux shows the countergradient behaviour at the cold boundary.

In the case of a variable-density flame, countergradient scalar flux induced by heat release can statistically overwhelm turbulent diffusion under certain conditions, but does not annihilate it locally and does not make $\overline{W}$ independent of turbulent diffusion at the leading edge. Turbulent eddies can throw flamelets far into unburned gas even if $\overline{\rho \mathbf{u}'' c''}$ points to the opposite direction. Indeed, flamelet motion is controlled by the difference in S_L and the velocity of unburned gas before the flamelet, while $\overline{\rho \mathbf{u}'' c''}$ given by Eq. (1) is controlled by the difference in the velocities conditioned on burned and unburned mixture. The two differences can have opposite signs in a general case.

Direction of Turbulent Scalar Flux at the Cold Boundary

Application of Eq. (24) to a fully-developed, statistically stationary, planar and one-dimensional flame that propagates from right to left yields

$$\frac{d}{dx}\overline{\rho u'' c''} = \overline{W} - \bar{\rho}\tilde{u}\frac{d\tilde{c}}{dx} = \rho_u \overline{u_c|\nabla c|} - \rho_u U_t \frac{d\tilde{c}}{dx} = -\rho_u \left[\frac{\bar{u}_c}{\bar{n}_x} + \frac{\rho_u \rho_b}{\bar{\rho}^2} U_t\right] \frac{d\bar{c}}{dx} \tag{13}$$

in the coordinate framework attached to the flame provided that the well-known BML Eqs. (6) and $\rho_b \bar{c} = \bar{\rho}\tilde{c}$ and the standard definition of the mean unit normal vector $\mathbf{n}$ [25]

$$\bar{\mathbf{n}} = \frac{\overline{\mathbf{n}|\nabla c|}}{\overline{|\nabla c|}} = -\frac{\overline{\nabla c}}{\overline{|\nabla c|}} = -\frac{\nabla \bar{c}}{\overline{|\nabla c|}} \tag{14}$$

are invoked. Here, n_x is the x-component of the unit normal vector $\mathbf{n} = -\nabla c/|\nabla c|$ and $\bar{u}_c = \overline{u_c|\nabla c|}/\overline{|\nabla c|}$ is the surface-averaged consumption velocity u_c, which is equal to S_L in the asymptotic case of infinitely thin flamelets.

[2] Scaling $U_t \propto u'$, which results from this claim, does not hold in a typical premixed turbulent flame [24] due to a number of physical mechanisms disregarded above, e.g. flame development, local flamelet stretching and quenching, influence of heat release on turbulence, etc. However, the disregarded physical mechanisms do not make $\overline{W}(\tilde{c} \to 0)$ independent of large-scale velocity fluctuations.

If $\sigma(\bar{u}_c/|\bar{n}_x|)_{\bar{c}\rightarrow 0} > U_t$, then, Eq. (13) results straightforwardly in countergradient scalar flux at the LE of turbulent flame brush, because $\bar{n}_x < 0$ in a flame that propagates from right to left. Therefore, Eq. (13) yields the following criterion

$$N_{B,LE} = \frac{\sigma S_L}{\Psi U_t} = 1, \tag{15}$$

which resembles the well-known Bray number [9, 10] (especially if $U_t \propto u'$), but addresses the LE, rather than the entire flame brush. Turbulent scalar flux shows countergradient (gradient) behavior at the LE if $N_{B,LE}$ larger (smaller) than unity.

In the considered asymptotic case of infinitely thin flamelets, the function Ψ is simply equal to $|\bar{n}_x| \leq 1$. If, based on purely geometrical reasoning and on DNS data reported by Lee and Huh [26], we assume that $|\bar{n}_x| \rightarrow 1$ as $\tilde{c} \rightarrow 0$, then, $\Psi \rightarrow 1$ and turbulent scalar flux shows the countergradient behaviour at the LE of a fully-developed, statistically planar and one-dimensional, turbulent flame brush provided that $\sigma S_L > U_t$.

If we consider flamelet of a finite thickness, then, the local flamelet structure and burning rate can be affected by turbulent stretching [23], with such phenomena being of substantial importance for highly curved flamelets at the LE of a turbulent flame brush [27]. In particular, $\bar{u}_c$ can be larger (lower) than S_L for mixtures characterized by the Lewis number Le lower (larger) than unity, with a ratio of u_c/S_L can be of the order of ten in very lean hydrogen-air flames [27]. Therefore, the function $\Psi = \Psi(\mathrm{Le}, D_F/D_O)$ can depend on the Lewis number and a ratio D_F/D_O of molecular diffusivities of fuel and oxygen and can be both larger and smaller than unity. However, it is worth remembering that, in the case of a finite flamelet thickness, the molecular diffusion flux can play a role in Eq. (13) if $\tilde{c}$ is very small. Such an eventual effect is beyond the scope of the present paper. Moreover, contrary to the asymptotic case of $\gamma \rightarrow 0$, the equality of $\overline{W} = -\rho_u \bar{u}_c \bar{n}_x^{-1}(\partial\bar{c}/\partial x)$ used in Eq. (13) is not exact if the probability γ is finite.

Contrary to the above criterion obtained within the framework of the BML paradigm, solely gradient flux was observed at the cold boundary of a fully-developed, statistically planar, one-dimensional premixed turbulent flame in numerical studies performed within the framework of the same paradigm, e.g. see the classical paper by Libby and Bray [4], which is widely cited in order to support the disputed hypothesis of $\overline{\rho u'' c''} \cdot (\partial\tilde{c}/\partial x) < 0$ at $\tilde{c} \rightarrow 0$. It is worth remembering, however, that more recent contributions to premixed turbulent combustion revealed certain limitations of the model developed in the cited paper, which was pioneering for those days. In particular, in Ref. [4] and in other early BML papers, term $\overline{c''(\partial p'/\partial x)}$ was neglected in the balance equation for $\overline{\rho u'' c''}$ and computed countergradient scalar flux resulted solely from a stronger acceleration of "high-temperature, low-density products relative to the cold, high-density reactants" by the mean pressure gradient. Indeed, if we assume that $|\bar{u}_b| < |\bar{u}_u|$ at the cold boundary via an analogy with non-reacting turbulent flows, then, the different acceleration can result in $|\bar{u}_b| > |\bar{u}_u|$ only at certain distance Δx downstream the LE [28]. Therefore, if the correlation $\overline{c''(\partial p'/\partial x)}$ is neglected, then, turbulent scalar flux should show the gradient behaviour at sufficiently low $\tilde{c}$, in line with the seminal work by Libby and Bray [4]. However, as theoretically argued by Kuznetsov [29, 30] and well-supported by currently available DNS data reviewed elsewhere [6], the neglected term $\overline{c''(\partial p'/\partial x)}$ can dominate in the $\overline{\rho u'' c''}$-balance equation. This term serves to cause countergradient scalar flux at the LE of a turbulent flame brush. Indeed, when a flamelet arrive at a point x associated with $\tilde{c} \ll 1$, we have $c''(x,t) > 0$ and the local pressure gradient caused by heat release is negative if $\partial\tilde{c}/\partial x > 0$. Therefore, $(\partial p'/\partial x)(x,t) < 0$ and, hence, $-\overline{c''(\partial p'/\partial x)} > 0$ at the LE, thus, contributing to positive $\overline{\rho u'' c''}$ and $\overline{\rho u'' c''}(\partial\tilde{c}/\partial x)$.

The fact that, contrary to the above criterion, solely $\overline{\rho \mathbf{u}'' c''} \cdot \nabla \tilde{c} < 0$ was documented at $\tilde{c} \to 0$ in DNS studies of premixed turbulent flames that were claimed to be fully-developed, statistically planar and one-dimensional, e.g. see Ref. [12], can be explained as follows.

First, because those studies addressed flamelets of a finite thickness, Eq. (13) is not exact in this case, as noted above.

Second, the speed and thickness of a premixed turbulent flame obtained in a DNS study of combustion in statistically stationary, planar, one-dimensional turbulent flow can exhibit strong oscillations in time, e.g. see dependencies of $U_t(t)$ shown in Fig. 3.1 in Ref. [31], or Fig. 3 in Ref. [32], or Fig. 3 Ref. in [33]. Accordingly, such flames can be subject to transient effects not addressed by Eq. (13).

However, transient effects can promote gradient transport at the LE. For instance, if the mean flow is statistically stationary and one-dimensional, but the studied flame develops, i.e. its mean thickness grows, then, the unsteady term in Eq. (24) serves to impede countergradient scalar flux, but, moreover, $\bar{\rho}\tilde{u} \neq \rho_u U_t$. To allow for and compare the two effects, let us assume that, in line with numerous experimental data analyzed elsewhere [24, 34], the mean structure of the flame is self-similar, i.e. $\tilde{c}(x,t) = \tilde{c}(\xi)$, where the normalized distance ξ is determined by Eq. (8). Then, substitution of $\bar{\rho} = \bar{\rho}[\tilde{c}(\xi)]$ into the Favre-averaged continuity equation yields

$$\frac{\partial}{\partial \xi}\left(\bar{\rho}\tilde{u}\right) = \frac{d\bar{\rho}}{d\xi}\left(-U_t + \xi \frac{d\Delta_t}{dt}\right). \tag{16}$$

Integrating this equation from $-\infty$ to ξ, substituting the result and $\tilde{c}(\xi)$ into Eq. (24), and invoking the well-known BML Eqs. (6) and $\rho_b \bar{c} = \bar{\rho}\tilde{c}$, we arrive at

$$\frac{\partial}{\partial \xi}\overline{\rho u'' c''} = -\rho_u \left(\frac{\bar{u}_c}{\bar{n}_x} + \frac{\rho_u \rho_b}{\bar{\rho}^2} U_t\right)\frac{d\bar{c}}{d\xi} - \frac{\rho_u \rho_b}{\bar{\rho}^2}\frac{d\bar{c}}{d\xi}\frac{d\Delta_t}{dt}\left(\int_{-\infty}^{\xi} \zeta \frac{d\bar{\rho}}{d\zeta} d\zeta - \bar{\rho}\xi\right). \tag{17}$$

The sole difference between Eqs. (13) and (17) consists of the fact that the latter equation involves an extra unsteady term (the last term on the RHS), which is negative at the LE of the flame brush, because both ξ and $d\bar{\rho}/d\xi$ are negative therein. For instance, invoking Eq. (8), one can easily show that

$$-\int_{-\infty}^{\xi} \zeta \frac{d\bar{\rho}}{d\zeta} d\zeta - \bar{\rho}\xi \to \rho_u \xi < 0 \tag{18}$$

as $\xi \to -\infty$. Therefore, the growth of Δ_t impedes countergradient scalar flux at the cold boundary.

If $\tilde{c}(\xi) \to 0$ as $\xi \to -\infty$, but $\tilde{c}(\xi) > 0$ for any finite ξ, then, one can always find such a $\varepsilon \ll 1$, that the negative unsteady term dominates on the RHS of Eq. (17) for $\tilde{c} < \varepsilon$, because $|\xi|$ is large. Accordingly, if $\tilde{c} < \varepsilon$, then, $\overline{\rho u'' c''}(d\tilde{c}/d\xi) < 0$ in line with the hypothesis of gradient diffusion.

Non-uniformities of the oncoming mean flow can also impede countergradient scalar flux at the cold boundary. For instance, if the mean flame brush is strained by spatially nonuniform mean flow, as occurs e.g. in a turbulent premixed flame stabilized in an impinging jet, then, the magnitude of the mean flow velocity increases with decreasing $\tilde{c}$. Accordingly, at the LE of the flame brush, Eq. (13) should be substituted with the following equation

$$\nabla \cdot \overline{\rho \mathbf{u}'' c''} = -\rho_u \left(\frac{\bar{u}_c}{\bar{n}_x} + \frac{\tilde{u}}{\sigma}\right)\frac{d\bar{c}}{dx}, \tag{19}$$

where $\tilde{u}(\tilde{c} \ll 1)$ is significantly larger than U_t, as reviewed elsewhere [6]. For instance, experimental data by Bourguignon et al. [35] indicate that a ratio of $\tilde{u}(\tilde{c} \ll 1)/U_t$ can be as large as four in a premixed turbulent flame stabilized in an impinging jet. Tangential derivatives of the transverse components of the vector $\overline{\rho \mathbf{u}'' c''}$ on the left hand side of Eq. (19) can also affect the normal flux $\overline{\rho u'' c''}$ at $\tilde{c} \ll 1$, but the available numerical data [36, 37] imply that such effects are of minor importance as compared with the difference in $\tilde{u}(\tilde{c} \ll 1)$ and U_t. Consequently, a ratio of $\sigma \bar{u}_c/U_t$ should be significantly larger than unity in order for turbulent scalar flux to show the countergradient behaviour at low $\tilde{c}$ in a flame strained by a mean flow.

Conclusions

Exact analytical results and physical reasoning discussed in the paper prove that a constraint of the gradient behaviour of turbulent scalar flux at the leading edge of a premixed flame brush is not a necessary condition for the flame to propagate into the unburned gas. A premixed turbulent flame can propagate even if turbulent scalar flux shows the countergradient behaviour at the cold boundary. This peculiarity of turbulent combustion is associated with the straightforward influence of turbulent diffusion on the mean rate of product creation at $\tilde{c} \ll 1$.

In the case of a fully-developed, statistically planar, one-dimensional premixed flame, turbulent scalar flux shows the countergradient behaviour at the cold boundary if a ratio of σS_L to the turbulent flame speed is larger than a number Ψ. In the asymptotic case of infinitely thin flamelets, Ψ is equal to the limit ($\tilde{c} \to 0$) value of the averaged component $\bar{n}_x \leq 1$ of the unit vector locally normal to flamelet, i.e. Ψ appears to be equal to unity. In the case of a finite flamelet thickness, the number Ψ appears to be increased when the diffusivity of the deficient reactant is decreased, but investigation of the latter case is beyond the scope of the present paper.

Both flame development and straining of a premixed turbulent flame by a divergent mean flow suppress countergradient scalar flux at the leading edge of the flame brush.

Acknowledgments

The first author (AL) was supported by the Swedish Energy Agency and the second author (VS) was supported by ONERA.

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