Simple criterion of importance of laminar flame instabilities in premixed turbulent combustion of mixtures characterized by low Lewis numbers

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Simple criterion of importance of laminar flame instabilities in premixed turbulent combustion of mixtures characterized by low Lewis numbers

Jerzy Chomiak and Andrei N. Lipatnikov◉

Department of Mechanics and Maritime Sciences, Chalmers University of Technology, Gothenburg SE-412 96, Sweden

By (i) highlighting the mitigation effect of strain rates on laminar flame instabilities and (ii) comparing peak growth rates of laminar flame instabilities with strain rates generated by small-scale turbulent eddies, a simple criterion of importance of the influence of the instabilities on an increase in premixed flame surface area in turbulent flows is suggested. The criterion implies that, even in lean hydrogen-air mixtures, laminar flame instabilities can significantly affect the flame area only in weak or moderate turbulence (the Karlovitz number defined using laminar flame speed, thermal flame thickness, and Kolmogorov time scale is on the order of 10 or less under room conditions).

I. INTRODUCTION

Interaction of a turbulent flow and a reaction wave is a highly nonlinear and multiscale phenomenon relevant to various processes ranging from combustion [1–8] and deflagration-to-detonation transition [9–11] under terrestrial conditions to evolution of thermonuclear Ia supernovae [11–13] in the Universe. If molecular transport coefficients of major reactants and heat are close to each other, the governing physical mechanisms of the influence of turbulence on a reaction wave are sufficiently well understood [14–20], e.g., turbulence accelerates the wave propagation by stretching reaction zones and increasing their surface area. If, however, molecular diffusivity D of a deficient reactant, e.g., H_2 in a lean hydrogen-air mixture, is significantly larger than molecular heat diffusivity κ of the mixture, i.e., the Lewis number $Le = D_{H_2}/\kappa$ is significantly smaller than unity, the physics of the influence of turbulence on a reaction wave is substantially enriched by a set of local and global phenomena that stem from local variations in mixture enthalpy and composition [21–23]. For instance, as reviewed elsewhere [22–25], abnormally high ratios of turbulent and laminar burning rates were reported in experimental studies of premixed turbulent flames where H_2 was the deficient reactant, with (i) the effect being documented even in very intense turbulence [26,27] and (ii) the effect magnitude being extremely large in certain measurements, e.g., see a recent paper by Yang *et al.* [26]. This effect is still one of the greatest fundamental challenges to the turbulent-reacting-flow community.

From a qualitative perspective, the effect is widely accepted to stem from local variations in enthalpy, mixture composition, and, hence, reaction rates due to imbalance of molecular fluxes of reactants and heat to/from reaction zones curved and strained by turbulent eddies [22–24]. Similar local phenomena are well known to occur in laminar flows and to trigger thermodiffusive instability of lean hydrogen-air laminar flames [21]. More specifically, due to focusing (defocusing) of highly diffusive H_2 in curved reaction zones convex (concave) to fresh mixture, the local enthalpy, equivalence ratio, and burning rate are increased (decreased) in such zones, which propagate faster (slower), thus, increasing amplitude of flame surface perturbations and making the flame unstable [21]. This instability manifests itself in the growth of laminar flame surface area and bulk burning rate, as investigated in detail in several recent numerical papers [28–34].

Since thermodiffusive instability is well known in laminar flows [21,28–34], it is expected to significantly contribute to an increase in flame surface area in turbulent media also, at least in the case of a weak turbulence. For this reason and because both (i) the abnormally high ratios of turbulent and laminar burning rates and (ii) thermodiffusive instability of laminar flames are governed by the same physical mechanism (local burning rate variations due to differential diffusion), the former phenomenon is often claimed to result from the instability [35–42]. Nevertheless, the present authors are not aware of evidence that thermodiffusive instability plays an important role in highly turbulent flows. Since another well-known instability of laminar premixed flames, i.e., hydrodynamic or Darrieus-Landau (DL) instability caused by density drop at the flame [43], was shown to substantially affect the bulk burning rate in weakly turbulent flows only [23,44–46] (if $Le \geqslant 1$), one may expect that the influence of thermodiffusive instability on the bulk burning rate is of great (minor) importance in sufficiently weak (strong, respectively) turbulence. However, under conditions of $Le < 1$, there is the lack of a criterion that allows researchers to estimate under what conditions joint contribution of DL and thermodiffusive in-

stabilities to an increase in flame surface area and, hence, the bulk burning rate is substantial when compared to turbulence contribution. The present communication aims at bridging this gap in fundamental knowledge by suggesting a simple criterion of this kind by taking advantage of recently published results [28–34] of numerical simulations of unstable laminar lean H_2−air flames.

II. CRITERION

For purely hydrodynamic instability of laminar premixed flames [21,43], i.e., under conditions of Le $\geqslant$ 1, such criteria were obtained using different reasons by the present authors [23] and, later, by Chaudhuri *et al.* [45]. Since the goal of this communication is to extend those criteria to turbulent combustion of thermodiffusively unstable mixtures characterized by a low Lewis number, it is worth briefly reminding of the key points of the earlier analyses [23,45] of flames characterized by Le $\geqslant$ 1.

In the former paper [23], the known fact that flame straining mitigates flame instabilities [47–49] was highlighted and the simple equation

$$\frac{d\zeta}{dt} = (\omega - a_t)\zeta \qquad (1)$$

was used to model the mitigation effect. Here, ζ designates the amplitude of the flame surface disturbance, ω is the growth rate of the unstrained flame instability, measured in 1/s, and the negative term $-a_t$ is the normal strain rate. If $a_t = 0$, Eq. (1) describes the exponential growth rate of the perturbation amplitude, i.e., $\zeta = \zeta_0\exp(\omega t)$, in line with classical theories of laminar premixed flames [21,43]. If $a_t > 0$, the term $a_t\zeta$ describes an increase in the normal flow velocity with distance from the flame, e.g., from the left branch of a laminar premixed flame stabilized in two identical counterflows (this is a classical model problem [21,22] research into which was pioneered by Klimov [50]). If this increase is sufficiently large, i.e., $\omega < a_t$, the flow pushes the disturbance back, the amplitude ζ decreases with time, and the instability is suppressed. Note that the instability is suppressed by the negative normal strain rate, whereas the positive tangential strain rate a_t (the sum of the normal, i.e., $-a_t$, and tangential, i.e., a_t, strain rates vanishes at the flame leading edge due to the continuity equation) stretches the flame surface in the tangential direction. In a turbulent flow, tangential straining increases the flame surface area [19,20] even if DL instability cannot arise, e.g., in a constant-density case.

In Ref. [23], the maximal DL instability growth rate $\omega_{\max} = \max\{\omega(k)\}$ was compared with the mean turbulent strain rate $\bar{a}_t$, i.e., the instability was considered to substantially increase flame surface area and, hence, bulk burning rate if $\omega_{\max} > \bar{a}_t$. Otherwise, the instability is mitigated by high normal strain rates created by small-scale turbulent eddies. Here, k is the disturbance wave number and $\bar{a}_t$ is controlled by the smallest eddy time scales. Within the framework of the Kolmogorov theory of turbulence [43,51], the mean strain rate scales as $\bar{a}_t = (\sqrt{15}\tau_K)^{-1}$, where τ_K is the Kolmogorov time scale. For instance, direct numerical simulation (DNS) data by Girimaji and Pope [52] show that $\bar{a}_t = 0.28\tau_K^{-1}$, with 0.28 being close to $1/\sqrt{15} \approx 0.26$.

Alternatively, to assess the importance of a flame instability, its growth rate may be compared with the rate of growth of an isoscalar surface due to tangential straining of the surface by turbulent eddies. If the peak (for various k) rates are compared, we arrive at the same criterion of $\omega_{\max} > \bar{a}_t$. Alternatively, Chaudhuri *et al.* [45] compared the DL instability growth rate $\omega(k)$ with the rate of growth of an isoscalar surface due to tangential straining of the surface by turbulent eddies characterized by the same wave number k, i.e., $a_t(k) \propto (\varepsilon k^2)^{1/3}$ within the framework of the Kolmogorov theory [43,51]. Here, ε designates the mean rate of dissipation of turbulent kinetic energy [43,51]. The two criteria, i.e., $\omega_{\max} > \bar{a}_t$ [23] and $\omega(k) > (\varepsilon k^2)^{1/3}$ [45], look basically similar, as they assume that the DL instability contributes substantially to an increase in flame surface area in a turbulent flow if the instability growth rate is sufficiently large when compared to a time scale of the turbulence. Nevertheless, the two criteria are not identical. The present authors [23] compared $\omega_{\max}$ with the highest strain rate $a_t \propto \tau_K^{-1}$ created by the smallest turbulent eddies, whereas Chaudhuri *et al.* [45], for each wave number k, compared $\omega(k)$ and the strain rate $a_t(k)$ created by eddies characterized by this wave number. Moreover, the former approach emphasizes the influence of turbulence on the instability, whereas the latter approach considers evolutions of flame surface due to (i) DL instability and (ii) turbulence to be independent from one another.

As far as the joint action of DL and thermodiffusive instabilities of low Lewis number flames is concerned, neither the criterion of $\omega_{\max} > \bar{a}_t$ [23] nor the criterion of $\omega(k) > a_t(k)$ [45] has yet been adapted due to the lack of data on the dispersion relation $\omega(k)$ in such a case. A theory of thermodiffusively unstable laminar premixed flames, developed by Sivashinsky [53,54], is restricted to a limiting case of a small density drop at the flame. Other theories, e.g., see Ref. [55], allow for realistic density variations but (i) are restricted to a small difference between Le and unity and (ii) significantly overpredict $\omega(k)$ at $k\delta_L = O(1)$ [33, Fig. 9]. Nevertheless, recent progress in DNS studies of unstable laminar flames [28–34], which allowed for both DL and thermodiffusive instabilities simultaneously, offers the opportunity to arrive at a simple criterion of importance of the instabilities in a turbulent flow.

In particular, complex-chemistry two-dimensional simulations [30–34] of lean H_2−air laminar flames (the equivalence ratio $\phi = 0.6$ [29,30], $0.5 \leqslant \phi \leqslant 2.0$ [31], $\phi = 0.4$ [32,34], or $0.4 \leqslant \phi \leqslant 1.0$ [33]) have shown that, under room conditions, the normalized maximal growth rate $\delta_L\omega_{\max}/S_L$ is close to 1.3 [30, Fig. 1], 1.1 [31, Fig. 1(g)], or 1.7 [32, Fig. 5; 33, Fig. 5(a)], with these maximal rates being reached at $\delta_L k_{\max} \approx 1$; see some of these data compiled in Fig. 1. Here, following Refs. [30–34], the laminar flame thickness $\delta_L = (T_b - T_u)/\max\{|\nabla T|\}$, with T_u and T_b designating temperatures of unburned reactants and combustion products, respectively, and $\omega_{\max}$ is controlled by the joint action of DL and thermodiffusive instabilities. Thus, the criterion of $\omega_{\max} > \bar{a}_t$, i.e., the instability growth rate is sufficiently high, reads

$$\mathrm{Ka} = \frac{\delta_L}{S_L\tau_K} < \mathrm{Ka}_{\mathrm{cr}} = \sqrt{15}\Omega(\phi, T_u, P), \qquad (2)$$
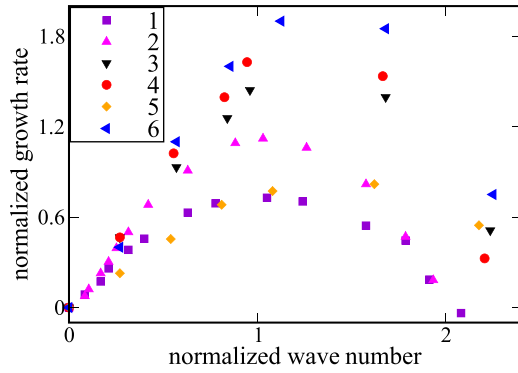
FIG. 1. Compilation of published numerical data on the growth rate of unstable lean hydrogen-air two-dimensional laminar flames. $1 - \phi = 0.75$, $T_u = 300$ K, $P = 1$ bar [30]; $2 - \phi = 0.5$, $T_u = 300$ K, $P = 1$ bar [30]; $3 - \phi = 0.5$, $T_u = 298$ K, $P = 1$ bar [32]; $4 - \phi = 0.4$, $T_u = 298$ K, $P = 1$ bar [31,32]; $5 - \phi = 0.5$, $T_u = 500$ K, $P = 1$ bar [32]; $6 - \phi = 0.5$, $T_u = 298$ K, $P = 5$ bar [32].

where the Karlovitz number Ka arises due to comparison of the time scale τ_K, which characterizes the highest turbulent strain rate, with the time scale $\tau_f = \delta_L/S_L$, which characterizes the instability growth rate. If one defines another Karlovitz number $\delta_Z/(S_L\tau_K)$ using Zel'dovich flame thickness $\delta_Z = \kappa_u/S_L$, the critical value of that number will be significantly less, because of the molecular heat diffusivity in the unburned mixture $\kappa_u \ll S_L\delta_L$ in moderately lean ($0.4 < \phi < 1.0$) hydrogen-air mixtures. An issue of the best definition of a Karlovitz number is beyond the scope of the present paper.

The function $\Omega(\phi, T_u, P) \equiv \tau_f \omega_{max}$, which describes dependence of the normalized peak growth rate on the equivalence ratio ϕ, unburned gas temperature T_u, and pressure P, is still unknown, as ω_{max} is significantly underpredicted [34] by theories [53,54] that do not allow for DL instability or is significantly overpredicted [34] by theories [55] that allow both instabilities but are restricted to $1 - \text{Le} \ll 1$. Nevertheless, currently available numerical data [28–34] show that the normalized ω_{max} (i) is of unity order under room conditions, see symbols 1–4 in Fig. 1, (ii) is decreased with increasing T_u [32, Fig. 5; 33, Fig. 5(b)], e.g., cf. down-pointing triangles and diamonds in Fig. 1, and (iii) is slowly increased with increasing P [33, Fig. 5(c)], e.g., cf. down-pointing and left-pointing triangles in Fig. 1. If we take the highest value of $\Omega(\phi, T_u, P)$ simulated under room conditions [28–34], i.e., $\Omega = 1.7$ [32, Fig. 5; 33, Fig. 5(a)], then $\text{Ka}_{cr} \approx 6.6$. Since this critical number has been estimated using results of numerical simulations of the two-dimensional laminar flames [29–34], the number could be larger, because instability growth rate is often expected to be higher in the three-dimensional case. However, Fig. 14 in a review article by Kadowaki and Hasegawa [28] shows that dispersion relations $\omega(k)$ obtained from two-dimensional and three-dimensional laminar premixed flames, with all other things being equal, are close to one another. Even for the nonlinear stage of flame instability development, only a modest quantitative difference between flame surface areas evaluated in two-dimensional and three-dimensional simulations was recently reported for lean hydrogen-air mixtures [34], with major qualitative trends being similar in both cases.

If, following Chaudhuri *et al.* [45], one compares ω_{max} with the strain rate $\varepsilon^{1/3}/(2\pi\delta_L)^{2/3}$ generated by eddies of the length scale $2\pi/k_{max} = 2\pi\delta_L$, an alternative criterion reads

$$\text{Ka} < 2\pi \left(\frac{S_L\delta_L}{\nu_u}\right)^{1/2} \Omega^{3/2}. \qquad (3)$$

In this case, the instability can play an important role in a wider range of conditions, because $S_L\delta_L/\nu_u > 10$ for lean hydrogen-air mixtures if $\phi > 0.4$.

It is worth stressing, however, that Eqs. (2) and (3) result from comparison of the instability growth rate with the rates of two different processes, i.e., (i) mitigation of the instability-induced flame surface perturbations by normal strain rates created by turbulent eddies upstream of the flame and (ii) the growth of the flame surface due to turbulent tangential strain rates, respectively. If the rate of any of these two processes is higher than the instability growth rate, the influence of DL and thermodiffusive instabilities on the flame surface area and bulk burning rate appears to be relatively weak. Therefore, the choice of the lower Ka_{cr} given by Eq. (2) appears to be appropriate. Moreover, the right-hand side (RHS) of Eq. (3) is significantly larger than the RHS of Eq. (2), because strain rates associated with turbulent eddies whose length scales are close to $2\pi\delta_L \gg \eta_K$ are significantly less (if $\text{Ka} \approx \text{Ka}_{cr}$) than strain rates created by the smallest turbulent eddies whose length scale is on the order of the Kolmogorov length scale η_K [43,51]. Accordingly, even if the instabilities substantially contribute to an increase in a flame surface area at a large scale $O(2\pi\delta_L)$, this contribution to the overall increase in the flame surface area could be low when compared to the turbulence contribution, which is controlled by significantly smaller eddies. For the above reasons, the criterion given by Eq. (2) appears to be more appropriate. It is worth noting, however, that the earlier studies [47–49] of mitigation of laminar flame instabilities by strain rates dealt with large-scale (when compared to perturbation wavelengths) flow nonuniformities. Accordingly, investigation of the influence of small-scale flow nonuniformities on unstable laminar premixed flames is required to better specify turbulent strain rate that should be compared with ω_{max}.

III. DISCUSSION

To the best of the present authors' knowledge, currently available experimental or DNS data do not offer an opportunity to straightforwardly test the suggested criterion. Nevertheless, in the DNS literature, there is indirect evidence that the joint action of DL and thermodiffusive instabilities is overwhelmed by sufficiently intense turbulence in premixed flames.

First, DNS data by Day *et al.* [56, Fig. 6] show that both length scales and magnitudes of fuel consumption rate perturbations are very different in unstable laminar and turbulent lean H_2-air flames. Therefore, scales of the former flame [56, Fig. 6(a)] appear to be useless for predicting scales of the latter flame [56, Fig. 6(b)].

Second, by analyzing other DNS data obtained from two weakly turbulent V-shaped flames (the normalized rms turbulent velocity $u'/S_L = 0.72$ and 2.8), Day *et al.* [57, p. 1043] have noted that "with increasing turbulence levels …

fluctuations, at even the lowest intensity levels, appear to suppress to some extent the growth and propagation of the spherical burning cells characteristic of the thermo-diffusive instability."

Third, results of a recent DNS study by Berger *et al.* [58, Figs. 13, 17, 19–21, 23–25] show significantly different statistics of various local flame characteristics (curvature, strain rate, stretch rate, equivalence ratio, scalar gradients, displacement speed, and fuel consumption rate) sampled from unstable laminar and moderately turbulent lean hydrogen-air flames.

Thus, both the proposed criterion and the cited DNS results consistently indicate that DL and thermodiffusive instabilities are unlikely to control abnormally high ratios of turbulent and laminar burning rates in intense (Ka ≫ 1) turbulence. Since, at high Ka, such ratios were documented in experimental studies [26,27,59] and were reported in recent DNS papers [60,61], another approach should be developed to predict the phenomenon. A leading point concept discussed in detail elsewhere [22–24] appears to be the most promising approach to solving the problem, but discussion of the concept is beyond the scope of the present paper. The readers interested in recent developments of the concept are referred to [62,63].

IV. CONCLUSION

The suggested simple criterion, i.e., Eq. (2), implies that DL and thermodiffusive instabilities of laminar premixed flames can substantially (when compared to turbulent straining) contribute to an increase in a low-Lewis-number-flame surface area and burning rate in a turbulent flow only at small and moderate Karlovitz numbers, i.e., $\mathrm{Ka} = \tau_f/\tau_K = O(10)$ or less.

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