Transition from Gradient to Countergradient Scalar Transport in Developing Premixed Turbulent Flames

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Abstract

It is well known that scalar transport in premixed turbulent flames may occur not only in the direction of the decreasing mean value of a scalar (the so-called gradient diffusion), but also in the opposite direction (countergradient diffusion). The present paper addresses the issue of transition from gradient to countergradient diffusion as a premixed turbulent flame develops. Using three different methods, basically the same simple criteria of the transition are derived and an important role played by flame development is highlighted. The criteria are qualitatively assessed by comparing them with available experimental and DNS data.

Introduction

In a constant-density, non-reacting flow, turbulent flux of a scalar quantity q is commonly modeled invoking turbulent diffusion approximation [1], i.e.

$$\overline{u_i' q'} = -D_t \frac{\partial \overline{q}}{\partial x_i}, \qquad (1)$$

where x_i and u_i are the spatial coordinates and flow velocity components, respectively, D_t is the turbulent diffusivity, and over-bars designate Reynolds averaging with $q' = q - \overline{q}$. The minus sign on the right hand side (rhs) of Eq. (1) is associated with the fact that a scalar quantity q is transported by turbulent eddies in the direction of decreasing the mean value of q.

In premixed flames, however, the signs of the flux $\overline{u_i' q'}$ and the gradient $\partial \overline{q} / \partial x_i$ may be the same, i.e. turbulent transport of q may occur in the direction of increasing $\overline{q}$. This phenomenon, predicted theoretically by several authors [2-4] and documented first by Moss [5], is often called countergradient diffusion in order to stress the same directions of the scalar flux and the gradient $\nabla \overline{q}$.

Since, in a typical premixed turbulent flame, reactants and products are separated by a wrinkled reaction zone (flamelet), which is much thinner than the mean flame brush [6]; one may assume that the probability of finding intermediate (between the reactants and products) states of the burning mixture is much less than unity everywhere within the flame brush [2,7] and, therefore [4],

$$\overline{\rho u_i'' c''} = \overline{\rho} \tilde{c} (1 - \tilde{c})(\overline{u}_{ib} - \overline{u}_{iu}). \qquad (2)$$

Here, ρ is the density, c is the combustion progress variable introduced by Bray and Moss [7], subscripts u and b designate unburned and burned gas, respectively, $\tilde{q} \equiv \overline{\rho q} / \overline{\rho}$ is the Favre-averaged value of a quantity q, with $q'' = q - \tilde{q}$. Based on Eq. (2), countergradient and gradient transports are often associated with $|u_{ib}|>|u_{iu}|$ and $|u_{ib}|<|u_{iu}|$, respectively.

For the past decades, countergradient transport (or $|u_{ib}|>|u_{iu}|$) was documented in many premixed turbulent flames (e.g. see Ref. [8] as a recent example), while gradient transport (or $|u_{ib}|<|u_{iu}|$) was also documented in many other premixed turbulent flames. By varying the equivalence ratio F, Frank et al. [9,10] obtained (i) countergradient transport from near-stoichiometric methane- and propane-air Bunsen flames, but (ii) gradient transport from lean flames. Kalt et al. [11] investigated 11 lean and stoichiometric, methane- and propane-air flames stabilized in impinging jets and obtained $|u_{ib}|<|u_{iu}|$ from five moderately turbulent flames, while $|u_{ib}|$ was higher than $|u_{iu}|$ in one moderately turbulent flame and five weakly turbulent ones. Furthermore, both regions with gradient transport and regions with countergradient transport were observed in the same flame, e.g. in a confined flame stabilized by a rod [12], or in an open flame stabilized by a bluff body [13], or in a swirl-stabilized flame [14].

Such experimental data as briefly reviewed above call for a criterion that would allow a researcher to predict the direction of turbulent scalar flux under particular conditions. The first criterion of that kind was proposed to be used by Bray et al. [15,16]. According to this criterion, turbulent scalar flux is countergradient (gradient) if the Bray number

$$N_B = \frac{\tau S_L}{2 \alpha u'} \qquad (3)$$

is larger (lower) than unity. Here, $\tau=\rho_u/\rho_b-1$ is the heat release parameter, S_L is the laminar flame speed, u' is the rms turbulent velocity, and the function $\alpha=\alpha(L/\delta_L)$ depends on a ratio of an integral length scale L of turbulence to the laminar flame thickness δ_L. According to the DNS data reported in Ref. [16], α is typically less than unity and is increased by L/δ_L.

After the pioneering work by Bray et al. [15,16], several authors [11,17-21] undertook attempts to improve the Bray-number criterion. Despite substantial contributions made by the cited papers, an important issue was beyond the focus of research into turbulent scalar transport in premixed combustion. This is the

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influence of flame development on the direction of turbulent scalar flux.

The point is that, contrary to combustion in a laminar flow, a typical turbulent premixed flame is a developing flame, as discussed in details elsewhere [22,23]. The most clear manifestation of the development of a premixed turbulent flame consists of the growth of the mean flame brush thickness Δ_t either with time t (in the case of an expanding flame) or with distance X from flame-stabilization zone (in the statistically stationary case).

If a flame develops, the direction of turbulent scalar flux within the flame brush may depend not only on τ, S_L, u', etc., but also on flame-development time t. The time-dependence of the direction of turbulent scalar flux was theoretically hypothesized in a few papers [24,25] and DNS data reported by Veynante et al. [16] clearly show gradient scalar flux during an early stage of flame development, followed by the transition to countergradient flux as the flame develops (see Figs. 10a-10c in the cited paper). However, the above criteria [11,15-21] do not involve flame-development time.

The goal of the present work is to advance a simple criterion for estimating the direction of turbulent scalar flux, that emphasizes an important role played by the development of premixed turbulent flames.

In the next section, three such criteria, which are basically similar to one another, are obtained using three different analytical approaches. These criteria are compared with available experimental and DNS data in Sect. 3. Conclusions are briefly summarized in the last section.

Analysis

Equation for the variance of the progress variable

A decade ago, Swaminathan et al. [26] developed an interesting approach to predicting the direction of turbulent scalar flux. They analyzed a statistically planar, one-dimensional premixed, turbulent flame and considered the following well-known balance equation

$$\frac{\partial}{\partial t}\overline{\rho c''^2} + \frac{\partial}{\partial x}\left(\tilde{u}\,\overline{\rho c''^2}\right) + \frac{\partial}{\partial x}\overline{\rho u'' c''^2} = -2\overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x} + 2\overline{c'' W} + 2\overline{c''\frac{\partial}{\partial x_k}\left(\rho D \frac{\partial c}{\partial x_k}\right)} \quad (4)$$

for the variance $\overline{\rho c''^2}$ of the combustion progress variable. Here, W is the mass rate of product creation, D is the molecular diffusivity, and the summation convention is applied to the repeated index k.

Integration of Eq. (4) from $x=-\infty$ to $x=\infty$ yields [26]

$$\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\overline{\rho c''^2}dx + 2\int_{-\infty}^{\infty}\overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x}dx = 2\int_{-\infty}^{\infty}\overline{c''\left[W + \frac{\partial}{\partial x_k}\left(\rho D\frac{\partial c}{\partial x_k}\right)\right]}dx. \quad (5)$$

Using the mass

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_k}\left(\rho u_k\right) = 0 \quad (6)$$

and combustion progress variable

$$\frac{\partial}{\partial t}\left(\rho c\right) + \frac{\partial}{\partial x_k}\left(\rho u_k c\right) = \frac{\partial}{\partial x_k}\left(\rho D\frac{\partial c}{\partial x_k}\right) + W \quad (7)$$

balance equations, one can easily show that

$$\frac{\partial u_k}{\partial x_k} = \frac{\tau}{\rho_u}\left[\frac{\partial}{\partial x_k}\left(\rho D\frac{\partial c}{\partial x_k}\right) + W\right]. \quad (8)$$

Accordingly, Eq. (5) reads {26}

$$\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\overline{\rho c''^2}dx + 2\int_{-\infty}^{\infty}\overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x}dx = 2\frac{\rho_u}{\tau}\int_{-\infty}^{\infty}\overline{c''\frac{\partial u_k}{\partial x_k}}dx. \quad (9)$$

Swaminathan et al. [26] used Eq. (9) in order to highlight a link between the direction of turbulent scalar flux (see the second term on the lhs) and the internal structure of flamelets in a turbulent flow (see the term on the rhs and note that $\nabla \mathbf{u} = 0$ outside flamelets). To highlight this link, Swaminathan et al. [26] discussed a hypothetical fully-developed flame, i.e. skiped the unsteady term on the lhs, while DNS data analyzed by them indicated the importance of the unsteady term.

Let us look at Eq. (9) from another perspective, i.e. (i) retain the unsteady term and (ii) consider transition from gradient to countergradient scalar transport (or vice versa). As the flux $\overline{\rho u'' c''}$ and the derivative $\partial \tilde{c}/\partial x$ have the same (opposite) signs in the case of countergradient (gradient) scalar transport, it appears to be natural to propose to use the following equality

$$\int_{-\infty}^{\infty}\overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x}dx = 0 \quad (10)$$

as a global criterion of the transition referred to. Then, Eqs. (9) and (10) yield the following simple criterion

$$\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\overline{\rho c''^2}dx = 2\frac{\rho_u}{\tau}\int_{-\infty}^{\infty}\overline{c''\frac{\partial u_k}{\partial x_k}}dx. \quad (11)$$

The integral on the lhs of Eq. (11) may be estimated as follows

$$\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\overline{\rho c''^2}dx = \frac{d}{dt}\left(\Delta_t \Xi\right), \quad (12)$$

where

$$\Xi \equiv \int_{-\infty}^{\infty}\overline{\rho c''^2}d\xi \quad (13)$$

and

$$\xi \equiv \frac{x - x_f(t)}{\Delta_t(t)}. \quad (14)$$

To estimate the integral Ξ, let us (i) invoke the following well-known equation

$$\overline{\rho c''^2} = \bar{\rho}\tilde{c}(1-\tilde{c}), \quad (15)$$

which is valid in the flamelet regime of premixed turbulent combustion [6,7], and (ii) note that the mean structure of a typical premixed turbulent flame is self-similar and universal, i.e. $\tilde{c}(t,x) = \tilde{c}(\xi)$, with the function $\tilde{c}(\xi)$ being approximately the same in various flames, as discussed in details elsewhere [22,23]. Finally, as $\bar{\rho} = \bar{\rho}(\tilde{c},\tau)$ in the flamelet regime of premixed turbulent combustion [7], we may assume that (i) the integral Ξ does not depend on t and (ii) Ξ/ρ_u is a decreasing non-dimensional function of the heat release parameter τ, see the mean density in Eq. (15).

To estimate the integral on the rhs of Eq. (11), let us note that both the mean dilatation $\overline{\nabla \mathbf{u}}$ and the mean rate of product creation $\overline{W}$ are controlled by the structure of reaction zones in premixed turbulent flames. Furthermore, $\overline{\nabla \mathbf{u}} = \rho_u \overline{W}/\tau$, see Eq. (8), provided that the probability of finding intermediate states of the mixture does not depend on c and, hence, the molecular diffusion term on the rhs of Eq. (8) vanishes after integration over the reaction zones (i.e. from $c=\varepsilon$ to $1-\varepsilon$, where $\varepsilon \to 0$). In this case, the turbulent burning velocity U_t is equal to

$$U_t = \frac{1}{\rho_u}\int_{-\infty}^{\infty} \overline{W} dx. \quad (16)$$

Based on the above reasoning, we may estimate the discussed integral as follows

$$\frac{\rho_u}{\tau}\int_{-\infty}^{\infty} \overline{c''\frac{\partial u_k}{\partial x_k}} dx = \int_{-\infty}^{\infty}\left(\frac{\overline{c\nabla \mathbf{u}}}{\overline{\nabla \mathbf{u}}} - \tilde{c}\right)\overline{W} dx = \Omega U_t, \quad (17)$$

where

$$\Omega \equiv \frac{\rho_u \int_{-\infty}^{\infty}\left(\frac{\overline{c\nabla \mathbf{u}}}{\overline{\nabla \mathbf{u}}} - \tilde{c}\right)\overline{W} dx}{\int_{-\infty}^{\infty} \overline{W} dx}. \quad (18)$$

Finally, Eqs. (11), (12), and (17) yield

$$\frac{1}{U_t}\frac{d\Delta_t}{dt} = \Psi_1, \quad (19)$$

where

$$\Psi_1 \equiv \frac{2\Omega}{\Xi(\tau)} \quad (20)$$

is an increasing function of the density ratio. Using u', L, and the turbulent time scale $\tau_t = L/u'$ to normalize U_t, $\delta_t = \Delta_t/L$, and $\theta = t/\tau_t$, respectively, Eq. (19) reads

$$\Gamma(\theta) \equiv \frac{1}{u_t}\frac{d\delta_t}{d\theta} = \Psi_1 \frac{U_{t,\infty}}{u'}, \quad (21)$$

where the turbulent burning velocity $u_t = U_t/U_{t,\infty}$ is normalized using the fully-developed burning velocity $U_{t,\infty} = U_t(t \to \infty)$. The lhs of this equation is controlled by the development of the flame, thus, highlighting the influence of premixed turbulent flame development on the direction of turbulent scalar flux.

Integration by parts

In the previous subsection, the integral on the lhs of Eq. (10) was estimated by analyzing the balance equation for the variance of the combustion progress variable. The present subsection is aimed at evaluating the same integral using another method.

To do so, let us (i) integrate the lhs of Eq. (10) by parts and (ii) invoke the following well-known balance equation [4,7,15,16]

$$\frac{\partial}{\partial t}(\bar{\rho}\tilde{c}) + \frac{\partial}{\partial x_k}(\bar{\rho}\tilde{u}_k\tilde{c}) = -\frac{\partial}{\partial x_k}\overline{\rho u_k'' c''} + \overline{W} \quad (22)$$

for the Favre-averaged combustion progress variable, supplemented with the Favre-averaged mass-conservation equation

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial}{\partial x_k}(\bar{\rho}\tilde{u}_k) = 0. \quad (23)$$

Then, we obtain

$$\begin{aligned}
\int_{-\infty}^{\infty} \overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x} dx &= -\int_{-\infty}^{\infty} \tilde{c}\frac{\partial \overline{\rho u'' c''}}{\partial x} dx \\
&= \int_{-\infty}^{\infty} \tilde{c}\left(\bar{\rho}\frac{\partial \tilde{c}}{\partial t} + \bar{\rho}\tilde{u}\frac{\partial \tilde{c}}{\partial x} - \overline{W}\right) dx \\
&= \frac{1}{2}\int_{-\infty}^{\infty}\frac{\partial}{\partial t}(\bar{\rho}\tilde{c}^2) dx + \frac{1}{2}\rho_b U_b - \int_{-\infty}^{\infty}\tilde{c}\overline{W} dx \quad (24) \\
&= -\frac{1}{2}\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\bar{\rho}\tilde{c}(1-\tilde{c}) dx + \frac{1}{2}\int_{-\infty}^{\infty}\frac{\partial}{\partial t}(\bar{\rho}\tilde{c}) dx \\
&\quad + \frac{1}{2}\rho_b U_b - \int_{-\infty}^{\infty}\tilde{c}\overline{W} dx
\end{aligned}$$

for a flame that moves from right to left. Here, U_b is the mean flow velocity at $x \to \infty$.

Since integration of Eq. (22) yields

$$\int_{-\infty}^{\infty}\frac{\partial}{\partial t}(\bar{\rho}\tilde{c}) dx + \rho_b U_b = \rho_u U_t, \quad (25)$$

we finally obtain

$$\begin{aligned}
&\int_{-\infty}^{\infty} \overline{\rho u'' c''}\frac{\partial \tilde{c}}{\partial x} dx \\
&= -\frac{1}{2}\frac{\partial}{\partial t}\int_{-\infty}^{\infty}\bar{\rho}\tilde{c}(1-\tilde{c}) dx + \int_{-\infty}^{\infty}\left(\frac{1}{2} - \tilde{c}\right)\overline{W} dx
\end{aligned} \quad (26)$$

using Eq. (16). Based on the reasoning already discussed in the previous subsection, Eqs. (10) and (26) may be rewritten as follows

$$\Gamma(\theta) \equiv \frac{1}{u_t}\frac{d\delta_t}{d\theta} = \Psi_2 \frac{U_{t,\infty}}{u'}, \quad (27)$$

where

$$\Psi_2 \equiv \frac{\rho_u \int_{-\infty}^{\infty} (1-2\tilde{c})\overline{W} dx}{\int_{-\infty}^{\infty} \overline{W} dx \cdot \int_{-\infty}^{\infty} \overline{\rho}\tilde{c}(1-\tilde{c}) d\xi} \qquad (28)$$

is an increasing function of the density ratio, as the second integral in the denominator involves $\overline{\rho}$.

Equation (27) is basically similar to Eq. (21). Moreover, Eqs. (27) and (28) emphasize the following point: transition from gradient to countergradient transport may occur if $\Psi_2 \geq 0$, i.e. if the dependence of the mean rate of product creation on the Favre-averaged combustion progress variable is asymmetric with respect to $\tilde{c} = 0.5$, with the maximum of the function $\overline{W}(\tilde{c})$ being shifted to reactants. This point was early emphasized by Zimont et al. [24] based on other arguments.

Combustion progress variable balance equation

Obtained in the two previous subsections are two basically similar ***global*** criteria, see Eqs. (21) and (27) for predicting the direction of turbulent scalar flux. In the present subsection, a basically similar ***local*** criterion will be derived by analyzing the balance equation for the Favre-averaged combustion progress variable in the case of a developing flame with a self-similar mean structure.

To do so, let us (i) consider a flame that moves from right to left, (ii) use such Galilean framework that $\overline{u}(x \to -\infty) = 0$, (iii) substitute arbitrary functions $\overline{\rho}(\xi)$ and $\tilde{c}(\xi)$ into Eqs. (22)-(23), and (iv) integrate the two obtained equations from $-\infty$ to ξ. We have [27,28]

$$\overline{\rho}\tilde{u} = S_t(\rho_u - \overline{\rho}) + \frac{d\Delta_t}{dt} \int_{-\infty}^{\xi} \zeta \frac{d\overline{\rho}}{d\zeta} d\zeta \qquad (29)$$

and

$$\overline{\rho u'' c''} = \frac{d\Delta_t}{dt} \int_{-\infty}^{\xi} \Phi \frac{d\tilde{c}}{d\zeta} d\zeta + \int_{-\infty}^{\xi} \left(\overline{W}\Delta_t - \rho_u S_t \frac{d\tilde{c}}{d\zeta} \right) d\zeta, \qquad (30)$$

where

$$\Phi(\xi) \equiv \overline{\rho}\xi - \int_{-\infty}^{\xi} \zeta \frac{d\overline{\rho}}{d\zeta} d\zeta \qquad (31)$$

and the magnitude of the flame speed $S_t = -dx_f/dt$ depends on the choice of a surface $x = x_f(t)$ associated with the mean flame position. As argued elsewhere [28], a physically meaningful definition of x_f is given by the following equation

$$\int_{-\infty}^{\infty} \Phi \frac{d\tilde{c}}{d\zeta} d\zeta = 0. \qquad (32)$$

If Eq. (32) holds, then, $S_t = U_t$ [28] and Eq. (30) reads

$$\frac{\overline{\rho u'' c''}}{\rho_u}(\xi) = \frac{d\Delta_t}{dt} \Lambda(\xi) + U_t \int_{-\infty}^{\xi} \left(\omega - \frac{d\tilde{c}}{d\zeta} \right) d\zeta, \qquad (33)$$

where

$$\Lambda(\xi) \equiv \rho_u^{-1} \int_{-\infty}^{\xi} \Phi \frac{d\tilde{c}}{d\zeta} d\zeta \qquad (34)$$

and

$$\omega \equiv \frac{\Delta_t \overline{W}}{\rho_u U_t}. \qquad (35)$$

First, Eq. (31) shows that $d\Phi / d\xi = \overline{\rho} > 0$ and $\Phi(\xi \to -\infty) < 0$, i.e. the function Φ increases monotonically with ξ. Second, Eq. (32) indicates that the function Φ should change its sign somewhere within the flame brush. Accordingly, Eq. (34) shows that the derivative $d\Lambda/d\xi$ is negative and positive at the leading and trailing edges of the flame brush, respectively, with $d\Lambda/d\xi = 0$ in a single point within the flame brush. Since the function Λ tends to zero both at the leading and trailing (due to Eq. (32)) edges, this function is negative everywhere within the flame brush, i.e. the first term on the rhs of Eq. (33) is negative and serves to cause gradient transport. Therefore, countergradient transport may exist only in the second term on the rhs of Eq. (33) is positive, i.e. if $\omega > d\tilde{c} / d\xi$ in the leading half of the flame brush. In other words, the maximum of the function $\overline{W}(\tilde{c})$ should be shifted to reactants as compared with $d\tilde{c} / d\xi$, in line with the discussion in the end of the previous subsection.

Finally, let us assume that the normalized reaction rate ω depends solely on ξ with $\int_{-\infty}^{\infty} \omega d\xi = 1$ due to Eq. (16). Then, transition from gradient to countergradient scalar transport occurs locally (i.e. $\overline{\rho u'' c''} = 0$) if

$$\Gamma(\theta) \equiv \frac{1}{u_t} \frac{d\delta_t}{d\theta} = \Psi_3(\xi) \frac{U_{t,\infty}}{u'}, \qquad (36)$$

where

$$\Psi_3(\xi) \equiv \frac{\int_{-\infty}^{\xi} \left(\omega - \frac{d\tilde{c}}{d\zeta} \right) d\zeta}{|\Lambda(\xi)|} \qquad (37)$$

is an increasing (see Eqs. (31) and (34)) function of the density ratio.

Equation (36) is basically similar to Eqs. (21) and (27), but, contrary to the latter equations, yield a local criterion of the transition referred to. The above global criteria should not be recovered by integrating Eq. (36) from $-\infty$ to ∞, because, at the same instant, not only zones characterized by $\overline{\rho u'' c''} = 0$, but also zones characterized by both $\overline{\rho u'' c''} > 0$ and $\overline{\rho u'' c''} < 0$ may exist within a flame brush.

Discussion

Equations (21), (27), and (36) clearly show that turbulent flame development affects the direction of turbulent scalar flux, as the function $\Gamma(\theta)$ determined by the lhs's of these equations depends on the normalized flame-development time $\theta=t/\tau_t$. Let us consider this dependence in more details.

It is well known that the thickness of a typical premixed turbulent flame grows linearly with time t (for an expanding flame) or with distance X from flame holder (for a stabilized flame) during an early stage of flame development (at low θ or in the vicinity of the flame holder, respectively), but the rate of the growth of $\Delta_t(t)$ slowly decreases with t or X, respectively [2,22]. In particular, as reviewed elsewhere [22,23], the growth of $\Delta_t(t)$ follows the well-known Taylor turbulent diffusion law [1,29,30] for many premixed turbulent flames, i.e.

$$\delta_t^2 = 4\pi\left(\theta - 1 + \mathrm{e}^{-\theta}\right). \qquad (38)$$

Equation (38) shows that the derivative $d\delta_t/d\theta$ decreases with flame-development time. Since the normalized turbulent burning velocity u_t increases during flame development [31] (while this process is much less pronounced than the growth of the thickness [2,22,32]), the function $\Gamma(\theta)$ decreases with θ. Accordingly, if the functions Ψ_i are positive and countergradient transport may exist in principle, the equalities given by Eqs. (21), (27), and (36) may be satisfied at a normalized time θ sufficiently long in order for the function $\Gamma(\theta)$ to decrease to Ψ_i.

To support the above scenario, let calculate the function $\Gamma(\theta)$ using Eq. (38) and the following theoretical expression

$$u_t = \left[1 + \frac{1}{\theta}\left(\mathrm{e}^{-\theta} - 1\right)\right]^{1/2}, \qquad (39)$$

which (i) has been obtained [22] combing the so-called Zimont model [33] of turbulent burning velocity with the aforementioned Taylor theory [29,30] of turbulent diffusion and (ii) has been validated [22] against a

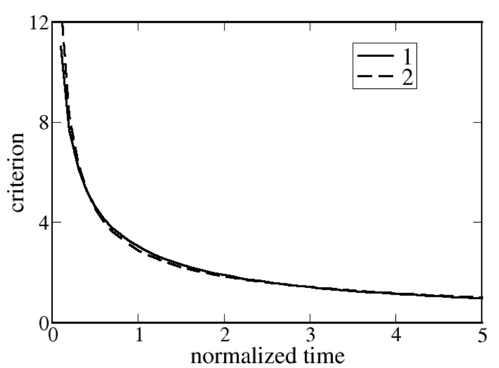

Figure. 1. Effect of flame development on transition from gradient to countergradient transport. Curve 1 shows the function $\Gamma(\theta)$ computed using Eqs. (21), (38), and (39). Curve 2 has been calculated using the following approximation $\Gamma(\theta)=2.9\theta^{-0.65}$.

number of experimental data obtained from expanding spherical flames.

Curve 1 in Fig. 1 has been computed using Eqs. (21), (38), and (39). These results show that the function $\Gamma(\theta)$ is high at low θ so that criteria given by Eqs. (21), (27), and (36) cannot be satisfied and turbulent transport is gradient during an early stage of flame development, in line with the hypothesis put forward by Zimont et al. [24] based on other reasoning. As the flame develops, the function $\Gamma(\theta)$ drops, the aforementioned criteria may be satisfied, and transition to countergradient transport happens at certain instant t_{tr}.

A similar behavior of the flux $\overline{\rho u'' c''}$ was documented by Veynante et al. [16] in DNS. Figures 10a-10c in the cited paper indicate that turbulent transport is gradient during an early stage of flame development, followed by transition to countergradient transport. The normalized transition time θ_{tr} is weakly increased by the rms turbulent velocity. For instance, as far as the global transition is concerned, $0.7\leq\theta_{tr}<1$ if $u'/S_L=2$, $1.3\leq\theta_{tr}<1.7$ if $u'/S_L=3$, and $1.6<\theta_{tr}<2.2$ if $u'/S_L=5$.

This trend agrees with the criteria obtained in the present paper. Indeed, various experimental data reviewed elsewhere [22,34,35] indicate that turbulent flame speed scales as u'^q with the power exponent q being less than unity. Accordingly, a ratio of $U_{t,\infty}/u'$ on the rhs's of Eqs. (21), (27), and (36) appears to be decreased when u' is increased. Therefore, the magnitude of the function $\Gamma(\theta)$, calculated at the transition instant, is also decreased when u' is increased, i.e., θ_{tr} is increased by u', in line with the aforementioned DNS data [16].

Since the ratio of $U_{t,\infty}/u'$ is increased by the laminar flame speed [22,34,35] and the functions Ψ_i are increased by the density ratio, the rhs's of Eqs. (21), (27), and (36) decrease when a near-stoichiometric mixture becomes leaner and leaner. Accordingly, if flame-development time is kept constant, the above criteria may predict transition from countergradient to gradient transport when the equivalence ratio F decreases from unity. Such a trend agrees well with the experimental data reported by Frank, Kalt, and Bilger [9,10].

However, another trend documented by the same group [9,10] from Bunsen flames challenges the criteria obtained in the present paper. When a grid used to produce turbulence in these experiments was moved upstream, the rms velocity u' in the combustion zone (measured in the non-reacting flow) was decreased and the values of $|u_b|$ (measured in the same zone but in the reacting case) became substantially higher than $|u_u|$, whereas $|u_b|\approx|u_u|$ for the standard position of the grid (cf. flames B and C either in Ref. [9] or in Ref. [10]). In other words, the decrease in u' promoted counter-gradient transport.

Curve 2 in Fig. 1 shows that the function $\Gamma(\theta)$ is well approximated as follows $\Gamma(\theta)=2.9\theta^{-0.65}\sim u'^{-0.65}$. Thus, if flame-development time t is kept constant and the ratio of $U_{t,\infty}/u'$ scales as $u'^{1/4}$ [22,34], then a factor of $\Gamma u'/U_{t,\infty}$

is decreased when u' is increased (scales as $u'^{-0.3}$), i.e. an increase in u' promotes countergradient transport (if t=const), contrary to the aforementioned measurements.

Thus, the simple criteria obtained for a hypothetical statistically planar, one-dimensional flame in the present paper may yield wrong results for laboratory flames. Multi-dimensional effects may play a substantial role, as discussed elsewhere [36]. Moreover, the movement of the grid in the above experiments may affect the mean flame position (due to variations in u'), the mean flow velocity, and, hence, the flame-development time t.

Furthermore, the mean pressure gradient is well known to substantially affect turbulent scalar flux [17,18]. The criteria obtained in the present paper do not involve the pressure gradient straightforwardly. Certainly, both $d\delta_t/dt$ and $U_{t,\infty}$ are likely to depend on the pressure gradient. However, the lack of models that address such a dependence does not allow us to apply the aforementioned criteria to premixed turbulent flames subjected to a sufficiently strong pressure gradient. In particular, the criteria may yield wrong results for premixed flames stabilized in impinging jets, as the study of Kalt et al. [11] showed substantial influence of the mean pressure gradient on the direction of turbulent scalar flux in such flames.

Conclusions

Three basically the same simple criteria (see Eqs. (21), (27), and (36)) of transition from gradient to countergradient scalar transport in premixed turbulent flames have been obtained theoretically using three different methods.

The criteria indicate that the direction of the turbulent scalar flux depends substantially on flame-development time, with gradient transport dominating during an early stage of flame development.

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