Gradient, counter-gradient transport and their transition in turbulent premixed flames

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Abstract

We theoretically and numerically analyse the phenomenon of counter-gradient transport in turbulent premixed flames with pressure distribution across the flame brush mainly controlled by heat release. The focus is on the transition from counter-gradient to gradient transport obtained when increasing the turbulence intensity/laminar flame speed ratio, a phenomenon recently found in open laboratory flame experiments by Frank \textit{et al} (1999 \textit{Combust. Flame} \textbf{116} 220). The analysis is based on the turbulent flame closure combustion model for the simulation of turbulent premixed flames at strong turbulence ($u' \gg s_L$). In this case, earlier work suggests that turbulent premixed flames have non-equilibrium increasing flame brush width controlled in the model only by turbulence and independent from the counter-gradient transport phenomenon which has gasdynamic nature, and equilibrium turbulent flame speed which quickly adapts to the local turbulence. Flames of this type have been called intermediate steady propagation flames. According to the present analysis, transport in turbulent premixed flames is composed of two contributions: real physical gradient turbulent diffusion, which is responsible for the growth of flame brush thickness, and counter-gradient pressure-driven convective transport related to the different acceleration of burnt and unburnt gases subject to the average pressure variation across the turbulent flame. The original gasdynamics model for the pressure-driven transport which is developed here shows that the overall transport may be of gradient or counter-gradient nature according to which of these two contributions is dominant, and that along the flame a transformation from gradient to counter-gradient transport takes place. Reasonable agreement with the mentioned laboratory experimental data strongly support the validity of the present modelling ideas. Finally, we explain why this phenomenon is also highly probable in large-scale industrial burners at much larger turbulent Reynolds numbers.

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Nomenclature

\begin{align*}
  c & \quad \text{progress variable} & u' & \quad \text{turbulent fluctuation} \\
  \bar{c} & \quad \text{Favre averaged } c & U_t & \quad \text{turbulent flame speed} \\
  \tilde{c} & \quad \text{Reynolds averaged } c & U_t, \delta_t & \quad \text{thickened flamelet parameters} \\
  D_t & \quad \text{turbulent diffusion coefficient} & W & \quad \text{rate of product formation} \\
  Da & \quad \text{Damköhler number} & \delta_t & \quad \text{flame brush thickness} \\
  \text{g}_{\text{cr}} & \quad \text{critical velocity gradient} & \epsilon & \quad \text{k dissipation rate} \\
  G & \quad \text{stretch factor} & \epsilon_{\text{cr}} & \quad \text{critical extinction } \epsilon \\
  k & \quad \text{turbulent kinetic energy} & \Theta & \quad \text{density ratio } \rho_u/\rho_b \\
  L & \quad \text{turbulent integral scale} & \nu & \quad \text{kinematic molecular viscosity} \\
  p & \quad \text{mean pressure} & \xi & \quad \text{non-dimensional spatial variable} \\
  p_{u}, p_{b} & \quad \text{conditional mean pressures} & \bar{\rho}_u, \bar{\rho}_b & \quad \text{unburnt, burnt gas densities} \\
  Re_t & \quad \text{turbulent Reynolds number} & \sigma & \quad \text{standard deviation} \\
  s_{\text{L}}, \delta_{\text{L}} & \quad \text{laminar flame parameters} & \tau_{\text{ch}} & \quad \text{chemical timescale} \\
  \bar{S}/S_0 & \quad \text{non-dimensional flamelet area} & \phi & \quad \text{equivalence ratio} \\
  u & \quad \text{velocity component} & \chi & \quad \text{heat transfer coefficient}
\end{align*}

1. Introduction

Counter-gradient transport of reacting chemical species is a phenomenon commonly occurring in turbulent premixed flames burning with the flamelet combustion mechanism. It has been observed, for example, in experiments on open [1], impinging [2] and swirl [3] stabilized flame and also in direct numerical simulations [4, 5]. Recent experimental data by Frank et al [6] in open premixed flames demonstrate a transition from counter-gradient to gradient transport at a given distance from the burner inlet when the ratio between the turbulent velocity fluctuation and the laminar flame speed $u'/s_{\text{L}}$ increases. This result seems physically natural as, for $u'/s_{\text{L}} \to \infty$, the flame transforms into a non-reacting jet with only gradient turbulent diffusion.

The counter-gradient phenomenon derives its name from the fact that the averaged transport $\bar{u}'\bar{c}$ of the progress variable $c$ (representing the generalized reacting concentration equal to zero in reactants and unit in products) is oriented in a direction opposite to the normal gradient turbulent diffusion in non-reacting turbulent flows. In this case the averaged flux of the combustion products takes place in the direction from the fresh mixture to burnt gas (i.e. along the gradient of $\bar{c}$) and at the same time the averaged flux of the reactants takes place in the direction from the burnt gas to fresh mixture. Obviously this counter-gradient transport cannot be predicted using the usual two-parameter turbulence models with positive turbulent diffusion coefficient.

The universally accepted term for this phenomenon is 'counter-gradient turbulent diffusion in premixed flames' and a common method of its analysis is the development of model equations by 'turbulent-type' empirical modelling of unclosed terms in exact second-order moment or PDF equations [7–9]. Though these statistical unclosed equations are obviously valid irrespective of the actual physical nature of the transport and counter-gradient transport can be predicted with ad hoc semi-empirical modelling, we believe that such approach is not adequate for the counter-gradient transport phenomenon. In fact, this phenomenon, as has been known for long time, is connected with a dominant contribution in the total transport
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We develop here a gasdynamic model for this pressure-driven counter-gradient contribution to total transport in turbulent premixed flames based on the direct estimation from the gasdynamic equations of the difference of the conditional averaged velocities of burnt (products) and unburnt (reactants) gas $\overline{\rho u^\prime c^\prime}$ directly related to the transport $\overline{\rho u c' \delta}$.

The main physical assumption of the model is the reactants’ total pressure conservation inside the flame that makes it possible to close the problem without any empirical constants. In this paper, we develop this model for the case of a 1D stationary combustion front and show that the problem has in this case an analytical solution.

We then analyse the progress variable transport in open flames and compare the results obtained with our model with the experimental data presented in [1, 6]. In our analysis there are two contributions to the total transport: the first connected with the mentioned gasdynamic effect and the second with the turbulent pulsations of the velocity. We therefore assume that $\overline{\rho u c' \delta}$ can be expressed as the sum of two terms that could be estimated independently: counter-gradient pressure-driven and gradient turbulent diffusion components which we estimate, from the gasdynamic model developed here and from the standard two parameter ‘$k$–$\epsilon$’ turbulence model, respectively. When the first term prevails over the second, we observe the counter-gradient transport, but if, because of variations of the system parameters, the second term prevails over the first, we have the transition phenomenon, i.e. change of the flux direction in a fixed point of the flame.

Observations demonstrate that open flames with a strongly wrinkled flamelet sheet have practically constant turbulent combustion velocity $U_t$ (the inclination of the flame to the flow is constant) and flame brush width $\delta_t$ increasing along the flame. This is readily illustrated, for example, by results of the instantaneous flamelet sheet visualization (figure 3 in [11]). The physical reason is that real open flames are transient flames with quasi-equilibrium small-scale flamelet wrinkles mainly controlling the flame sheet area and $U_t$ and with non-equilibrium large-scale wrinkles mainly controlling $\delta_t$. This is because the flame sheet structure in complete equilibrium corresponding to the 1D stationary front is unattainable (the flame crosses at the flow axis and combustion is completed before the 1D steady front can be obtained). For the simulation of open flames analysed in this paper, we use the turbulent flame closure (TFC) combustion model developed in [12–14], which describes such transient flames. These transient flames have been called intermediate steady propagation (ISP) flames [14, 15].

In the TFC combustion model, the apparent contradiction between increasing flame brush width and the dominating counter-gradient transport is automatically resolved. This is because only the turbulent diffusion component of the transport controls the brush width while the pressure-driven transport component is included with the actual source term in a single model source term. The gasdynamic model developed here for counter-gradient transport makes it possible to numerically extract the actual source term from the model one. We compare this result with the estimation from measurements of the flamelet crossing frequency (to which the actual source term is proportional) performed in [3, 16].

The paper is organized as follows. Section 2 contains the general description of the counter-gradient transport and transition phenomenon. In section 3 we develop the gasdynamic model for counter-gradient transport for 1D stationary combustion front and outline its invariant generalization on the case of arbitrary 3D flames. In section 4, we present expressions for the turbulent combustion velocity $U_t$ for flames with increasing brush width at the flamelet combustion mechanism (flamelets which are thickened by small-scale turbulence as estimated in [12] for the large Reynolds and moderately large Damköhler numbers characterizing combustion in industrial burners). Finally, in section 5, we compare results of theoretical
predictions of the flux across the flame based on the 1D gasdynamics theory with existing experimental data on the counter-gradient transport and the transition phenomenon, and on the distribution of flamelet crossing frequency across the flame brush.

2. Kinematic equation: the counter-gradient transport phenomenon

The TFC combustion model kinematic equation describing premixed flames with brush width increasing according to the turbulent dispersion law corresponding to the ISP regime \([13,14]\)) is given by

\[
\bar{\rho} \frac{\partial \tilde{c}}{\partial t} + \bar{\rho} \bar{u} \frac{\partial \tilde{c}}{\partial x} = \frac{\partial}{\partial x} \left( \bar{\rho} D_t \frac{\partial \tilde{c}}{\partial x} \right) + \rho_i U_i \left| \frac{\partial \tilde{c}}{\partial x} \right|.
\]

It is important to remember the exact result that \(U_i\) in (1) is the volumetric consumption rate of reactants per unit area of the flame \([14]\) but not, for example, the speed of the front edge of widening flame sometimes measured in experiments.

Prudnikov was probably the first who established, during the late 1950s, that turbulent premixed flames have the peculiarity to develop with growing brush thickness. He showed experimentally that turbulent premixed flames stabilized in uniform duct flows with strong artificial turbulence had temperature profiles corresponding to the integral probability distribution, i.e. close to the normal flamelet probability density function in space, and increasing flame brush width which was controlled mainly by the cold flow turbulent diffusion coefficient (this result was openly presented in the book \([17]\)). Using an original optical-diffusion method he also showed that, for strong cold flow turbulence, the transport of a non-uniform passive concentration inside the premixed flame is gradient turbulent diffusion with \(D_t\) coefficient comparable to the one calculated at the cold flow conditions \([18]\) (despite strong counter-gradient transport of reacting species discovered later in similar flames).

The estimation of \(U_i\) at practical turbulence and chemistry conditions is a challenging problem as it is controlled by the unresolved small-scale coupling of turbulence and chemistry \([19]\). The TFC model, which corresponds to the widened flamelet combustion mechanism, makes use of the theoretical expression for \(U_i\) in terms of resolved large-scale parameters based on the assumption of equilibrium small-scale structure of the wrinkled flame sheet (resulting in constant \(U_i\)) for increasing flame brush width \([12, 14]\). Equation (1) models the exact but unclosed equation

\[
\bar{\rho} \frac{\partial \tilde{c}}{\partial t} + \bar{\rho} \bar{u} \frac{\partial \tilde{c}}{\partial x} = -\frac{\partial (\bar{\rho} \bar{u} c' c')}{\partial x} + \bar{\rho} \tilde{W},
\]

where \(\bar{\rho} \bar{u} c' c'\) is the progress variable flux that, as a rule, is generally attributed a counter-gradient nature and \(\bar{\rho} \tilde{W}\) is the averaged rate of products formation. In equations (2) and (1), the molecular diffusion contribution has been neglected because at sufficiently large Reynolds numbers it is negligible in comparison to the turbulent one.

As we have mentioned in the introduction there is no contradiction between the gradient transport in the model equation (1) resulting in increasing flame brush width and counter-gradient transport in the exact unclosed equation (2). Now subtracting equation (1) from (2), we obtain the following relation:

\[
\rho_i U_i \left| \frac{\partial \tilde{c}}{\partial x} \right| = \bar{\rho} \tilde{W} - \frac{\partial}{\partial x} \left[ \bar{\rho} \bar{u} c' c' + \bar{\rho} D_t \frac{\partial \tilde{c}}{\partial x} \right] = \bar{\rho} \tilde{W} - \frac{\partial (\bar{\rho} \bar{u} c' c')_{p-d}}{\partial x},
\]

where \((\bar{\rho} \bar{u} c' c')_{p-d}\) is the pressure-driven component of transport. Equation (3) shows that the model source term in (1) not only contains the actual source but also the difference between
the total $\tilde{\rho}u\tilde{c}$ and turbulent diffusion flux $-\tilde{\rho}D_t\tilde{c}/\partial x$. We interpret this difference in the model source term as the pressure-driven gasdynamic counter-gradient transport component. Obviously

$$\rho_u \int_{-\infty}^{+\infty} U_t \left| \frac{\partial \tilde{c}}{\partial x} \right| dx = \int_{-\infty}^{+\infty} \tilde{\rho} \tilde{W} \, dx,$$

i.e. this procedure does not change the integral combustion rate.

This interpretation of the model equation (1) was formulated by Zimont in [20] in answer to a question from F Williams about the possibility of predicting counter-gradient transport within the framework of this model (see also [14]). It should also be remarked that Lipatnikov first realized in [21] that the interpretation of the turbulent diffusion term in the model equation (1) used as approximation of the transport term in the unclosed equation (2) and the model source term as approximation of actual source $\rho \tilde{W}$ is incorrect. A ‘joint closure’ in fact took place while developing the model given by equation (1), such that the sum of the total transport term and the source term in (2) is instead approximated by the sum of turbulent diffusion and model source term in the model equation (1). At the same time Lipatnikov’s idea in [22] to predict the counter-gradient transport in the context of the TFC model by developing a special turbulent equation directly for $\tilde{\rho}u\tilde{c}$ is quite different from the one developed by us.

It is significant to remember that equations (1) and (3) are valid only for flames with increasing flame brush width controlled by turbulent diffusion, i.e. in the case of flames in the ISP regime of combustion. In turbulent flames composed by laminar flamelets, this regime takes place in the case of $u' > s_L$ and for time $t < \tau (u'/s_L)^2$ when turbulent transport by pulsation velocities $u'$ prevails over the transport connected with the flamelets local propagation with velocity $s_L$. In turbulent flames composed by thickened flamelets, at $Re_t \gg 1$ and $Da > 1$ with flamelet velocity $U_L > s_L$, this regime takes place for $t < \tau Da$ (these estimations are presented in [14, 15]).

For larger times (which are practically unattainable at strong turbulence, premixed flames would propagate according to the 1D stationary combustion front, and instead of the model equation we can write the exact kinematic equation of the running wave (where the actual source and the total transport terms are clubbed together under a single term, the model source):

$$\tilde{\rho} \frac{\partial \tilde{c}}{\partial t} + \tilde{\rho}u \frac{\partial \tilde{c}}{\partial x} = \rho_u U_t^{st} \left| \frac{\partial \tilde{c}}{\partial x} \right| \left( = \tilde{\rho} \tilde{W} - \frac{\partial (\tilde{\rho}u\tilde{c}^o)}{\partial x} \right).$$

(5)

In accordance with the ideas of Damköhler [23] and Shchelkin [24], in the case of strong turbulence ($u' > s_L$ or $u' > U_L$) we have $U_t^{st} \approx u'$, as the front speed is controlled by the averaged velocity of the most advanced volumes of products thrown into the reactants by large-scale eddies, i.e. $U_t^{st}$ is equal to the speed of the front edge of the flame. We therefore connect the observed dependence of the turbulent combustion velocity (the volume consumption rate) on chemistry and molecular properties of the fuel/air mixture (which is significant but nevertheless weaker than for laminar flames) with the fact that real combustion takes place in flames with increasing flame brush width. In this case, according to [12, 14], $U_t$ depends on the flamelet sheet area which is controlled by the equilibrium between its production by turbulence (generation of sheet wrinkles) and destruction by flamelet movement (smoothing of sheet wrinkles) that, in the ISP regime, results in $U_t = A u' Da^{1/4}$, $A \sim O(1)$, velocity which is less than the front edge speed and in fact does not depend on it.

In the case of combustion occurring in the flamelet regime (and therefore with negligible probability to find burning mixture), the total transport term can be expressed as [25]

$$\tilde{\rho}u\tilde{c}^o = \tilde{\rho}c(1 - \tilde{c})(\tilde{u}_b - \tilde{u}_a),$$

(6)

where $\tilde{u}_a$ and $\tilde{u}_b$ are conditioned averaged velocities of unburnt and burnt volumes.
According to the ideas which were originally proposed in [14, 15], there are two mechanisms giving opposite influence to the difference \( \Delta \bar{u} = \bar{u}_b - \bar{u}_a \) in the flame: (a) connected with the turbulent pulsations of the velocity and (b) with more strong gasdynamic acceleration of hot gas than cold gas, resulting, respectively, into \( \Delta \bar{u} < 0 \) and \( \Delta \bar{u} > 0 \). We assume therefore that the total flux is split into two contributions which can be analysed independently:

\[
\bar{\rho} \bar{u} \bar{c}' = \bar{\rho} \bar{u} \bar{c}_a'' + \bar{\rho} \bar{u} \bar{c}_{b-d}'',
\]

(7)

where the first term (the turbulent diffusion controlling the increment of the flame brush width due to dispersion of the flamelet sheet by the turbulent velocity pulsation) is described by the usual gradient approximation with positive turbulent diffusion coefficient \( D_t \), and the second term (the pressure-driven transport which has counter-gradient nature and is of convective type) is generated by the pressure drop across the turbulent flame brush. Because of its convective nature, it is assumed here that the pressure-driven component of the flux does not affect the flame brush thickness.

The gasdynamic model developed in this paper for the pressure-driven component in the case of 1D stationary combustion front (where the pressure drop is connected only with combustion) is applied together with the gradient approximation for the turbulent diffusion component (we use for the estimation of \( D_t \) the standard ‘\( k-\epsilon \)’ turbulence model) to the quantitative analysis of the phenomenon of transition from gradient to counter-gradient transport experimentally observed in open flames [6] (where the pressure distribution across the flame brush is mainly determined by heat release).

The connection of counter-gradient transport with the averaged pressure drop across the flame is well known. In recent DNS work, it has been shown, in the framework of the unclosed transport equation for \( \bar{\rho} \bar{u} \bar{c}' \) that the term containing the pressure pulsation gives a well-defined contribution [26]. In our interpretation, this term contributes to both components of the reacting scalar flux. Turbulent diffusion is closely connected with the pressure pulsations forming the eddies structure, and the approximation based on \( D_t \) in fact takes into account the effect of these turbulent pressure pulsations. In the gasdynamic component of transport, this effect is connected with the instantaneous pressure drop across the flamelet. In our gasdynamic model developed below, the conditional averaged pressures \( \bar{p}_u \) and \( \bar{p}_b \) are used and therefore the difference \( \bar{p}_u - \bar{p}_b \) should be taken in consideration in order to account for the pressure pulsation effect. In our simulations we nevertheless ignore this effect as, in open flames with strong turbulence, the pressure drop across the flamelet is usually much smaller than the drop across the flame brush (in contrast, for example, to impinging flames where this effect could be substantial).

There are two possible ways for predicting the pressure-driven component \( (\bar{\rho} \bar{u} \bar{c}'')_{b-d} \) in the framework of the TFC model.

(a) To model the actual source term \( \bar{\rho} \bar{W} \) and estimate \( (\bar{\rho} \bar{u} \bar{c}'')_{b-d} \) from equation (3). This was done in [15] and corresponds to qualitative rough approximation of the counter-gradient effect in 1D flames. We give anyway, in this section, a short resume of this idea as it illustrates our general point of view on the problem.

(b) To develop a physically reasonable model for the pressure-driven component. In this case the total flux \( (\bar{\rho} \bar{u} \bar{c}'') \) and the actual source can be estimated using (7) and (3). It is remarkable that this estimation can be done during the post-processing stage. This method, which demonstrates good agreement with experimental data, is developed in the next sections.

In [15] approach (a) was followed. It was assumed that the actual averaged chemical source term in equation (2) is proportional to the probability of finding the flamelet at a given
position \( p_b(x) \); this probability is related to the probability of finding products \( P_b(x) \) at the given position by the relation

\[
P_b(x) = \int_{-\infty}^{\xi} p_t(\xi) \, d\xi \quad \Rightarrow \quad p_t(x) = \frac{\partial P_b(x)}{\partial x},
\]

(8)

where, without loss of generality, it has been assumed the flame propagating in the negative \( x \) direction (\( \partial P_b/\partial x > 0 \)). Note also the following expression for the averaged progress variable (as flamelets are thin, the probability to have \( \xi < 0 \) is small):

\[
\bar{c} = P_b1 + (1 - P_b)0 = P_b.
\]

(9)

Therefore we can write

\[
\bar{\rho} \bar{W} = \text{const} \frac{\partial \bar{c}}{\partial x},
\]

(10)

where the constant is equal to \( \rho_u U_i \) as can be shown by replacing the modelling expression (10) for the pressure-driven flux points toward higher \( \bar{c} \). This relation explicitly gives evidence of the two contributions composing \( \bar{\rho} \bar{c} \frac{\partial \bar{c}}{\partial x} \): (a) real turbulent transport, (b) a contribution which is proportional to the integral of the difference between the actual chemical source \( \bar{\rho} \bar{W} = \rho_u U_i \frac{d\bar{c}}{dx} \) and the model source \( \rho_u U_i \frac{d\bar{c}}{dx} \). This contribution can be expressed with simple algebraic manipulations as

\[
\rho_u U_i (\bar{c} - \bar{\bar{c}}) = \rho_u U_i \bar{c} (1 - \bar{\bar{c}}) \frac{\Theta - 1}{1 + \bar{\bar{c}}(\Theta - 1)}.
\]

(13)

with \( \Theta = \rho_u / \rho_b \). We note that it does not depend on the flame brush width.

As already mentioned, the nature of the progress variable transport (gradient or counter-gradient) can be determined by analysing the difference between the average conditional velocities \( \bar{u}_0 \) and \( \bar{u}_b \), in the cold and hot gases, respectively, as shown by relation (6). Solving the system of equations given by (6), (12) and the mass conservation equation in the two unknowns \( \bar{u}_0 \) and \( \bar{u}_b \),

\[
\bar{\rho} \bar{c} (1 - \bar{\bar{c}})(\bar{u}_b - \bar{\bar{u}}_b) = \rho_u U_i (\bar{c} - \bar{\bar{c}}) - \bar{D}_1 \frac{\partial \bar{c}}{\partial x},
\]

(14)

\[
(1 - \bar{\bar{c}})\bar{u}_0 + \bar{\bar{c}} \bar{u}_b = [1 + (\Theta - 1)\bar{\bar{c}}] U_i,
\]

(15)

we obtain the following expression for the contributions of the turbulent diffusion and the pressure-driven transport to the difference of the conditional velocities \( \Delta \bar{u} = \bar{u}_b - \bar{u}_0 \):

\[
\Delta \bar{u} = \Delta \bar{u}_\text{td} + \Delta \bar{u}_\text{p-d} = - \frac{D_1}{\bar{c}(1 - \bar{\bar{c}})} \frac{\partial \bar{c}}{\partial x} + (\Theta - 1) U_i.
\]

(16)

This relation shows that turbulent dispersion gives \( \Delta \bar{u}_\text{td} < 0 \) which corresponds to normal turbulent diffusion where the flux takes place in the direction from higher to lower \( \bar{c} \) while different gasdynamic acceleration of hot and cold gases gives \( \Delta \bar{u}_\text{p-d} > 0 \), which means that the pressure-driven flux points toward higher \( \bar{c} \).
In the case of a steady flame \((t \to \infty, \text{ constant thickness and velocity})\) the flame is described by equation (5), i.e. in (16) we must use \(D_t = 0\), resulting in the two conditional velocities being constant across the flame brush (in the coordinate system travelling with the flame \(\bar{u}_a = \bar{u}_{-\infty} = U_1^a\) and \(\bar{u}_b = \bar{u}_{+\infty} = \Theta U_1^b\)). It is interesting to note that in the DNS of turbulent premixed flames for the case of low turbulence levels \((u'/s_L < 1)\) performed by Veynante et al [5], the two conditional velocities have been found approximately constant across the flame brush and equal to \(s_L\) and \(\Theta s_L\), respectively.

This situation which is non-real for strongly wrinkled flamelets also implies constant conditional pressures \((\bar{p}_a(x) = \bar{p}_{-\infty} = \text{const}_1, \bar{p}_b(x) = \bar{p}_{+\infty} = \text{const}_2)\) as will be shown in the next section. Such a value of velocities in fact corresponds to the assumption that cold and hot volumes move inside the flame without mutual force interaction. Obviously this strongly overestimates the counter-gradient transport (for typical conditions approximately three times, see below) and corresponds to the hypothetical upper boundary of this phenomenon. We will call this analysis ‘the upper estimation of the counter-gradient phenomenon’. More realistic modelling must take into account this interaction, i.e. the acceleration of relatively slowly cold volumes by faster hot volumes and vice versa. Such a model based on the gasdynamic equations is developed in the next section and we will call it ‘the gasdynamic model’.

3. The gasdynamic model for counter-gradient transport

3.1. The pressure-driven component of the 1D stationary combustion front

The starting equation for the present analysis is the kinematic expression (6) which is exact for infinitely thin flamelets. To estimate the pressure-driven component of the total flux, we assume here that the conditional velocities in (6) are controlled only by the gasdynamic, i.e. we ignore the effect of turbulent dispersion. This gives us the opportunity to determine \(\bar{u}_a\) and \(\bar{u}_b\), giving their contribution to \(\Delta\bar{u}_{p-d}\) and \((\bar{p}u^2c^2)_{p-d}\) (here in non-dimensional form \(\bar{u}_a/U_1^a\), \(\bar{u}_b/U_1^b\) and \((\bar{p}u^2c^2)/\rho_0U_1^a\nu\); note that in the figures we do not use the subscript ‘\(p-d\)’ as a function of \(\tilde{c}\) or \(\bar{c}\). These functions are universal and do not depend on spatial distributions of \(\tilde{c}(x)\) or \(\bar{c}(x)\).

We consider the conservation equations for mass, momentum and additionally we assume conservation of the reactants total pressure across the flame brush. The equations are written here in the coordinate system travelling with the flame front, in non-dimensional form using \(U_1^a\), \(\rho_0\) and \(\rho_0(U_1^a)^2\) as the quantities to normalize velocity, density and pressure, respectively:

\[
(1 - \hat{c})\bar{u}_a + \hat{c} \bar{u}_b = 1, \tag{17}
\]

\[
\bar{p} + (1 - \hat{c})[\bar{u}_a^2 + \bar{u}_{\tilde{c}}^2] + \hat{c} \bar{u}_b^2 = \bar{p}_{-\infty} + 1 + u^2_{-\infty}, \tag{18}
\]

\[
\bar{p}_b + \frac{1}{2}[\bar{u}_a^2 + \bar{u}_{\tilde{c}}^2] = \bar{p}_{-\infty} + \frac{1}{2}(1 + u^2_{-\infty}), \tag{19}
\]

where the average pressure \(\bar{p}\) is given by \(\bar{p} = (1 - \hat{c})\bar{p}_a + \hat{c}\bar{p}_b\).

The analytical expressions for \(\bar{u}_a\) and \(\bar{u}_b\) obtained below do not depend on the turbulent velocity pulsations which implies special behaviour of \(\bar{u}_a^2\) and \(\bar{u}_{\tilde{c}}^2\) across the flame. If in fact we subtract equation (19) from equation (18) and require the fluctuating terms to cancel, we end up with the following two relations:

\[
(1 - \hat{c})\bar{u}_a^2 + \hat{c} \bar{u}_b^2 = \frac{1}{2}, \tag{20}
\]

\[
\left(\frac{1}{2} - \hat{c}\right)\bar{u}_{\tilde{c}}^2 + \frac{1}{2} \bar{u}_{\tilde{c}}^2 = \frac{1}{2} \bar{u}_{-\infty}^2. \tag{21}
\]
Assuming, in first approximation, $\alpha_a^2$ and $\alpha_b^2$ are constant across the flame brush, the second of these relations gives $\alpha_b^2 = \Theta \alpha_a^2$ which, without further discussion here, we consider reasonable (obviously $\alpha_a^2(\tilde{c})$ and $\alpha_b^2(\tilde{c})$ could be arbitrary but in this case there is no analytical solutions).

In the particular case of the upper estimation $\tilde{u}_a(x) = 1$ and $\tilde{u}_b(x) = \Theta$ analysed in the previous section, equation (17) becomes redundant and the system therefore reduces to two equations in two conditional pressures as yet unknown. It results that $\hat{p}_a(x) - \hat{p}_b(x) = (\Theta - 1)$, i.e. $\hat{p}_a(x) = \hat{p}_{-\infty}$ and $\hat{p}_b(x) = \hat{p}_{+\infty}$, which obviously is a hypothetical unrealistic case. We will instead analyse here the case where $\hat{p}_a = \hat{p}_b = \hat{p}$, i.e. ignoring the small difference of conditional pressures that cannot exceed the pressure drop across the flamelet and in experiments is usually much less than the overall pressure drop across the flame brush. In fact applying the momentum equation locally across the moving flamelet (assuming laminar flamelets) and across the overall combustion front, we have $(U_1^a \simeq u')$

$$\hat{p}_a(x) - \hat{p}_b(x) = \left( \frac{\dot{S}_L}{U_1^a} \right)^2 (\Theta - 1), \quad \hat{p}_{-\infty} - \hat{p}_{+\infty} = (\Theta - 1)$$

$$\Rightarrow \hat{p}_a(x) - \hat{p}_b(x) = \left( \frac{\dot{S}_L}{u'} \right)^2 \left[ \simeq Da^{-3/2} \right] \ll 1. \quad (22)$$

The estimate $Da^{-3/2}$ in the square brackets is obtained in the case of ISP flames by ignoring the influence of the flame brush broadening on the pressure drop across the flame and using expressions (32) and (33) for $U_1$ and $U_t$ presented in the next section. This shows that for Damköhler numbers typical of an industrial burner $Da \simeq 10$, the widened flamelet pressure drop is approximately 3% of the total one across the flame brush.

Under the assumption $\hat{p}_a - \hat{p}_b = 0$, we have from equation (20)

$$\left( \frac{1}{2} - \tilde{c} \right) \tilde{u}_a^2 + \frac{1}{\Theta} \tilde{c} \tilde{u}_b^2 = \frac{1}{2}. \quad (23)$$

The system given by (17) and (23) can be solved with simple algebraic manipulations. This yields the following expressions for $\tilde{u}_a$ and $\tilde{u}_b$:

$$\tilde{u}_b = -\beta + \sqrt{-4\alpha \gamma + \beta^2} \frac{2\alpha}{2\alpha}, \quad \tilde{u}_a = \frac{1 - \tilde{u}_b \tilde{c}}{1 - \tilde{c}}, \quad (24)$$

$$\alpha = \frac{\tilde{c}}{\Theta} \left[ \frac{0.5 - \tilde{c}}{1 - \tilde{c}} \right] \tilde{c} + 1 \right], \quad \beta = -2 \tilde{c} \frac{0.5 - \tilde{c}}{\Theta (1 - \tilde{c})^2}, \quad \gamma = \frac{0.5 - \tilde{c}}{(1 - \tilde{c})^2} - 0.5. \quad (25)$$

Formulae (24) and (25) also obviously follow from (18) and (19) when omitting all turbulent terms, i.e. they correspond to the interpretation of counter-gradient transport as conceptually a gasdynamic phenomenon and not a turbulent one.

The expression obtained now for the conditional velocities gives the opportunity to calculate the distribution of the average rate of product formation $\tilde{\rho} \tilde{W}$ within the combustion front using equation (5). In this equation the flux $\tilde{\mu} \tilde{W} \tilde{c}'$ includes both counter-gradient gasdynamic and gradient turbulent components, but simple estimates demonstrate that in the 1D stationary front with $\delta_{st}^\alpha \gg L$ at $\Theta \gg 1$, the gasdynamic component strongly prevails such that for the analysis of $\tilde{\rho} \tilde{W}$ in this paragraph we ignore the effect of turbulent diffusion.

Figure 1(a) shows a distribution of the theoretical conditional velocities non-dimensionalized with $U_1^a$ such that $\tilde{u}_b > \tilde{u}_a$, that corresponds to counter-gradient flux. The counter-gradient flux $\tilde{\mu} \tilde{W} \tilde{c}'$ corresponding to this case is presented in figure 3 when comparing theoretical results with the experimental data.
Figure 1. Distribution of (a) non-dimensional conditional velocities and (b) non-dimensional progress variable source terms across the flame brush. Gaussian distribution is assumed for $\bar{d}\bar{c}/dx$ ($\rho_u/\rho_b = 5$).

In connection with these results, we make three remarks.

(a) The pressure-driven transport does not depend (in contrast to turbulent diffusion) on the gradient of $\bar{c}$. So it cannot be interpreted as a specific turbulent diffusion flux inside the premixed flame $\bar{\rho}u'c'' = -\rho D_{\text{eff}}\nabla \bar{c}$ with negative effective turbulent diffusion coefficient $D_{\text{eff}} < 0$. It is instead the averaged convective component of transport caused by the coupling of bimodal density pulsations and averaged pressure gradient (more exactly conditional averaged pressure gradients in reactants and products).

(b) The pressure-driven transport is not inevitably counter gradient: in flows with a divergence of streamlines, the pressure increment connected with the increment of flow tube area can, in principle, prevail over the decrease due to combustion (e.g. in flames impinging on a wall with relatively small heat release). This obviously results in $\bar{u}_b < \bar{u}_u$ and gradient pressure-driven transport.

(c) It is obvious that the term ‘counter-gradient turbulent diffusion’, is a misnomer from the physical point of view. But the term ‘counter-gradient transport’, implying in fact that the mean flux has the opposite direction to the normal turbulent diffusion described in the gradient form, also does not seem a very appropriate one. We think that a more correct term is ‘non-gradient pressure-driven component of the flux’ (that does not depend directly on the mean progress variable gradient) keeping in mind that the total flux also contains the gradient (i.e. proportional to the gradient) turbulent component.

To determine the distribution of the actual combustion rate $\bar{\rho}\bar{W}$, we assume that in 1D stationary flames the spatial PDF of the flamelet $p_1 = \text{d}\bar{c}/\text{d}x$ is distributed as a Gaussian function (this distribution follows, for example, from the detailed Prudnikov measurements of the mean temperature profiles in premixed flame [17]):

$$\frac{\text{d}\bar{c}}{\text{d}x} = \frac{1}{\sqrt{2\pi} \sigma^2} e^{-(x-a)^2/2\sigma^2}.$$  

(26)

where $a$ is the position of $\bar{c} = 0.5$ and $\sigma^2$ is the the spatial dispersion of the random flamelet sheet. Introducing the non-dimensional spatial variable $\xi = (x-a)/\sqrt{2\sigma^2}$, the average heat
release non-dimensionalized with \( \rho_u U^{n}_{\infty} / \sqrt{2\sigma^2} \) is given by

\[
H = \frac{\bar{\rho} W}{\rho_u U^{n}_{\infty} / \sqrt{2\sigma^2}} = \frac{1}{\sqrt{\pi}} e^{-x^2} \frac{d}{dx} \left[ \tilde{c} - \hat{c}(1 - \hat{c}) \frac{\tilde{u}_b - \tilde{u}_a}{U^{n}_{\infty}} \right]. \tag{27}
\]

Figure 1(b) follows directly from (27) and shows the non-dimensional actual source term \( H_{ac} = |d\tilde{c}/dx| \) corresponding to the hypothetical case of the constant conditional velocities across the flame brush (i.e. based on the upper estimation of the transport), the actual source \( H_{gm} \) based on gasdynamic estimation of the conditional velocities (24) and the TFC model source term \( |d\tilde{c}/dx| \). The actual source term \( H_{gm} \) is shifted towards the front part of the flame with respect to the model source but it remains non-symmetrical in this coordinate. Note that using the strong overestimation of pressure-driven transport results in the symmetric distribution of \( H_{ac} \).

It is interesting to note that the shift of \( \bar{\rho} W \) to the back part of the flame brush for the case of the accurate estimation of transport has been observed experimentally in [3] and [16] where a large dataset of experimental data on turbulent premixed flames of various types (v-., conical, stagnation and swirl stabilized flames) was analysed. The experiments from [3, 16] will be considered in section 5 for a qualitative validation of the result obtained with the present gasdynamic model.

### 3.2. Extension to the general case

The extension and application of analytical results obtained in the previous section for 1D flames to the quantitative analysis of open 2D (as performed in the next section) and 3D flames deserve some explanation.

The exact unclosed transport equation for the progress variable in the general case is given by (steady situation)

\[
\bar{\rho} \mathbf{u} \cdot \nabla \tilde{c} = -\nabla \cdot (\bar{\rho} \mathbf{u} \tilde{c}^n) + \bar{\rho} W. \tag{28}
\]

This equation projected in direction \( n \) orthogonal to the flame (therefore parallel to \( \nabla \tilde{c} \) and oriented from reactants toward products) becomes

\[
\bar{\rho} \tilde{u}_n \frac{\partial \tilde{c}}{\partial n} = -\frac{\partial}{\partial n} (\bar{\rho} \tilde{u}_n \tilde{c}^n) + \bar{\rho} W. \tag{29}
\]

Assuming the theory developed in the previous sections to be still valid in the direction \( n \), we can write equation (29) as

\[
\bar{\rho} \tilde{u}_n \frac{\partial \tilde{c}}{\partial n} = \bar{\rho} W - \frac{\partial (\bar{\rho} \tilde{u}_n \tilde{c}^n)_{p=0,q}^{d=0,q} + \partial}{\partial n} \left[ \bar{\rho} D_{\xi} \frac{\partial \tilde{c}}{\partial n} \right]. \tag{30}
\]

We use here the expression for \( U_{\xi} \) corresponding to ISP flames with increasing brush width which is different from \( U^{n}_{\infty} \) (see next section). The estimation of the actual combustion rate distribution \( \bar{\rho} W \) from the splitting of the model source term \( \rho_u U_{\xi} \partial \tilde{c} / \partial n \) given in equation (30), is obtained using for \( \bar{\rho} \tilde{u}_n \tilde{c}^n_{p=q} \) the results of previous paragraph for 1D stationary front. The reason for this is that for \( \Theta \gg 1 \) and for realistic turbulence levels, the pressure drop of the 1D stationary front and ISP flame with increasing brush width are very close.

Finally we want to point out that the gasdynamic model can be extended to 2D and 3D cases as an invariant method for simulation of the pressure-driven flux \( (\bar{\rho} \tilde{u} \tilde{c}^n)_{p=0,q}^{d=0,q} \) by assuming reactants total pressure conservation and collinearity of the pressure-driven components \( (\tilde{u}_n)_{p=0,q} \) and \( (\tilde{u}_b)_{p=0,q} \) (at the same time common \( \tilde{u}_n \) and \( \tilde{u}_b \) containing both gasdynamic and turbulent contributions are in general not collinear). This gives the opportunity to calculate the fields...
of \( \bar{\rho} u \vec{c}(x) \) and \( \bar{\rho} \tilde{W}(x) \), i.e. to calculate all terms of the unclosed equation (28) in the post-processing stage of 3D simulations of premixed flames using the TFC combustion model equation:

\[
\tilde{\rho} \tilde{u} \cdot \nabla \tilde{c} = \nabla \cdot (\tilde{\rho} D \nabla \tilde{c}) + \rho_0 U_t \nabla |\tilde{c}|. \tag{31}
\]

These results will be published elsewhere.

4. Modelling of the turbulent combustion velocity \( U_t \) in ISP flames

The turbulent combustion velocity in the 1D and 3D equations (1) and (31) is the volumetric consumption rate \( U_t = U_t(\tilde{S}/S_0) \) that in the ISP flames with increasing brush width is smaller than the speed of the front edge of the flame. The widened flamelet parameters and the non-dimensional flamelet sheet area of the ISP flames at large \( Re_t \) and \( Da \) numbers have been estimated in [12] (see also [14]) based on the Kolmogorov methodology: the equilibrium fine-scale turbulence from the inertial interval controls the thickened flamelet combustion velocity giving \( U_t > S_0 \), the flamelet width \( \delta_f > \delta_L \) and the micro-turbulent heat transfer coefficient inside the flamelet \( \chi_f > \chi \), while the equilibrium small-scale flame wrinkles are the physical reason for practically constant flame surface area \( \tilde{S}/S_0 \) and hence \( U_t \). Finally non-equilibrium large-scale wrinkles control the flame brush width which increases similarly to a non-reacting mixing layer. The following expressions have been obtained:

\[
U_t \approx u' Da^{-1/2}, \quad \delta_f \approx L Da^{-3/2}, \quad \chi_f \approx D_s Da^{-2}
\]

\[
\Rightarrow \left( \frac{\tilde{S}}{S_0} \right) \approx Da^{3/4} \left( \frac{u'}{U_t} \right)^{3/2} \left( \frac{L}{\delta_t} \right)^{1/2} \tag{32}
\]

Though the ratio \( \delta_f/L \) increases with the turbulent velocity fluctuation \( u' \), according to estimations presented in [14], combustion extinction due to flamelet stretch takes place long before the formation of distributed volume combustion with \( \delta_f/L \geq 1 \) (corresponding to \( Da \leq 1 \)). Existing experimental data demonstrate that real flamelet widening does not change the combustion mechanism: direct measurements in [27] show that \( \delta_f = (3-5) \delta_L \), experimental estimations in [28] show that even if \( \delta_f \) is smaller than the thickness of the normal 1D laminar flame, at the same time it is larger than the width of the stretched laminar flame subject to the strain corresponding to small-scale turbulence, a typical estimation of \( \delta_t \) being of the order of 1 mm.

From relations (32), the following expression is obtained for the turbulent combustion velocity:

\[
U_t = U_t \left( \frac{\tilde{S}}{S_0} \right) = A u' Da^{1/4} = A u' \delta_L^{1/2} \chi^{1/4} L^{1/4}, \tag{33}
\]

where \( A \sim 1 \) is an empirical coefficient (practically \( A = 0.5 [13] \)), the molecular heat transfer coefficient \( \chi \) refers to the unburnt mixture in the simulations, i.e. \( \chi = \chi_u \). It is worth emphasizing that all of these expressions were derived from the physical model and, with the exception of the coefficient \( A \), they do not contain any quantitative empirical parameter. From (33) we can see that the turbulent flame speed depends on chemistry according to \( U_t \sim \tau_{ch}^{-1/4} \) in a much weaker way than the case of laminar combustion where \( s_L \sim \tau_{ch}^{-1/2} \).

Previous formulae are based on the Kolmogorov theory of the inertial micro-turbulence that in fact ignore pulsations of the dissipation rate that are responsible for the random flamelet stretch that reduces the local flamelet velocities and even cause at large strain rates their extinction. In the numerical analysis below, we take into account this effect applying a methodology based on the assumption of a log-normal PDF distribution of the dissipation rates
Gradient, counter-gradient transport in turbulent premixed flames

developed in [29] which was used in [13] for the simulation of combustion in the spherical bomb with artificial turbulization. According to this model, a stretch factor $G$, which represents the probability for the instantaneous turbulent kinetic energy dissipation rate $\epsilon$ to be less than the critical extinction value $\epsilon_{cr}$, has been introduced. Assuming a log-normal distribution for $\epsilon$ [29], the following expression for the stretching factor is obtained:

$$G = 0.5 \operatorname{erfc} \left[ -\frac{1}{\sqrt{2}\sigma} \left( \ln \left( \frac{\epsilon_{cr}}{\epsilon} \right) + \frac{\sigma}{2} \right) \right],$$

(34)

where erfc denotes the complementary error function, $\sigma = \mu \ln(L/\eta)$ the standard deviation ($\mu = 0.28$ being a constant). The final expression for $U_t$ is therefore given by

$$U_t = AGu' Da^{1/4},$$

(35)

It must be observed that an accurate estimation of $\epsilon_{cr}$ is necessary to correctly account for the ‘bending’ of $U_t$ in the dependence $U_t = f(u')$; in fact a reduction in $U_t$ results at large turbulent intensities.

Validation of the combustion model based on expression (35) for the turbulent flame speed and for the bending effect can be found in [13, 21, 30]. Application of the model to industrial premixed combustion can be found in [31] and validation for the case of a high-speed 2D turbulent premixed flame in [15].

5. Validation

The modelling ideas proposed here to account for transport of reacting components in turbulent premixed flames have been validated with three sets of experimental data.

The first of these consists of the experiments performed by Moss [1] for an open LPG/air turbulent premixed flame at stoichiometric conditions which are used here to validate the model for counter-gradient transport.

The second part of the validation is presented here only on a qualitative basis in order to show that the present model for counter-gradient transport gives the combustion rate distribution $\bar{\rho} W$ across the flame brush which is consistent with available experimental observations. The experimental data considered for this validation are from Cheng and Sheperd [3] and Cheng [16] who have analysed turbulent premixed flames in various configurations.

The third set of data consists of the experiments recently performed by Frank et al [6] for an axisymmetric natural gas/air open turbulent premixed flame stabilized in a co-flowing air stream by a pilot jet of hot gases. In this case, the conditional average velocities $\bar{u}_n$ and $\bar{u}_b$ have been measured for different values of the ratio $u'/S_L$ (mainly by changing the fuel/air mixture stoichiometry) and a transition from gradient to counter-gradient transport has been observed for decreasing $u'/S_L$. These data are therefore of particular interest for the overall validation of the model for the total flux developed here.

5.1. Validation with Moss experiments [1] for an open turbulent premixed flame

The schematic of the experiments for this open turbulent premixed flame is shown in figure 2(a). The burner is composed of a pipe, 5 cm in diameter and 60 cm in length. The pipe is fed with air and LPG (approximately 70% propane and 30% butane) which becomes fully premixed at the pipe exit. The velocity at the burner exit is $\bar{u} \simeq 4 \text{ m s}^{-1}$, approximately ten times the laminar flame speed. The rms velocity $\sqrt{\bar{u}^2}$ at the centre of the pipe exit is $0.5 \text{ m s}^{-1}$. 
Measurements have been taken across the flame brush along a line inclined at $23^\circ$ with the pipe axis. The angle observed between the flame and the burner axis was approximately $\theta - \phi = 16^\circ$. Concentration (the progress variable) was measured by a light scatter technique and velocities by laser Doppler velocimetry. Moss experimental data were subsequently post-processed in [7]; the data reported in this last reference have been adopted here for validation.

A full CFD analysis of this case has not been performed. Using the available information, we have estimated that the order of magnitude of the turbulent diffusion component within the total progress variable flux (the gradient component of transport) is small in comparison to the counter-gradient component. For these experiments we have in fact a turbulent flame speed of the order of $U_t \sim \overline{u} \sin (\theta - \phi) = 4 \sin(16) = 1.1 \text{ m s}^{-1}$. Assuming the integral turbulent length scale at the pipe exit to be in the range of $L \simeq 1.5 \text{ mm}$, we have $\epsilon \simeq 0.37\overline{u}'^2/L = 31 \text{ m}^2 \text{s}^{-3}$ and the turbulent diffusion coefficient $D_t \simeq 0.09(3/2\overline{u}^2)^2/\epsilon = 0.0004 \text{ m}^2 \text{s}^{-1}$. The flame brush increases in thickness according to the turbulent dispersion law, i.e. $\delta_f \simeq \sqrt{2D_tH/\overline{u}}$ where $H$ is the height from the pipe exit at which measurements have been taken, i.e. $H \simeq 0.0351 \text{ m}$ (assuming the flame to be flat). We have therefore $\overline{u}' \simeq \sqrt{2D_tH/\overline{u}}$ and assuming the maximum progress variable gradient to be approximately $1/\delta_l = 1/2.65E - 3m^{-1}$ at $\tilde{c} = 0.5$, we have from (16) a contribution to $(\tilde{u}_a - \tilde{u}_b)/U_t$ from turbulent dispersion roughly equal to $4E - 4/2.65E - 4/0.25/1.1 = 0.55$. The experimental value of the difference $(\tilde{u}_b - \tilde{u}_a)/U_t$ at this location is $\simeq 3$, i.e. the contribution to the conditional velocities by real turbulent transport is small in comparison to the contribution by counter-gradient transport. We compare, therefore, only the contribution to the transport generated by the pressure drop across the turbulent flame brush.

Figure 3 shows the non-dimensional averaged flux $\rho \overline{u}' \overline{c}'/\rho_a U_t$ and the non-dimensional conditional velocities $\tilde{u}_a/U_t$ and $\tilde{u}_b/U_t$ calculated using the gasdynamic model presented in this work. It can be seen that the upper estimation gives two to four times larger values of the flux compared to the experiments. At the same time the gasdynamic model gives instead,
good agreement with the experimentally determined progress variable flux in general, the small overestimation probably related to having neglected here the opposite effect of gradient diffusion. The conditional velocities calculated with this model are also in good agreement with the experimental data as shown in figure 3(b). Note that the predicted $\bar{u}_u$ and $\bar{u}_b$ are on average slightly smaller and larger, respectively, than the corresponding measured values which might be again the consequence of having neglected the effect of the turbulent diffusion component (which increases $\bar{u}_u$ and decreases $\bar{u}_b$).

5.2. Qualitative validation of actual source $\rho \dot{W}$ distribution with Cheng and Shepard experimental data

Cheng and coworkers [3, 16] have studied turbulent premixed flames in various configurations, including impinging, swirl stabilized and v-flames. These experiments include measurement of the flamelet crossing frequency $\nu(x)$ to which the averaged actual source distribution across the flame brush is proportional. In fact in the flamelet regime the averages source can be expressed as [32]

$$\rho \dot{W} = \rho_u n_1 \frac{\nu(x)}{U_n(x)},$$

where $\nu(x)$ is the flamelet crossing frequency (average number of flamelet crossings for unit of time) and $U_n(x)$ the mean flamelet crossing speed within the turbulent flame in a laboratory frame. The experimental results show that the flamelet crossing frequency is skewed toward values of $\dot{c} > 0.5$ with respect to the symmetrical heat release distribution predicted by the BML theory and proportional to $\bar{c}(1-\bar{c})$. Under the assumption of constant $U_n(x)$ and laminar flame speed $s_\lambda$, this also indicates that the averaged heat release is skewed in a similar fashion. Our gasdynamic model has been applied to the case of a turbulent premixed flame with $\Theta = 3$ to emulate the increment of velocity experimentally found by Cheng [16] in swirl stabilized flame ($u_{b_{\infty}}/u_{u_{\infty}} \simeq 3$) and assuming $d\bar{c}/dx$ and $d\bar{u}/dx$ to be distributed according to the symmetrical distribution $\bar{c}(1-\bar{c})$, consistently with the assumption in [16]. This distribution,
Figure 4. Qualitative comparison of the source $\bar{\rho} \tilde{W}$ across the flame brush with mean flamelet crossing frequency from Cheng experiments [3]. (——) symmetric BML distribution, (⊙) experimentally measured crossing frequency, (----) $\bar{\rho} \tilde{W}$ based on the gasdynamic model for counter-gradient transport developed here.

in fact, does not differ substantially from the Gaussian one previously assumed at the end of section 3. Figure 4 shows the distribution of the non-dimensional actual source term versus $\bar{c}$. The symmetric distribution from the BML theory is also shown. The figure clearly shows the skewness of the experimental heat release with respect to the theoretical symmetric distribution. As shown in figure 4, the model gives an actual source distribution which is skewed with respect to the symmetric one in a fashion similar to the experimental data. More detailed and quantitatively accurate analysis of these interesting experimental data will be the subject of future work. We believe that this experimental data and the successful agreement predicted by our model support the main ideas of the TFC combustion model, i.e. the clubbing in the model source term of the actual source $\bar{\rho} \tilde{W}$ and the gasdynamic component of the transport.

5.3. Validation with experiments by Frank et al [6] for turbulent premixed flames

The schematic of these experiments is shown in figure 2(b). The fuel used is natural gas and the fuel/air mixture flows out fully premixed from a 36 mm diameter piloted axisymmetric burner. The pilot gases have a very small flow rate (approximately 1.5% by volume of the main flow rate) and the co-flowing air has a velocity of 2.4 m s$^{-1}$. A perforated plate with 4 mm diameter holes spaced 7 mm apart is placed inside the burner to generate turbulence. Experimental measurements have been taken for five different flames corresponding to five different values of the ratio $u'/s_L$. This ratio has been adjusted generally changing the equivalence ratio of the fuel/air mixture (changing the laminar flame speed $s_L$). The characteristic data of these five flames are reported in table 1.

In these experiments digital particle image velocimetry (PIV) and planar laser-induced fluorescence (PLIF) are used to provide simultaneous 2D measurements of the velocity and the OH radicals concentration. Sharp gradients in the OH concentration are used to determine the instantaneous location of the flame front.
Table 1. Flame parameters in experiments by Frank et al [6].

| Flame | $\phi$ | $\Theta$ | $u'$ (m s$^{-1}$) | $u'/\bar{u}$ | $u'/\eta_L$ |
|-------|--------|----------|------------------|---------------|--------------|
| A     | 1.00   | 6.5      | 0.86             | 0.16          | 2.3          |
| B     | 0.70   | 5.2      | 0.53             | 0.10          | 3.1          |
| C     | 0.70   | 5.2      | 0.83             | 0.17          | 4.9          |
| D     | 0.61   | 4.6      | 0.79             | 0.17          | 8.8          |
| E     | 1.30   | 5.9      | 0.85             | 0.17          | 3.6          |

The radial and axial components of the conditionally averaged velocities and the Reynolds average progress variable $\bar{c}$ have been measured at 40 locations (5 axial $\times$ 8 radial) within a $2.8 \times 4.2$ mm$^2$ region of the flame located approximately 2.7 cm downstream of the burner mouth (see figure 2(b)).

The flames are generally parallel to the burner axis (exception applies mainly to flame A which is characterized by the largest $U_t$ and which is not considered in the present analysis). This is confirmed by the grouping of experimental data points at eight different values of $\bar{c}$ corresponding to eight radial locations within the imaged area. The 1D theory developed in the present work for modelling the pressure-driven transport component of the flux has therefore been applied here in the radial direction (to which the flame is approximately orthogonal). We therefore have

$$\rho u'c'' = \bar{\rho}c(1 - \bar{c})U_t \frac{\bar{u}_b - \bar{u}_a}{U_t} - \bar{\rho}D_t \frac{\partial \bar{c}}{\partial r},$$

(37)

where $\rho u'c''$ represents here the radial component of the total flux, and $\bar{u}_b/U_t$ and $\bar{u}_a/U_t$ are calculated using relations (24).

Flames B, C, D and E have been simulated using a commercial finite volume code where the TFC combustion model and the post-processing for extracting the total progress variable flux have been implemented. Modelling of turbulent transport is based on a standard ‘$k-\epsilon$’ model to calculate the eddy viscosity ($\mu_t = c_\mu \bar{\rho}k^2/\epsilon$ with $c_\mu = 0.09$). The computational domain has been discretized with approximately 15 000 computational cells. The hybrid central-upwind differencing scheme has been used for all transport equations. Model (34) has been used for calculating the flame stretch factor $G$ in the turbulent flame speed. The critical turbulent kinetic energy dissipation is given by $\epsilon_{cr} = 15\nu g_{cr}$, where $g_{cr}$ is the critical velocity gradient estimated here as the inverse of the chemical timescale: $g_{cr} = \tau_{ch}^{-1} = s_L^2/\chi u$.

Transport both of gradient and counter-gradient type was previously observed using DNS. Counter-gradient transport was observed, for example, in the simulations in [4] where $u'/\eta_L = 1$ while gradient transport was obtained by Trouvé and Poinsot [33] with $u'/\eta_L = 10$. Frank et al’s work is the first experimental evidence of the possibility to have a transition from counter-gradient to gradient transport in turbulent premixed flames. In general, the experiments show that this transition occurs when the ratio $u'/\eta_L$ increases. This is valid, for example, in the case of flames B, C, D as can be seen from figure 5 and table 1. According to the modelling ideas proposed here, it should be observed anyway that the $u'/\eta_L$ ratio is not sufficient to determine the type of transport and in what extent one component of the total flux dominates over the other. This is clear from (12) which shows that the gradient part of transport depends also from the actual thickness of the turbulent flame brush (i.e. from $d\bar{c}/dn$). This is also confirmed by the experiments: flame E is in fact characterized by a ratio $u'/\eta_L$ which is (slightly) larger than in flame B but with the total flux which is more biased toward the counter-gradient type. An observation of the experimental data shows that flame E is characterized by a substantially larger flame thickness and therefore smaller $d\bar{c}/dn$ than flame B. This explains why, despite
the slightly larger value of $u'/s_{11}$, flame E has an overall transport substantially more biased toward counter-gradient than flame B.

The transition from counter-gradient to gradient type of transport occurs in the experiments corresponding to flame C, between flame B (counter-gradient) and flame D (gradient). This transition occurs instead in the numerical simulations between flame B (mainly counter-gradient) and flame C (mainly gradient). The predicted flux in flame D is totally of gradient type and it overpredicts the gradient flux shown by the experiments.

Figures 6 and 7 show the comparison of conditional velocities. These data give the same information provided by the flux under a different point of view, $\bar{u}_b > \bar{u}_u$ meaning counter-gradient type of transport and $\bar{u}_b < \bar{u}_u$ meaning gradient type.

The transition from counter-gradient to gradient type of the transport is finally emphasized in figure 8. This figure shows progress variable contours and the region where the flux is of gradient type for flames E, B, C, D. The figure shows that transport in flame E is of the counter-gradient type nearly everywhere with only a small gradient type region existing near the splitter plate where the progress variable attains its maximum gradient. The region of gradient transport increases in size for flames B and C and becomes very broad in the case of flame D because of the very lean conditions of this flame.

To close this section we want to emphasize two important points. The first one is that, according to the idea presented here, the transport term $\rho u'u''c''$ can have overall gradient nature even in the case of flamelets combustion. As already mentioned, the gradient component in fact depends not only on the turbulent diffusion coefficient but also on the local gradient of the progress variable which is very large in the locations from where the turbulent flame starts developing (theoretically infinite in these locations). Therefore, according to this point there will always exist a region around the flame attachment locations where the gradient component is dominant over the counter-gradient one (see figure 8) and then $\rho u'u''c''$ has overall gradient.

**Figure 5.** Comparison with experiments from Frank et al [6]. Progress variable radial flux. ($\triangle$) flame D, (○) flame C, (□) flame B, (♦) flame E. Left: experiments; right: present calculations based on the gasdynamic model.
nature. In particular we can expect the size of this region to be controlled by the ratio $u'/s_L$. This idea is different from the one presented in [34] where, by analysing direct numerical simulation data, it has been proposed that in the laminar flamelet regime transport can be only of counter-gradient type and gradient transport is present only when the flame front is greatly perturbed from that of a laminar flamelet, i.e. the flamelets become so widened that combustion takes place with the distributed volumetric mechanism instead that the thin flamelet one.

The second point is that the theory of the TFC combustion model also predicts the existence of thin flamelets (not necessarily laminar) at much higher $Re_f$ numbers typical in large-scale and high velocities industrial burner. We therefore also expect counter-gradient transport and the transition to gradient in this case. Regarding this, numerical simulation of Moreau experiments [35] for high velocity premixed combustion performed in [15] based on the upper estimation of the pressure-driven transport component have shown the presence of counter-gradient transport practically along the whole length of the flame. We did not focus in that paper on the transition effect but it is pertinent to note here that in those simulations gradient transport took place only in a small region at the beginning of the flame and transition to counter-gradient transport occurred at distance $\sim 1$ cm from the splitter plate anchoring the flame (obviously using the upper estimation of the counter-gradient component yields the lower estimation of this distance).
6. Conclusions

The phenomena of counter-gradient transport and gradual transition from gradient to counter-gradient transport in turbulent premixed flames burning according to the flamelet combustion mechanism have been analysed theoretically and numerically in the present paper. We have the following conclusions.

1. The analysis starts from the assumption that the averaged flux of the progress variable can be presented, as shown by (7), as the sum of two contributions which can be described independently: a gradient turbulent diffusion component (due to the spatial dispersion of the flamelet sheet by the velocity pulsations) and a counter gradient pressure-driven one (due to different gasdynamic acceleration of cold and hot volumes in a non-uniform mean pressure field). The total transport is therefore counter gradient when the second contribution prevails over the first one and vice versa.

Using our TFC combustion model, we have predicted counter-gradient transport and its transition to gradient (when this becomes the dominant contribution) experimentally observed in [6]. In particular, our model, consistent with experiment, predicts a turbulent flame brush width which increases in the same fashion of a non-reacting mixing layer as it is controlled only by the turbulent diffusion component of the flux (independently from the total transport being of gradient or counter-gradient nature). In such a way we have resolved the observed apparent contradiction between increasing brush width and the counter-gradient transport. For estimation of the turbulent diffusion component \((\bar{\rho}u\tilde{c}')_{ld}\) in our numerical simulations we have used the standard ‘\(k–\varepsilon\)’ turbulence model.
2. A gasdynamic model has been developed for the pressure-driven component of the total flux \( \bar{\rho} \tilde{u} (\epsilon) \). This model describes this component directly in terms of the conditional averaged velocities \( \bar{u}_a \) and \( \bar{u}_b \) which obey the gasdynamic equations. The key point of this model is an assumption that the total pressure of reactants flow inside the flame is constant. Furthermore, this model does not contain empirical constants. In the case of the 1D stationary combustion front for physically reasonable behaviour of turbulence the model yields an analytical solution, equation (24), which demonstrates that the pressure-driven component does not depend directly on the gradient of the mean \( \tilde{c} \) field but on the pressure drop across the flame brush.

3. We have applied this gasdynamic model to the analysis of transport in open flames, in the direction orthogonal to the flame. We have compared our results with classical data from Moss [1] where counter-gradient nature of the transport is clearly shown and with recent experiments of Frank et al. [6] where at some fixed distance from the burner inlet counter-gradient transport transforms into gradient one when the ratio between the turbulent velocity fluctuation \( \tilde{u} \) and the laminar flame speed \( s_L \) increases as consequence of the variation in the fuel/air mixture equivalence ratio. In Moss data the turbulent diffusion component of the flux has been estimated small compared to the counter-gradient one and then neglected. To describe the transition phenomenon experimentally observed in [6], the turbulent diffusion component has been estimated using the standard ‘\( k-\epsilon \)’ turbulence model.

Numerical results demonstrate quite reasonable quantitative agreement with both these experimental data.

4. In the TFC combustion model the actual source term \( \bar{\rho} \tilde{W} \) and pressure-driven transport component are clubbed together under a single model source term, equation (30). The gasdynamic model for pressure-driven transport makes possible extracting the actual source
term from the model source term determined via numerical simulation during post-processing stage. Because of the counter-gradient nature of the pressure-driven component in open flames, the actual source term is shifted to the front part of the flame with respect to the model source. The skewed distribution of the actual source term is qualitatively confirmed by the experimental data in [3, 16] where the flamelet crossing frequency was determined.

5. We think that the terminology ‘counter-gradient turbulent diffusion in premixed flames’ is inappropriate for the counter-gradient phenomenon which has gasdynamic nature. We have used instead the term ‘the counter-gradient transport’ which we also think is not fully adequate as the gasdynamic transport is not proportional to the progress variable gradient. A possible more correct terminology is ‘non-gradient pressure-driven transport’ (this flux is in fact ‘counter-gradient’ when the pressure field is mainly generated by combustion but it could be ‘gradient’ for sufficiently large external disturbance of the pressure field).

6. Finally, from the practical point of view, the present analysis shows that quantitatively correct modelling of the counter-gradient transport is possible without invoking the second-order moment transport models. The combination of the ‘$k$–$\epsilon$’ model for the gradient turbulent transport component and the developed gasdynamic model for counter-gradient component results in satisfactory predictions of the counter-gradient phenomenon and its transition into the gradient transport and can also describe the actual source term, i.e. all terms of the unclosed equations (2) and (28) could be predicted.

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