CRS4
Center for Advanced Studies, Research and Development in Sardinia
Cagliari - Italy

Development of the TFC Turbulent Premixed Combustion Model: the Combustion Rates and the Counter-gradient Transport Phenomenon.

Vladimir Zimont

Abstract.

We analyze two "bottlenecks" of turbulent premixed combustion modeling at developed turbulence corresponding to modern lean-premixed large-scale industrial gas-turbine combusters when instantaneous combustion (heat release) takes place in thin strongly wrinkled surfaces (flamelet sheets). They are as follows: 1) fundamental failure of statistical combustion models in terms of moment or PDF equations to resolve the structure flamelet reaction zones and small-scale sheet wrinkles controlling actual combustion rates (so called challenge of turbulent combustion); 2) well-known the counter-gradient turbulent transport phenomenon that renders direct application of existing turbulence model together with combustion models impossible at simulation of premixed combustion.

The first problems was solved in analyzed our TFC combustion model by assumption of equilibrium small-scale structure if these flamelet sheet structure, i. e. in fact by using implicitly Kolmogorov methodology developed for simulation non-reactive turbulent flows. In this report we develop Kolmogorov ideas for combustion modeling, analyze their possibilities and restrictions, these results provide the necessary background to the TFC model.

For solving the second problem we developed in this report some gas-dynamics model of pressure-driven counter-gradient component of transport that gave an opportunity together with the combustion model and traditional "k-e" turbulence model to simulate the counter-gradient transport phenomenon and it transition to the gradient transport at variations on the combustion operation regime.

So the report contains:

1. Statement and analysis of the method of attack to the problem of correct description in context of model statistical equations combustion rates at strong turbulence and fast chemistry when combustion rates are controlled by unreserved small-scale coupling of turbulence and chemistry, some answer at practical combustion simulations to the "challenge of turbulent combustion";

2. Analysis of the counter-gradient transport phenomenon observed in premixed turbulent flames, description of developed gas-dynamics model of the pressure-driven mechanism of it, results of numerical simulations of turbulent flames with quantitative analysis of this phenomenon and its transition to the gradient turbulent diffusion, comparison with recently published experimental data where this transition was the first observed.

We discussed this results from the industrial standpoint at combustion simulations of real ecologically clean lean premexed combustion chambers.

1. Introduction

We analyze two "bottlenecks" of turbulent premixed combustion modeling at developed turbulence corresponding to modern lean-premixed large-scale industrial gas-turbine combusters when instantaneous combustion (heat release) takes place in thin strongly wrinkled surfaces (flamelet sheets). They are as follows: 1) fundamental failure of statistical combustion models in terms of moment or PDF equations to resolve the structure flamelet reaction zones and small-scale sheet wrinkles controlling actual combustion rates (so called challenge of turbulent combustion); 2) well-known the counter-gradient turbulent transport phenomenon that renders direct application of existing turbulence model together with combustion models impossible at simulation of premixed combustion.

I. The term "challenge" has been used last years in scientific papers [1]-[2] to emphasize that there is the fundamental obstacle on the way of developing general-purpose turbulent combustion models for solving practical problems, i. e. the models are intended for actual turbulent combustion simulations of real burners that would give not only quantitative agreement with experimental data (it is in itself is very difficalt problem) but also to have ability to predict the turbulent combustion process trends at variations of compositions, the operation conditions, burner geometry and so on. The reason of it is connected with the fact that at real turbulence and chemical combustion reaction rates at real conditions are controlled by such small-scale coupling of chemistry, microturbulent mixing and molecular diffusion that cannot be resolved in frames of moment, PDF and LES combustion models[2]. So "the accurate prediction of mean reaction rates, which can be influenced strongly by molecular diffusion caused by small-scale turbulent mixing, represents the central problem and challenge of turbulent combustion"[2]. This problem is especially pressing in the case of premixed combustion as for nonpremixed combustion at sufficiently fast chemistry the problem of flame aerodynamics and combustion rates simulation often can be reduced to analysis of mixing, i. e. this coupling becomes the secondary importance factor.

This challenge connected with the inability to describe mentioned small-scale coupling is the most distinct at premixed combustion at real Reynolds Re and Damköhler Da numbers when takes place the flamelet combustion mechanism with very thin sheet-type reaction zones. It means in fact that turbulent combustion models cannot predict, for example, the local distribution of the averaged combustion rate across the flame $\overline{\rho W(x)} = \rho_u U_f \overline{\Sigma}(x)$ or the turbulent combustion speed $U_t = U_f \overline{(S/S_0)} = (1/\rho_u) \int \overline{\rho W(x)}\, dx$ in relation to the fuel, the air excess coefficient, initial temperature of a mixture, the pressure and so on over more or less range of these parameters. Here U_f is the flamelet velocity (at theoretical estimations U_f often assumed to be the laminar combustion velocity U_l), $\overline{\Sigma}$ is the flamelet surface density, $\overline{(S/S_0)}$ is the dimensionless flamelet surface area, ρ_u is the density of unburned mixture.

It should be noted that Professor Ken Bray who worked many years in the field of turbulent (mainly premixed) combustion and have fundamental results in this field titled his review Hottel Planary lecture "Challenge of turbulent combustion" instead of, for example, "Achevements of turbulent combustion" because as he wrote "the challange appearing in the title of this paper is directed to the whole combustion research community". Proposed paper is an attempt to give a constractive answer to this appeal and to invoke some idea that gives some necessary information about this nonresolved coupling that can be used at designing of turbulent combustion models.

At first glance, this problem seems intractable: we cannot resolve in the frames of combustion model equations space and time scales that are necessary for a correct description of chemical kinetics, molecular transfer processes and their coupling. If it were so it would mean that to design physically reasonable combustion model containing the key combustion mechanisms (and between them one of the main is mentioned coupling) would be impossible. It would mean that every possible combustion model even if it reproduces some limited experiments (by tuning empirical constants) could not describe a large ensemble of experimental data and, in general, could not give correct trends.

One of the aims of this paper is to show that from the application-oriented point of view this problem does not look unresolved, and that there are ideas that could give positive applied results. Our approach is based on an assumption of universal small-scale turbulent structure of reaction zones that is in equilibrium with large-scale turbulent movement. We try to formulate a practicable line of inquiry to answer this challenge and analyze its practical application to simulation of premixed combustion for large Re and Da numbers. The character of this challenge and the main idea of our answer in fact is not new. A similar challenge was in past that of practical turbulence modeling as existed methods could not predict accurately the turbulent dissipation rates, which was influenced strongly by small-scale vortexes. Kolmogorov gave a practical answer to this challenge and it was in fact the cornerstone of all turbulence models for large Re numbers. In both cases, turbulence and turbulent combustion, controlling processes take place in such small scales that could not be resolved by model equations.

Kolmogorov answer[3] was based on the assumption of equilibrium fine-scale turbulence (where actual dissipation took place) due to the Richardson cascade mechanism. This assumption yields that the turbulent dissipation rate is controlled by large scale turbulence and at large Reynolds numbers the averaged dissipation rate $\overline{\varepsilon}$ does not depend on the molecular viscosity coefficient: $\overline{\varepsilon} = Cu'^3/L$, where $C \sim 1$. Next year Kolmogorov designed using these ideas the first two parametric turbulence model ("$k - \omega$")[4], where the dissipation rate was described not in terms of the molecular viscosity coefficient, but through this physical mechanism.

In this paper we extend this Kolmogorov methodology to model turbulent combustion at large Reynolds and Damköhler numbers. We in fact used these ideas in past[5] and TFC model that has some application (in "Fluent 5", for example) is based on them but we did not formulate is as a principle. Here we formulate and analyze the feasibility of these ideas for combustion modeling, this ideas give and opportunity to understand better some limitation of existing combustion models and provide the necessary background to the TFC model.

II. Counter-gradient transport is a phenomenon which commonly occurs in turbulent premixed combustion. It is connected with the differential acceleration of hot and cold fluid volumes under the flame pressure drop generated by heat release. It has been experimentally observed for example in open [1] and impinging [2] flames. This phenomenon is closely connected with the flamelets combustion mechanism in turbulent premixed flames, when combustion takes place in thin and strongly wrinkled layers called flamelet sheets. In laboratory flows with relatively low turbulence Reynolds numbers $Re_t = u'\, L/\nu$ these flamelet sheets are laminar flames with speed s_L and thickness δ_L highly wrinkled by small scale turbulence.

A new phenomenon consisting in the transition from counter-gradient to gradient transport has been recently experimentally observed [3]. According to these experiments in open premixed flames, counter-gradient transport observed at some fixed distance from the burner inlet transforms into gradient transport when the ratio between the turbulent velocity fluctuation u' and the laminar flame speed s_L increases as consequence of the variation in the fuel/air mixture equivalence ratio. In the present paper we focus the main attention on the theoretical analysis of this phenomenon, its numerical simulation and comparison with these laboratory experimental data.

Another issue addressed in the report is the existence of counter-gradient transport and the phenomenon of transition to gradient transport in large-scale high velocity flames in industrial combustors where direct measurements are very difficult. Our point of view in fact is that in industrial burners characterized by much larger Reynolds and Damköhler ($Da\tau_t/\tau_{ch}$ ratio between the integral turbulence and chemical times scales) numbers, presence of counter-gradient transport is highly probable. At large turbulent Reynolds numbers in fact, turbulence has a very wide continuous spectrum of eddies including very small dissipative vortices. At Damköhler numbers in industrial burners ($Da \simeq 10$) the size of the smallest Kolmogorov eddies $\eta = L\,Re_t^{-3/4}$ (where L is the integral length scale of turbulence) can be comparable or less than the laminar flame thickness, i.e. $\eta \leq \delta_L$. In this case we have wrinkled flamelets which can have thickness δ_f larger than laminar one δ_L because of the intensification in the transport processes by small-scale eddies with size less than δ_f. If at the same time the turbulent integral length scale L is much larger that δ_f, it means that combustion takes place in a strongly wrinkled sheet that is not laminar but appears broadened by small scale turbulence.

As the counter-gradient phenomenon is closely connected with the flamelet combustion mechanism, it is important to stress that the successive falling of more and more large eddies in in the expanding broadened flamelet has a limit. Equilibrium thickness is in fact achieved when convection, diffusion and heat release intensity have the same order of magnitude [4, 5]. Such expanded equilibrium flamelet, in accordance with the theoretical estimations at $Da >> 1$, is always characterized by $\delta_f > \delta_L$. Existing direct experimental data demonstrate that $\delta_f \simeq (3-5)\,\delta_L$ [6] such that the typical order of magnitude of δ_f is $\simeq 1mm$.

Unfortunately, there is no direct evidence that combustion takes place in thin and strongly wrinkled sheets also in industrial combustors, where u' can be quite large, as there are not direct measurements of the flamelet parameters in these kind of burners. In spite anyway of the increase in the ratio δ_f/L for increasing u' (which gives a decrease in Da), estimations presented in [5] show that extinction due to flamelet stretch takes place long before the formation of distributed combustion zones with $\delta_f/L \geq 1$. We therefore believe that counter-gradient transport and the phenomenon of transition to gradient transport exist also in flames in industrial burners. We must nevertheless mention that a different point of view, which we do not share, is often found in literature. According to this, industrial combustion occurs with the distributed volume mechanism corresponding to the model of stirred reactor. Clearly, in this case a well-defined counter-gradient phenomenon is not likely to be possible.

The effect of small-scale turbulence with size larger than δ_f is to make the flamelet sheet wrinkled. This increases the flamelet surface/turbulent flame cross sectional areas ratio $\overline{S}/S_0$ which, together with the expanded flamelet combustion velocity U_f, controls the turbulent burning velocity, i.e. $U_t = U_f\,\overline{S}/S_0$. It is very significant to emphasize here that, for the case of fully developed turbulence, the main contribution to the increase in $\overline{S}/S_0$ is given by small-scale wrinkles generated by eddies with sizes $\delta_f \leq d << L$, while large-scale wrinkles caused by eddies with $d \sim L$ mainly control the flame brush width and give only relatively small contribution to $\overline{S}/S_0$.

The consequence of this situation is that immediately after combustion has started, we have not only equilibrium broadened flamelets with $U_f = const$ but also equilibrium small-scale wrinkles structure of the flamelet sheet. On the other hand, in the case of $u' >> U_f$ and during a relatively long period of time, the large scale wrinkles structure will not be in equilibrium (according to estimations in [7] during a time scale of $t \sim \tau_t (u'/U_f)^2$). The practical result of this is that during this relatively long period of time, an intermediate propagation combustion regime, proceeding the formation of a stationary combustion front structure with constant U_t^{st} and δ_t^{st}, takes place. Premixed combustion within this intermediate regime takes place therefore in flames with increasing flame brush width controlled by turbulent diffusion and with practically constant turbulent combustion velocity $U_t \sim const$ (more exactly with slowly increasing U_t due to relatively small contribution to the flamelet sheet area of non-equilibrium large wrinkles). Strictly speaking this arguments are valid only in the hypothetical case of combustion at constant density. Leaning upon experimental observations, we have postulated the same physical picture and the possibility to use the associated combustion model also in the real case of variable density.

Prudnikov was probably the first who established this peculiarity of premixed combustion during the late '50s. He in fact 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, and flame brush with increasing width which was controlled mainly by the cold flow turbulent diffusion coefficient. This result was first published in 1959 in a nowadays hard-to-find preprint and successively presented in [8]. The analysis of the latest experimental data in the paper [32] confirms also that turbulent premixed combustion occurs in flames with increasing brush width.

This particular regime of turbulent premixed combustion has been named Intermediate Steady Propagation (ISP) regime [5, 7] (intermediate asymptotic between the initial stage of formation of an equilibrium small-scale wrinkled structure and final stage of a turbulent flame completely in equilibrium with $U_t = const$ and $\delta_t = const$) in order to emphasize the concept of flames characterized by approximately constant turbulent flame speed (constant in the case of homogeneous turbulence and equilibrium in more complex situations) and a flame brush thickness which grows according to the turbulent dispersion law. The reason of ISP flames prevailing within an industrial combustor is given by the the fact that combustion in this burners is still well far from the final stage of fully steady conditions (constant U_t and δ_t) as the mechanism which is responsible for compensating the turbulent growth of the flame brush thickness becomes effective on a time scale much larger than the average residence time.

In the case of open axisymmetric premixed flame analyzed in this paper, this concept means that a premixed flame is simply a turbulent mixing layer with increasing width (similarly to the mixing layer of a non reactive flow), propagating in the fresh mixture with equilibrium combustion velocity U_t. This propagating mixing layer crosses at the combustor axis long before the formation of the flame with complete equilibrium structure.

Given that a flame in complete equilibrium with $U_t = const$ and $\delta_t = const$ is unattainable in practical devices under strong turbulence conditions, it is reasonable to develop a turbulent premixed combustion model which describes only the ISP regime and does not contain the mechanisms responsible for reaching the final full equilibrium flame structure. This model which was proposed and investigated in the papers [4, 5] and called in [9] the Turbulent Flame Closure (TFC) combustion model, has been further developed and used here for the description of counter-gradient and gradient transport as well as their transition in correspondence of variations in the fuel/air mixture equivalence ratio.

The reasons of experimentally observed turbulent premixed flames with increasing width might not be completely clear at this stage, deserving then further comments. A contradiction in fact apparently arises between the idea of a turbulent flame brush which grows in thickness according to the turbulent dispersion law (i.e. according to a positive turbulent diffusion coefficient) and the experimentally observed counter-gradient nature of the progress variable transport which has been so far the subject of many theoretical and numerical studies. For example Moss [1] has observed this phenomenon in his early experiments for an open premixed flame; subsequently counter-gradient transport has been observed in the experiments performed in [2, 10] and in direct numerical simulations of turbulent premixed flame [11]. It has been consequently argued that the assumption of gradient transport for the progress variable or the reactive components is in general not appropriate for turbulent premixed flames and sometimes also that, as a consequence of "counter-gradient diffusion", the flame brush thickness decreases instead than increasing [11]. The possibility to model the turbulent transport of reactive components in turbulent premixed combustion using models based on gradient transport and a conveniently estimated turbulent diffusion coefficient has been therefore abandoned a long time ago and the attention has been totally concentrated on second order moment closure methods [12, 13].

The point of view developed in this paper is that the term "counter-gradient diffusion" often given in the literature to describe transport of the progress variable in flows with heat release has its origin in the interpretation of this transport as a pure turbulent diffusion flux and in the attempts sometimes to connect it to a turbulent diffusion coefficient, that for agreement with experimental data must be negative (counter-gradient, i. e. negative diffusion). We believe instead that this transport is controlled both by turbulence (turbulent diffusion) and by specific gas-dynamics displacement of hot and cold volumes driven by pressure variation across the flame brush (in open flames the pressure is decreasing because of heat release). We will therefore use hereafter the term *transport* instead than *turbulent diffusion* especially in the case of the counter-gradient phenomenon in order to emphasize the presence of an additional transport mechanism, usually prevailing in turbulent premixed flames, which is not directly associated with the turbulent pulsations of velocities.

It will be shown that the progress variable transport in open turbulent premixed flames has gradient character at the beginning of the flame, where turbulent diffusion prevails over the gas-dynamics transport component (counter-gradient) because of the small flame brush width, and has counter-gradient character at larger distance where the gasdynanamics effect starts to dominate. It is worth also mentioning that, if we create in a combustor turbulent premixed flow a nonuniform distribution of very small passive contaminants, there are strong grounds to believe that the nature of its transport would be of the gradient type controlled by the turbulent diffusion coefficient. This approach was in fact used in his experimental work by Prudnikov[14] where measurements of turbulent intensity and turbulent diffusion coefficient were performed using the optical diffusion method based on photo registration of the diffusion wake of luminous particles behind a point source placed in premixed flames. These experimental results did not show any "counter-gradient turbulent diffusion" at least at the back part of the flames where measurements were made.

In the TFC combustion model there is no contradiction between the experimentally observed increase of the flame brush width and the measured counter-gradient transport of the progress variable. The convective nature of the counter-gradient component does not directly affect in the model the flame brush thickness which, similarly to a non reactive mixing layer, is controlled by physical turbulent diffusion. In the present simulations of the counter-gradient phenomenon and its transition into gradient transport we have estimated the turbulent diffusion component using the standard $k-\epsilon$ model and the counter-gradient transport using a novel gas-dynamics model developed here. These simulations demonstrate good agreement with the trend shown by the experimental data in the transition from counter-gradient to gradient transport.

A similar idea was developed earlier in the paper [7] where high Reynolds number turbulent premixed combustion in a channel was simulated using the TFC model. In that case anyway a very rough and less quantitatively accurate, estimation of the gasdinamics transport component was used. This was based on the assumption of constant conditionally averaged velocities in the unburnt and burnt mixtures and gave the possible upper boundary for the counter-gradient transport component. It must be mentioned that conclusions consistent with this assumption followed also from the analysis of DNS results of turbulent premixed flames for the case of low turbulence levels ($u'/s_L < 1$) performed by Veynante *et al* [11] when the two conditional velocities were found approximately constant across the flame brush and respectively equal to s_L and $\Theta\, s_L$, where $\Theta = \rho_u/\rho_b$ is the ratio between the unburnt and burnt gases densities.

In our case of strong turbulence ($u'/s_L >> 1$) such assumption is not valid. As it will be seen below in fact the assumption of constant conditional velocities across the turbulent flame brush results in a much larger effect. Comparing with experimental data available from Moss [1] for an open turbulent premixed flame, it is shown that the model developed in [7] overestimates the counter-gradient part of the total turbulent scalar flux by approximately three times.

In the present work a new improved model for the pressure-driven transport component is developed. This is based on the assumption of equal conditional average pressure $\overline{p_u}$ and $\overline{p_b}$ respectively in the unburnt and burnt fluid volumes. This model gives excellent agreement with Moss experimental data.

It is furthermore demonstrated that the capability to correctly model the pressure-driven transport component directly affects the accuracy with which we can estimate the heat release distribution. Consistently with the experimental observation made in [10, 15], it is shown, in fact, that the improved model for counter-gradient transport developed here yields an average heat release which is skewed towards value of the progress variable $\overline{c} > 0.5$ with respect to the symmetric theoretical distribution of the Bray-Moss-Libby (BML) model (also predicted by our previous assumption of constant conditional velocities across the flame brush).

Experimental evidence supporting the present analysis is represented by the recent experimental data from [3]. According to these experiments in open premixed flames a transition from counter-gradient to gradient transport is observed at a given distance from the burner inlet when the ratio between the turbulent velocity fluctuation and the laminar flame speed u'/s_L increases, suggesting that physical turbulent diffusion becomes dominant on the gas-dynamics pressure-driven mechanism. These experiments have been considered here for assessing the feasibility of the proposed modeling idea for the counter-gradient transport phenomenon.

2. Kinematics picture of premixed combustion flame

In the case under review combustion takes place in thin highly wrinkled flamelet sheets that separates the reactants from the products and propagates relative to the reactants with velocity U_f. In our model the flamelet is not the laminar flame with the combustion velocity U_l , but is thickened by small scale turbulence flamelet with the combustion velocity $U_f > U_l$.

As a preliminary we assume that gas density $\rho = const$, i. e. combustion does not change hydrodynamic flow and turbulence. Let denote P_u, P_b and P_f the probabilities of unburned mixture (reactants), burned mixture (products) and flamelet compositions in every point of the turbulent flame. For kinematics description of the flame mechanism we assume that $P_u + P_b = 1$ as $P_f << 1$ (the flamelet combustion on mechanism). We will use the combustion product probability $P_b(\vec{x}, t)$ or the averaged progress variable $\overline{c}(\vec{x}, t)$ (at $\rho = const$, $\overline{c} = P_b$) for description of turbulent flames.

At first we will analyze a 1-D non-stationary flame front in gas moving along the x-axis at $u' >> U_f$. Assume that at the initial time $t = 0$ for $x < 0$ $\overline{c} = 1$ and for $x > 0$ $\overline{c} = 0$. For $t > 0$ the plane boundary becomes wrinkled at $t > 0$ and its dimensional area $\overline{(S(t)/S_0)}$ grows. So at the beginning we have a combustion front with increasing turbulent combustion velocity $U_t(t)$ and increasing flame brush width $\delta_t(t)$.

Let $F(k, t)$ be a spectrum of the flamelet sheet thought as being a random surface $x = h(y, z, t)$, where k is the wave-number ($k = 2\pi/\Lambda$ and Λ is the wave length). The dispersion of the flamelet sheet is determined by the large waves of the random flamelet sheet (small k): $\sigma^2(t) = \overline{(x - \overline{x})^2} = \int_0^\infty F(k, t)\, dk$, whereas the flamelet area is determined by the small waves (large k): $\overline{(S(t)/S(0))} = const \int_0^\infty k^2 F(k, t)\, dk$, where the $const$ is order unity (in the case of Gauss random surface it can be calculated exactly). Without combustion $\overline{(S(t)/S(0))}$ increases very fast (approximately exponentially), but in the case of combustion the flamelet progress suppresses this fast increase. So after some time, that probably is the order of so called Gibson time $\tau_G = L_G/U_f = \tau_t(U_f/u')^2$, where $\tau_t = L/u'$[9], we would have $U_t \approx const$ (more exactly very slow increasing of U_t).

For $\tau_G < t < \tau_*$, when suppression of large wrinkles of the sheet by moving flamelet is negligible, we have flame with increasing brush width δ_t that is growing in accordance with the turbulent diffusion law $\delta_t \sim (\sigma^2)^{1/2} = (2D_t t)^{1/2}$, where σ^2 is the dispersion of the flamelet sheet and D_t is the turbulent diffusion coefficient. We estimated the time $\tau_* \approx 2D_t/U_f^2 \approx 2\tau_t(u'/U_f)^2$ from the condition $(\sigma^2(\tau_*))^{1/2} \approx U_f \tau_*$, i. e. when transport due to turbulent diffusion and due to the flamelet progress are of the same order (obviously this is a lower estimation, since due to fluctuation of the flamelet angles, the averaged flamelet velocity in some direction less U_f.

So for times $\tau_G < t < \tau_*$ we have a flame with increasing brush width and practically constant turbulent combustion velocity. We call these combustion fronts Intermediate Steady Propagation (ISP) flames and believe that they should be recognized as a special class of flames. They correspond to a combustion regime which is intermediate between the initial stage at $t < \tau_G$, when we have intensive forming of wrinkles on the flamelet sheet and fast increasing of $U_t(t)$ and the final stage corresponding to $t >> \tau_*$, when we have stationary turbulent combustion fronts with $U_t^{st} = const.$ and $\delta_t^{st} = const.$

Simple estimations show that for real industrial combustors, as a rule, the time τ_* is larger that the residence time (that could be $\sim 10\tau_t$), i. e. in real combustors a flame reaches a wall and combustion is completed long before forming a flame with constant brush width. As τ_G is much less than the residence time in many cases we can assume that ISP regime takes place at $t < \tau_*$. We will see below that properties of ISP and stationary flames and their controlling mechanisms are quite different.

The 3-D ISP flame kinematics equation for $\rho = const$ (as the special case of the model equation for the partially premixed combustion) was proposed in [6]:

$$\partial(P_b)/(\partial t) + \nabla \cdot (\overline{\mathbf{u}} P_b) = \nabla \cdot (D_t \nabla P_b) + U_t |\nabla P_b|, \tag{1}$$

where D_t is the turbulent diffusion coefficient.

The statement of this section: Premixed combustion at intensive turbulence $u' >> U_f$ *and* $\tau_t (U_f/u')^2 < t < \tau_t (u'/U_f)^2$ *takes place in intermediate steady propagation (ISP) flames, i. e. in flames with* $U_t \approx const$ *(as approximate equilibrium between generation and dissipation of small-scales wrinkles of the flamelet sheet controlling its area is reached) and increasing by turbulent diffusion of the flame brush width (as at these times the regime is far from the equilibrium between formation of large-scale sheet wrinkles controlling this width and their consumption due to flamelet movement. Combustion models that based directly or indirectly on the only time* τ_t *in fact does not contain this regime.*

3. Mean reaction rates and TFC combustion model

The main attention in this section would be devoted to the physical equilibrium mechanisms at developed turbulence which enable us to introduce into the TFC model equation the coupling between turbulence and chemistry. We will analyze the integral combustion rate U_t dependence on controlling parameters and the local combustion rate $\overline{\rho W}$ distribution across the premixed flame. It would be shown that in flames with known integral characteristics U_t and δ_t the distribution of $\overline{\rho W}$ and counter-gradient transport phenomenon are closely connected.

The main physico-chemical mechanism controlling U_t :

The main controlling mechanism responsible for much weaker U_t dependence on chemistry in comparison with the laminar flame velocity U_l (more exactly with the actual flamelet velocity U_f) is the smoothing of wrinkled flamelet sheet due to moving of their elements with the velocity U_l. It means, for example, that a more fast chemistry increases U_l, but at the same time implies a decrease of the flamelet sheet area $\overline{(S/S_0)}$ due to consumption by more fast flamelet of additional small-scale wrinkles and makes it more smooth. We have what we call hydrodynamic "flamelet combustion self-compensation mechanism".

For stationary flames at $u' >> U_l$ in accordance with Damköhler[10], Shchelkin[11] ideas U_t^{st} does not depends on chemistry ($U_t^{st} \sim u'$), i. e. complete compensation: $\overline{(S/S_0)} \sim 1/U_l$. This situation is similar to turbulent dissipation at large Re numbers [3], when the effect of the kinematic viscosity increase is completely compensated by a decrease of instantaneous velocity gradients.

We will see below that for ISP flame, that takes place at $t < \tau_t (u'/Uf)^2$, takes place a partial combustion velocity compensation: U_t depends on chemistry but much weaker than U_l or U_f and this flame takes place at $t < \tau_t (u'/Uf)^2$. At the same time the most part of combustion model equations give at $t > \tau_t$ the stationary flame (see, for example, result of simulation in [12]) and this 1-D flame dependence on chemistry in fact corresponds to laminar flames. It is worth noting that the model that contains the result as an limiting case $U_t^{st} \sim u'$ was developed recently by Peters[13].

The parameters of thickened flamelet and its area, U_t dependence:

a. In accordance with [5] transfer process inside the thickened flamelet depend on vortices from equilibrium inertial interval and the value of the relevant transfer coefficient follows directly from dimensional analysis $\chi_f \approx \varepsilon^{1/3}\delta_f^{4/3}$, which is in fact the well-known Richardson law of the turbulent diffusion for scales inside the inertial interval (δ_f is the flamelet width). U_f and δ_f are function of χ_f and of the characteristic chemical time τ_{ch}, and by using dimensional analysis considerations (for laminar flames $U_l \approx (\chi/\tau_{ch})^{1/2}$, $\delta_l \approx (\chi\tau_{ch})^{1/2}$), we obtain:

$$U_f \approx (\chi_f/\tau_{ch})^{1/2} \approx u'(Da)^{-1/2}, \quad \delta_f \approx (\chi_f\tau_{ch})^{1/2} \approx L(Da)^{-3/2}, \chi_f \approx D_t(Da)^{-2} \quad (2)$$

This formulas can be derived straightforward using the inertial interval spectrum $E(k) = Ck^{-5/3}$[5]. The relationships (2) are equivalent to the fact that, in a coordinate system where the thickened flamelet is fixed, the heat fluxes in the front due to heat transfer and convection are of the same order of magnitude of the heat release due to chemical reactions [5]. Notice that chemical dependence of U_f in (2) is identical to that of the laminar flame.

b. For the estimation of $\overline{(S/S_0)} >> 1$ dimensional analysis is not sufficient and it is necessary to use also some general property of random surfaces $x = h(y, z, t)$. Our estimation is as follows:

$$\overline{(S/S_0)} = \overline{(1 + |grad\, h|^2)^{1/2}} \approx \overline{|grad\, h|} \approx \overline{(|grad\, h|^2)^{1/2}} \approx \int k^2 F(k)\, dk \approx \sigma/\lambda, \quad (3)$$

where $\sigma^2 = \overline{(x - \overline{x})^2} = \int F(k)\, dk = 2D_t\, t$ is the dispersion, λ is the micro-scale of the length of the random surface and $F(k)$ is the spectrum of the flamelet surface disturbances. We see, that σ^2 is defined by large scale and $\overline{(S/S_0)}$ by small scale disturbances of the flamelet surface. In ISP flames the spectrum $F(k,t))$ a small wave number k part is non-steady (increasing of large wrinkles by turbulence) and at the same time a large wave number part is steady (equilibrium between generation and consumption of small wrinkles).

The micro-scale λ, due to the assumed equilibrium, is a function of large scale turbulence characteristics L, u', flamelet parameters δ_f, U_f and time, t. Applying then the Π-theorem of a dimensional analysis yields $\lambda/\delta_f = f_1(u't/\delta_f, u'/U_f, L/\delta_f)$. Taking into account expressions (2) and using the condition of $\overline{(S/S_0)}$ stationary, we obtain that $\lambda/\delta_f = (u't/\delta_f)^{1/2} f_2(Da) \approx (u't/\delta_f)^{1/2} f_2(\infty) \approx (u't/\delta_f)^{1/2}$. Hence using (3), the expression for the averaged flamelet sheet area reads:

$$\overline{(S/S_0)} \approx (Da)^{3/4} \approx (u'/U_f)^{3/2} \approx (L/\delta_f)^{1/2} >> 1. \quad (4)$$

We see that assumption of physical equilibriums made possible to express parameters of U_f, δ_f and $\overline{(S/S_0)}$, controlled mainly by small-scale turbulence, in terms of large-scale turbulent characteristics, as shown by formulas (2) and (4).

c. Expressions (2) and (4) and well known formula for the chemical time $\tau_{ch} = \chi/U_l^2$ give finally the expression of the turbulent combustion velocity for the ISP flames:

$$U_t = U_f\overline{(S/S_0)} = Au'(Da)^{1/4} = Au'^{3/4}U_l^{1/2}\chi^{-1/4}L^{1/4}, \quad (5)$$

where $A \sim 1$ is an empirical parameter. It is worth emphasizing that all powers have been derived from the physical model and they don't contain any quantitative empirical information.

The chemistry dependence of U_t that is given by (5) ($U_t \sim \tau_{ch}^{-1/4}$) is much weaker than for laminar combustion ($U_l \sim \tau_{ch}^{-1/2}$). The reason is mentioned above partial flamelet combustion self-compensation mechanism: increasing of U_f decreasing in accordance with (4) $\overline{(S/S_0)}$ and vice-versa. Comparison of this prediction with empirical correlations for U_t and the range of applicability of (5) are presented in [8]. We have to mention that, for the hypothetical case of thickened but not wrinkled flame at $L << \delta_l$ proposed by Damköhler [10] the turbulent flame speed is $U_t \sim (D_t/\tau_{ch}$. This flame has laminar flame chemistry dependence, i. e. more strong than ISP flame. This regime is contained as a limiting case in the new Peters model [13].

It should be particularly emphasized that the thickened flamelet in our model have no quasi-laminar structure: in accordance with analysis of temperature pulsation balance presented in [5] the temperature pulsations inside the thickened flamelet are high, i. e. instantaneous reaction sheet strongly wrinkled inside the flamelet. It seems that this model closely correspond to the thin reaction zone regime[13]. In our analysis we ignore the temperature pulsation inside a flamelet. From a methodological point of view it is similar to ignore the dissipation rate pulsations in the Kolmogorov theory of the fine-scale turbulence[3].

Turbulent flame closure (TFC) combustion model:

TFC combustion model is based on the kinematics equation (1) modified of $\rho \neq const$[7]. This equation is as follows (the conventional and Favre averaging are symbolized $\overline{a}$ and $\tilde{a} = \overline{\rho a}/\overline{\rho}$,):

$$\partial(\overline{\rho}\tilde{c})/(\partial t) + \nabla \cdot (\overline{\rho}\tilde{\mathbf{u}}\tilde{c}) = \nabla \cdot (\overline{\rho} D_t \nabla \tilde{c}) + (\rho_u U_t)|\nabla \tilde{c}|. \tag{6}$$

Eq. (6) describes only ISP flames and does not contain limiting case of 1-D stationary flames, it strongly simplifies the combustion model and the same time does not impose a limitation on its practical applications for simulation combustion at strong turbulence.

In TFC model the theoretical expression (5) for U_t as a function of the physico-chemical properties of the combustible mixture and turbulent parameters is substituted in the Eq. (6). In other words we introduce directly in the model equation the properties of the ISP flames (their combustion velocity and width dependences) that why we name it the turbulent flame closure (TFC) equation. TFC model equation simulated together with fluid dynamics Reynolds equations and "$k - \varepsilon$" turbulence (at simulations it was assumed $\chi = \chi_u$ and local u' and L expressed in terms of k and ε). This set of equations simulated only large-scale processes but in accordance with the foregoing it describes the small-scale coupling between turbulence, molecular transport and chemistry. It is interaction between turbulence and molecular viscosity (dissipation rate), between turbulence and instantaneous reaction zone (flamelet parameters), between turbulence and flamelet sheet (its area). To describe experimental bending for U_t at regimes close to a blow-out boundary (Lipatnikov proposal) Bray model[14] was used of the stretch effect in terms of flamelet critical velocity gradient based in fact on the assumption of universal equilibrium PDF for instantaneous dissipation (i. e. for instantaneous characteristic velocity gradients in small scales controlling flamelet extinction). It was in fact the forth equilibrium mechanism used in our combustion simulations.

Good agreement with a great body of Karpov experimental data in spherical bombs with artificial turbulence for different fuels, air excess coefficients and turbulence[15] (i. e. at large variation of kinematics, molecular transport and turbulent properties of mixtures) is good indirect evidence that their coupling using idea of physical equilibrium is fruitful. The comparison with Moreau data on high velocity combustion in a channel[16] confirm existence of ISP flames with increasing brush width.

$\overline{\rho W}$ and the counter-gradient transport in the TFC model:

In the equation (6) the transport and the source terms are not real transport and source terms of the unclosed equation, i. e. $\nabla \cdot (\overline{\rho} D_t \nabla \tilde{c}) \neq -\nabla \cdot (\overline{\rho \mathbf{u}" c"})$, and $(\rho_u U_t)|\nabla \tilde{c}| \neq \overline{\rho W}$. In TFC model equation transport term has gradient nature, while the transport term $\nabla \cdot (\overline{\rho \mathbf{u}" c"})$ has in many turbulent premixed flames counter-gradient nature. It means that in the model equation transport term we include only the gradient physical diffusion part (to describe flames with increasing flame brush width) whereas the counter-gradient part was included in the model source term $(\rho_u U_t)|\nabla \tilde{c}|$. So though TFC model equation describe physical distributions of $\tilde{c}$ and connected with it $\overline{\rho}$, $\overline{T}$ and concentration of species, for extraction of the physical source term $\overline{\rho W} = \rho_u U_f \overline{\Sigma}$ from the model source term it is necessary, as it would seen below, to have additionally some hydrodynamic model for the progress variable transport term.

4. Kinematics equation. The counter-gradient transport phenomenon.

The modeling kinematics equation for the Favre averaged progress variable in the case of flames in the ISP regime is given by:

$$\overline{\rho}\,\frac{\partial \tilde{c}}{\partial t} + \overline{\rho}\,\tilde{u}\,\frac{\partial \tilde{c}}{\partial x} = \frac{\partial}{\partial x}\left(\overline{\rho}\,D_t\,\frac{\partial \tilde{c}}{\partial x}\right) + \rho_u\,U_t\left|\frac{\partial \tilde{c}}{\partial x}\right|, \tag{7}$$

where $\tilde{c}$ is the averaged progress variable ($\tilde{c} = 0$ and $\tilde{c} = 1$ correspond respectively to the cold reactants and hot products), U_t is the turbulent combustion velocity, D_t is the physical turbulent diffusion coefficient. The model source term in this equation determines propagation of the flame with velocity U_t with respect to the unburnt fluid mixture while the gradient diffusion term determines the thickening of the turbulent flame brush according to the turbulent dispersion law.

This equation models the exact but unclosed equation:

$$\overline{\rho}\,\frac{\partial \tilde{c}}{\partial t} + \overline{\rho}\,\tilde{u}\,\frac{\partial \tilde{c}}{\partial x} = \frac{\partial(-\overline{\rho}\,\widetilde{u'' c''})}{\partial x} + \overline{\rho}\widetilde{W}, \tag{8}$$

where $-\overline{\rho}\,\widetilde{u'' c''}$ is the progress variable flux that as a rule is generally attributed a counter-gradient nature and $\overline{\rho}\,\widetilde{W}$ is the averaged rate of products formation. As we have mentioned in the introduction there is no contradiction between the gradient transport term in the model equation (7) and the as a rule counter-gradient transport term in the exact unclosed equation (8). This because the transport term in the model equation (7) contains only the turbulent diffusion component of the total transport term $-\overline{\rho}\,\widetilde{u'' c''}$, while the second gas-dynamics convective component connected with the differential acceleration of hot and cold volumes in a nonuniform pressure field (the pressure-driven transport) is integrated together with the actual source term $\overline{\rho}\widetilde{W}$ in the model source term $\rho_u U_t|\partial \tilde{c}/\partial x|$ in equation (7). Obviously

$$\rho_u \int_{-\infty}^{+\infty} U_t\,\left|\frac{\partial \tilde{c}}{\partial x} i\right|\,dx = \int_{-\infty}^{+\infty} \overline{\rho}\widetilde{W}\,dx, \tag{9}$$

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

This interpretation of the model equation (7) was formulated by Zimont in [16] answering to a question from F. Williams about the possibility to predict counter-gradient transport within the framework of this model. The methodology for the analysis of the counter-gradient transport phenomenon based on this interpretation was subsequently developed in [7, 5] and now in this paper in more advanced form. It should also be remarked that Lipatnikov was the one who first realized in [23] that the interpretation of the turbulent diffusion term in the model equation (7) used as approximation of the transport term in unclosed equation (8) and the model source term as approximation of real source $\rho\widetilde{W}$ is non correct. A "joint closure" took place in fact while developing the model given by equation (7), such that the sum of the total transport term and the source term in (8) is instead approximated by the sum of turbulent diffusion and model source term in the model equation (7). At the same time Lipatnikov idea presented in [17] about the possibility to model counter-gradient transport is quite different from the one developed here.

It is significant to remember that equation (7) (and the following equations (13) and (17)) 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 $t << \tau_t(u'/s_L)^2$ when turbulent transport by pulsation velocities u' prevails over the transport connected with flamelets local propagation with velocity s_L. In the case of turbulent flames composed by thickened flamelets at $Re_t >> 1$ and $Da >> 1$ with flamelet velocity $U_f > s_L$ this regime takes place for $t < \tau_t\, Da$ [7].

For larger times $t > \tau_t(u'/s_L)^2$ (or $t > \tau_t Da$) turbulent premixed flames propagate according to the 1-D stationary propagating combustion front and instead of the model equation we can write exact kinematics equation of the running wave (in coordinate system where the wave is motionless)

$$\overline{\rho}\, \tilde{u}\, \frac{\partial \tilde{c}}{\partial x} = \rho_u\, U_t^{st} \left| \frac{\partial \tilde{c}}{\partial x} \right|, \tag{10}$$

where in accordance with the ideas of Damköhler [18] and Shchelkin [19], in the case of strong turbulence we have $U_t^{st} \approx u'$, i.e. in the case of $u' >> s_L$ (or $u' >> u_F$) the turbulent combustion velocity does not depends on chemistry and molecular properties of the fuel/air mixture.

Comparison of equations (8) and (10) yields

$$\rho_u\, U_t^{st} \left| \frac{\partial \tilde{c}}{\partial x} \right| = \overline{\rho}\widetilde{W} - \frac{\partial(\overline{\rho}\, \widetilde{u"c"})}{\partial x} \tag{11}$$

demonstrating that the transport and source terms in the exact kinematic equation are clubbed under a single term given in our model equation (7) by $\rho_u\, U_t\, |d\tilde{c}/dx|$, i.e. they are intimately coupled for the 1-D stationary flame.

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:

$$-\overline{\rho}\, \widetilde{u'' c''} = -\overline{\rho}\, \tilde{c}\, (1 - \tilde{c})\, (\overline{u}_b - \overline{u}_u), \tag{12}$$

where $\overline{u}_u$ and $\overline{u}_b$ are conditioned averaged velocities of unburned and burned volumes. Simple estimation shows that for $\Theta >> 1$ (strong heat release) and $\delta_t^{st} >> L$, the ratio of the turbulent diffusion flux to the pressure-driven flux is $\sim (u'L/\delta_t^{st})/(U_t^{st}\Theta) << 1$, i. e. the counter-gradient transport component and the actual source term are clubbed under the single term $\rho_u\, U_t\, \partial\tilde{c}/\partial x$. In the case of the ISP combustion regime where flames have increasing flame brush width and $\delta_t < \delta_t^{st}$ we have extracted the turbulent diffusion component (controlling in our model the brush width) into a separate term and left the remaining gas-dynamics transport and the chemical source clubbed into the model source term.

According to these ideas which were originally proposed in [5, 7]), the progress variable flux can be split into two contributions: one of gradient nature generated by real turbulent diffusion transport which has the property to increase the flame brush thickness (according to the turbulent dispersion law) and the second of counter-gradient nature which is of convective type and is generated by the pressure drop across the turbulent flame brush. The first component of the transport is controlled by positive turbulent diffusion coefficient that can be estimated using usual turbulence models and the second component can be estimated from some gas-dynamics model that is strongly connected with the nonuniform averaged pressure distribution (in our case of open flames, with the averaged pressure drop across the flame and resulting in the counter-gradient gas-dynamical component).

The possibility of such decomposition can be better understood observing the exact expression for the progress variable flux (12). This expression shows that the turbulent scalar flux $-\overline{\rho}\,\widetilde{u''\,c''}$ has gradient (counter-gradient) nature when $u_b < u_u$ ($u_b > u_u$). In absence of a negative pressure gradient which can accelerate the products more than the reactants, the effect of turbulent dispersion will be to generate penetration of burnt mixture inside the unburnt one at the cold boundary and penetration of unburnt mixture inside the burnt one at the hot boundary. According to this description the conditional velocities will be such that $u_b < u_u$. In case presence of a pressure drop across the turbulent flame brush, the light burnt gases are accelerated more than the unburnt ones. This may reduce the gradient transport or even transform it into the counter-gradient phenomenon when $u_b > u_u$.

The connection of the counter-gradient transport with the averaged pressure gradient is well known. In recent DNS work, it has been shown a well-defined effect of pressure pulsations

(BRAY cite?).

This effect is connected with the instantaneous difference of conditional averaged pressures of unburned $\overline{p}_u$ and burned gases $\overline{p}_b$ as the boundary between these gases is a moving flamelets that results $\overline{p}_u > \overline{p}_b$. Therefore, for example, even at $\overline{p} = const$ there is some counter-gradient component. In our simulations below we ignore this effect.

It must be observed that, because of the convective nature of the pressure-driven transport, while developing the TFC combustion model we have assumed that it doesn't have the capability to change the thickness of the flame boundary layer, a property which is typical of turbulent diffusion phenomena.

Subtracting now equation (7) from (8) we obtain the following relation:

$$\rho_u\, U_t \left| \frac{\partial \tilde{c}}{\partial x} \right| = \overline{\rho}\, \widetilde{W} + \frac{\partial}{\partial x} \left[-\overline{\rho}\, \widetilde{u''\,c''} - \overline{\rho}\, D_t\, \frac{\partial \tilde{c}}{\partial x} \right] \tag{13}$$

which formally shows that the model source term in (7) accounts at the same time for real heat release and the pressure-driven (counter-gradient in our case of open premixed flames) transport component. In order to extract the counter-gradient transport term from (13) two approaches may be used: a) give a model for the average heat release $\overline{\rho}\widetilde{W}$, b) model directly the counter-gradient transport component (in this case relation (13) will give an expression for $\overline{\rho}\,\widetilde{W}$).

In [7] approach a) has been followed. It was assumed that the actual averaged chemical source term in equation (8) is proportional to the probability to find the flamelet at a given position $p_f(x)$; this probability is related to the probability of finding products $P_b(x)$ at the given position by the relation:

$$P_b(x) = \int_{-\infty}^{x} p_f(\xi)\, d\xi \Rightarrow p_{fl}(x) = \frac{\partial P_b(x)}{\partial x} \tag{14}$$

Note also the following expression for the averaged progress variable (as flamelets are thin, the probability to have $0 < c < 1$ has been neglected):

$$\overline{c} = P_b\, 1 + (1 - P_b)\, 0 = P_b \tag{15}$$

Therefore we can write:

$$\overline{\rho}\widetilde{W} = const \left| \frac{\partial \overline{c}}{\partial x} \right| \tag{16}$$

where the constant is equal to $\rho_u\, U_t$ as can be shown by integrating equation (13) from $-\infty$ to $+\infty$.

Using (16) in (13) the following expression for the second order velocity-progress variable correlation is obtained (where it has assumed $d\overline{c}/dx > 0$ without loss of generality):

$$\frac{\partial(-\overline{\rho}\,\widetilde{u''\,c''})}{\partial x} = \rho_u\, U_t\, \frac{\partial}{\partial x}\,(\tilde{c} - \overline{c}) + \frac{\partial}{\partial x}\left[\overline{\rho}\, D_t\, \frac{\partial \tilde{c}}{\partial x}\right] \tag{17}$$

which integrated from $-\infty$ to x yields:

$$-\overline{\rho}\,\widetilde{u''\,c''} = \rho_u\, U_t\,(\tilde{c} - \overline{c}) + \overline{\rho}\, D_t \frac{\partial \tilde{c}}{\partial x} \tag{18}$$

This relation explicitly gives evidence that the second order Favre correlation between the progress variable and velocity fluctuations is composed of two contributions:

a) real turbulent transport (modeled here with an eddy diffusivity assumption) which is responsible for the thickening of the ISP flame brush;

b) a contribution which is proportional to the integral of the difference between the model $\rho_u\, U_t\, d\tilde{c}/dx$ and the real chemical source term $\overline{\rho}\,\widetilde{W} = \rho_u\, U_t\, d\overline{c}/dx$. This contribution can be expressed with simple algebraic manipulations as:

$$\rho_u\, U_t\,(\tilde{c} - \overline{c}) = -\rho_u\, U_t\, \tilde{c}\,(1 - \tilde{c})\, \frac{\Theta - 1}{1 + \tilde{c}\,(\Theta - 1)} \tag{19}$$

It should be again emphasized that the point of view adopted here is that the turbulent flame brush increases (here this is assumed to be an effect of turbulent dispersion), independently on the nature of the progress variable flux (of gradient or counter-gradient type). Nevertheless it sometimes stated that in case of a total scalar flux of counter-gradient type the flame brush becomes thinner whereas in case of gradient type it becomes thicker [11].

We think that this point of view is obtained as result of interpreting the progress variable flux in turbulent premixed flames only as a turbulent diffusion flux of passive concentration. The term "countegradient turbulent diffusion" is in fact often associated with the phenomenon of counter-gradient transport which, we stress here, is not connected with a negative value for the turbulent diffusion coefficient.

We believe that in open flames a decreasing flame brush width is highly improbable. Numerous experiments show in fact that the flame brush width increases with distance along the flame similarly to a diffusing wake despite of the commonly observed counter-gradient nature of the transport in flames and in principle this thickness increases until a "statistically steady" finite value which is unattainable in practical combustion devices. At the same time, in presence of strong external pressure gradient (combustion in a nozzle, for example) the qualitative picture could be more complicated, but we do not analyze this case here.

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

$$-\overline{\rho}\,\tilde{c}\,(1-\tilde{c})\,(\overline{u}_b-\overline{u}_u) = \rho_u\,U_t\,(\tilde{c}-\overline{c})+\overline{\rho}\,D_t\frac{\partial\tilde{c}}{\partial x} \tag{20}$$

$$(1-\tilde{c})\,\overline{u}_u+\tilde{c}\,\overline{u}_b = [1+(\Theta-1)\,\tilde{c}]\,U_t \tag{21}$$

we obtain the following expression for the conditional velocities:

$$\overline{u}_u = \frac{D_t}{1-\tilde{c}}\frac{d\tilde{c}}{dn}+U_t \tag{22}$$

$$\overline{u}_b = -\frac{D_t}{\tilde{c}}\frac{d\tilde{c}}{dn}+\Theta\,U_t \tag{23}$$

These relations show that each of the conditional velocities is composed by two contributions: one related to turbulent dispersion and the other to convection.

In the case of a steady flame ($t \to \infty$, constant thickness and velocity) the flame is described by the equation (10), i.e. in 22 and 23 we must put $D_t = 0$, resulting in the two conditional velocities being constant across the flame brush, $\overline{u}_u = U_t^{st} = u(-\infty)$ and $\overline{u}_b = \Theta U_t^{st} = u(+\infty)$. This also implies constant conditional pressures ($\overline{p}_u = p(-\infty)$, $\overline{p}_b = p(+\infty)$) as it will be shown in the next paragraph. Such value of velocities correspond in fact 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 possible upper boundary of this phenomenon. Such model for estimation of the counter-gradient phenomenon based on the expression (16) for actual combustion rates and further using of the equation (13) for estimation of the transport $\overline{\rho}\,\widetilde{u''\,c''}$ was developed in [7] where it was used for estimation of the counter-gradient phenomenon in well known standard Moreau high velocity premixed combustion experimental data in a channel. We will call below this model "a rough model" or "upper estimation model".

More realistic modeling must take into account this interaction, i. e. the acceleration of relatively slow cold volumes by more fast hot volumes and vice versa. Such gas-dynamics model that estimates directly the pressure-driven counter-gradient component (described in (13) by an expression between the brackets is developed in the next section. For estimation of actual combustion rate it is necessary to use the equation (13). We will call this model for estimation of the counter-gradient phenomenon and actual combustion rates " an accurate model" or "a gas-dynamics model".

5. A gas-dynamics model of the counter-gradient transport phenomenon

In this section we analyze 1-D stationary premixed flames. These results then will be applied to open flames with increasing flame brush width. The basis of it is an assumption that at real relatively slow increasing of the brush width the pressure gradient across open flames bringing about the counter-gradient phenomenon can be assumed the same as in 1-D stationary flames. Good accord between results of numerical simulations of this phenomenon and experimental experimental data demonstrated below confirm the truth of this hypothesis at least as a first approximation.

According to the idea presented in section 4, the progress variable transport in open turbulent premixed flames is composed by two fundamental contributions: real gradient turbulent diffusion which is responsible for the growth of flame brush thickness and counter-gradient transport related to the pressure drop across the flame brush due to heat release. In this section we will present a gasdymamics model for the pressure driven component of the progress variable transport in the case of 1-D stationary flames which will be applied to real open flame with increasing flame brush width.

The basis for this analysis is represented by equations (10)-(12). For estimating the pressure-driven component we assume here that the conditional velocities in (12) are controlled only by gas-dynamics, i. e. we ignore the effect of turbulent dispersion. In this case the term $-\overline{\rho}\,\widetilde{u''\,c''}$ includes only the counter-gradient transport component, and in fact the model source (11) of the equation (10) term contains the actual source term and the pressure-driven counter-gradient component. The results obtained under these conditions can therefore be extended to ISP flames by using equation (7). In this equation in fact the gradient transport term is controlled by turbulent diffusion and the model source term contains the actual source term and the pressure-driven counter-gradient component.

Consider therefore the conservation equations for mass, momentum and reactants total pressure (iso-entropic condition for reactants). The equations have been written here for $U_t^{st} = 1\,m/s$ and $\rho_u = 1\,kg/m^3$ (or equivalently in normalized form):

$$(1-\overline{c})\,\overline{u}_u + \overline{c}\,\frac{\overline{u}_b}{\Theta} = 1 \tag{24}$$

$$\overline{p} + (1-\overline{c})\,[\overline{u}_u^2 + \overline{u_u'^2}] + \frac{1}{\Theta}\,\overline{c}[\overline{u}_b^2 + \overline{u_b'^2}] = p_{-\infty} + 1 + \overline{u'^2}_{-\infty} \tag{25}$$

$$\overline{p}_u + \frac{1}{2}\,[\overline{u}_u^2 + \overline{u_u'^2}] = p_{-\infty} + \frac{1}{2}(1 + \overline{u'^2}_{-\infty}) \tag{26}$$

where the average pressure $\overline{p}$ is given by $\overline{p} = (1-\overline{c})\,\overline{p}_u + \overline{c}\,\overline{p}_b$. In the case introduced in the previous paragraph we have seen that $\overline{\rho}\widetilde{W} = \rho_u\,U_t\,dP_b/dn$ implies uniform conditional velocities across the turbulent flame brush and equal to $\overline{u}_u = 1$ and $\overline{u}_b = \Theta$ in the present case. In this case equation (24) becomes redundant and the system therefore reduces to two equation in the two conditional pressures as unknowns.

If we subtract equation (26) from equation (25) and require the fluctuating terms to cancel, we end up with the following two relations:

$$\overline{c}\,(\overline{p}_b - \overline{p}_u) + \left(\frac{1}{2} - \overline{c}\right)\,\overline{u}_u^2 + \frac{1}{\Theta}\,\overline{c}\,\overline{u_b}^2 = \frac{1}{2} \tag{27}$$

$$\left(\frac{1}{2}-\overline{c}\right)\overline{u'_u}^2+\frac{1}{\Theta}\,\overline{c}\,\overline{u'_b}^2=\frac{1}{2}\,\overline{u'^2_u}_{-\infty} \tag{28}$$

The second of these relations gives a turbulent velocity fluctuation at the hot boundary which is equal to $\Theta/2\,\overline{u'^2_u}_{-\infty}$. This result is consistent with several experimental works where the turbulent velocity fluctuation has been found increasing across the turbulent flame brush. For example, Moreau [26] has found in oblique high velocity planar methane/air flames at equivalence ratio 0.8 that the axial turbulent fluctuating velocity increases inside the flame, reaching a value about three times that measured in the cold flow.

In the particular case of $\overline{u}_u=1$ and $\overline{u}_b=\Theta$ analyzed in the previous section we have $\overline{p}_u-\overline{p}_b=(\Theta-1)$. Obviously this is an hypothetical unrealistic case. We will instead analyze here the case with strong interraction between hot and cold volumes with an assumption $\overline{p_u}=\overline{p_b}=\overline{p}$, i. e. with ignoring the small difference in the presuure before and behind flamelet sheet that is moving boundary between cold and hot volumes as we already mentioned above.

Under this assumption we have from equation (27):

$$(\frac{1}{2}-\overline{c})\,\overline{u}_u^2+\frac{1}{\Theta}\,\overline{c}\,\overline{u}_b^2=\frac{1}{2} \tag{29}$$

The system given by (29) and (24) can be solved with simple algebraic manipulations. This yields the following expressions for $\overline{u}_u$ and $\overline{u}_b$:

$$\overline{u}_b=\frac{-\beta+\sqrt{-4\,\alpha\,\gamma+\beta^2}}{2\,\alpha},\qquad \overline{u}_u=\frac{1-\overline{u}_b\,\overline{c}/\Theta}{1-\overline{c}} \tag{30}$$

$$\alpha=\frac{0.5-\overline{c}}{(1-\overline{c})^2}\,\frac{\overline{c}^2}{\Theta^2}+\frac{\overline{c}}{\Theta},\qquad \beta=-2\,\frac{\overline{c}}{\Theta}\,\frac{0.5-\overline{c}}{(1-\overline{c})^2},\qquad \gamma=\frac{0.5-\overline{c}}{(1-\overline{c})^2}-0.5 \tag{31}$$

The expression obtained now for the conditional velocities gives the opportunity to calculate the average heat release within the turbulent flame brush using equation (13). If we assume $\partial\overline{c}/\partial x$ to be distributed as a Gaussian function (validation of this hypothesis has been performed in [8] containing very careful measurement of the temperature profiles in premixed flames and also in [32] containing analysis some literature experimental data that demonstrate the complementary error function for the profiles):

$$\frac{\partial\overline{c}}{\partial x}=\frac{1}{\sqrt{2\,\pi\,\sigma^2}}\,\mathrm{e}^{-(x-a(t))^2/2\,\sigma^2} \tag{32}$$

introducing the non-dimensional spatial variable $\xi=(x-a(t))/\sqrt{2\,\sigma^2}$, the average heat release nondimensionalised with $\rho_u\,U_t/\sqrt{2\,\sigma^2}$ is given by:

$$\frac{\overline{\rho}\widetilde{W}}{\rho_u\,U_t/\sqrt{2\,\sigma^2}}=\frac{1}{\sqrt{\pi}}\,\mathrm{e}^{-\xi^2}\,\frac{\partial}{\partial\overline{c}}\left[\tilde{c}-\overline{c}\,(1-\tilde{c})\frac{\overline{u}_b-\overline{u}_u}{U_t}\right] \tag{33}$$

In the hypothetical case of conditional velocities constant across the flame brush, this expression yields the result already introduced at paragraph, i.e. $\overline{\rho}\widetilde{W}=\rho_u\,U_t\,|\partial\overline{c}/\partial x|$. Figure 1 shows the distribution of the conditional velocities non-dimensionalised with U_t and non-dimensional heat release, $\partial\overline{c}/\partial\xi$ and $\partial\tilde{c}/\partial\xi$. The heat release is shifted toward the front part of flame brush with respect to the model source term, the maximum shift obtained for the model based on uniform distribution of conditional pressures and velocities across the turbulent flame brush (when $\overline{u}_b-\overline{u}_u$ attains its maximum value). In the case of more realistic gas-dynamics model taking into account force interaction between high velocity hot and low velocity cold volumes that we analyzed assuming $\overline{p}_u = \overline{p}_b$ the heat release is less shifted toward the front part of the flame brush.

It is interesting to note that such situation has been observed experimentally by [10] and [15] who analyzed a large dataset of experimental data on turbulent premixed flame of various types (v-, conical, stagnation and swirl stabilized flames). The experiments from [10, 15] will be considered in paragraph 7 for a qualitative validation of the result obtained with the present model.

6. Modeling of the turbulent flame speed

The turbulent flame speed U_t of a 1-D flame is given by the product between the local propagation velocity U_f and the surface area for unit of flame cross sectional area $(\overline{S}/S_0)$ of the thin flamelet, i. e. $U_t = U_f(\overline{S}/S_0)$. These two quantities have been estimated by asymptotic analysis at large turbulent Reynolds Re_t and Da numbers based on the Kolmogorov methodology: the equilibrium fine-scale turbulence from the inertial interval controls the thickened flamelet combustion velocity giving $U_f > s_L$ and flamelet width $\delta_f > \delta_L$, the equilibrium small scale flamelet wrinkles control the practically constant flame surface area and finally non-equilibrium large-scale wrinkles control the flame brush width which increases similarly to a non-reactive mixing layer [4, 7].

In this model the micro-turbulent transport of heat or concentrations $(\chi_f \simeq D_f)$ within the flamelets thickness is described by the well-known Richardson diffusion law $\chi_f \simeq \varepsilon^{1/3}\delta_f^{4/3}$ where ε is the averaged rate of turbulent kinetic energy dissipation. The following expressions have been obtained:

$$\begin{aligned} &U_f \approx u'\, Da^{-1/2},\ \delta_f \approx L Da^{-3/2},\ \chi_f \approx D_t\, Da^{-2} \Rightarrow \\ &(\overline{S}/S_0) \approx Da^{3/4} \approx (u'/U_f)^{3/2} \approx (L/\delta_f)^{1/2}. \end{aligned} \tag{34}$$

According to the first of these relations the local propagation velocity of the thickened flamelet increases with decreasing Damköhler number (for example, reducing of the chemistry rate). This can be explained considering that a reduction in Damköhler number produces an increase in the flamelet thickness (the second relation). It results therefore an increase in the micro-turbulent transport coefficient (the third relation) because of small vortices in the inertial range falling inside the flamelet which being dominant over the reduction in chemistry rate produces an overall increase in the local propagation velocity. The fourth expression shows that there is connection between the flamelet area and the flamelet velocity: an increase in the flamelet velocity (or increasing of the flamelet width) produce a decrease in the flamelet sheet area. The physical reason of this effect is self-smoothing of the wrinkled sheet but its movement (or due to increasing of the flamelet width): an increase in U_f (or decrease in δ_f) "consumes" the smallest existing wrinkles and it decreases the area. This self-compensation mechanism is responsible for the relatively weak chemistry dependence of U_t shown by experiments.

The final expression for the turbulent combustion velocity is as follows:

$$U_t = U_f(\overline{S}/S_0) = A\, u'\, Da^{1/4} = A\, u'^{3/4}\, s_L^{1/2}\, \chi^{-1/4}\, L^{1/4}, \tag{35}$$

where $A \sim 1$ is an empirical coefficient, χ is the molecular heat transfer coefficient that (referred to the unburned mixture in the simulations, i. e. $\chi = \chi_u$). It is worth emphasizing that all 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 (35) we can see that the f the turbulent flame speed depends from chemistry according to $U_t \sim \tau_{ch}^{-1/4}$ which is much weaker than the case of laminar combustion where $s_L \sim \tau_{ch}^{-1/2}$. We expect that in the case of laminar flamelets the chemistry dependence for U_t would be also weaker due to self-compensation mechanism mentioned above but for laminar flamelets similar formulas do no exist. It seems obvious that this self-compensation mechanism is responsible for many quantitative peculiarities of turbulent combustion occurring according to the flamelet mechanism and in particular the relatively weak dependence of U_t on chemistry.

This is direct experimental conformation of existing of thickened flamelets, direct measurements in [33] demonstrated $\delta_f/\delta_L = 2 - 5$ with the turbulence dependence $\delta_f(u')$ close to the presented above formula. But situation is not simple here as we have two tendency here: broadening of flamelets by fine-scale turbulence and narrowing of flamelets by stretch. So quite probable situation when flamelets with fine-scales eddies into it has width more than normal laminar flame. It means that we must strictly speaking compare thickened flamelets width not with normal laminar flame but with laminar flame with similar stretch. My be this is a reason why in experiments with developed fine-scale turbulence flamelet thickness was less in comparison with normal laminar flame, see for example **this paper of German authors are also in 27 Symposium**. In our formulas (28)flamelet stretch was not taken into account and probably in experiments in [33] this effect was not significant. In our simulation we took into account the stretch effect only for correction directly expression for U_t (35) but separately expressions for U_f, δ_f and $\overline{S}/S_0$, see below.

Expression (35) is an accordance with many old Russian experimental data (see [4, 5]). Bradley [20] have correlated recently many West experimental results for premixed turbulent flames in order to obtain an expression for the turbulent flame speed as function of the relevant physico-chemical parameters. This expression, assuming that all molecular transport coefficients are equal, is given by:

$$U_t \simeq u' \, Da^{0.3} \, Re_t^{-0.15} \tag{36}$$

This empirical expression is close to the theoretical one (35). The weaker dependence of U_t on u' was connected in [23] with the effect of the flamelet stretch which is present in (36) and not in (35). When the strain rate becomes large local flamelet quenching may occur. This is not taken in account in the theoretical expression (35) which is therefore valid if the local strain rate is sufficiently small. x The effect of large strain rates x is to reduce the local flamelets velocities and even cause their extinction. This effect has been incorporated in the expression for U_t in [29] using a model developed by Bray [22] for the stretch-effect. According to this model, a stretch factor G which represent the probability for the instantaneous turbulent kinetic energy dissipation rate ϵ to be less than the critical extinction value ϵ_{cr} has been introduced. Assuming a log-normal distribution for ϵ [22] the following expression for the stretching factor is obtained:

$$G = 0.5 \, erfc \left[-\frac{1}{\sqrt{2}\,\sigma} \left(\ln(\epsilon_{cr}/\epsilon) + \frac{\sigma}{2} \right) \right] \tag{37}$$

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

$$U_t \simeq G \, u' \, Da^{1/4} \tag{38}$$

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

Validation of the combustion model based on the expression (38) for the turbulent flame speed and for the bending effect can be found in [23, 27]. Application of the model to industrial premixed combustion can be found in [9] and validation for the case of a high speed 2D turbulent premixed flame (experiments from [26]) in [7].

7. Validation

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

The first of these consists in the experiments performed by Moss [1] for an open LPG/air turbulent premixed flame at stoichiometric conditions. These data correspond therefore to a situation with the largest possible laminar flame speed, such that the effect of gradient diffusion is likely to be small. This is favorable for validating the model developed here for the counter-gradient component of the total scalar 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 a heat release distribution across the flame brush which is consistent with available experimental observations. The experimental data considered for this validation are from Cheng and Sheperd [10] and Cheng [15] who have analyzed turbulent premixed flames in various configurations.

The third set of data consists in the experiments recently performed by Frank *et al* [3] 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 $\overline{u}_u$ and $\overline{u}_b$ 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 of u'/s_L. These data are therefore of particular interest for the overall validation of the model for the total scalar flux developed here.

It should be stressed that developed in the section the counter-gradient transport gas-dynamics model refers to one-dimensional stationary flames so its application for estimation of the counter-gradient component of the transport to real flames with increasing brush width is an approximation. For open flames we estimate it as a good approximation as flames have a simple structure and in most part of real flames the counter-gradient component dominate in transport so controlling flame brush width turbulent diffusion transport in many case is relatively small.

But for at real industrial burner where flames have no simple structure and the pressure gradient are connected not only with heat release but also with complicated hydrodynamics such approach is not applicable. The author develops now more general invariant gas-dynamics model base on similar ideas that could be applied to real 3-D combustors. This model after validation will be published elsewhere.

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

The schematics of the experiments for this open turbulent premixed flame is shown in figure 2 a). The burner is composed by a pipe, 5 cm in diameter and 60 cm in length.

The pipe is fed with air and LPG (approximately 70 percent propane and 30 percent propene) which becomes fully premixed at the pipe exit. The velocity at the burner exit is $\overline{u} \simeq 4\,m/s$, approximately ten times the laminar flame speed. The velocity rms $\sqrt{\overline{u'^2}}$ at the center of the pipe exit is $0.5\,m/s$.

Measurements have been taken across the flame brush along a line inclined 23^o with the pipe axis. The angle observed between the flame and the burner axis was approximately $\theta - \phi = 16^o$. Concentration (the progress variable) was measured by light scatter technique and velocities by laser Doppler velocimetry. Moss experimental data have been subsequently post-processed in [12]; the data reported in this last reference have been adopted here for validation.

A full CF-D analysis of this case hasn't been performed. Using the available information, we have estimated that the order of magnitude of the turbulent diffusion component with the total scalar 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 $U_t \sim \overline{U}\,\sin(\theta - \phi) = 4\,cos(16) = 1.1\,m/s$. Assuming the integral turbulent length scale at the pipe exit to be in the range of $l_t \simeq 1.5\,mm$ we have $\epsilon \simeq 0.37\,\overline{u'^3}/l_t = 31\,m^2/s^3$ and the turbulent diffusion coefficient $D_t \simeq 0.09\,(3/2\,u'^2)^2/\epsilon = 0.0004\,m^2/s$. The flame brush increases in thickness according to the turbulent dispersion law, i.e. $\delta_f \simeq \sqrt{D_t\,H/\overline{U}}$ where H is the height from the pipe exit at which measurements have been taken, i.e. $H \simeq 0.28\,m$ (assuming the flame to be flat). We have therefore $\delta_f \simeq \sqrt{0.0004\,0.28/4} = 0.0053\,m$ and assuming the maximum progress variable gradient to be approximately $1/\delta_f = 1.0/0.0053\,m^{-1}$ at $\tilde{c} = 0.5$ we have a contribution to $\overline{u}_u$ from turbulent dispersion roughly equal to $0.0008/0.0053 = 0.15\,m/s$. The experimental values of $\overline{u}_b/U_t$ and $\overline{u}_u/U_t$ at this location are respectively equal to 6 and 3, i.e. the contribution to the conditional velocities by real turbulent transport is small compared to the contribution by counter-gradient transport.

We compare therefore here only the contribution to the transport generated by the pressure drop across the turbulent flame brush.

Figure 3 shows the non-dimensional turbulent scalar flux $\overline{\rho}\,\widetilde{u''c''}/\rho_u\,U_t$ and the non-dimensional conditional velocities $\overline{u}_u/U_t$ and $\overline{u}_b/U_t$ calculated using the two models presented in this work: the rough (upper estimation model) in fact corresponding to constant conditional velocities $\overline{u}_u$ and $\overline{u}_b$ across the flame and accurate (gas-dynamics) model that takes into account force interaction between cold and hot volumes. The rough model results in an overestimation of the progress variable flux of about three times compared to the experimental values. The accurate gas-dynamics model gives instead very good agreement with the experimentally determined progress variable flux, the small over-prediction 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 $\overline{u}_u$ and $\overline{u}_b$ are on average respectively slightly larger and smaller than the corresponding measured values which might be again the consequence of having neglected the effect of gradient transport (which increases $\overline{u}_u$ and decreases $\overline{u}_b$).

7.2. Qualitative validation of heat release distribution with Cheng experimental data

Cheng *et al* [10, 15] have studied turbulent premixed flames in various configurations, including impinging, swirl stabilized and v-flames. During these experiments the average heat release distribution across the flame brush via the measurement of the flamelets crossing frequency $\nu(x)$ has been determined. In the flamelet regime in fact the average heat release can be expressed as [30]:

$$\overline{\rho}\widetilde{W} = \rho_u \, s_L \, \frac{\nu(x)}{U_n(x)} \tag{39}$$

where $\nu(x)$ is the flamelet crossing frequency (average number of flamelet crossing 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 $\overline{c} > 0.5$ with respect to the symmetrical heat release distribution predicted by the BML theory and proportional to $\overline{c}\,(1-\overline{c})$. Under the assumption of constant $U_n(x)$ and laminar flame speed s_L, this indicates that also the averaged heat release is skewed in a similar fashion. The model for counter-gradient transport based on $\overline{p}_u = \overline{p}_b$ has been applied here to the case of a turbulent premixed flame with a ratio of conditional velocities consistent with Cheng data for a swirl stabilized flame [15] $(u_{b+\infty}/u_{u-\infty} = 3)$ and assuming $d\overline{c}/dx$ and $d\overline{u}/dx$ to be distributed according to the symmetrical distribution $\overline{c}\,(1-\tilde{c})$, consistently with the assumption in [15]. This distribution, in fact, doesn't differs substantially from the Gaussian one previously assumed here. Figure 4 shows the heat release distributions versus $\overline{c}$. 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 a heat release 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 subject of future work.

We believe this is a remarkable result which, even if on a qualitative basis, shows that that the capability to correctly account for counter-gradient transport directly affect the accuracy we can estimate heat release. In some sense this experimental data and the successful agreement predicted by our model strongly support our idea that heat release and counter-gradient transport are intimately connected.

7.3. Validation with experiments for a confined turbulent premixed flame by Frank et al

The schematics 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 (approx. 1.5 % by volume of the main flow rate) and the co-flowing air has a velocity of 2.4 m/s. 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 two-dimensional 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.

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

The flames are generally parallel to the burner axis (exception applies mainly to flame A which is characterized by the largest U_t). This is confirmed by the grouping of experimental data points at eight different values of $\overline{c}$ corresponding to eight radial locations within the imaged area. The 1-D theory developed in the present work has been therefore applied here in the direction normal to the flame which in good approximation corresponds to the radial direction.

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. Modeling of turbulent transport is based on a standard $k - \epsilon$ model. The computational domain has been discretised with approx. 15,000 computational cells. Model (37) has been used for calculating the flame stretch factor G in the turbulent flame speed. The critical strain rate has been assumed equal to the inverse of the chemical time scale, i.e. $g_{cr} = s_L^2/\chi$.

Turbulent scalar transport both of gradient and counter-gradient type was previously observed using DNS. Counter-gradient transport was observed for example in the simulations by [25] where $u'/s_L = 1$ while gradient transport was obtained by Trouvé and Poinsot [31] with $u'/s_L = 10$. According to Frank *et al* [3], their work is the first experimental evidence of the possibility to have a transition from counter-gradient to gradient scalar transport in turbulent premixed flames. In general the experiments show that this transition occurs when the ratio u'/s_L decreases. 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 modeling ideas proposed here, it should anyway be observed that the u'/s_L ratio is not sufficient to determine the type of transport and in what extent one component of the total turbulent scalar flux dominates over the other. This is clear from (18) which shows that the gradient part of transport depends also from the actual thickness of the turbulent flame brush (i.e. from $d\tilde{c}/dn$). This is also confirmed by the experiments: flame E in fact is characterized by a ratio u'/s_L which is (slightly) larger than in the case of flame B but with a turbulent scalar 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 $d\tilde{c}/dx$ than flame B. This explain why, despite the slightly larger value of u'/s_L, flame E has an overall gradient transport more biased toward counter-gradient than flame B.

The transition from counter-gradient to gradient type of transport occurs in correspondence of 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 over-predicts the gradient flux shown by the experiments.

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

Table 1. Flame parameters in experiments by Frank *et al* [3].

Flame	ϕ	Θ	u' (m/s)	$u'/\overline{u}$	u'/s_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 transition from counter-gradient to gradient type of scalar transport is finally emphasized in figure 8. This figure shows progress variable contours and region where the scalar flux is of gradient type for flames E, B, C, D. The figure shows that transport in Flame E is nearly everywhere of the counter-gradient type 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 flame 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 that the theory of the TFC combustion models predicts the existence of thin flamelets (not necessarily laminar) also at much higher Re_t numbers typical in large-scale and high velocities industrial burner. We expect therefore counter-gradient transport and the transition to gradient also in this case. Regarding this, numerical simulation of Moreau experiments [26] for high velocity premixed combustion performed in [7] using the the rough model which gives the upper estimation of the counter-gradient 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 $\sim 1cm$ from the splitter plate anchoring the flame. At real burners with combustion stabilization by recirculation zones the gradient transport and transition effect we think is unlikely.

8. Conclusions

Without answer the turbulent combustion challenge is how to describe interaction between turbulence and chemistry, coupling between reaction and diffusive processes on the small turbulent scales that cannot be resolved by accessible methods impossible to develop physically correct combustion models that could describe a great body of experimental data and predict correct trends. It is especially important for premixed case where the combustion rates are completely controlled by this coupling. For non-premixed case the most part of the combustion processes is connected with dissipation rate of a passive concentration, that is physically reasonable described by turbulence models so improper coupling in a model is not so noticeable in simulations.

We see the answer to this challenge in using and developing for turbulent combustion at developed turbulence and fast chemistry Kolmogorov ideas of existing of equilibrium small-scale hydrodynamic structures at combustion modeling. Of course the equilibrium process used must exist in reality, but we believe that such is in the case of real industrial full-scale large velocities combustors. The experience of using

TFC model suggested that this approach could be useful for real applications.

In summary we can say that if a turbulent combustion model expresses combustion rates at $Da >> 1$ and $Re_t >> 1$ in terms of chemical kinetic equations, this model is physically incorrect just as incorrect is a turbulence model that expresses at $Re_t >> 1$ the dissipation rate in terms of the molecular viscosity coefficient.

The phenomenon of scalar transport in turbulent premixed flames has been analyzed in the present paper. The analysis starts from the idea (which is consistent with experimental observations) that turbulence has the effect to increase the thickness of the turbulent flame brush in the same fashion it can increase the thickness of a non reactive mixing layer. This happens independently of the nature of the overall progress variable flux (gradient or counter-gradient). Starting from this assumption, a theory on turbulent premixed flames has been developed in a very natural way in the framework of the Turbulent Flame Closure (TFC) model. This theory shows that, despite what commonly believed, the use of gradient transport (based on $k-\varepsilon$ modeling for example) to model turbulent diffusion component of the progress variable flux in premixed flames is not in contradiction with the phenomenon of counter-gradient transport. It has been demonstrated in fact that this type of transport is characterized by two components: real turbulent diffusion which is responsible for thickening the flame brush and gas-dynamics transport which is connected with the pressure variation across the turbulent flame brush (counter-gradient in the case of analyzed here open flames).

In particular, the analysis shows that the common terminology "counter-gradient turbulent diffusion in premixed flames" and consequently the possibility to have flames with decreasing thickness are improper and mainly related to a (we believe non correct) interpretation of this transport as pure turbulent diffusion flux.

Two models have been developed for the counter-gradient component using assumptions on the distribution of conditional pressures within the unburnt and burnt mixtures. The first (quite unrealistic) is based on assuming the conditional velocities to be uniform within the turbulent flame brush, i. e. this model did not take into account mutual interaction of cold and hot volumes. This model gives principally the possible upper estimation of counter-gradient transport that in typical situations is approximately three times larger than experimentally determined counter-gradient fluxes [1]. The second much more realistic model took into account the actual interaction (acceleration of slower cold volumes by faster hot volumes and vice versa) via the assumption that conditional pressures are equal $p_u(x) = p_b(x) = \overline{p}$. This model gives counter-gradient transport which is in good agreement with Moss experimental data [1].

An important result of the present analysis is that the distribution across the turbulent flame brush of the average heat release is strongly linked with the counter-gradient transport component. The first model proposed for counter-gradient transport, for example, gives flamelets with uniform distribution across the turbulent flame brush, i.e. the heat release distribution versus the Reynolds averaged progress variable is symmetrical in the case of $d\overline{c}/dx$ distributed according to a Gaussian function. The second model instead gives averaged heat release distribution that is skewed toward $\overline{c} > 0.5$. Such skewed heat release distribution is confirmed by the experimental data of [10, 15] where the flamelet crossing frequency has been analyzed.

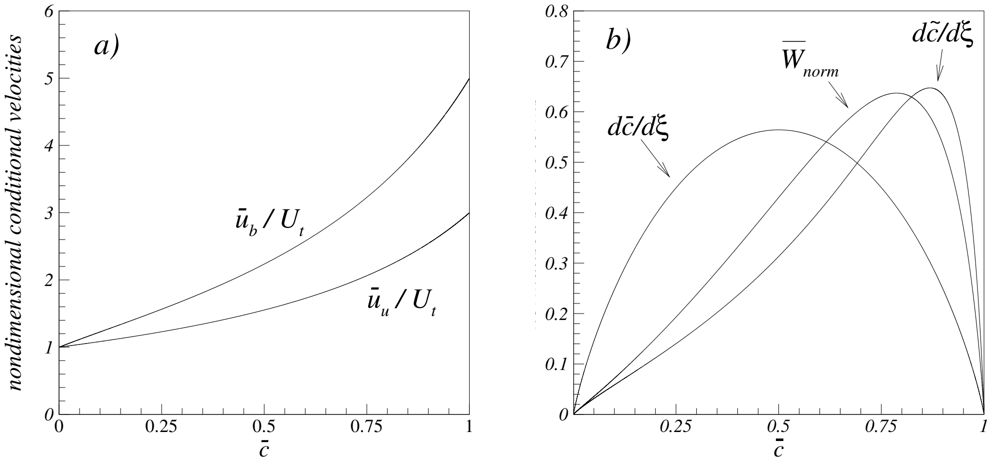

Figure 1. Distribution of a) non-dimensional conditional velocities and b) normalized progress variable source terms across the flame brush. Gaussian distribution assumed for $d\overline{c}/dx$. ($\rho_u/\rho_b = 5$).

Finally, from the practical point of view, the present analysis shows that, for quantitatively correct modeling of the counter-gradient transport it is possible to avoid using of the second order moment closures transport models and that combination of $k - \varepsilon$ model for gradient turbulent transport component and gas-dynamics model for counter-gradient component results quite satisfactory prediction of the counter-gradient transport phenomenon and in transition into the gradient one at variation of experimental conditions.

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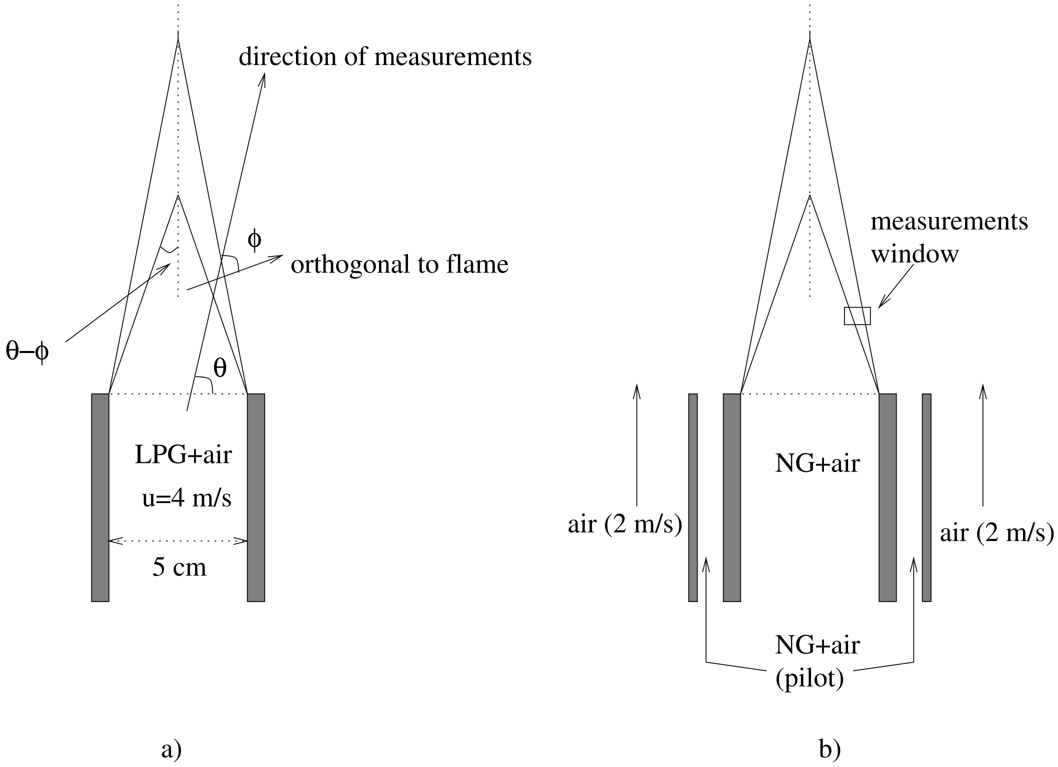

Figure 2. Schematics of combustor in [1] experiments (left) and [3] experiments (right).

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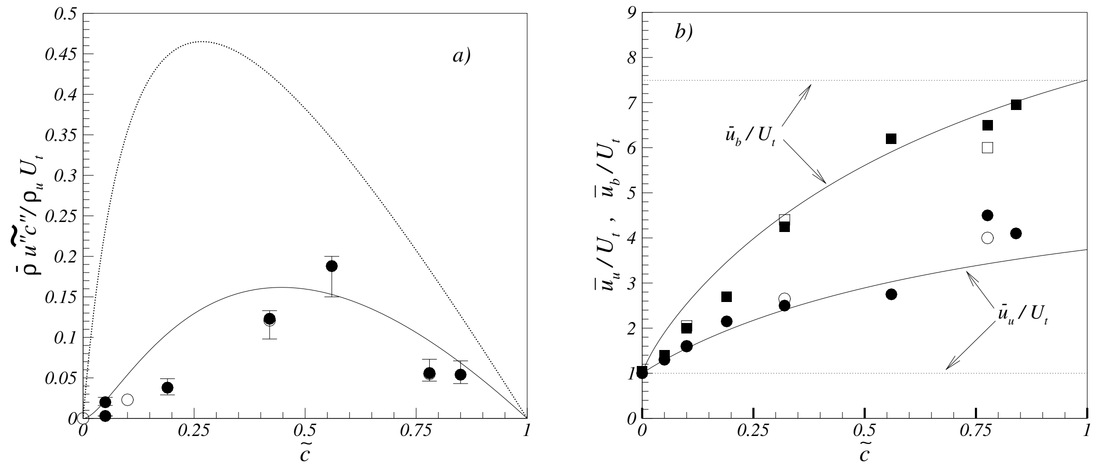

Figure 3. Comparison with Moss experimental data. a) $\cdots\cdots\overline{\rho}\,\widetilde{u''\,c''}$ the rough (upper estimation) model; ——$\overline{\rho}\,\widetilde{u''\,c''}$ the accurate (gas-dynamics) model; ● calculated in [12] from Moss measured conditional mean velocities, ○ calculated in [12] from Moss measured $\overline{\rho}\,\widetilde{u''\,c''}$. b) $\cdots\cdots$the rough model; ——u_u and u_b the accurate model; ● measured experimental u_u, ○ u_u from measured $\overline{\rho}\,\widetilde{u''\,c''}$, ■measured experimental u_b, □t pap u_b from measured $\overline{\rho}\,\widetilde{u''\,c''}$

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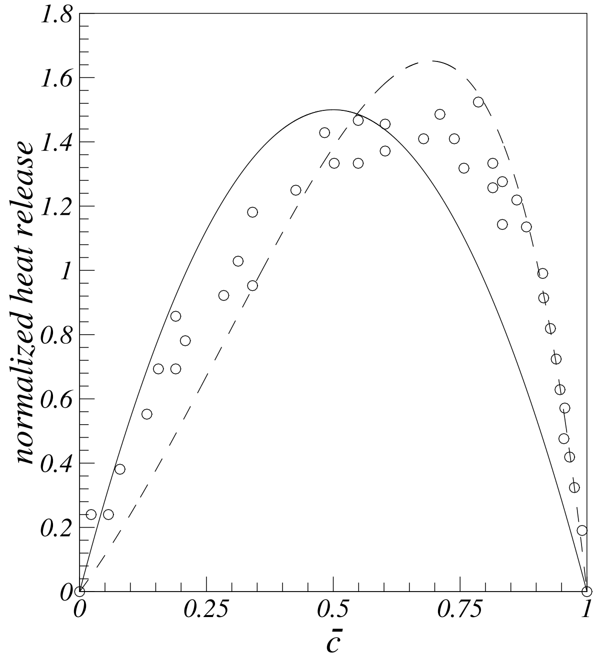

Figure 4. Distribution of heat release across flame brush. ——symmetric BML distribution, ● from experimentally measured crossing frequency [10], - - - -the accurate (gas-dynamics) model.

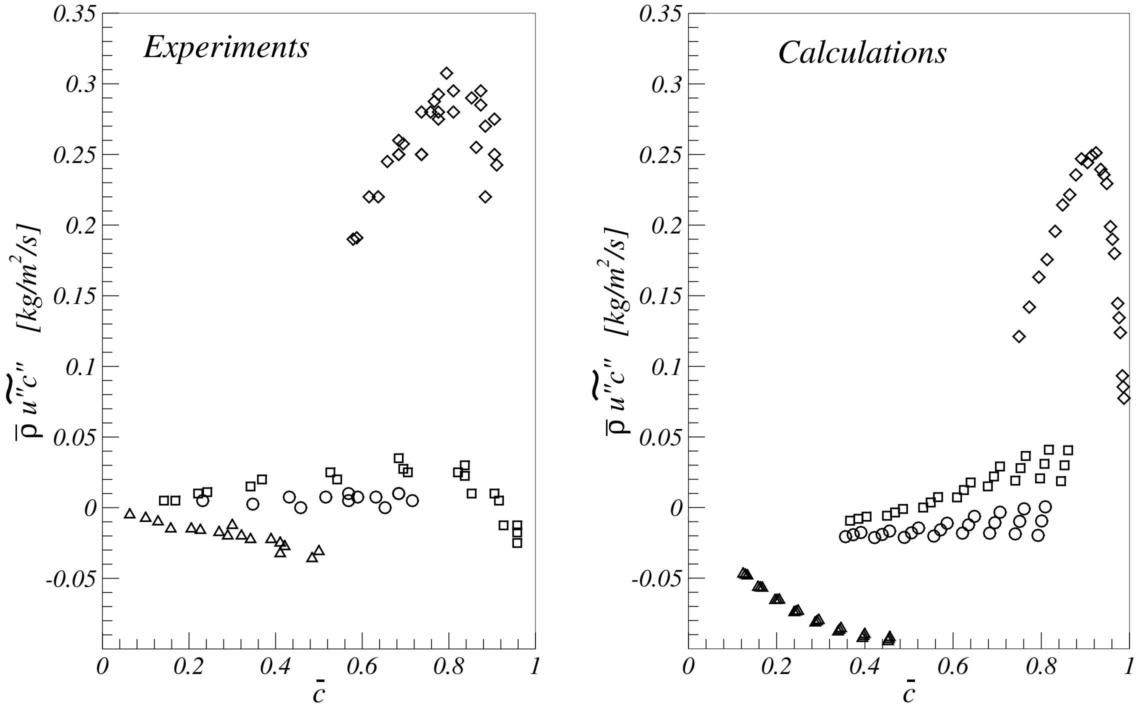

Figure 5. Progress variable turbulent fluxes in Frank *et al* experiments [3]. △Flame D, ○ Flame C, □Flame B, ◇Flame E. Left: experiments. Right: the calculations on the base of the accurate (gas-dynamics) model.

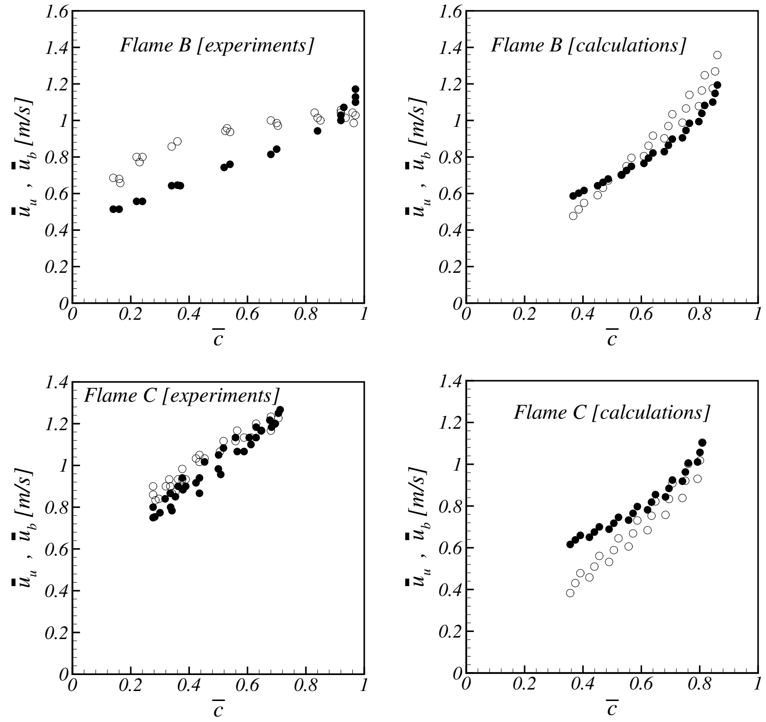

Figure 6. Conditional velocities in flames B and C. Left: experiments; right: the predictions based on using the accurate model.

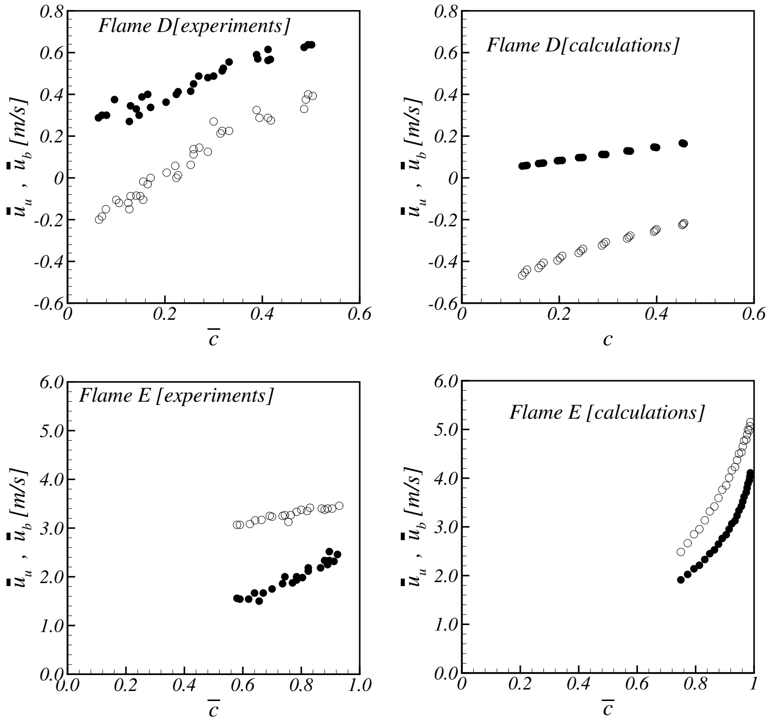

Figure 7. Conditional velocities in flames C and D. Left: experiments; right: present predictions based on using the accurate model

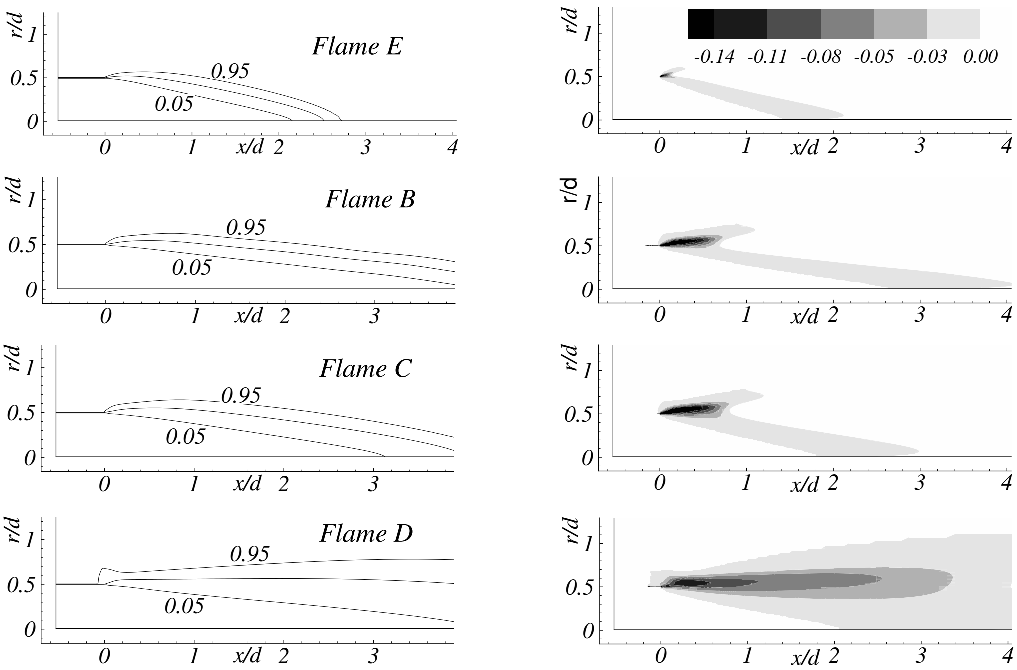

Figure 8. Right: calculated contours of progress variable in flames B, C, D and E in [3] experiments; dark area indicates region of gradient diffusion.