

Stretch Effects Extracted from Propagating Spherical Premixed Flames with Detailed Chemistry

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This paper is concerned with extracting stretch effects from outward propagating spherical flames with full chemistry. It is a continuation of a recently published study from this laboratory where it is shown that single-step chemistry is insufficient to explain the experimental results on methane, propane and hydrogen-air systems. Comparisons of the present full chemistry calculations with the experimental results for several fuel/air systems are good/excellent, with the exception of rich propane-air for which it is argued that soot chemistry with attendant radiation loss need to be invoked. The reversal in trends of stretch effects due to change in pressure, initial temperature and N_2 dilution (observed in experiments) can be predicted well by full chemistry but not with simple single-step chemistry. Analysis of the results shows the important role played by many intermediate species and explains why full chemistry is needed to predict stretch effects.

INTRODUCTION

Most practical flames—laminar or turbulent—experience both curvature and local unsteadiness of the flame surface. This causes stretch which affects the propagation speed in premixed flames. Stretch effects in premixed flames have been studied experimentally with several geometries involving stagnation plane, spherical propagation, and Bunsen burners. Stagnation plane flames experience positive stretch and the flames at Bunsen burner tips experience negative stretch. Spherical flames experience positive stretch in outward propagation and negative stretch in inward propagation. Several asymptotic studies [1–5] have been devoted to extracting the stretch effects. Of these, the theories by Frankel and Sivashinsky [4] and Clavin and Joulin [5] show that the IPF (Inward Propagating Flame, negative stretch) exhibits stretch effects different from those of OPF (Outward Propagating Flame, positive stretch). The reasons for this can be related to the fact that the flame speeds with reference to the hot and cold regions cannot be related simply by the density ratio due to the unsteadiness in the continuity equation. Experiments

on OPF have been presented by Palm-Leis and Strehlow [6], Smith et al. [7], Dowdy et al. [8] and Kwon et al. [9]. The results of stagnation of flames are presented by Mendes-Lopes and Daneshyar [10], Law et al. [11]. Results for negative stretch have been presented by Echekki and Mungal [12] from measurements on Bunsen flames.

Experimental results are presented in different formats. Palm-Leis and Strehlow reported the results of dr_f/dt versus r_f , where r_f is the flame radius and t is the time coordinate. Kwon et al. [9] used a different normalization in their presentation of the results. Since basic experimental data have been reported, it is possible to reduce the data into a standard format, namely flame speed versus stretch. Numerical calculations for extracting stretch effects have been few. Those on the stagnation plane flames are motivated by the extinction considerations rather than stretch effects. Those on spherical flames [8, 13–16] have presented the variation of dr_f/dt versus r_f , mostly for single-step reactions, without further extraction of data in terms of flame speed versus stretch. In a recent study, Dowdy et al. [8] have presented experimental and computational results on OPF aimed at obtaining the planar burning velocities. They also show a comparison of the Markstein lengths (which is proportional to the slope of the flame speed vs stretch curve) with experimental data.

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With this background, Mishra et al. [17] have studied propagating spherical flames with single-step chemistry computationally, and obtained the variation of flame speed ratio (ratio of the spherical flame speed to planar flame speed) with nondimensional stretch, $\tilde{\kappa} = \kappa \delta_0 / S_u^0$ (where $\kappa = (2/r_f)(dr_f/dt)$, $\delta_0 = \lambda_u / (\rho_u c_{pu} S_u^0)$, δ_0 is the planar flame thickness, λ_u , ρ_u , and c_{pu} are the conductivity, density, and constant pressure specific heat of the unburned gas, respectively). A comparison of the results of various asymptotic theories, numerical results with single-step chemistry, and experimental results shows that the results of the theories of Frankel and Sivashinsky [4] and Clavin and Joulin [5] compare well with the numerical results, but the comparison with experiments is not satisfactory for most cases except for lean CH₄/air system. The motivation for the present work is to explore the possibility of reproducing the experimental results with full chemistry (FC) and realistic diffusion.

COMPUTATIONAL ASPECTS

The code used for the present calculations is the same as in Mishra et al. [17] and has been validated in several earlier studies [18, 19]. The chemistry used in the calculations is taken from Warnatz [20]. The reactions have been chosen from a single large set for the C-H-N-O system, with 65 reversible steps and 23 species: 8 species and 18 reactions for the H₂-air system, 14 species and 40 reactions for the CH₄-air system, and the full set for the C₃H₈-air system. The chemistry includes the C₂ chemistry for the C₃H₈-air system. Soot chemistry is not included, since well founded details are not available. Transport properties are obtained in same manner as in Lakshmisha et al. [18, 19]. The chemistry is validated by computation of the planar burning velocities over a wide range of mixture ratios for the above compositions. This includes the C₃H₈-air system with C₂ chemistry [21].

Calculation of Flame Speed and Stretch

The flame stretch parameter, κ is defined by the rate of change of a Lagrangian flame sur-

face element per unit area [22] and is given by $\kappa = (2/r_f)(dr_f/dt)$ for spherical flames. Here dr_f/dt is identified as the flame speed with respect to the burnt gases, S_b . The flame speed ratio, s , is S_b/S_b^0 where S_b^0 is the planar flame speed with respect to burned gases. The nondimensional stretch, also called Karlovitz number is given by

$$\tilde{\kappa} = \frac{2}{r_f} \frac{dr_f}{dt} \frac{\delta_0}{S_u^0} \quad (1)$$

This normalization is widely used, except in a recent study by Kwon et al. [9].

The spherical flame is initiated at the center of the combustible mixture of fuel and air and propagates outwards towards the cold mixture. The initial profile (at $t = 0$) for temperature and species is a linear distribution from equilibrium values at the hot boundary to fresh mixture values at the cold boundary. After the flame overcomes the initial transients, during which adjustment of the profiles takes place, the propagational features can be used to extract the stretch effects. Stretch is large at small r_f and as the flame moves outwards it decreases gradually.

The systems considered are (1) CH₄/air, (2) H₂/air, (3) H₂/O₂/N₂, and (4) C₃H₈/air. The predicted results are compared with existing experimental data and the results of single-step chemistry. The effects of pressure, initial temperature, and N₂ dilution on s versus $\tilde{\kappa}$ relationship are also investigated for H₂/O₂/N₂ system. The values of δ_0 and S_u^0 of various mixtures considered in the present work are given in Table 1.

RESULTS AND DISCUSSION

The results of s versus $\tilde{\kappa}$ for CH₄/air at $\phi = 0.6$ are presented in Fig. 1. The results for full chemistry (FC) with the flame being tracked at the two isotherms— $0.5(T_{ad} + T_0)$ and $(0.8T_{ad} + 0.2T_0)$ —are close to one another. This confirms the earlier finding with single-step reaction (SS) [17] concerning the relative independence of the results with the choice of the flame location. The results with FC are close to the experimental values from Palm-Leis and Strehlow [6], unlike those with

TABLE 1

Values of δ_0 and S_u^0 of Fuel Oxidizer Mixtures Used in This Study

Fuel/Oxidizer	ϕ	S_u^0 (cm/s)	δ_0 (mm)
CH ₄ /Air, P = 1 atm	0.6	10.1	0.23
	1.0	39.6	0.053
	1.5	8.2	0.25
C ₃ H ₈ /Air, P = 1 atm	0.775	30.5	0.073
	1.0	47.5	0.0442
	1.5	16.1	0.124
H ₂ /Air; P = 1 atm	0.513	64.1	0.0536
	1.65	330	0.0167
	3.27	238.3	0.0303
H ₂ /N ₂ /O ₂ , P = 3 atm O ₂ /(O ₂ + N ₂) = 0.125	1.0	87.2	0.0138
	1.41	150.3	0.0091
	1.8	155.7	0.0097
	2.78	118	0.0155
	3.27	101	0.0195

single-step chemistry. The improvement of the results with the allowance for individual and actual Lewis numbers is distinct, but not significant. The results for rich and stoichiometric methane-air are presented in Fig. 2. The experimental data include those for OPF from Ref. 6 and for the stagnation plane flame of Mendes-Lopes and Daneshyar [10]. There is little difference in the experimental data from the two sources. The calculated results from

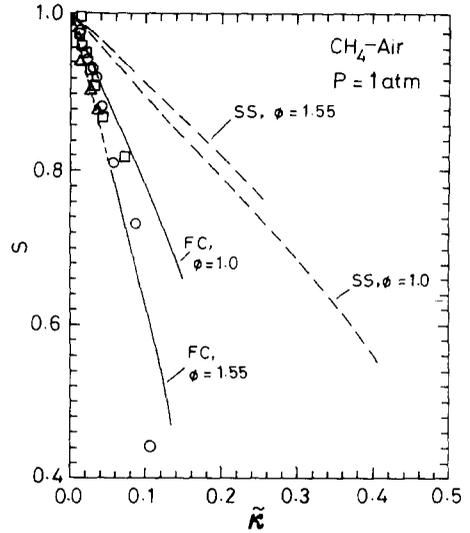


Fig. 2. Comparison of present FC predictions of s versus $\bar{\kappa}$ for CH₄/air flames with SS prediction and experimental results. (Legend: \square Ref. 6 and \triangle Ref. 10 for $\phi = 1$; \circ Ref. 6 for $\phi = 1.55$).

FC are in the same range and the comparison appears very good. The results from single-step chemistry are very different from the other results.

Results in the form of s versus $\bar{\kappa}$ for H₂/air flames, covering lean and rich mixtures, are presented in Fig. 3. Based on experimental

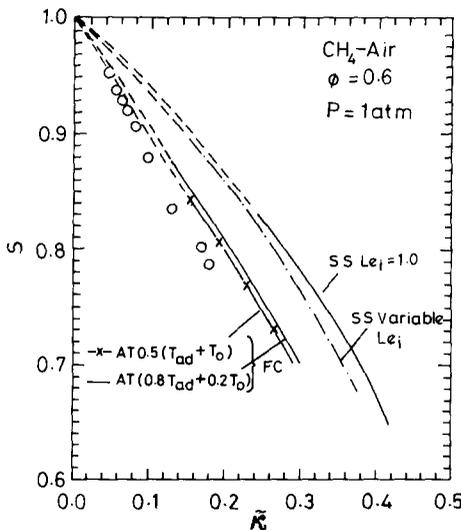


Fig. 1. Sensitivity of s versus $\bar{\kappa}$ to the flame location for present FC prediction for CH₄/air flames and comparison of s versus $\bar{\kappa}$ with SS results and experimental data for $\phi = 0.6$. (Legend: \circ Ref. 6).

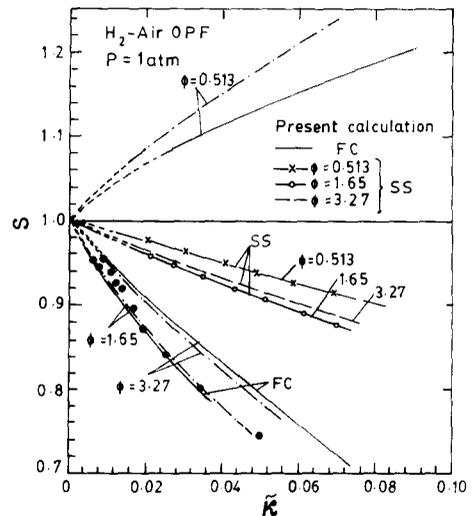


Fig. 3. Comparison of present FC and SS predictions of s versus $\bar{\kappa}$ with experimental and numerical results of Dowdy et al. [8] for H₂/air flames (Legend: \bullet Expt. for $\phi = 1.65$ and $---$, Numerical, Dowdy et al. [8]).

data of flame radius versus time in Ref. 8, results of s versus $\tilde{\kappa}$ were deduced for $\phi = 1.65$ and are shown in the figure. The results indicate that in the case of lean flame ($\phi = 0.513$), the flame speed increases with stretch, whereas for the other mixtures ($\phi = 1.65$ and 3.27), it decreases with stretch. The slope of s versus $\tilde{\kappa}$ curve is marginally higher for $\phi = 1.65$ than for $\phi = 3.27$. The curves for all cases are nearly linear as the range of stretch covered in these flames is small.

Kwon et al. [9] have reported results for $H_2/O_2/N_2$ mixtures from stoichiometric to rich at 3 atm and an initial temperature of 298 K, with the majority of the results at $O_2/(O_2 + N_2) = 0.125$. The experimental data reported in this work are not free from the effects of instabilities, particularly for $\phi = 1.0$ to 1.41 due to preferential diffusion, as stated by the authors. The flames for $\phi = 1.8-3.27$ are free from instability. The results of the computations and the experiments for $\phi = 1.41, 1.8, 2.78,$ and 3.27 are presented in Fig. 4. The results at $\phi = 1.41$ are included because part of the data is free of the effects of instabilities. Figure 4 shows that the present numerical results match well in the small stretch range; it is believed that the data at small radius (higher $\tilde{\kappa}$) are not free from transients.

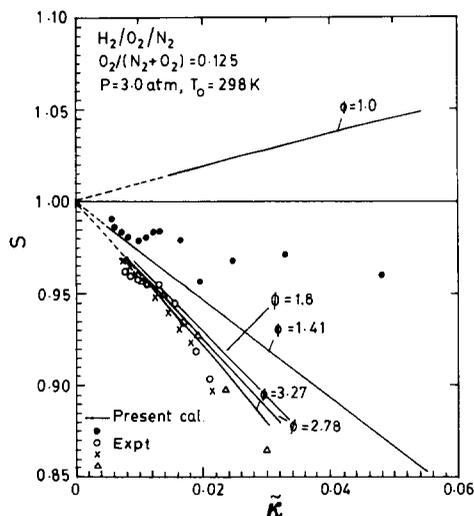


Fig. 4. Comparison of present FC predictions of s versus $\tilde{\kappa}$ for $H_2/O_2/N_2$ flames with SS prediction and experimental results of Kwon et al. [9]. (Legend: ● for $\phi = 1.41$, △ for $\phi = 1.8$, × for $\phi = 2.78$ and ○ for $\phi = 3.27$).

From Fig. 4, it is seen that for $\phi = 1.0$ at 3 atm and $O_2/(O_2 + N_2) = 0.125$, s increases with $\tilde{\kappa}$. The present prediction displays the same qualitative trend as do the experiments of Kwon et al. [9] (not shown in Fig. 4). But, the result of dr_f/dt versus r_f from the work of Dowdy et al. [8] for $\phi = 1.0$ and $O_2/(O_2 + N_2) = 0.21$ indicates that the flame speed increases to S_b^0 as the flame front moves away from the center. In other words, s decreases with increasing $\tilde{\kappa}$, which is the opposite in trend to the above. The two cases differ in terms of pressure and N_2 dilution. The effects of pressure and N_2 dilution are, therefore, studied for this case. Calculations were made for $\phi = 1.0$ at pressures of 1.0, 3.0 and 10.0 atm at $O_2/(N_2 + O_2) = 0.125$. The results of s versus $\tilde{\kappa}$ as in Fig. 6 show the s versus $\tilde{\kappa}$ curve for 1 atm in the lower quadrant, and the curves for 3 and 10 atm in the upper quadrant much like the results of Dowdy et al. and Kwon et al., respectively. These effects cannot be predicted from single-step chemistry unless artificial reaction rate models are invoked.

The effects of N_2 dilution are studied for this case ($\phi = 1.0$) at a pressure of 3 atm. The results of s versus $\tilde{\kappa}$ at $O_2/(O_2 + N_2) = 0.125$ and 0.14 in Fig. 6 show opposing trends in the

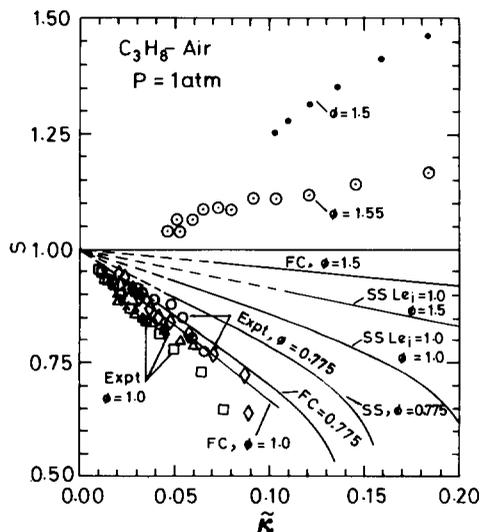


Fig. 5. Comparison of present FC predictions of s versus $\tilde{\kappa}$ for C_3H_8 /air flames ($\phi = 0.775, 1.0, 1.5$) with SS prediction and experimental results (Legend: ● Ref. 7 for $\phi = 1.5$ and ○ Ref. 6 for $\phi = 1.55$; □ Ref. 6, × Ref. 23, and △ Ref. 7 for $\phi = 1.0$; ○ Ref. 6 and ◇ Ref. 7 for $\phi = 0.775$).

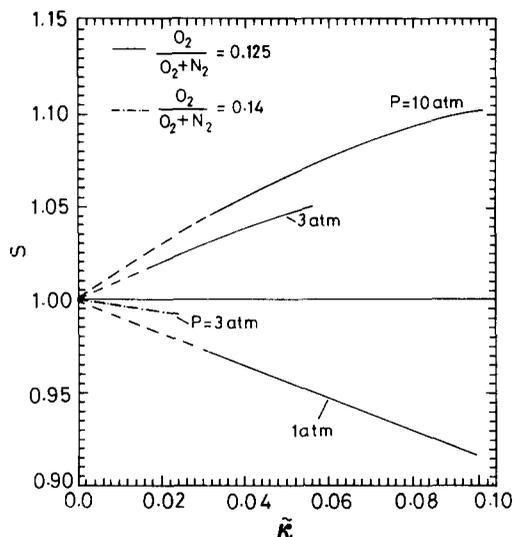


Fig. 6. Effects of pressure and nitrogen dilution of mixture on s versus $\tilde{\kappa}$ relationship for $\text{H}_2/\text{O}_2/\text{N}_2$ flames at $\phi = 1.0$.

effects of stretch on flame speed. The effect of nitrogen dilution is of the same nature as that of pressure.

Propane-air flames are considered next because there are a number of experimental studies on these flames by Fristrom [23], Palm-Leis and Strehlow [6], Smith et al. [7], and Deshaies and Cambay [24]. In all except the last, OPFs are studied. Calculations are made with a kinetic scheme involving 23 species and 128 elementary reactions, taken from a larger set of 28 species and 198 reactions of Warnatz [20]. Calculations have been made for $\phi = 0.775, 1.0$, and 1.5 ; the last case is close to the soot limit and the reaction scheme may be incomplete. This case is chosen since the Lewis number of the deficient species, O_2 is 1.02, for which single-step chemistry predicts a decrease in flame speed with stretch, while the experiments show an increase. It is of interest to see if FC, though approximate, resolves this anomaly.

The predictions along with the experimental results are plotted in Fig. 5. While the experimental results for $\phi = 0.755$ and 1 in the lower quadrant are close to one another, the results for the rich mixture occupy the upper quadrant. There is a difference between the results from the two sources of Palm-Leis and Strehlow [6] and Smith et al. [7], but the trends

are the same; stretch enhances flame propagation. The results of the SS chemistry are all in the lower quadrant and are far removed from the experimental results. These results are similar to what may be expected of the CH_4/air system. The FC predictions for lean and stoichiometric flames are close to the experimental data, but the predictions for $\phi = 1.55$ are in the lower quadrant albeit with a smaller slope. The departure from the experimental data, though less than for SS chemistry, is significant.

The Role of Full Chemistry

The parameters in SS model are $q = (T_{\text{ad}}/T_0 - 1)$, $\beta (= E/RT_{\text{ad}})$ and Le_i , where T_{ad} and T_0 are adiabatic and initial temperatures, respectively, E/R is the activation temperature of the equivalent single-step reaction, and Le_i is the Lewis number of the i th species. The choice of q is based on T_{ad} and obtained easily. The value of β is obtained such that the heat release rate (H_r) variation with τ , the nondimensional temperature ($= [T - T_0]/[T_{\text{ad}} - T_0]$) of a planar flame with the full chemistry calculation is reproduced as closely as possible, particularly the temperature at which the heat release rate peaks. The values of Le_i are obtained on the basis of deficient species, or from individual trace diffusion coefficients. It is expected that the SS model will perform well when the heat release rates obtained by the SS and FC models match and the various cases considered in this work are examined from this view point.

For the lean CH_4/air flame, $\beta \approx 10.6$, and it is large enough for asymptotic theories to be valid. This implies that the reaction zone thickness ($\sim 1/\beta$) is small compared with the overall thickness. Hence, the details in the reaction zone are not crucial. This is the reason that the SS model performs well, particularly when diffusivities of all species are calculated and the values of Le_i are considered variable. The situation is not the same for $\phi = 1$, where $\beta \approx 6.2$. In this case the details in the reaction zone do matter and many details, such as the hot tail of the temperature profile, are not reproduced properly by the SS model. Hence predictions with the SS model are not so good and FC is

required. For the case of CH_4/air flame at $\phi = 1.55$, SS predictions are bad even though β is large. It is not obvious why it performs so poorly.

The results for lean and stoichiometric $\text{C}_3\text{H}_8/\text{air}$ flames have features similar to CH_4/air system and similar explanations are also valid here. But, for the rich case the predictions of FC calculation are not satisfactory. The basic reason for this is, possibly, the inferior FC model which neglects soot chemistry. Soot as a participant can change the energy distribution and can also cause a significant radiation loss to the environment. To check the effect of the former is beyond the scope of the present work since the appropriate chemical kinetic models are not available. However, in a recent work Fendell et al. [25] have addressed the effect of radiative loss due to soot formation in spherical flames using an approximate technique. The results show that the flame speed does not vary with r_f in the absence of soot, but in the presence of soot it decreases with r_f . This implies that the s increases with $\bar{\kappa}$, as found in the experiments. Presumably more accurate calculations with soot fraction will support this result.

The H_2/air systems have a very low value of β at all equivalence ratios [26], typically 1.0–2.7, and hence the stretch effects predicted with the SS model for various equivalence ratios are all close to one another. They show flame speed decreasing with stretch and the results of FC show similar trends for all the equivalence ratios considered except at $\phi = 0.513$ which alone shows flame speed enhancement (Fig. 3). Similar anomalies are also noticed in the effect of pressure and nitrogen dilution. This is in spite of the fact that the heat release profile obtained with the SS model is similar to that obtained with FC chemistry as can be seen from Fig. 7. The cause for the differences in these cases are more subtle and finer details of the reaction zone have to be examined to unravel it.

An examination of the importance of the various species in influencing the stretch effects was made by obtaining the sensitivity of the heat release rate to the mass fraction of the species. This quantity is the Jacobian of the volumetric heat release rate with respect to

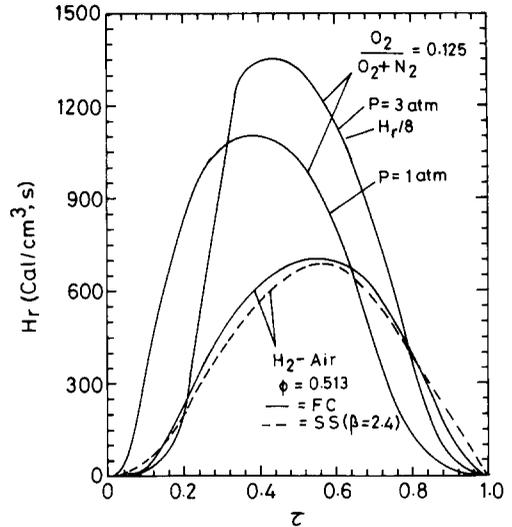


Fig. 7. Predicted H_r versus $\tau = (T - T_0)/(T_{ad} - T_0)$ for H_2/air flames at $\phi = 0.513$ and pressure of 1 atm with FC and SS, and for $\text{H}_2/\text{O}_2/\text{N}_2$ flame at $\phi = 1$ and pressures of 1 atm and 3 atm with FC.

the species mass fraction ($J_{H_i} = \partial H_r / \partial Y_i$). Table 2 shows those Jacobians of significantly large magnitude for nine of the cases studied. It has many interesting features. Even for the lean methane/air flame ($\phi = 0.6$) the largest Jacobians are of CHO and H and the relative importance shifts as the equivalence ratio is varied. Since these species have widely different Lewis numbers (Le_i), it is not surprising that the response of flames to stretch also varies with mixture ratio even though the relevant major reactants (like deficient reactant) have Lewis numbers close to unity.

In the light of this observation, the behavior of hydrogen flames with regard to the effect of mixture ratio, pressure, and nitrogen dilution on the s versus $\bar{\kappa}$ relationship can be examined. It can be seen from Table 2 that the controlling species in the case of $\text{H}_2/\text{O}_2/\text{N}_2$ flame is HO_2 with $Le = 1.5$ at $p = 1$ atm and H with $Le = 0.27$ at $p = 3$ atm. In addition, enhanced pressure shifts the heat release rate versus τ curve (shown in Fig. 7) to the right, implying that the effective β increases. If it is assumed that the effective Lewis number of the mixture is the same as that of the species having the largest J_{H_i} , then the value of β ($Le - 1$) at $p = 1$ atm is 0.4 and at $p = 3$ atm is -0.8 . Thus the behavior can be qualita-

TABLE 2
Large Jacobians ($J_{H_i} = \partial H_r / \partial Y_i$, cal/cm³s) of the Heat Release Rate with
Respect to Species Mass Fraction for Various Flames

CH ₄ /Air, $p = 1$ atm						
ϕ		CHO	H	CH ₃	O	H ₂ O ₂
0.6	J_{H_i}	-0.55×10^{10}	0.44×10^{10}	-0.18×10^{10}	-0.13×10^{10}	
	Le	1.46	0.19	1.00	0.71	
1.0	J_{H_i}	-0.16×10^{11}	-0.51×10^{11}	-0.30×10^{10}	-0.67×10^{10}	0.65×10^{10}
	Le	1.47	0.19	1.00	0.71	1.11
1.55	J_{H_i}		-0.13×10^{11}		-0.11×10^{11}	0.13×10^{11}
	Le		0.19		0.71	1.11
H ₂ /Air, $p = 1$ atm						
ϕ		H	H ₂	HO ₂	OH	H ₂ O ₂
0.513	J_{H_i}	0.83×10^{10}	-0.67×10^9	-0.58×10^{10}	-0.12×10^{10}	
	Le	0.26	0.43	1.45	0.95	
1.65	J_{H_i}	-0.13×10^{11}		-0.57×10^{11}	-0.56×10^{10}	-0.40×10^{10}
	Le	0.37		1.86	1.24	1.88
H ₂ /O ₂ /N ₂ , O ₂ /(O ₂ + N ₂) = 0.125, $\phi = 1.0$						
p		H	H ₂	HO ₂	OH	H ₂ O ₂
1.0	J_{H_i}	-0.83×10^{10}	-0.53×10^9	0.15×10^{11}	-0.14×10^{10}	
	Le	0.27	0.46	1.50	0.98	
3.0	J_{H_i}	-0.49×10^{11}	-0.16×10^{10}	-0.24×10^{11}	-0.49×10^{10}	0.42×10^{10}
	Le	0.27	0.46	1.50	0.98	1.51
C ₃ H ₈ /Air, $p = 1.0$ atm						
ϕ		<i>i</i> -C ₃ H ₇	<i>n</i> -C ₃ H ₇	C ₂ H ₄	C ₂ H ₅	CH
0.775	J_{H_i}	0.52×10^{12}	0.14×10^{12}	-0.19×10^{13}	0.17×10^{12}	-0.22×10^{12}
	Le	1.94	1.94	1.39	1.40	0.92
1.0	J_{H_i}	-0.91×10^{12}	0.23×10^{12}	-0.22×10^{13}	0.32×10^7	-0.23×10^{12}
	Le	1.92	1.92	1.38	1.39	0.91

tively explained within the framework of single-step chemistry, the predictions of which shows increasing $(ds/d\bar{\kappa})_{\bar{\kappa}=0}$ with decreasing β (Le - 1). However, the magnitude of slope is not predicted. Similar explanations can be extended to the effects of mixture ratio and nitrogen dilution variations [21].

CONCLUDING REMARKS

This paper is devoted to the study of the stretch effects on the flame speed and the detailed structure of flames using FC model. Based on the availability of experimental data in the literature, three fuel-air systems are explored.

Plots of s versus $\bar{\kappa}$ obtained using FC for CH₄/air, covering both lean and rich mixtures, are different from those with the SS model. On further comparison with the existing experimental data, the results of FC are shown to

reproduce these data for all the cases considered here.

Calculations of H₂/air flames at 1 atm and H₂/O₂/N₂ at 3 atm with the FC model show that the plots of s versus $\bar{\kappa}$ with FC compare excellently with the existing numerical and experimental data for all cases. The present predictions for H₂/O₂/N₂ at 3 atm and O₂/(O₂ + N₂) = 0.125 also agree well the experimental data.

The atmospheric C₃H₈/air flames studied with FC show marked improvements over those of SS results for all cases. The present predictions match the experimental data for lean and stoichiometric mixtures but are not satisfactory for rich ones. The reason for the poor performance is attributed to the inadequacy of the FC scheme in not taking into account soot chemistry and associated radiation heat losses.

The effects of pressure, initial temperature,

and N_2 dilution are also investigated for the $H_2/O_2/N_2$ systems. These indicate that these parameters change even the trend of s versus $\bar{\kappa}$ for the cases considered.

Single-step chemistry, the principal approximation around which analytical studies (asymptotic analyses) are constructed, is not satisfactory in predicting stretch effects. The situation may be different with the reduced kinetic models currently under development in various laboratories. One recent study of wet CO/air flames by Wang and Rogg [27], where heat release profiles with reduced kinetics and full chemistry are compared, seems to show that reduced one-step models may be inadequate. A further point is that the heat release profiles of most hydrocarbons are such that their prediction from reduced single-step models is not simple. These are issues that need to be addressed in the future.

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