Twenty-Third Symposium (International) on Combustion/The Combustion Institute, 1990/pp. 433–440

ON THE FLAMMABILITY LIMIT AND HEAT LOSS IN FLAMES WITH DETAILED CHEMISTRY

K. N. LAKSHMISHA, P. J. PAUL AND H. S. MUKUNDA
Department of Aerospace Engineering
Indian Institute of Science
Bangalore 560012 INDIA

This paper reports a computational study of one-dimensional, planar, premixed, nonadiabatic flames near lean flammability limits, considering detailed chemistry and variable properties. Calculations made for CH_4/air mixtures of varying leanness between 4.5–6.0% of CH_4 show that (1) a steady propagation is obtained for *all* lean mixtures when heat loss is not considered, and (2) with the inclusion of a distributed heat loss a steady propagation is predicted above a certain CH_4 fraction; below this critical CH_4 fraction the solution decays, indicating nonflammability. The limit so determined varies from 5.18–5.6% of CH_4 with the magnitude of heat loss raised by a factor of 3. This small change in the limit for a large variation in heat loss factor is argued to indicate the fundamental nature of limit. The limiting fuel composition and flame speed found are nonzero and compare well with the results of zero-gravity experiments. The critical heat loss factor and flame speed at the limit obtained from the present work confirm the results of asymptotic theory. The principal effect of detailed chemistry is argued to be the correct reproduction of heat release rate vs. mixture ratio.

Introduction

The subject of flammability limits of gaseous mixtures occupies an important role in combustion theory. Whether the limits arise out of external factors like heat loss and stretch, or a *fundamental limit* based on adiabatic flame and detailed chemical kinetics can be obtained, has been debated in the literature. A summary position on the subject in recent times can be gleaned from an examination of the book by Williams.[1] While the possibility of chemical kinetics as a source of the limit is not ruled out, it is suggested that successful explanations usually invoke heat loss. That stretch could be an important factor for causing the limit in only upward propagating flames, has been a more recent contribution.[2] The fact that the more engineering combustion literature treats the limits as a function of mixtures alone without explicit dependence on any apparatus has also been debated by Williams.[1] He argues that the chemical kinetic behavior of the limit mixtures may be such that the overall heat release drops significantly over a narrow range of mixture ratios such that 'a very small heat loss may produce the same limit as a large loss.' These perceptive conjectures need, as yet, to be clearly demonstrated for their validity. The primary purpose of the present paper is to treat this issue. It appears to the present authors that, while a single-step reaction and simple heat loss model are adequate to provide an explanation for the existence of flammability limits,[3] it would be necessary to obtain accurate heat release rate variations with mixture ratio, particularly near the limits in order to examine the sensitivity of the limit to the magnitude of the heat loss. This cannot be accomplished within the framework of a single-step reaction. Hence calculations of the flame structure considering detailed kinetics and variable properties with heat loss are needed to be undertaken.

Methane-air system has been the subject of experimental flammability studies by a large number of workers (see Ref. 4 for a recent review). In order to determine the limit, (1) Flammability tubes of 25, 50, 75 mm diameters,[2,5,6] (2) Flat flame,[7] (3) porous cylinder,[8] and (4) opposed jet flame burner[9] have been used. The results from various studies have been summarized in Table 1. The results of Yamaoka and Tsuji show the lowest value of flammability limit. The effects of asymmetrical geometry pointed out by Ishizuka and Law,[9] coupled with the possible slight differences in the concentration of the two streams—one from the wall and the other from the freestream, account for the lowest limit. The result of Ishizuka and Law[9] is also close to this value. The authors in their paper[9] cite a table similar to Table 1 and conclude the comparison to be satisfactory. This does not seem appropriate to the present authors. The lower value of limit observed by Ishizuka and Law is due to the fact that the

TABLE I
Experimental results on lean limits of methane/air flames by various authors

No.	Reference	Method	% CH_4
1	Yamaoka & Tsuji[8]	Porous Burner	4.7
2	Ishizuka & Law[9]	Binary Flame	4.8
3	Egerton & Thabet[7]	Flat Flame	5.1
4	Zabetakis[5]	Flame Tube	5.0
5	Strehlow et al[2]	Flame Tube (zero-g)	5.25
6	Ronney[13]	Spherical (μ-g)	5.11
7	Andrews & Bradley[12]	Spherical Flame	4.5

downstream region of the flame in their apparatus is nearly adiabatic. The asymptotic theories[10,11] clearly show that the slow decay in the temperature downstream of the flame over a thickness of the order of activation parameter (E/RT_b) is responsible for the limit. In the above geometry,[9] the downstream region is nearly adiabatic as recognized already in Ref. 9, and hence one should expect a limit lower than that in other cases, for instance a flammability tube. The other experiments in flat flame configuration, flame tube and spherical flame show a limit between 5.0 and 5.3%. We ignore the suggestion by Andrews and Bradley[12] that the limit in their result is 4.5% with the criterion of zero pressure rise, because it is not clear what physical processes occur in this case (these do not seem to have been discussed adequately in their work). The two recent results of zero-g experiments which may be considered more appropriate, are by Ronney[13] for spherical flame and Strehlow et al[2] for flame tube, and they indicate 5.1% and 5.25% respectively.

The present authors in a previous contribution[14] showed that the adiabatic equations with full kinetics do not predict extinction, and a steady flame propagation is observed much beyond the known limits of flammability for CH_4—O_2—N_2 mixtures under N_2 dilution. After noting that this is demonstrated even in the present work on CH_4-air flames, it may be taken that the issue of *chemical kinetics alone as a possible explanation for the limits* has been resolved; kinetics alone cannot be an explanation. What heat losses do, needs exploration.

While Gerstein and Stine[15] studied the single-step kinetics problem with heat losses and showed some consistency of the results with observation, in a recent work, Sibulkin and Frendi[16] have treated the spherical flame problem with a single-step reaction and radiant heat loss model. The significant conclusion from this work is that above a critical mixture strength the flame propagates with heat loss and below this value, enhancement of ignition energy even by large amounts does not help propagation. The critical mixture strength is likened to flammability limit. The flame propagation problem with full chemistry and heat losses does not seem to have been treated till now. The present work treats the problem within the framework of one-dimensional flame and radiant heat loss model, much similar to Sibulkin and Frendi.[16]

The Equations and Solution Technique

The assumptions made and the basic differential equations solved in computation of one-dimensional planar adiabatic flames are now standard. Hence reference is made to the literature[17] for basic theoretical aspects which are not given here. In the present analysis, however, a heat loss term is added to the energy equation while the species equations remain unaltered. Consequently, the final equations take the form:

$$\textit{Species:}\ \frac{\partial Y_i}{\partial t} = -\frac{\partial J_i}{\partial \psi} + \frac{\dot{\omega}_i'''}{\rho}(i = 1,\ldots,N_s) \quad (1)$$

$$\textit{Energy:}\ \frac{\partial H}{\partial t} = \frac{\partial}{\partial \psi}\left(\frac{\lambda\rho}{C_p}\frac{\partial H}{\partial \psi}\right) - \sum_{i=1}^{N_s}\frac{\partial}{\partial \psi}\left[h_i\left(J_i + \frac{\lambda\rho}{C_p}\frac{\partial Y_i}{\partial \psi}\right)\right] - \frac{\dot{q}_L'''}{\rho} \quad (2)$$

where usual notation is followed. $\dot{q}_L'''$ is the volumetric rate of heat loss from the gases to the ambience. The corresponding boundary conditions are,

$$\psi \to -\infty:\ \frac{\partial Y_i}{\partial \psi} \to 0,\ \frac{\partial T}{\partial \psi} \to 0 \quad (3)$$

$$\psi \to +\infty:\ Y_i \to Y_{iu},\ T \to T_u \quad (4)$$

Modeling of Heat Loss Term:

The heat loss term $\dot{q}_L'''$ is modeled within the spirit of 1-d formulation. Two modes of heat transfer from the hot gases to the ambience are conduction and radiation. In the present analysis radiant heat loss alone is treated as the basic loss, because in most experiments this is unavoidable and we would examine if a flammability limit can be predicted based on such a loss alone. The radiation emission from nonoverlapping bands of the combustion products CO_2 and H_2O are considered, and the burning gases are assumed to be optically thin. This approximation simplifies the numerical solution of the source term nevertheless yielding a reasonably good estimate of the radiant flux. For a unit volume of gases, the heat radiated per unit time is:

$$\dot{q}_L''' = 4\bar{a}_p\sigma(T^4 - T_u^4) \qquad (5)$$

where $\bar{a}_p$ is the overall Planck mean absorption coefficient of the mixture. For nonoverlapping bands of CO_2 and H_2O, this is given by

$$\bar{a}_p = (a_p)_{CO_2}P_{CO_2} + (a_p)_{H_2O}P_{H_2O} \qquad (6)$$

where P_{CO_2} and P_{H_2O} are the partial pressures of CO_2 and H_2O respectively. The coefficients a_p for CO_2 and H_2O as functions of temperature are obtained from curve-fits for values of Hubbard and Tien.[18] In the present study a heat loss factor α has been used, which is defined as the ratio of actual heat loss included in the equations to the amount calculated from Eq.(5). For $\alpha = 0$, the equations reduce to the adiabatic case, and $\alpha = 1$ refers to the nonadiabatic case, where a minimum heat loss is considered. For $\alpha > 1$ we have the nonadiabatic case with excess heat loss.

Method of Solution:

The eqns (1–4) are numerically solved using an *operator-split technique* which has been shown to be robust and efficient, and has been extensively validated for H/O, C/H/O and H/N/O systems.[14,17,19] A detailed description of the numerical procedure for the adiabatic flame may be found elsewhere.[19] The same method has been used in the present calculations.

The transport properties were calculated from formulas given in Brokaw's report[20] and using the trace-diffusion model. While initial calculations were made with the kinetic scheme of Tsatsaronis,[21] most of the results presented here are based on the data of Warnatz[22] for the C_1-chain of hydrocarbon combustion. This scheme has 15 species and 40 elementary, reversible, reaction steps. The computations were initially validated for the adiabatic, stoichiometric case (9.5% CH_4). The stoichiometric flame speed predicted was 35.5 cm/s for the first scheme while it was 26 cm/s using the second scheme. This underprediction in flame speed from the second scheme was traced to the uncertainty in rate parameters of the reaction

$$CH_3 + H + M' \rightarrow CH_4 + M'$$

A careful study of the data review by Warnatz[22] shows that the kinetic parameters for the above reaction are far from being settled. Therefore, the data for this reaction from Seshadri and Peters[23] (k_f = 6.3×10^{14} $cm^6 \cdot mol^{-2} \cdot s^{-1}$ *units*; $b = 0$; $E = 435$ kJ/*mole*) was tried, which yielded predictions of flame speed over the entire lean side consistent with the known experimental results.

Computation of Nonadiabatic Flames; Criterion for Extinction

The addition of a heat loss term to the energy equation affects the downstream boundary condition for temperature. In a nonadiabatic flame, the temperature increases from the unburned state T_u to a maximum T_{max} (which is less than the corresponding adiabatic flame temperature T_b) before the hot gases are cooled down to the ambient temperature. In the present computations for a given mixture strength, first the adiabatic solution was obtained starting from *s*-shaped initial profiles and setting the heat loss factor $\alpha = 0$. Once the adiabatic flame attains steady-state, the heat loss was introduced by setting $\alpha = 1$, and time marching was continued therefrom. In our code, no special changes are required to be made for the downstream boundary condition, since sufficiently large number of grids are provided in the burned gas region, and the temperature and its gradient at the last grid are small enough to have any influence on the solution upstream. It was found that the computational time taken for the nonadiabatic flame to reach steady propagation was 8–10 times more compared to that for the adiabatic case. To reduce this computing time, a downward slope was given to the starting temperature profile between the point of peak heat release and the burned end, keeping the preflame region unaltered. It was found that the improvement was only marginal. The steady state was indicated by a constancy in the peak temperature and flame speed. The nonadiabatic flame was considered to be either propagating or extinguishing, depending on whether the flame speed $S_{u,na}$ and the peak temperature T_{max} reached a constant, or were found to decay continuously with time.

Results and Discussion

Figure 1 shows the plot of temperature profiles at various times for 5.18% CH_4 flame, with radiant

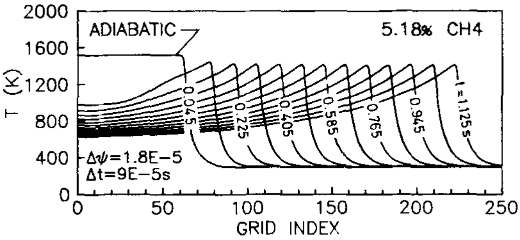

FIG. 1. Evolution of temperature profile for propagating flame with heat loss ($\alpha = 1$).

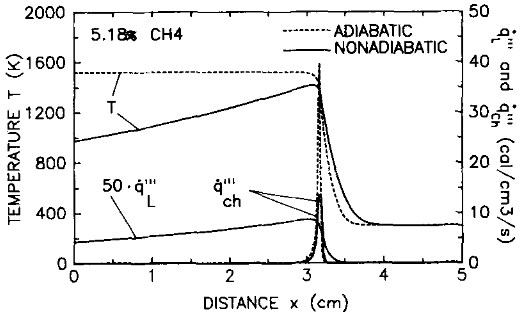

FIG. 3. Detailed thermal structure of a propagating limit-flame (5.18% CH_4).

heat loss. It can be noticed that the time required to establish the steady state is large (~0.5s) and more than 5000 computational steps are needed. In contrast, steady state for the adiabatic flame is obtained in less than 500 timesteps. This implies that the time taken to relax from adiabatic to nonadiabatic state is significant. It may be attributed to the fact that the burned gas region cools down at a small rate. Figure 2 shows the plot of temperature profiles for 5.13% CH_4 flame with heat loss. In this case, the drop in peak temperature is significant and the temperature profile shows no tendency to stabilize. It was observed that the flame is quenched when the peak temperature drops below less than 10% of the adiabatic value. Figure 3 shows the detailed thermal structure of the steadily propagating flame near extinction (5.18% CH_4). Heat loss broadens the flame, which is already thick (~8 mm, based on 1% to 99% of temperature profile) by 20%. The chemical heat release profile is substantially altered. The actual magnitude of the heat loss is not large (~5–10%) compared to the heat release. The calculations made for various compositions are summarized in Fig. 4 in terms of peak temperature vs. time. It is clearly seen that the steady state is reached for both 5.18% and 5.27% CH_4. Equally clearly seen is the continuous drop in the peak temperature for $CH_4 \leq 5.13\%$. These imply that the limit is between 5.13 and 5.18% CH_4. This value compares well the experimentally observed limit lying between 5.11 and 5.25% CH_4 (as indicated earlier in the introduction). The plots of flame speed, peak temperature and flame thickness are shown for both adiabatic and nonadiabatic cases as a function of mixture ratio. Ronney and Wachman[24] argue that the cause of the limit could not be heat loss, since there is no indication of S_u vs. composition curve to "bend towards the vertical" while approaching the limit. However, present calculations show that heat loss does not produce such a behavior (see Fig. 5). The predicted curve is nearly a straight line down to a methane concentration which is less than 0.05% away from the limit. Further, Ronney does not seem to continue to hold the same position with regard to the cause of the limit as seen in his later work,[13] where heat loss is considered as the principal cause. Figure 5 indicates that the limit speed is about 2 cm/s, temperature 1405 K and thickness 3.8 mm. These values compare well with limit values observed experimentally;[13] 1.7 cm/s for flame speed and 1373 K for the peak temperature. The variation of mean heat release with and without heat loss and the mean heat loss with mixture strength are shown in Fig. 6. The heat release rate with heat loss departs from the adiabatic case more and more as the limit is approached. The average heat loss calculated as

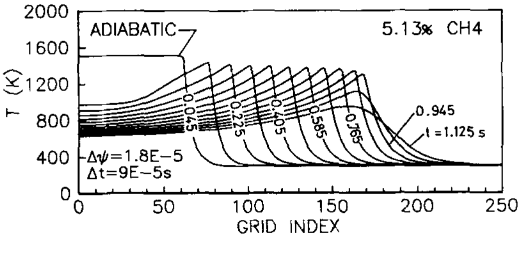

FIG. 2. Evolution of temperature profile for extinguishing flame with heat loss ($\alpha = 1$).

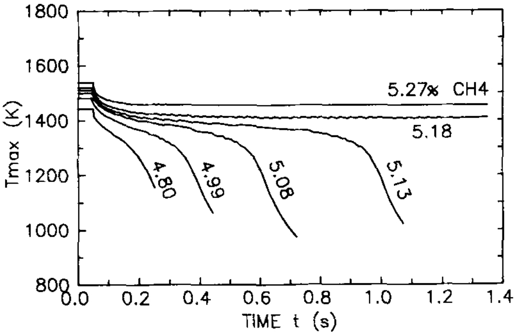

FIG. 4. Plot of peak temperature vs. time for different mixtures ($\alpha = 1$).

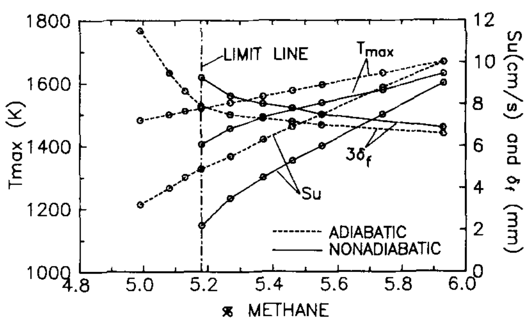

FIG. 5. Plots of steady-state flame temperature, flame speed and flame thickness.

$$\bar{q}_L = \frac{1}{(x_2 - x_1)} \int_{x_1}^{x_2} \dot{q}_L''' \cdot dx$$

where the limits x_1 and x_2 are chosen such that the entire flame zone is included (1% to 99% in temperature profile), does not vary much with composition. The fraction of the heat loss is about 5.5% of the heat release rate. This implies that a relatively small heat loss is able to extinguish the flame, a feature recognized in the asymptotic theories on flammability. Jarosinski[4] based on approximate calculations concludes that extinction is produced if the ratio of heat loss to heat release exceeds 8%, independent of all other conditions. Present calculations do show that this ratio varies from 5–10% for the various cases tested (higher mixture ratios with enhanced heat loss, by way of setting $\alpha > 1$). Therefore, a unique value of the ratio of heat loss to heat release at extinction does not appear to exist.

All the above results have been obtained with the basic radiant heat loss ($\alpha = 1$). It was intended to explore the effect of enhanced heat loss—due to possible convection to the walls—on the limits. In order to get an appreciation about the magnitude of convective heat loss, an estimate of the heat loss was made. For this purpose, it is assumed that a boundary layer grows from the leading edge of the propagating front, for the case with the burned-gas end open. The heat loss is obtained from the relation

$$\dot{q}_{conv}''' = \frac{4}{D(x_2 - x_1)} \int_{x_1}^{x_2} h(T - T_u) \cdot dx$$

where $(x_2 - x_1)$ is about 8 mm for 5.18% CH_4, and D is the tube diameter in cms. The above expression is the heat loss to the walls per unit volume of the flame. The heat transfer coefficient is obtained from the laminar case relation[25]

$$h = 0.332\lambda_g \sqrt{\frac{U_\infty}{\nu_g x}} \cdot Pr^{1/3}$$

where U_∞ is taken as S_b, and x is measured from the leading edge of the flame or the boundary layer. Using local properties for λ_g and ν_g as a function of x and allowing 20% enhancement due to flow acceleration along x, the loss is obtained as $\dot{q}_{conv}''' = 0.39/D$ with D in cms. The mean radiant heat loss is found to be nearly constant with mixture ratio, this being $0.14\ cal \cdot cm^{-3} \cdot s^{-1}$ at the limit. The total heat loss is therefore

$$\dot{q}_L''' = \dot{q}_{rad}''' + \dot{q}_{conv}''' \approx 0.14 + \frac{0.39}{D} (cal \cdot cm^{-3} \cdot s^{-1})$$

The above relation suggests that the convective loss accounts for 53%, 35% and 26% of the total heat loss for tube diameters of 2.5 cms, 5.0 cms and 7.5 cms respectively. One can estimate an equivalent heat loss factor α by including convective loss, for the purpose of examining the influence of variation in heat loss on the limit. From the above values, the equivalent α can be taken as 2.0, 1.6 and 1.4 for the three tube diameters mentioned. These calculations imply that the convective loss is small for tube diameters of 7.5 cms and above, as also qualitatively recognized by White.[6] Allowing for the uncertainties in the above calculations, the maximum variation in α can be by a factor of 3. In the present study, calculations were made at various mixture strengths to determine the critical value of α at which flame can be extinguished. The results are presented in Fig. 7. It can be seen that as the methane fraction is increased, the heat loss required to extinguish the flame is very large. Typically at 5.5% CH_4, the heat loss required is 3 times the radiant heat loss. At 6%, more than 5 times the radiant heat loss is required to extinguish the flame. In other words, near the limit composition, variation of heat loss by several factors of the basic loss

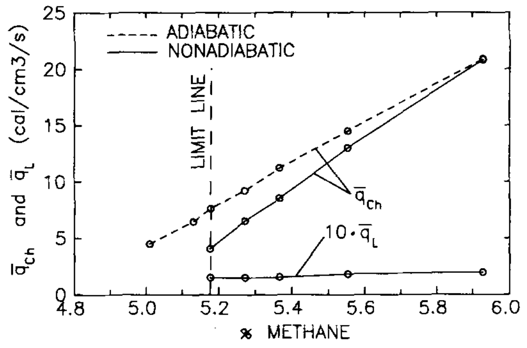

FIG. 6. Mean heat release and heat loss for different mixtures.

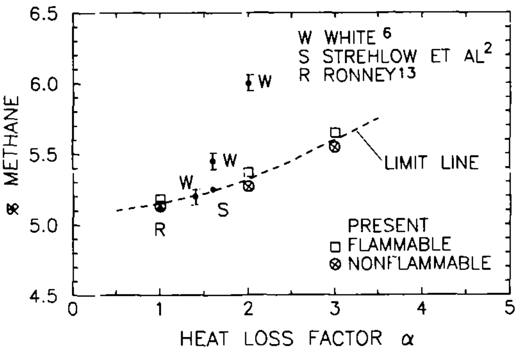

FIG. 7. Variation of Limit with heat loss factor.

results in only a relatively small change in the limit. This is precisely the conjecture made by Williams.[1]

The results of Ronney[13] corresponding to radiant heat loss ($\alpha = 1$) and Strehlow et al[2] for tube diameter of 5.0 cm ($\alpha = 1.6$) are shown in Fig. 7. It may be seen that the results compare very well with those from the present computations. The experimental results of White[6] for the limit observed in horizontal propagation for tube diameters of 2.5, 5.0 and 7.5 cms are also plotted to compare the results of enhanced heat loss on the limit, based on the heat loss estimates as indicated earlier. The values of equivalent α and the limit value are set out in Fig. 7. The experiments of White are with both tube ends closed. These gave limit as 6.2%, 5.65% and 5.4% of methane. It is expected that this limiting methane composition to be higher than the case with the burned end open because of effect due to pressure rise. An estimate of the pressure effect can be obtained from the work of Ronney and Wachman[24] as 0.2% CH_4 per 100 Torr. Taking that the pressure rise effects are accounted by reducing the limit by 0.2% uniformly (therefore a pressure rise of 100 Torr), the limits obtained are 6.0%, 5.45% and 5.2%. These are as shown in Fig. 7. Though the results of 7.5 and 5.0 cms tube data are close to the predicted, the trend of the results including that for 2.5 cms diameter indicate that factors other than simple heat loss (for example, fluid mechanics) may be dominant in these experiments. Statements to this effect are also noted by White.[6]

Finally, it was thought useful to compare the predictions of the present work with those of asymptotic theories. The asymptotic theories[10,11] use conductive heat loss model given by $\dot{q}'''_L = K(T - T_u)$. In order to obtain an equivalent K value, the heat loss in our calculations is equated to

$$(\dot{q}'''_L)_{peak} = K^*(T_{max} - T_u)$$

where T_{max} is the peak temperature in the nonadiabatic flame. The asymptotic theories yield[26] the maximum possible heat loss factor K^* at any mixture ratio for steady propagation as

$$P = \frac{K^*}{S^2_{u,ad}} = \frac{(\rho \cdot C_p)^2}{\lambda} \cdot \frac{e^{-1}}{2\beta}$$

In Table 2, the values of P obtained from the present work are compared with those from the above relation. The average activation energy for methane/air mixture of 29 kcal/mole leading to $\beta = 7.8$, has been used in this comparison. It can be seen that though the values of P differ by a factor (of nearly 1.5), the variation with mixture ratio is a constant to within 10%. The asymptotic theory[11] also predicts that the flame speed at extinction is $e^{-1/2}(\approx 0.61)$ times the adiabatic flame speed. In the present computations, it is obtained as 0.56. This reasonable comparison indicates that the asymptotic theories capture the physics of the flammability limit properly. This also implies that to predict the limit a relatively simple chemical model retaining the correct heat release profile for different mixture ratios as does the detailed kinetic model, may be satisfactory.

Conclusions

Summarizing, the present work shows that

1. The flammability limit can be predicted accurately based on radiant heat loss from the

TABLE II
Comparison of present results with Asymptotic Theory[26]

% CH_4	$S_{u,a}$(cm/s)	$P_{present}$	$P_{asymptotic}$	$\frac{K^*_{asymptotic}}{K^*_{present}}$
5.18	4.90	7.49×10^{-6}	$1.28 \times 10\ ^{5}$	1.646
5.37	6.27	8.35×10^{-6}	1.27×10^{-5}	1.506
5.55	8.00	$7.61 \times 10\ ^{6}$	1.21×10^{-5}	1.585

flame using a realistic flame model. Such a limit for methane/air compares well with the zero-gravity experimental results.

2. The limit varies slightly (from 5.18–5.6% of CH_4) for a fairly large heat loss variation (by a factor of 3). This feature conforms to the observation that closely matching limits are obtained from different apparatus.
3. The comparison of results with predictions from asymptotic theory seems fair. The primary function of full chemistry is to correctly replicate the average heat release profile for different mixture ratios.
4. A single value of ratio of heat loss to heat release at extinction is untenable. A small value of this ratio between 5–10% is observed for the methane/air mixtures.

Nomenclature

$\bar{a}_p$	Planck mean absorption coefficient
C_p	specific heat of the mixture, $\Sigma_{i=1}^{N_s} c_{p,i} Y_i$
D_i	mass diffusion coefficient
H	enthalpy of the mixture, $\Sigma_{i=1}^{N_s} h_i Y_i$
h	heat transfer coefficient
h_i	enthalpy, $h_i^0 + \int c_{p,i} dT$
h_i^0	heat of formation
J_i	diffusional mass flux, $-\rho D_i \frac{\partial Y_i}{\partial x} + Y_i \Sigma_{j=1}^{N_s} \rho D_j \frac{\partial Y_j}{\partial x}$
N_s	total number of species in the mixture
P_i	partial pressure
Pr	Prandtl number
$\dot{q}_L'''$	volumetric rate of heat loss
$\bar{q}_L$	mean heat loss rate
$\bar{q}_{ch}$	mean chemical heat release rate
$S_{u,ad}$	adiabatic flame speed
$S_{u,na}$	nonadiabatic flame speed
T	temperature
t	time
x	physical distance coordinate
Y_i	mass fraction
α	heat loss parameter
β	nondimensional activation energy
δ_f	flame thickness
λ	thermal conductivity of mixture
ν_g	mixture viscosity
ψ	transformed distance coordinate, $\int \rho dx$
ρ	mass density of mixture
σ	Stefan-Boltzmann constant, 1.355×10^{-12} $cal \cdot s^{-1} \cdot cm^{-2} \cdot K^{-4}$
$\dot{\omega}_i'''$	reaction rate

Subscripts

i	i-th species
u	unburned end condition
b	burned end condition

REFERENCES

1. WILLIAMS, F. A.: Combustion Theory, p. 271, Benjamin/Cummings, Calif. (1985).
2. STREHLOW, R. A., NOE, K. A., AND WHERLEY, B. L.: Twenty-First Symposium (International) on Combustion, p. 1899, The Combustion Institute, 1988.
3. SPALDING, D. B.: Proc. Roy. Soc. London A240, 83 (1957).
4. JAROSINSKI, J.: Prog. Energy Comb. Sci. 12, 81 (1986).
5. ZABETAKIS, M. G.: U.S. Bur. Min. Bulletin 627, 1965.
6. WHITE, A. G.: J. Chem. Soc., 125, 2387 (1924).
7. EGERTON, A. C. AND THABET, S. K.: Proc. Roy. Soc. London A211, 445 (1952).
8. YAMAOKA, I. AND TSUJI, H.: Seventeenth Symposium (International) on Combustion, p. 843, The Combustion Institute, 1979.
9. ISHIZUKA, S. AND LAW, C. K.: Nineteenth Symposium (International) on Combustion, p. 327, The Combustion Institute, 1983.
10. JOULIN, G., AND CLAVIN, P.: Acta Astronautica 3, 223 (1976).
11. BUCKMASTER, J.: Comb. & Flame 26, 151 (1976).
12. ANDREWS, G. E., AND BRADLEY, D.: Fourteenth Symposium (International) on Combustion, p. 1119, The Combustion Institute, 1973.
13. RONNEY, P. D.: Twenty-Second Symposium (International) on Combustion, p. 1615, The Combustion Institute, 1989.
14. LAKSHMISHA, K. N., PAUL, P. J., RAJAN, N. K. S., GOYAL, G., AND MUKUNDA, H. S.: Twenty-Second Symposium (International) on Combustion, p. 1573, The Combustion Institute, 1989.
15. GERSTEIN, M. AND STINE, W. B.: Fourteenth Symposium (International) on Combustion, p. 1109, The Combustion Institute, 1973.
16. SIBULKIN, M. AND FRENDI, A.: Submitted to Comb. & Flame (1989).
17. GOYAL, G., PAUL, P. J., MUKUNDA, H. S., AND DESHPANDE, S. M.: Comb. Sci. Tech. 60, 167 (1988).
18. HUBBARD, G. L. AND TIEN, C. L.: ASME J. Heat Transfer 100, 235 (1978).
19. GOYAL, G.: Ph.D Thesis, Indian Institute of Science, Bangalore, India (1989).
20. BROKAW, R. S.: Alignment charts for transport properties, viscosity and diffusion coefficients for nonpolar gases and gas mixtures at low density, NASA TR R-81, 1961.
21. TSATSARONIS, G.: Comb. & Flame 33, 217 (1978).
22. WARNATZ, J.: Rate Coefficients in the C/H/O

System, Combustion Chemistry, (W. C. Gardiner, Jr.: Ed.), p. 197, New York, Springer-Verlag, 1984.

23. SESHADRI, K. AND PETERS, N.: Inner Structure of Methane Flame, to appear in Comb. Sci. Tech., (1989).

24. RONNEY, P. D., AND WACHMAN, H. Y.: Comb & Flame 62, 107 (1985).

25. SCHLICHTING, H.: Boundary Layer Theory, 4th Ed, New York, McGraw-Hill, 1965.

26. JOULIN, G., AND CLAVIN, P.: Comb. & Flame 35, 139 (1979).