On the Structural Sensitivity of Purely Strained Planar Premixed Flames to Strain Rate Variations

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The effects of aerodynamic straining on the structure and response of adiabatic, unrestrained, equidiffusive, planar premixed flames were experimentally and computationally studied via the counterflow, twin-flame configuration formed by oppositely directed identical jets of nitrogen-diluted, near-stoichiometric methane/air mixtures. Experimentally, the velocity, temperature and major species concentration profiles were determined as functions of the applied strain rate by using LDV and spontaneous Raman scattering. Computationally, the experimental situation was simulated with detailed reaction mechanisms and transport properties. Both the experimental and computational results show that the temperature and species structure of the flame in the direction normal to the flame surface remains largely similar in response to variations in strain rate as long as the flame is sufficiently far away from the stagnation surface so that incomplete reaction is minimal. These results substantiate the concepts that the scalar structure of the flame, and thereby the flame thickness, are insensitive to strain rate variations for these purely strained flames, and that these flames cannot be extinguished by straining alone. The computed results are further shown to agree quantitatively with the experimental data, hence supporting the usefulness of the computational model for the simulation of strained flames. Implications of present findings on the concept of the local flow time, the extinction of strained flames, the modeling of turbulent flames through the concept of laminar flamelets, and flame stabilization and blowoff, are discussed.

INTRODUCTION

The structure and response of laminar premixed flames under the influence of stretch have attracted much attention because not only most practical laminar flames are stretched, but a major effort in turbulent flame modeling is also based on the concept of (stretched) laminar flamelets. Recognizing that flame stretch can be induced either individually or collectively by aerodynamic straining, flame curvature, and flame/flow unsteadiness, a mature qualitative and quantitative understanding of the structure and response of stretched flames requires systematic theoretical and experimental investigations of each of these three aspects, and the eventual synthesis of their individual effects.

In this study we shall focus on effects due to aerodynamic straining. In order not to be unduly cluttered by other factors, we shall consider the simplest and cleanest situation, namely an unrestrained, freely propagating, adiabatic, equidiffusive, planar flame with complete reaction and subjected to aerodynamic straining imposed by a divergent (or convergent) flow field. Hereafter this flame will be designated as a purely strained flame. It is of interest to note that, even for such a simple flame, there still exist conflicting interpretations of its response to straining, especially regarding its scalar structure consisting of the temperature and species profiles. For example, Lewis and von Elbe [1] argued that increasing (positive) straining tends to flatten the temperature profile, reduce the flame temperature, and consequently decrease the flame speed, while the concept of Klimov [2, 3] indicated that such an increase actually steepens the temperature gradient and hence decreases the flame thickness. Chung and Law [4, 5], on the other hand, suggested that the flame response is minimally affected by changes in straining. However, the most frequently mentioned interpretation at present [3, 6] is that the flame becomes thinner with increasing stretch.
Let us now consider in more detail the reasoning behind each of the above concepts. Lewis and von Elbe [1] indicated that positive strain increases the volume of flow in the direction of flame propagation. Consequently, it generates less chemical heat release and loses more heat from the burned to the unburned gases, causing the species and temperature gradients of the flame to be flatter and the flame temperature to be lower than those of the one-dimensional freely-propagating flame. The inadequacy of this argument is that while it takes into account of the negative effect of strain on the flame temperature due to the diffusive transport of heat away from the reaction zone, it overlooks the simultaneous, positive effect of reactant diffusion into the reaction zone. Indeed, flame structure analyses [7, 8] allowing for both heat and mass diffusion with equal diffusivity have shown that the flame temperature remains at the adiabatic flame temperature, and therefore is invariant to stretch.

The concept of Klimov is based on the reasoning that strain-induced convection tends to thin the temperature and concentration profiles within the flame, and that the nature of the solution depends only on the velocity gradient, \( \kappa \), experienced by the flame. This concept, however, does not account for the fact that the flame, being unrestrained, can freely adjust its location to accommodate changes in strain [8]. Consequently the response of the flame structure to strain rate variations is expected to be much less sensitive than if the flame remains stationary. We shall return to this point later.

We also note that theoretical studies of counterflow flames have sometimes indicated that the system Damköhler number decreases with increasing strain rate, hence promoting flame extinction. These solutions, however, are expressed in terms of a similarity variable which scales with \( \kappa^{1/2} \). Thus if the solution were transformed back to the physical domain, the response of the flame structure could become independent of strain.

The concept of strain insensitivity, advanced through the analyses of Chung and Law [4, 5], shows that, to leading order considerations, the flame temperature, flame thickness, and the burning flux at the upstream boundary of a purely strained flame are unaffected by strain rate variations. The burning flux at the downstream boundary of the flame is reduced simply due to flow divergence [4, 5, 8, 9]. Subsequent thermocouple measurements of the temperature profile across flames with different strain rates also demonstrated that they are basically similar [8]. Furthermore, an examination of the numerical solutions of Kee et al. [10] shows that, for near-stoichiometric methane/air mixture, the flame structure and thereby the flame thickness remain almost unaffected for different strain rates—a point also noted by the authors. It may be noted that comparisons based on profiles are more stringent than those based on either the flame thickness or properties evaluated at certain locations, which are definition dependent.

Observations also have been made [4, 5, 11–15] that the flame burning flux decreases with increasing (positive) strain rate. The burning flux referred to in these studies is that associated with the reaction zone, at the downstream boundary of the flame. Such a decrease is to be expected and is a consequence of flow divergence, not reduced chemical reactivity, as mentioned above. Indeed, for an adiabatic, unrestrained, equidiffusive flame with complete reaction, which can be simulated by impinging the combustible stream against a hot inert stream at the flame temperature, the downstream burning flux will monotonically decrease to zero with increasing strain rate without exhibiting the abrupt, turning-point behavior characteristic of flame extinction at a finite burning flux [11–15]. Vanishing of the burning flux, however, does not appear to be an appropriate indication of the occurrence of flame extinction.

There are three objectives for the present investigation. The first is to provide further, and detailed, experimental and computational substantiation of the insensitivity of the flame structure and response to strain rate variations. The need for such a study is the following. Experimentally, the only systematic study of the effect of strain on the flame structure is the intrusively determined temperature profile
of Law [8]. The influence of the thermocouple on the flame structure, and thereby the accuracy of the data, cannot be easily assessed. Furthermore, a conclusive verification of the concept should require an investigation of the species profiles, at least for the major species. Finally, a systematic and comprehensive computational study based on profile comparisons also has not been conducted.

The second objective is to provide experimental data of high fidelity for the velocity, temperature, and major species concentrations of strained flames, obtained by using nonintrusive, laser-based techniques. In this regard it may be noted that while extensive computational studies have been conducted for strained flames [10, 12–19], corresponding experimental investigations have been meager. Data of high accuracy is needed to guide and scrutinize further flame studies. These data are also expected to be useful in the compilation of flamelet libraries for the simulation and modeling of turbulent flames and processes within practical combustion devices.

The third objective is to compare the independently determined experimental and computational flame structures. In view of the recent advances in flame kinetics and computational flame simulation, such a comparison is needed and in fact is overdue.

The present investigation adopted the counterflow, twin-flame configuration because it is steady, nearly adiabatic, planar, and has a fairly well defined strain rate. A near-stoichiometric methane/air mixture, which is essentially diffusionaly neutral, was used. Experimental temperature and major species concentrations were determined using spontaneous Raman scattering, while the flow field was mapped with LDV. Flame computation employed detailed property variations and chemical kinetic mechanisms. Through these studies we shall demonstrate that the structure of the purely strained flame is indeed insensitive to strain rate variations, and that the primary cause of flame extinction in high-strain situations is not a direct effect of strain but is due to strain-induced incomplete combustion.

In the following we shall sequentially present the experimental and computational aspects of the investigation, and then the comparison and discussion of the results.

**EXPERIMENTAL METHODOLOGY**

The stagnation flow field was established by impinging two symmetrical counterflowing combustible mixtures generated from two identical aerodynamically shaped high-contraction-ratio nozzles with 14 mm diameter. The nozzle separation distance was 11 mm and each combustible stream was surrounded by a shroud of nitrogen flow. The axial flow velocity profile along the centerline was measured by an argon-ion LDV system. The measuring volume was 0.1 mm diameter × 0.9 mm length, with seeding provided by 0.3-μm alumina particles. By fitting a straight line to the LDV measurements just upstream of the thermal mixing layer, the strain rate κ was defined as the negative of the measured axial velocity gradient, −du/dx. A mixture of methane, air, and nitrogen with an equivalence ratio of ϕ = 0.95 was used because its estimated Lewis number was close to unity. Here the Lewis number is defined as the ratio of thermal diffusivity of the mixture (α_mixture) to an appropriate controlling mass diffusivity (D_m), which is typically that between the deficient reactant species and the abundant inert species such as nitrogen. Furthermore, the chemical kinetic mechanism of lean methane/air mixtures is considered to be reasonably well established, as far as the prediction of such bulk flame properties as the flame speed and major species concentrations are concerned. Since the experimentally determined extinction strain rate of the ϕ = 0.95 methane/air mixture reported by Law et al. [20] is about 1770 s⁻¹, extremely high flow rates are needed for the 14-mm nozzle in order to reach near-extinction states. The mixtures therefore were diluted by nitrogen (N²/O₂ = 5) to achieve extinction strain rates which were within the capability of the flow system. Using α_mixture = 0.224 cm²/s and D_CH₄−N₂ = 0.222 cm²/s, the Lewis number of this mixture is estimated to be 1.01. The flames were also monitored by a cathetometer during the course of experiment and flickering of these flames was minimal, with the worst situation
being less than ±75 μm for the spatial displacement of the flame.

A schematic of the spontaneous Raman scattering setup is shown in Fig. 1. The excitation source was a frequency-doubled (532 nm) Nd:YAG laser operating at 20 Hz. The laser beam was focused with a 500-mm focal-length plano-convex quartz lens. Detection was accomplished by passing the collected light through a 0.5-m monochromator with a 2400 groove/mm grating onto a gated, 1024-element intensified photodiode array. The system magnification was unity, and the slit was 0.2 mm wide and 2 mm high. Data collection times ranged from 1 to 6 min, depending on the species concentrations at the location of the measurement in the flame. Spontaneous Raman spectra of nitrogen were used to determine the temperature profiles.

A code developed at Sandia [21] for coherent anti-Stokes Raman spectroscopy was modified to analyze the Raman spectra. The quick-fit approach of Hall and Boedeker [22] was used to determine the flame temperature. Specifically, a library of nitrogen spectra spaced 50 K apart were first generated by convolving the spectra with the experimental instrument function. Then the experimental spectra were least-square fitted to the library spectra. The fitting error was estimated conservatively by the temperature bounds where the experimental and theoretical spectra clearly differed [23]. The uncertainty in the fitted temperature was determined to be less than ±50 K [24] and the reproducibility of the measurements was within ±20 K.

While temperature measurements were derived from the shapes of the Raman spectrum, species measurements were obtained from the strength of the Raman signal. This signal is proportional to the number density of the Raman active species and its Raman cross section, the incident laser energy, optical collection efficiency and bandwidth factor. The bandwidth factor accounts for the temperature-dependent distribution of molecules in their allowed quantum states. The Sandia code is reliable for calculation of bandwidth factors of diatomic molecules such as N₂, O₂, and CO [25]. For polyatomic molecules, such as CO₂, H₂O, and CH₄, two types of calibrations were performed. For temperatures below 1400 K, the temperature-dependent calibration factor was determined in a heated mixture of known concentration. For temperatures above 1500 K, concentrations of a number of methane/air premixed flames stabilized on a flat flame burner were measured and calibrated with the predicted equilibrium values [25]. The gap between 1400 and 1500 K is caused by the limitations of the heating elements and the fact that the flat flame is not stable for flame temperatures below 1500 K. Results for these slightly disjoint series of calibrations, however, appear to be smooth and continuous [24]. To further improve the accuracy of species measurement, the bandwidth for each species was chosen in order to minimize the temperature uncertainties. Further details of the spontaneous Raman scattering experiments are discussed in the Appendix.

EXPERIMENTAL RESULTS

Cases A to D designate four experiments conducted with increasing strain rates; the corresponding global strain rates, obtained by dividing the mean exit flow velocity by half of the separation distance between nozzles, are 73, 118, 207, and 298 s⁻¹. Case D has the highest strain rate and is close to the extinction state, which was found to be around 400 s⁻¹. Since the velocity measurements inevitably have some scatter and some judgement of best fit is required to evaluate the velocity gradient, the computationally determined strain rate discussed in the next section will be used to characterize each experiment. All of these laboriously collected experimental data are presented in this paper because of their potential usefulness. Figure 2–5 thus show the profiles of temperature and mole fractions of the major species for each strain rate, in which “–5.5 mm” corresponds to the nozzle exit while “0 mm” corresponds to the stagnation surface. The trend of the variations of these quantities are as expected, with the flame being pushed towards the stagnation surface with increasing strain. The sum of the molar fractions of the major species is seen to be close to unity, ranging between 0.96 and 1.02. This provides an independent check of the experimental re-
Fig. 1. Schematic of the spontaneous Raman scattering setup.

Fig. 2. Comparison between experimental and computed profiles (represented by symbols and solid lines, respectively) of temperature and mole fraction of the major species and their sum in the laboratory coordinate, for Case A ($\kappa = 148$ s$^{-1}$).

Fig. 3. Comparison between experimental and computed profiles (represented by symbols and solid lines, respectively) of temperature and mole fraction of the major species and their sum in the laboratory coordinate, for Case B ($\kappa = 192$ s$^{-1}$).

Fig. 4. Comparison between experimental and computed profiles (represented by symbols and solid lines, respectively) of temperature and mole fraction of the major species and their sum in the laboratory coordinate, for Case C ($\kappa = 240$ s$^{-1}$).

Fig. 5. Comparison between experimental and computed profiles (represented by symbols and solid lines, respectively) of temperature and mole fraction of the major species and their sum in the laboratory coordinate, for Case D ($\kappa = 348$ s$^{-1}$).
Fig. 6. Experimental temperature profiles in the laboratory coordinate, for Cases A to D. Locations of the upstream boundary of the luminous zone and the maximum temperature gradient for each case are also indicated.

Results. Note that some CO data were discarded because their spectra were plagued by the O branch of N₂.

We next separately compare variations of the temperature and the individual species profiles for various strain rates. Figures 6 and 7 respectively show the temperature profiles in the laboratory coordinate and a shifted, flame coordinate. The shifting is based on collating the locations of the maximum temperature gradients determined for all cases (see Fig. 8), with the location for Case A fixed as the reference. All the following figures on profile comparisons in the flame coordinate are plotted in this way.

We first note from Fig. 6 that the locations of the maximum temperature gradient are very close to those of the upstream boundary of the luminous zone, with the spreading representing the uncertainties caused by flame flickering. Figures 6 and 7 further show that while the flame moves closer to the stagnation surface when subjected to increasing straining, in the flame coordinate the temperature profiles remain nearly insensitive to strain rate variations. The only slight deviation from the above observation is for the near-extinction Case D, for which the final, peak temperature at the stagnation plane is lower than those of the smaller strain rates sufficiently away from extinction. This is reasonable because of the reduced residence time, and hence the inability to achieve complete reaction, for this strongly strained flame whose movement is restrained by the adiabatic and impenetrable stagnation surface.

To further scrutinize the possible insensitivity of the flame structure to straining, the profiles of the temperature gradient in the flame coordinate are compared in Fig. 8. Here the original temperature data are first fitted with a cubic spline, and then smoothly interpolated to yield equally spaced data. The temperature gradient profile is obtained by taking the derivative of the fitted data. Recognizing that gradient comparison provides an even more stringent test of insensitivity than profile comparison, the results clearly show the existence of such an insensitivity.

Figures 9–11 show the mole fraction profiles for CH₄, O₂, and CO₂ under different strain rates, in the flame coordinate. It is seen that the overall profiles of these species are again
basic analysis and the experimental accuracy. The above results therefore demonstrate quite convincingly that the flame structure including the flame thickness are insensitive to strain rate variations, especially when the reaction is basically complete.

**COMPUTATIONAL RESULTS AND EXPERIMENTAL COMPARISONS**

Numerical formulation of the counterflow problem and the code employed here are those of Smooke [16]. Calculations were performed in the axisymmetric configuration with one atmosphere pressure and 300 K upstream temperature. Previous studies on counterflow diffusion flames [26, 27] have suggested that the experimental outer flow field is either a plug flow nor a potential flow. However, the flame structure is nearly the same for calculations employing either the plug-flow or the potential-flow boundary conditions. Hence, for simplicity, we have used the potential flow boundary conditions, with an appropriate adjustment to be described below. The reaction mechanism used is a complete C2 scheme [28] consisting of 28 species and 151 elementary reaction steps.

The computed profiles of the velocity, temperature, and concentrations of CH4, O2, CO2, H2O, and CO are shown in Figs. 2–5 and Figs. 12–15 for the four strain rates, as represented by solid lines. To reconcile the difference in the potential flow adopted in the calculation and the mixed flow in the experiment, and recognizing that there is one degree of free-
dom in specifying the boundary condition of the flow because the radial velocity profile at the inlet is not precisely described anyway, the mass flux at the nozzle exit was chosen such that the calculated velocity and temperature profiles (visually) align well with the measured profiles. The adjustment of the mass flux from the global value sometimes can be quite small, but can sensitively affect the calculated profiles because it affects the flame location. To demonstrate this point, consider Case B shown in Fig. 13, for which the mass flux based on the mean nozzle exit velocity is 0.073 g/cm²·s. Fine adjustment shows that 0.076 g/cm²·s yields a better agreement. Consequently, the computationally determined strain rates for Cases A to D are 148, 192, 240, and 348 s⁻¹, respectively. These are the values used in the calculations for comparisons with the experimental data. Finally, since the experimental and calculated velocity and temperature profiles are approximately matched in order to define the strain rate, the comparisons should be considered as those for the species profiles for given velocity and temperature environments.

Figures 2–5 show that the calculated species profiles agree well with the experimental measurements, with the possible exception of the H₂O profiles in Cases A and B. This may be attributed to the difficulty in the calibration of H₂O below 1400 K. For velocity measurements, two observations are to be noted. First, although a small portion of the flow immediately downstream of the nozzle exit is expected to be somewhat plug-flow like, the first experimental data points already exhibit a fairly linear velocity profile ahead of the thermal mixing layer. Since reasonable agreement of the flame structure can be obtained by fitting the linear velocity profile, this result therefore seems to corroborate previous conclusions [26, 27] that the structure of the mixing layer is insensitive to calculations employing either plug-flow or potential-flow boundary conditions. Second, the seeding particle appear to lag the gas flow in the thermal expansion region downstream of the minimum point in the velocity profile. Among the various possible effects that can act on the seeding particles to cause them to depart from the fluid motion [29], thermophoresis appears to be of particu-
lar significance due to the presence of the high-temperature gradient in the thermal expansion region. Here the seeding particles experience a thermophoretic force in the direction opposite to that of the temperature gradient, resulting in measured velocity being lower than the actual value. Using the fitting formula of Talbot et al. [30], the thermophoretic velocity of the alumina seeding particles used for the LDV measurements is estimated to be 8.5 cm/s for a temperature of 800 K and a temperature gradient of 1600 K/mm. This value is in the range of the velocity discrepancy between the calculated and measured values, implying that thermophoresis can account for the velocity lag. A detailed study [31] of the motion of LDV seeding particles under the influence of viscous and thermophoretic forces in the present flame configuration provides further support that the thermophoretic force can indeed induce significant lag between the fluid and particle velocities within a flame. Thus caution should be exercised when interpreting LDV data obtained in the active preheat zone, in that it is not reasonable to expect quantitative accuracy for LDV-determined velocities in the rapid thermal expansion region downstream of the velocity minimum point. In particular, determination of the flame speed based on measurements of flow velocities in this rapid thermal expansion region (i.e., the point of maximum velocity [32, 33] or the upstream boundary of the luminous zone [34]) is expected to be substantially less accurate than that based on the upstream boundary of the preheat zone. This velocity lag could also account for the results [34] that extrapolated data based on the velocity minimum as reference point yield higher values of the flame speed than those referenced to the downstream flow velocity. Finally, it was also found [35] that the smallest experimental error in the velocity profiles for the stagnation flow field was obtained for the flame segment up to the minimum velocity point, thereby further supporting the use of the data upstream of the velocity minimum for flame speed determinations [36].

Having established that the computational results can indeed simulate the behavior of the strained flame, we shall next study the sensitiv-

ity of the computed flame structure and response to varying strain. Clearly, a good degree of insensitivity is expected for the temperature and major species profiles because we have already shown that the computed profiles agree well with the experimental profiles, which have been found to be insensitive to straining. However, an additional, direct comparison among the computed profiles is still needed in order to demonstrate the extent of insensitivity. Furthermore, since minor species were not experimentally measured, the computed profiles provide the only information for the assessment of their sensitivity to strain-rate variations. Of particular interest is the response of the kinetically crucial and highly mobile hydrogen radical because our consideration of insensitivity requires the concept of equidiffusion.

Figure 16 shows the computed temperature profiles in the laboratory coordinate for the four different strain rates. Also indicated for reference are the three important peak locations, which respectively correspond to the maximum temperature gradient, maximum heat release, and maximum CH radical concentration. It is of interest to note that while the deep violet radiation associated with the luminosity of lean hydrocarbon–air mixtures is due to excited CH radicals, Figs. 6 and 16 show that the experimentally observed upstream boundary of the luminous zone is actually closer to the computed location of maximum heat release, with the maximum CH concen-

![Fig. 16. Computed temperature profiles in the laboratory coordinate, for Cases A to D. Locations of the maximum temperature gradient, maximum heat release and maximum CH radical concentration for each case are also indicated.](image-url)
Fig. 17. Computed temperature profiles in the flame coordinate, for Cases A to D. The stagnation surface for each case is indicated by (*).

Fig. 18. Computed temperature gradient profiles in the flame coordinate, for Cases A to D. The stagnation surface for each case is indicated by (*).

Fig. 19. Computed major species profiles in the flame coordinate, for Cases A to D. The stagnation surface for each case is indicated by (*).

Fig. 20. Computed CH$_2$O and CH$_3$ profiles in the flame coordinate, for Cases A to D.

tration occurring within the luminous zone.

Figures 17 and 18 show the profiles of the temperature and temperature gradient in the flame coordinate. The results again show that the bulk of the profiles are largely insensitive to the strain rate, with the possible exception of the near-extinction Case D, for which the flame is slightly thinner. Most of the deviations in the profiles occur either near the leading edge of the flame where temperature starts to increase from its freestream value, or in the downstream region where the flame structure is "truncated" by the stagnation surface. Since the reaction zone structure and its influence on the preheat zone are expected to be mostly influenced by the temperature gradient near the upstream boundary of the active heat release region, the bulk flame response is not expected to be significantly affected by the differences either around the leading edge or in the downstream region. Excessive downstream truncation and thereby incomplete reaction of course will lead to extinction.

Similar observation can also be made for the concentrations of the major species of CH$_4$, O$_2$, CO, CO$_2$, and H$_2$O, as shown in Fig. 19, and the minor species CH$_2$O and CH$_3$, as shown in Fig. 20. Of particular interest are the profiles of the H and OH radicals shown in Fig. 21. It is seen that while the profiles still match very well for most of the flame, their final concentrations, at the stagnation surface, can be noticeably different. This implies that equilibrium is actually not accomplished for these flames, even though the primary flame location, as indicated by that of the maximum temperature gradient, is sufficiently far from the stagnation surface. This lack of complete
equilibrium in the downstream region of the flame, however, appears not to have strong effect on the flame structure and response as long as the flame is not close to extinction. The continuously increasing leakage of the H radical with increasing straining will eventually cause flame extinction, as just mentioned and previously shown by Rogg [18].

Figure 22 shows that, although the degree of complete reaction decreases with increasing strain rate, the overall heat release profile is minimally affected. This is because incomplete reaction occurs downstream of the main heat release zone. Figure 23 further plots variations of the normalized maximum temperature, $T_{\text{max}}$, occurring at the stagnation surface, and the mass flux at the location of the maximum heat release rate, $f_{\text{max}}$, with the inverse of the strain rate; the normalizing values are those of the one-dimensional, unstrained flame. The plot exhibits the upper branch and a small portion of the middle branch around the extinction turning point of the ignition-extinction S-curve, corresponding to $k_{\text{ex}} = 637 \, \text{s}^{-1}$. This is to be contrasted with the experimental "fitted" extinction strain rate of about 425 s$^{-1}$, which is obtained by linearly extrapolating the computationally determined strain rates against the experimental global strain rates, and noting that the experimental global extinction strain rate is around 400 s$^{-1}$. The difference in the extinction strain rate is expected to be due to the approximate, potential description of the flow field, indicating that the flame response does become sensitive to the flow field description when the flame is close to extinction. Experimentation for these extremely weak, near-extinction flames has not been successful because of their propensity to extinguish during the prolonged period needed to conduct a complete experimental mapping.

Figure 23 also shows that the reduction in the maximum temperature is minimum until $k > 150 \, \text{s}^{-1}$, indicating that the burning is almost complete for smaller values of $k$. For this low-strain-rate, strongly burning situation, the mass flux at the reaction zone is still substantially smaller than one, indicating that the reduction is a consequence of flow divergence.
instead of weakened reaction, as mentioned earlier.

CONCLUDING REMARKS

In the present investigation we have successfully demonstrated the viability of the concept [8] that the flame structure and thereby thickness are largely insensitive to strain rate variations for adiabatic, equidiffusive, freely-propagating, planar premixed flames with complete reaction. Such a demonstration would not have been conclusive without the use of nonintrusive laser-based experimentation and computational simulation with detailed chemistry and transport. While additional, independent investigations by others clearly are needed to further scrutinize this concept of insensitivity, the comprehensive nature of the present study does allow us to discuss, with some degree of confidence, its implications on several important fundamental concepts in combustion.

The first implication is the identification of the characteristic flow time $\tau$ and flame thickness $\delta$ associated with a strained flame. Frequently these two values are assessed on the basis of the global strain rate in that $\tau = \kappa^{-1}$ while $\delta = (D/\kappa)^{1/2}$, where $D$ is the gas diffusivity. These relations imply that both $\tau$ and $\delta$ would decrease sensibly with increasing $\kappa$. However, the present experimental data clearly do not show that $\delta \sim \kappa^{-1/2}$ with suitable definitions of $\kappa$, even allowing for the slight variations in the flame profiles with different strain rates. Furthermore, recognizing that properties and responses of a premixed flame must depend on chemistry, it is significant to note that these relations do not contain any information on the reactive nature of the flame.

The intrinsic nature of chemistry is manifested by the ability of the flame to adjust its location, and hence the upstream velocity it experiences, in response to strain rate variations. Thus it appears logical to define the characteristic flow time and thickness of the flame based on the local values, with $\tau = D/\nu_u^2$ and $\delta = D/\nu_u$, where $\nu_u$ is the normal flow velocity at the upstream boundary of the flame. For a stagnation flow with $u = -\kappa x$ and $x = 0$ being the stagnation surface, $\nu_u = \kappa x_f$, where $x_f$ is the flame location. If we further apply the dynamic equilibrium relation that $\nu_u = s_u$, where $s_u$ is the upstream flame speed, then $\delta = D/s_u$. Thus the insensitivity of $\delta$ to strain rate variations implies a corresponding insensitivity of the upstream flame speed $s_u$ as well as the upstream flow velocity $\nu_u$. This in turn implies that with increasing $\kappa$, $x_f$ must decrease proportionally with the flame moving closer to the stagnation surface, as observed. Thus the use of the local characteristic time provides a consistent interpretation of the flame behavior observed herein.

The second implication is our understanding of the extinction mechanism of strained flames. The frequently accepted concept is that a premixed flame will extinguish with a sufficiently strong strain rate. What we have demonstrated is that, since the structure and hence temperature of the purely strained flame studied herein are insensitive to strain rate variations, consideration of strain alone is not sufficient to explain or predict extinction. Additional mechanisms or factors are needed to effect reduction of the flame temperature to the state of extinction. The most prominent mechanisms are system nonadiabaticity, mixture nonequidiffusion, and incomplete reaction due to flame restraining. Any of these factors can manifest its influence in subtle ways, and care is needed to identify them before assessment on the effect of strain on extinction can be made. For the present study flame restraining is imposed by the stagnation surface: close proximity of the flame to this surface induces incomplete reaction and reduces the flame thickness and temperature.

We note in passing that, unlike premixed flames, a strained diffusion flame has less flexibility to freely adjust itself in response to strain rate variations. It has been shown computationally [14] that the counterflow diffusion flame indeed becomes thinner with increasing strain rate. Consequently, the progressively higher leakage of fuel and oxidizer through the active reaction zone leads to a reduction in flame temperature, and eventually to flame extinction.

The third implication is on the modeling of turbulent flames through the concept of laminar flamelets. Here it is recognized that while positive straining imposed by the turbulent ed-
dies increases the flame surface area at the hydrodynamic scale and thereby increases the overall area-integrated burning rate, it can also affect the burning intensity of the flame through the various factors mentioned earlier. Thus effects at both the hydrodynamic and flame scales need to be taken into account. Furthermore, in assessing the flame thickness and comparing it with the turbulent eddy scales, it is necessary to use the local flow time instead of the global value based on the hydrodynamic strain rate. As demonstrated herein, use of the former will leave the flame thickness basically insensitive to strain rate variations for the purely strained flames, while use of the latter would (incorrectly) indicate a proportionate decrease in the flame thickness with increasing straining.

The fourth implication is on flame stabilization in a strained flow field. Previous explanations of flame blowoff have involved the concept of flame weakening with increasing straining. The present study has shown that straining alone cannot induce blowoff; additional mechanisms and factors are needed.

It is, however, also important to emphasize that these implications are based on steady-state considerations. They could be significantly modified by transient effects.

The present study has also yielded some additional observations of general interest. For example, it appears that, because of thermophoresis, the seeding particles for LDV measurements do not readily follow the flow in the rapidly accelerating, high-temperature-gradient region of the flame in which significant thermal expansion occurs over a very short distance. Thus use of LDV instrumentation should be limited to parameters upstream of the active preheat zone, such as the global strain rate of interest here. Furthermore, when determining the laminar flame speeds by using the counterflow twin flame technique [36], it is preferable to measure the upstream instead of the downstream values.

The computational simulation also showed that complete equilibrium for the minor species of H and OH is not attained at the downstream boundary for the present configuration. This, however, does not appear to affect the bulk flame structure and response. Experimentally, downstream equilibrium can be promoted by increasing the residence time there through the use of larger nozzles and correspondingly larger nozzle separation distances.

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APPENDIX: EXPERIMENTAL CONSIDERATIONS FOR SPONTANEOUS RAMAN SPECTROSCOPY

To obtain the detailed structure of stretched laminar flames, spontaneous Raman spectroscopy (SRS) was used to measure concentrations of major species and temperature. The experimental setup is shown schematically in Fig. 1. Detection of the vibrational Stokes Q-branch Raman signal was achieved with an optical multichannel analyzer (OMA) which was capable of integrating and storing entire spectral signatures, performing background subtraction, and providing direct digital output. The signal-to-noise (S/N) ratio of the OMA was enhanced by (1) employing a retroreflector opposite to the collection optics, (2) installing a depolarizer in front of the entrance slit of the monochromator to scramble the polarization of the Raman signal and improve the overall efficiency of the holographic gratings, (3) rejecting the background luminosity by gating the OMA, (4) mounting a high-pass color-glass filter inside the monochromator to reject elastically scattered light, and (5) collecting data for long periods of time, typically 1–6 min. This last point required that the flow system be carefully designed to ensure that the flames were steady.

The Raman signal is not affected by collisional quenching and is linearly dependent on laser power and number density [37, 38]. To ensure that the OMA detection system was linear as well, it was run in the accumulation mode in which consecutive scans of rather short duration (~10 s) are accumulated in memory. This mode avoids potential problems of saturation and improves the S/N. The linearity of the entire system was satisfactorily confirmed by measuring signal intensity as functions of incident laser power and accumulation time. Moreover, under the operating conditions of the OMA, nonuniformities in
diode sensitivity and photocathode spectral response were found to be negligible.

As discussed in the Experimental Methodology section, temperature was determined by least-squares fitting the measured spectrum for N$_2$ to a library of theoretical spectra at different temperatures. Here only relative intensities were considered. However, for the concentration measurements, determination of absolute integrated signals was necessary. The measured Raman signal is proportional to the species number density, Raman cross section, incident laser energy, optical collection efficiency and bandwidth factor [38]. To quantify the measurements, system calibration constants, with respect to that for N$_2$, were measured for each species of interest. The constants, which were determined at room temperature, identify the Raman cross section and optical collection efficiency for each species relative to that for N$_2$. Room temperature N$_2$ spectra were taken at the beginning and end of each experiment in order to monitor any variation in calibration during the experiment and correct for any day-to-day variations in the optical system.

The bandwidth factor accounts for the temperature-dependent distribution of molecules in their allowed quantum states and depends on spectral location, shape and bandwidth of the detection system, and on laser line width. Detection bandwidths were chosen so that the bandwidth factors were nearly constant over a wide range of temperatures, thereby reducing error associated with uncertainty in temperature. For diatomic molecules such as N$_2$, O$_2$, and CO, bandwidth factors were computed with the Sandia code. The product of the bandwidth factor and the calibration constant yielded a calibration factor that was used for data analysis. For polyatomic molecules, like CH$_4$, CO$_2$, and H$_2$O, calibration factors were empirically determined. Depending on the temperature range of interest, either the heated-gas or flat-flame method of calibration was conducted (see Experimental Methodology).

An inherent difficulty in making SRS measurements in counterflow flames is the strong gradients in refractive index $n$ experienced by the incident beam and scattered radiation. For the experimental configuration in Fig. 1, the propagation direction of the incident beam is normal to the gradient in $n$, leading to beam steering. In addition, the Raman-scattered radiation is collected in a direction that is near-normal to $n$, thus leading to a shadowgraph-type distortion of the probe volume. If perfect symmetry existed between incident and collection optics then while the apparent location of the probe volume may be shifted (less than 50 $\mu$m), the collection efficiency would not be affected. However, the outer flame region typically suffers from some asymmetry and the collection optics have a finite angle. Thus, some variation in collection efficiency could be observed. Furthermore, some blockage of the collection optics by the burner surfaces occurred for axial locations close to the nozzle exit. These effects on collection efficiency where collectively accounted for by assuming that the mole fraction of N$_2$ remained constant at the value in the cold mixture. This assumption, which is particularly appropriate for methane flames, allowed a correction factor to be obtained that accounted for variations in collection efficiency with axial position. From the calculation of one-dimensional, freely propagating flame of the mixture studied herein [24], the mole fraction variation of N$_2$ across the flame is indeed less than 2%.

The assumption of constant N$_2$ mole fraction was also needed to account for variations in collection efficiency for the calibration studies. For the heated-gas calibration method this

![Fig. 24. Experimental spectra of O$_2$ and CO$_2$ at $T = 1444$ K, showing an overlap of the O$_2$ spectrum by the wing of the CO$_2$ spectrum at high temperature. The area above the dotted line was integrated to determine the mole fraction of the corresponding species.](image-url)
required that a mixture of known concentration of N₂ and calibration gas be considered to be nonreactive. This assumption was found to be valid even for a mixture of CH₄ and N₂ at temperatures up to 1400 K, the reason being that characteristic reaction times for the reaction CH₄ + M are too long, compared to the available residence time, to allow for significant decomposition of CH₄. Based on this assumption, a correction factor for the heated-gas calibration data was obtained as a function of temperature. For the flat-flame calibration method an approach similar to that for the experiments was used. All calibration data and experimental data reported have been modified with the appropriate correction factors.

Additional corrections to the raw data were necessary for the CO and O₂ concentrations. At high temperatures the Q branch of CO overlaps the O branch of N₂. Although the O branch has a relatively small cross section, its contribution to the apparent CO spectrum cannot be neglected because N₂ was an abundant species [24]. Therefore, the theoretical prediction of the N₂ O branch spectrum was subtracted to obtain the CO spectrum. An overlap of the O₂ spectrum by the wing of the CO₂ spectrum also occurs at high temperature. The contribution from CO₂ was eliminated by assuming that the wing of the CO₂ spectrum was linear, as shown in Fig. 24. This assumption leads to an underestimate in O₂ concentration, which may account for the experimental mole fractions of O₂ being smaller than the computational values in the high temperature regions of the flames. Furthermore, it can also be seen from Fig. 24 that the interference from the low Raman shift component of CH₄ on the spectra of CO₂ and O₂ [25, 39] seems to be negligible in the high-temperature regimes.