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A NUMERICAL STUDY OF THE LAMINAR FLAME SPEED OF STRATIFIED METHANE/AIR FLAMES

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Freely propagating laminar methane/air flames were modeled for spatially stratified equivalence ratio conditions at atmospheric pressure. The GRI 2.11 mechanism was used with a modified version of the Lawrence Livermore National Laboratory HCT code to describe the reaction kinetics. Results showed that the laminar flame speed was strongly affected by the equivalence ratio gradient and by the burned gas composition and temperature. Production of molecular hydrogen from the original fuel and its transport to the reaction zone as well as heat transfer from the burned to the fresh gases are key factors in understanding the influence of stratification on laminar flame speed. Combinations of different mixture stratification conditions can either enhance or reduce the flame velocity when compared with homogeneous mixtures. Mixture stratification is also responsible for higher flame resistance to extinction both on the lean and rich sides of the equivalence ratio. The importance of heat and mass transfer on the observed results implies that their extrapolation to high pressure and to turbulent systems must be made with care.

Introduction

Combustion of stratified fuel/air mixtures has important practical interest in applications such as internal combustion engines, oil spill fires, and industrial furnaces. In these applications, spatially non-homogeneous fuel/air distributions may lead to unexpected combustion characteristics. To better understand these systems, it is important to learn how flames propagate through regions of varying equivalence ratios where mixing gradients are not negligible.

The laminar flame speed is one of the fundamental parameters in characterizing premixed combustion. Theoretical [1], experimental [2], and numerical [3] studies have previously been conducted to understand its behavior as a function of the surrounding environment. A thorough description of physical and chemical effects on laminar flame speed can be found in Ref. [4].

The mechanisms driving the velocity of laminar flames propagating in homogeneous mixtures are fairly well understood. It might be expected that flame propagation in a stratified environment exhibits changes in behavior. Several authors have studied laminar flame propagation within stratified mixtures. Experimental [5–7] and numerical [7,8] studies have shown that the flame propagating through stratified media has features different from what is observed within homogeneous systems. Currently, however, a clear understanding of the mechanisms leading to those differences does not exist.

The objective of this study was to increase the understanding of how flames propagate through stratified equivalence ratio mixtures and to search and identify differences in flame propagation characteristics of homogeneous and stratified media. It relied on numerical simulations of one-dimensional laminar methane/air flames using complex chemistry and molecular transport.

Methane was the fuel chosen for this study because methane/air flames are well characterized from experiments and numerical calculations. Their laminar flame speed and response to changes in physical parameters like temperature, pressure, or equivalence ratio are similar to those of heavier hydrocarbons. However, some aspects of methane/air flame propagation are different when compared with other hydrocarbon fuels [4].

Simulations were conducted using the Lawrence Livermore National Laboratory HCT code [9] with enhanced boundary condition treatment to increase code robustness [10]. The HCT code solves the time-dependent Navier-Stokes equations coupled with species transport and energy equations using detailed chemistry and transport. The ability to solve transient flame configurations is an important feature of this code and is essential when dealing with propagation through stratified media.

The GRI 2.11 [3] mechanism was used to represent the kinetics of the methane/air flames. The NO_x formation and reduction reactions were unnecessary

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in the current work and omitted without loss of accuracy on flame speed calculations. This reduced the GRI 2.11 mechanism from 279 reactions and 49 species to 177 reactions and 32 species.

The HCT simulations also included detailed multicomponent gas-phase molecular transport and heat conduction modeled with the help of the Sandia transport package [11] and transport library data.

Numerical Configuration and Setup

Computational Domain and Initial Conditions

The physical configuration studied in this work is representative of an adiabatic flame propagating in a pipe closed at the left end ($x = 0$) and open at the right end at ($x = 0.1$ m) with the flame moving from left to right. The flame was initialized by imposing burned gases from $x = 0$ to $x = 0.05$ m and fresh gases from $x = 0.05$ m to $x = 0.1$ m. An adaptive Eulerian grid with a high density of grid points in the region where the temperature and concentration gradients are higher was used to represent the domain and resolve the flame structure.

Only the major species present in the mechanism were initialized. The burned gas composition and temperature were those produced by a stabilized steady-state flame in chemical equilibrium for a given set of physical conditions. The initial fresh gas composition reflected the simulated mixture stratification. After the start of the calculation, a delay was needed for the kinetics to fit the physical configuration. During that period, radicals and intermediate species were produced in the flame region, and the flame began to consume the fresh mixture. For the stratified simulations, the flame was allowed to stabilize to a steady-state configuration before finding any mixture gradients.

Changes to HCT

The original HCT code was modified to meet the needs of this study. A translator between the CHEMKIN [12] reaction kinetics file format and the HCT format was built. The method exit boundary conditions handled by HCT were also improved. In the original version of the code, the values of the physical variables were extrapolated to the boundary. This kind of numerical boundary treatment, known as totally reflective, implies that perturbations generated by the system are not damped at its exit. They are reflected back inside the computational domain disturbing the calculation. The Navier-Stokes Characteristic Boundary Conditions (NSCBC) method of Poinsot and Lele [10] was implemented to deal with the exit boundary conditions and led to significant improvement in the stability and robustness of the calculation.

Setup Cases for the Stratified Simulations

Four model cases were considered. The first three were initialized as step distributions of the equivalence ratio. The fourth case corresponded to an alternatively rich/lean pocket distribution of the equivalence ratio:

1. Stoichiometric ($\phi = 1.0$) to lean mixture ($\phi = 0.35$)
2. Stoichiometric ($\phi = 1.0$) to rich mixture ($\phi = 2.5$)
3. Rich ($\phi = 1.5$) to stoichiometric mixture ($\phi = 1.0$)
4. Stoichiometric ($\phi = 1.0$), then rich ($\phi = 2.5$), then lean ($\phi = 0.0$), and then stoichiometric mixture ($\phi = 1.0$)

During the simulations, all the initial gradients in equivalence ratio were reduced due to molecular diffusion.

The fresh gas temperature and the pressure were kept the same for all cases at 300 K and 100 kPa, respectively.

Laminar Flame Speed Calculation in Stratified Mixtures

The objective of this work was the study of laminar premixed flame propagation in a stratified environment as a function of the mixture equivalence ratio. The laminar flame speed and the equivalence ratio characteristic of a premixed flame are well-defined quantities in a homogeneous steady-state system. The laminar flame speed is the velocity of the flame relative to the unburned mixture of fuel and oxidizer. The equivalence ratio is the fuel to air mass ratio normalized by its stoichiometric value $\phi = (F/A)/(F/A)_{st}$, where F is the fuel mass, A is the air mass, and the index st corresponds to the stoichiometric condition. When dealing with stratified systems, however, both quantities become poorly defined. If the flame is crossing a region where the mixture gradient is non-zero, there is not a straightforward way to define the flame speed relative to the changing unburned gas mixture characteristics.

The mathematical expression for determining the laminar flame speed of a steady-state laminar premixed one-dimensional planar flame is obtained by integrating the fuel density ρ_F transport equation. For example, consider a flame fixed at $x = 0$ and the unburned gases occupy $x > 0$ (flowing from left to right). Since as $x \to +\infty$ and $x \to -\infty$ the fuel density gradients are zero, and as $x \to -\infty$ the fuel density is also zero, it can be shown that

$$S_l = \frac{\int_{-\infty}^{+\infty} \dot{\omega}_F dx}{\rho_F^{+\infty}} \qquad (1)$$

The fuel reaction rate $\dot{\omega}_F$ and its density in the premixed unburned gases $\rho_F^{+\infty}$ are quantities available during the numerical calculation of the combustion

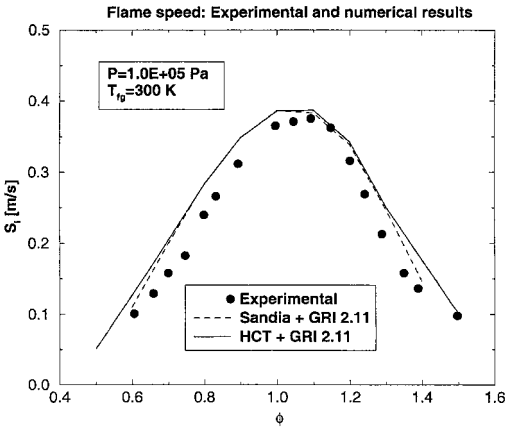

FIG. 1. Comparison between experimental measurements of laminar flame speed by Vagelopoulos and Egolfopoulos (1998) and numerical results given by the Sandia Flame code and the HCT code.

system. The laminar flame speed of a one-dimensional steady-state homogeneous laminar premixed flame is then easily determined.

In the stratified case, the relation between the laminar flame speed and a set of given unburned gas conditions is not straightforward. The normalizing term $\rho_F^{+\infty}$ appearing in equation 1 is no longer constant. As the flame moves toward the unburned gas, it experiences different ratios between the fuel and oxidizer concentrations. In this case, it is not clear how to relate the flame speed and the equivalence ratio in order to compare the behavior of the stratified configuration to the homogeneous case. In this study, for the stratified system, the equivalence ratio experienced by the flame at each instant is taken as corresponding to the equivalence ratio that would exist at the flame location in the absence of chemical reactions. Since the flame has a finite thickness, for calculation purposes, it is assumed that the flame location is at the point of maximum heat release. The decision to choose the point of maximum heat release is arbitrary but valid in the context of this study since the same method is used to evaluate the flame speed of both the stratified and the homogeneous configurations. The values of $\rho_F^{+\infty}$ in equation 1 and of the equivalence ratio of the stratified mixture are also determined according to this. The definition of the equivalence ratio, however, has no meaning inside the reaction zone because the fuel and the air have already been decomposed to intermediate species and radicals. To overcome this problem, nonreacting fuel and air mass concentrations at the point of maximum heat release were reconstructed from the gas composition at that point. A mixture fraction variable based on the element composition of the gas was used for this purpose [13]. If the following imaginary reaction between carbon, hydrogen, and oxygen atoms is considered (with nitrogen used as a diluent), $\nu_O C + \nu_H H + \nu_O O + \nu_N N \rightarrow P$, where ν_i is the stoichiometric coefficient of element i, then the mixture fraction Z is defined by

$$Z = \frac{\dfrac{Z_C}{\nu_C W_C} + \dfrac{Z_H}{\nu_H W_H} + 2\dfrac{Z_{O,2} - Z_O}{\nu_O W_O}}{\dfrac{Z_{C,1}}{\nu_C W_C} + \dfrac{Z_{H,1}}{\nu_H W_H} + 2\dfrac{Z_{O,2}}{\nu_O W_O}} \quad (2)$$

W_i is the molecular weight of element i, Z_i is the element i mass fraction, and $Z_{i,j}$ is the mass fraction of element i inside an imaginary stream j where fuel and oxidizer are not mixed. For a reaction between a general hydrocarbon fuel C_nH_m and oxygen, $\nu_C = n$, $\nu_H = m$ and $\nu_O = 2(n + m/4)$. Also, $Z_{C,1} = nW_C/(nW_C + mW_H)$, $Z_{H,1} = mW_H/(nW_C + mW_H)$ and $Z_{O,2}$ is the oxygen mass fraction in the air stream. The element mass fractions at a given position are determined by adding the contributions of all the species that contain the element at that location: $Z_i = (W_i/\rho)\,(\Sigma_{k=1}^{N_{species}} n_{i,k}C_k)$, where $n_{i,k}$ is the number of i atoms in species k, and C_k and ρ are, respectively, the mole concentration of species k and the gas density at the calculation point. The fuel and air mass fractions that would exist in the absence of a chemical reaction can now be calculated. By definition, $Y^{nr}_{C_nH_m} = Z$, $Y^{nr}_{O_2} = Z_{O,2}\,(1 - Z)$ and $Y^{nr}_{N_2} = Z_{N,2}(1 - Z)$, where the superscript nr corresponds to the non-reactive configuration. The equivalence ratio at the flame position in the absence of chemical reaction is then

$$\phi = \frac{Y^{nr}_{C_nH_m}}{Y^{nr}_{O_2} + Y^{nr}_{N_2}} \Big/ \left(\frac{F}{A}\right)_{st} \quad (3)$$

The fuel density in the fresh gases term that appears in equation 1 is determined by $\rho_F^{+\infty} = Y^{nr}_{C_nH_m}\rho^{nr}$, where ρ^{nr} is the density calculated at the pressure, temperature, and composition of the fresh non-reacting mixture previously determined.

Flame Simulations and Results

HCT Validation

The ability of HCT to predict the laminar flame speed of methane/air flames was first tested by simulating homogeneous configurations with known experimental and numerical results. The velocity of methane/air flames was calculated for steady-state conditions with a fresh gas temperature of 300 K and atmospheric pressure. Equivalence ratios ranging from $\phi = 0.35$ to $\phi = 2.5$ were used. The HCT results are shown in Fig. 1, where they are compared with the experimental results of Vagelopoulos and

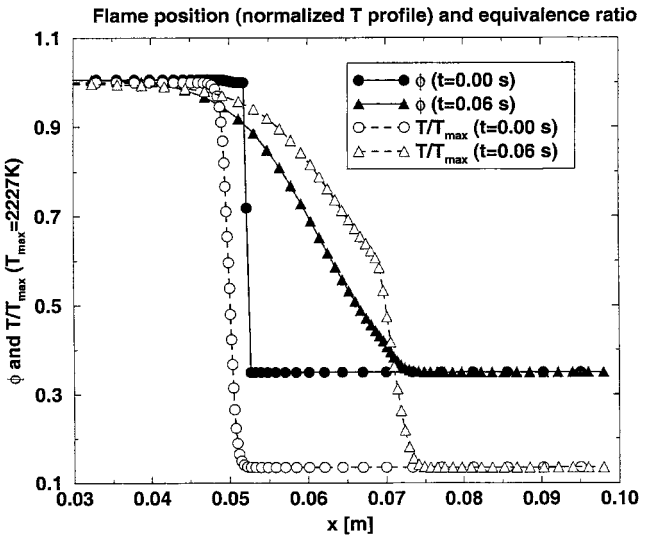

FIG. 2. Equivalence ratio and normalized temperature profiles corresponding to case 1 at $t = 0$ and $t = 0.06$ s. Initial step from $\phi = 1.0$ to $\phi = 0.35$.

Egolfopoulos [2] and with numerical results also calculated with the GRI 2.11 mechanism using the Sandia Flame code [14]. In homogeneous configurations, HCT is able to reproduce the values of the laminar flame speeds for a broad range of equivalence ratios.

The differences in predicted laminar flame speeds for very rich and very lean mixtures between the two codes do not affect the conclusions of this study because the homogeneous calculations compared with the stratified cases were all performed with HCT. They may be due to the way flame speed is calculated as well as to different numerical approaches used by each code. In the stationary Sandia code, the laminar flame speed is by definition the velocity of the fresh gases after convergence to a physical solution has been reached. In HCT, equation 1 is used to determine the flame velocity. The numerical calculation of $\int_{-\infty}^{+\infty} \dot{\omega}_F dx$ can lose accuracy for extreme values of ϕ due to low flame velocities leading to a thicker flame front [4] and, therefore, to a lower resolution of the reaction rate term. On the other hand, numerical convergence to a solution with the Sandia code is more difficult to obtain when the flame is close to extinction.

Case 1: Stoichiometric to Lean $\phi = 1.0 \rightarrow \phi = 0.35$

Figure 2 represents the equivalence ratio and normalized temperature profiles (T/T_{max}) in the computational domain at $t = 0$ and $t = 0.06$ s. The temperature profiles give a good estimation of the flame location relative to the equivalence ratio distribution. It can be seen initially ($t = 0$) that the flame is in a stoichiometric region. For later times, the flame starts to move and causes a rise in the temperature of the fresh gases being consumed. Since the pressure is constant and the domain is closed at $x = 0$, the density decreases and the volume occupied by the burned gases increases. The expansion of the burned gases pushes the fresh gases in front of it at a velocity related to the density decrease in the burned gases. At the same time, species molecular diffusion is attenuating the equivalence ratio gradient. This causes the flame not to jump from $\phi = 1.0$ to $\phi = 0.35$ but instead to cover part of the equivalence ratio range between the two extreme values. Before finding the equivalence ratio gradient, the flame had time to reach a steady state at $\phi = 1.0$.

A comparison between the laminar flame speeds of the homogeneous calculations (●) and the evolution of the laminar flame speed as a function of equivalence ratio calculated during case 1 (×) is shown in Fig 3. At $t = 0.06$ s, the transient flame has covered the range of equivalence ratios between $\phi = 1.0$ and $\phi = 0.43$. Further extension of the simulation showed that a very slow flame is sustained at $\phi = 0.42$ by the presence of hot burned gases. Differences in the behavior of the homogeneous and the stratified simulations are observed. The transient flame is faster than the homogeneous flame for most values of ϕ. The increased flame speed of stoichiometric to lean methane/air flames compared with homogeneous flame propagation has already been reported by Ra [7]. It has been related to chemistry momentum effects induced by the higher temperature of the stoichiometric combustion from which the flame was initiated. The equivalence ratio value $\phi = 0.42$ found as the extreme for flame propagation in the homogeneous and stratified simulations is lower than the experimental lean flammability

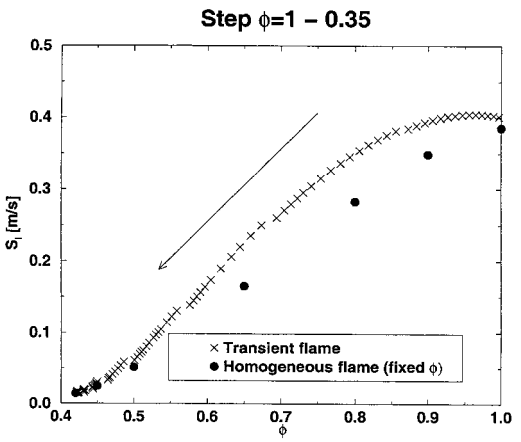

FIG. 3. Comparison between the evolution of the laminar flame speed during case 1 transient simulation as a function of equivalence ratio and the laminar flame speed of steady-state flames at fixed equivalence ratio.

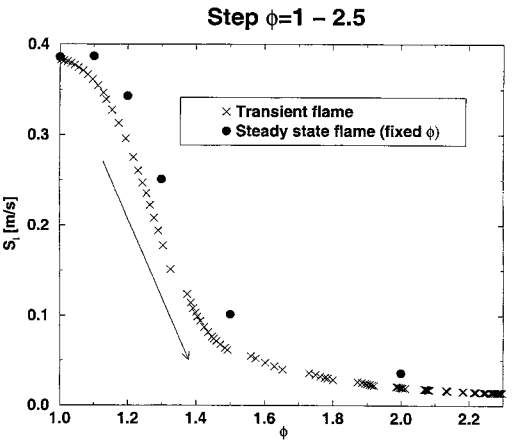

FIG. 5. Comparison between the evolution of the laminar flame speed during case 2 transient simulation as a function of equivalence ratio and the laminar flame speed of steady state flames at fixed equivalence ratio.

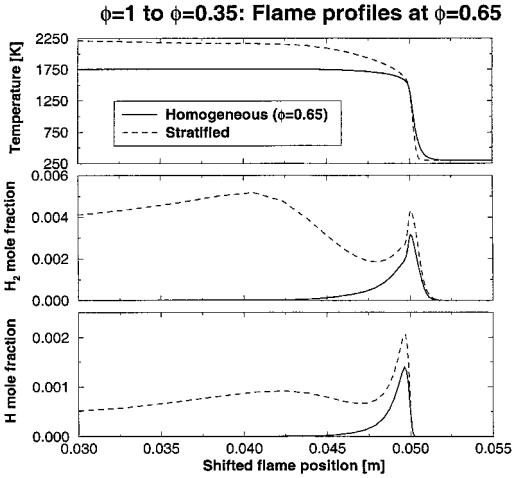

FIG. 4. Temperature, H_2, and H mole concentration profiles in homogeneous lean conditions ($\phi = 0.65$) and during the transient simulation 1 when $\phi_{\text{flame}} = 0.65$. $x = 0.05$ corresponds to the point of maximum heat release in both flames.

limit of the methane/air flames reported in the literature ($\phi = 0.5$) [4]. This may be attributed to the fact that numerically, the one-dimensional flame constantly fed by a heat source (the burned gases) will not extinguish if no heat losses are taken into account, even if the reaction rate becomes very low. Experimentally, such a lean flame is difficult to ignite, and adiabatic conditions are difficult to maintain. For a sufficiently long calculation time, the flame speed of the stratified system reaches an asymptotic value at $\phi = 0.42$, and the memory effects are lost.

To understand the differences between homogeneous and stratified flame propagation, the H_2, H, and temperature profiles of homogeneous ($\phi = 0.65$) and stratified lean methane flames are shown in Fig. 4. In the stratified case, the profiles were taken when the flame equivalence ratio defined in the previous section was 0.65. The shift of the flame position in both configurations to $x = 0.05$ m allows straightforward comparisons between all the profiles. It can be seen that the burned gas temperature is higher in the stratified case. The higher amount of heat conducted to the fresh gas results in faster fuel decomposition and increased formation of molecular and atomic hydrogen. These two species remain in the burned gases due to less oxygen present. The higher concentrations of molecular and atomic hydrogen in the burned gas can also contribute to an increase in the flame speed because of the high reactivity and diffusivity of these species.

Case 2: Stoichiometric to Rich $\phi = 1.0 \rightarrow \phi = 2.5$

Figure 5 shows that when the flame travels from stoichiometric to rich conditions, its velocity is reduced when compared with the equivalent homogeneous flame.

Temperature, H_2, and H profiles of homogeneous and stratified flames are plotted in Fig. 6 at an equivalence ratio of $\phi = 1.3$. The burned gas temperature is higher in the stratified simulation, but the flame temperature difference between the two systems is small compared with case 1, and, therefore, cannot explain the behavior of the stoichiometric to rich flame. The burned gases H_2 concentration is, on the other hand, much higher in the homogeneous flame

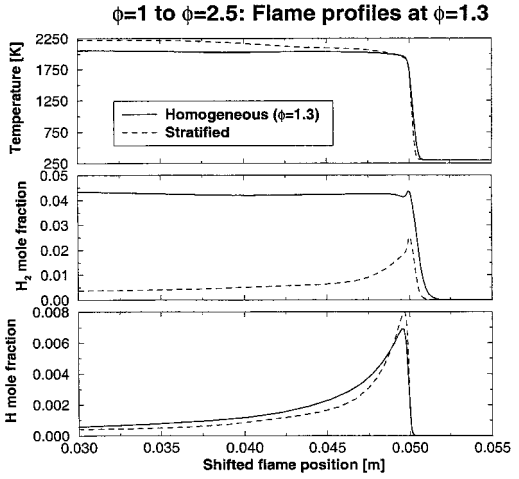

FIG. 6. Temperature, H_2, and H mole concentration profiles in homogeneous rich conditions ($\phi = 1.3$) and during the transient simulation 2 when $\phi_{\text{flame}} = 1.3$. $x = 0.05$ corresponds to the point of maximum heat release in both flames.

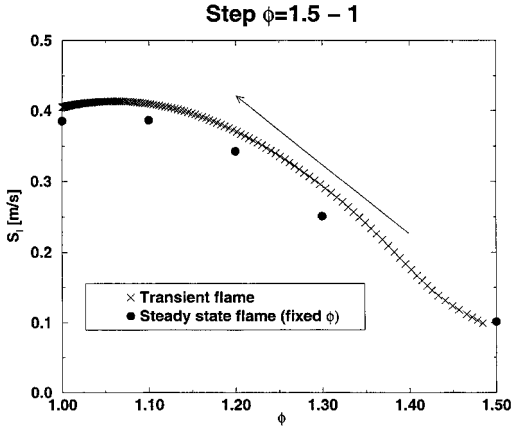

FIG. 7. Comparison between the evolution of the laminar flame speed during case 3 transient simulation as a function of equivalence ratio and the laminar flame speed of steady-state flames at fixed equivalence ratio.

with its lower oxygen concentration. Due to its high molecular diffusivity, hydrogen present in the burned gases diffuses to the flame front to be burned as long as oxygen is present. Since molecular hydrogen has a very high flame speed, its high concentration in the flame front leads to a faster homogeneous flame. The atomic hydrogen profiles are similar in both configurations, although its maximum concentration is slightly higher in the stratified system. This is probably due to a higher oxygen (atomic and molecular) concentration on the burned gases of the stratified stoichiometric to rich flame.

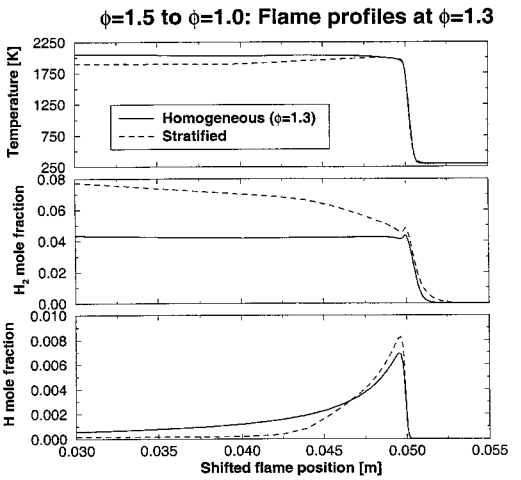

FIG. 8. Temperature, H_2, and H mole concentration profiles in homogeneous rich conditions ($\phi = 1.3$) and during the transient simulation 3 when $\phi_{\text{flame}} = 1.3$. $x = 0.05$ corresponds to the point of maximum heat release in both flames.

Case 3: Rich to Stoichiometric $\phi = 1.5 \rightarrow \phi = 1.0$

In this simulation, the flame was first stabilized at $\phi = 1.5$ and then propagated through a stratified region toward $\phi = 1.0$. Fig. 7 shows that, unlike the stoichiometric to rich case, the flame velocity is higher in the stratified mode in spite of its lower burned gas temperature. Fig. 8 again demonstrates that the significant difference between both systems is the amount of hydrogen (molecular and atomic) produced and left behind the flame. The maximum molecular and atomic concentrations of hydrogen are now higher in the stratified mode.

Case 4: Rich/Lean Pockets $\phi = 1.0 \rightarrow \phi = 2.5 \rightarrow \phi = 0.0 \rightarrow \phi = 1.0$

The objective of this model geometry was to represent a flame propagating through a mixture of changing equivalence ratios by alternating rich and lean pockets. The physical length of those pockets is an important parameter that can change the attained limit values of the equivalence ratio as well as its gradient and, consequently, the transient values of the laminar flame speed. The same kind of computations performed with different pocket lengths and pocket distributions have shown that the trend of the observed results does not change. The evolution of the laminar flame speed as a function of equivalence ratio is given in Fig. 9. These results show that the behavior of this complex system can be predicted from the combination of the simpler ones represented by the previous step function evolutions. The

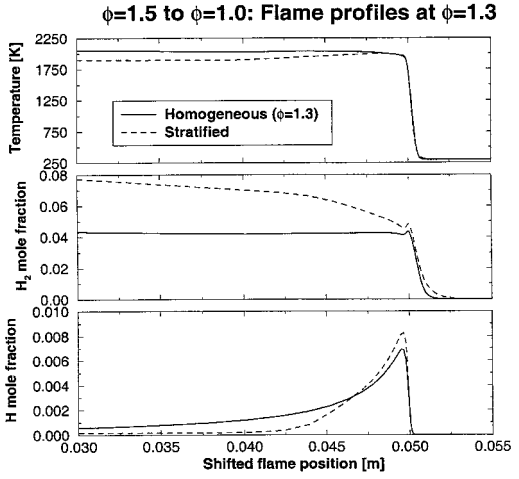

FIG. 9. Comparison between the evolution of the laminar flame speed during case 4 transient simulation as a function of equivalence ratio and the laminar flame speed of steady-state flames at fixed equivalence ratio. The flame follows path 1-2-3-4.

equivalence ratio path followed by the flame is indicated with arrows numbered from 1 to 4 in Fig. 9. The flame is initially stoichiometric, then travels toward a rich mixture (1). It then becomes stoichiometric again (2) on its way toward a lean mixture (3). At the end, it returns to a stoichiometric configuration (4). This fourth path has not been previously simulated. The behavior of the flame in this region was, however, expected since the high-temperature burned gases that accelerated the flame during its way toward the lean mixture still prevail. It is important to note that the higher stratified flame speed is maintained during the lean to stoichiometric evolution.

Conclusions

The laminar flame speed of methane/air flames at atmospheric pressure and ambient temperature as a function of the equivalence ratio has been studied through numerical studies using complex chemistry and transport. Differences in flame propagation through stratified and homogeneous systems have been observed. Stratified flames exhibit memory effects, which depend on the type of mixture stratification and are related to chemical kinetic effects as well as mass and heat transport. The following conclusions were mades:

- High burned gas temperature behind the flame controls lean stratified flames traveling from stoichiometric to lean conditions. These flames are faster than their equivalent homogeneous ones.
- The propagation of rich stratified flames is controlled by production and consumption of molecular hydrogen in the flame front and in the burned gases. If the fuel decomposition leads to high H_2 production that is not consumed because of insufficient oxygen, then the flame tends to accelerate if oxygen is available in the fresh gases. This causes the stoichiometric to rich flames to slow down and the rich to stoichiometric flames to accelerate compared with homogeneous propagation.
- The behavior of alternatively rich and lean flames can be predicted from the effects of stratification on each individual system.

Care should be taken in extending the above conclusions to other physical conditions or other fuel/air systems. The mechanisms of heat and mass transfer as well as the chemical kinetics are strongly linked to the type of system configuration in which the flame is evolving. This includes high pressure and turbulence effects. Turbulence increases the rate of mass and heat transport, whereas high pressure inhibits molecular transport. High pressure may then attenuate the rich stratified flame behavior but turbulence might, on the other hand, enhance the lean stratified flame speed due to a higher rate of heat transfer from the burned to the fresh gases.

From a combustion modeling perspective, the ratio of the characteristic times or length scales between the flame and the equivalence ratio gradients is an important aspect that should be addressed in future work.

Acknowledgment

We would like to thank Dr. C. K. Westbrook for the fruitful discussions regarding the setup and use of the HCT code.

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COMMENTS

Yiguang Ju, Princeton University, USA.

1. It seems that the reason for high H_2 concentration is due to the local quasi-state equilibrium via reaction ($H + CH_4 \Leftrightarrow CH_3 + H_2$). If so, the results are due only to the chemistry. As such, different fuel mixtures at different conditions may have different results. What are your comments?
2. I would like to examine both the chemical and transport properties by using one-step chemistry. It would be helpful for clarification of your results.

Author's Reply.

1. The results of the rich-flame calculations show a strong correlation between laminar flame speed and H_2 concentration. The pathways to H_2 production and consumption are therefore relevant. The dependence of those pathways on the type of fuel/air mixture indicated that the results may indeed be fuel dependent. The $H + CH_4 \Leftrightarrow CH_3 + H_2$ reaction is not in quasi-state equilibrium. The forward reaction prevails.
2. This suggestion can be checked in the future.