Laminar Flame Speeds of Hydrocarbon + Air Mixtures with Hydrogen Addition

G. YU

Department of Mechanical Engineering, Northwestern University, Evanston, Illinois 60201

C. K. LAW

Department of Mechanical Engineering, University of California, Davis, California 95616

and

C. K. WU

Institute of Mechanics, Chinese Academy of Sciences, Beijing, China

Using the symmetrical, adiabatic, counterflow arrangement, the laminar flame speeds of methane + air and propane + air mixtures, with and without the addition of stoichiometrically small amounts of hydrogen, have been determined by first measuring the flame speeds with stretch and then linearly extrapolating these values to zero stretch. The results show that the flame speed is substantially increased with hydrogen addition, and that it can be linearly correlated with the flame speed without hydrogen addition and a single parameter indicating the extent of hydrogen addition.

I. INTRODUCTION

The burning of lean hydrocarbon + air mixtures offers the potential of enhanced fuel economy, reduced pollutant formation, and improved thermal stress characteristics of the combustor hardware. However, lean mixtures are hard to ignite and more susceptible to extinguish. Thus their utilization imposes more stringent criteria on the operational reliability of the combustor.

A promising approach to enhance the combustion intensity of these lean mixtures is through the addition of hydrogen which has higher burning intensity. Indeed, because of the importance of the hydrogen radical in the overall hydrocarbon + air reaction scheme, it is reasonable to expect that the addition of hydrogen in the freestream should significantly increase the concentration of the radicals, which otherwise would have to be supplied through back diffusion from the active reaction region where they are generated.

An important parameter needed for the utilization of hydrocarbon + hydrogen + air

mixtures and the concurrent understanding of the underlying combustion mechanism is their laminar flame speed $S_L°$, which represents the reactivity and exothermicity of a given mixture. Furthermore, since $S_L°$ also contains the fundamental information regarding the reactive and diffusive properties of the mixture, the availability of an accurately determined $S_L°$ function also offers the potential for the extraction of the kinetic information by comparing the experimentally measured and numerically simulated values of $S_L°$.

Recently Milton and Keck [1] reported laminar flame speeds of hydrocarbon + hydrogen + air mixtures, as functions of the mixture temperature and pressure, by using the constant volume bomb technique. Because of the multi-parameter nature of the investigation, the data reported are limited to stoichiometric concentrations.

The main thrust of the present investigation is to provide accurately determined values of $S_L°$ over the complete concentration ranges of hydrocarbon and air, with a small amount of hydrogen addition. The novel feature here is the specific methodology adopted which allows the determination of the laminar flame speed without effects of heat loss and aerodynamic stretching, and thereby conforms most closely with the definition of $S_L°$. Our data also yield a simple empirical expression for the mixture laminar flame speed which is expected to be of practical utility.

In the next section we discuss the concepts of the experimental methodology and the specific arrangement adopted herein. This is then followed by the presentation and discussion of the experimental data in Section III.

II. EXPERIMENTAL METHODOLOGY

In Ref. [2] it is pointed out that most of the existing methodologies for the determination of $S_L°$ involve flames which suffer aerodynamic stretching. Examples are the positive stretch experienced by the outwardly expanding flame in a constant volume bomb, and the negative stretch experienced by the widely used Bunsen flame because of the flame curvature. Since the
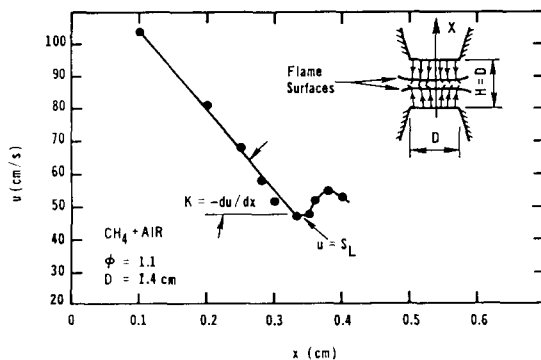

Fig. 1. Typical velocity profile across a stagnation flame, showing the definitions of K and S_L.

laminar flame speed $S_L°$ is defined as the propagation velocity of an adiabatic, one-dimensional planar flame, the flame speeds determined from such stretched flames obviously are not $S_L°$.

In response to this concern, Wu and Law [2] proposed a methodology in which stretch effects can be systematically subtracted out to yield an $S_L°$ which conforms closely to its definition. The methodology involves establishing a planar flame in the divergent stagnation flow produced by impinging a uniform stream of combustible onto a stagnation surface. The velocity profile of this stagnation premixed flame configuration can be determined by using, say, Laser Doppler Velocimetry. Figure 1 shows the axial velocity profile of such a flame, which can be basically interpreted as the superposition of those of a nonreacting stagnation flow and a planar, one-dimensional flame. Thus by approaching from the freestream we initially have the velocity profile of the stagnation flow characterized by the negative of its velocity gradient, K. As the upstream boundary of the preheat zone is approached, the velocity attains a minimum and then increases due to thermal expansion. Eventually, upon complete heat release, the velocity again decreases in accordance with the stagnation flow requirement.

The stretch experienced by the flame is therefore simply K, expressed in the unit of s^{-1}. Furthermore, if we define the flame speed S_L of this stretched flame to be the propagation veloc-
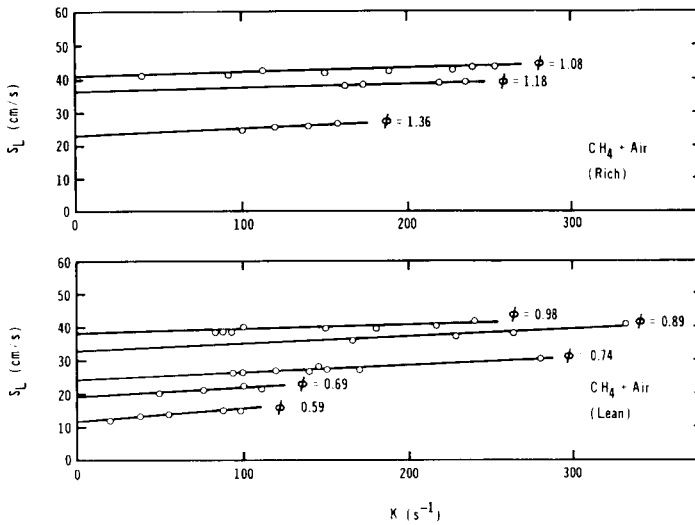
Fig. 2. Demonstration of the linear variation of the flame speed S_L with the stretch rate K, for methane + air mixtures.

ity of the upstream boundary of the preheat zone, then S_L can be identified as the minimum value of the velocity profile. Thus S_L as a function of K, for a given fuel/air equivalence ratio ϕ, can be determined by systematically varying the freestream velocities. For small values of K, both theory [3–5] and experiment [2] have shown that S_L varies linearly with K, as demonstrated in Fig. 2. Thus by linearly extrapolating $S_L(\phi; K)$ to $K = 0$, we obtain $S_L(\phi; 0) = S_L^\circ(\phi)$, which is the laminar flame speed by definition.

This technique is further improved in the present investigation by using a symmetrical counterflow instead of the plate-generated stagnation flow. This eliminates any possibility of downstream heat loss. Although effects on flame speed due to heat loss to the stagnation plate is generally very small unless the flame is close to the plate, by using the symmetrical counterflow we are assured of downstream adiabaticity under all situations. It may also be noted that since the present uniform flow is generated by nozzles instead of porous plates, upstream heat loss is also minimized. Indeed, our results show that the flame speed depends only on the local stretch rate instead of the

absolute nozzle separation distance, as long as the stagnation flow characteristics are preserved for such separation distances.

Figure 3 shows a schematic of the experimental setup. The symmetrical counterflow is generated by two identical aerodynamically shaped nozzles with diameters 10, 14, 20, and 30 mm. The nozzle separation distance is about one diameter. Each combustible stream is also surrounded by an external shroud flow of N_2. By varying its flow velocity, this shroud flow is found to be especially important in stabilizing flame disturbances for rich mixtures.

The flow velocity is measured by a TSI argon-ion LDV system in the back-scattering mode. The measuring volume is 0.1 mm diam. × 0.9 mm length, while seeding is accomplished by 1μm MgO particles.

Last, it is also important to emphasize the versatility of the present technique. That is, because our method is based on local measurements, the quality of the bulk flame, as well as the flow, is not crucial in that as long as the upstream velocity variation is linear for a reasonable distance ahead of the preheat zone, and the characteristic dimension of the flame curvature is large compared with that of the preheat
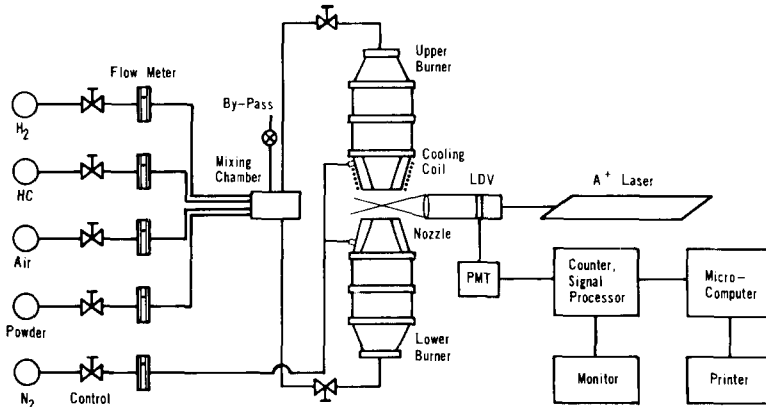
Fig. 3. Schematic of the experimental setup.

zone, then S_L as a function of K can be determined with good accuracy. This point is especially relevant for rich propane + air flames which tend to develop flame-front instabilities over certain concentration and stretch regions [6]. Since for most situations such instabilities appear only sporadically and infrequently, in the form of normally propagating straight ridges over the flame surface, the flame is smooth most of the time and its speed can be determined during these disturbance-free periods. Note also that these instabilities can be minimized/suppressed with increasing stretch [6, 7], which is a parameter that can be independently varied in the experiment.

III. RESULTS AND DISCUSSIONS

Validity of Linear Extrapolation

An important feature of the present methodology is that the flame speed S_L scales linearly with the stretch rate K for small values of K. In the present investigation, we have further confirmed this property. Figure 2 shows typical variations of S_L with K for methane + air mixtures. It is seen that the variation is indeed linear within the accuracy of the experimentation. Furthermore, since the variation is quite gentle and is spread out over a wide range of K, the viability of the linear behavior is further enhanced.

The final point to check is whether K is indeed

small. Theoretical development [3–5] has shown that the extent of correction in the flame speed (normalized to unity) due to stretch is proportional to a nondimensional stretch rate $\tilde{K} = DK/(S_L°)^2$, where D is the gas-phase diffusivity. Thus taking $D = 10°$ cm^2/s, $K = 200$ s^{-1}, and $S_L° = 40$ cm/s, we have $\tilde{K} \simeq 0.1$, implying that the stretch rate adopted in the experimentation is indeed small enough for the purpose of linear extrapolation. In fact, some of our data show that linearity holds even when $\tilde{K} = O(1)$. Since the theories are approximate ones, embodying such assumptions as incompressibility, it is conceivable that the property of linearity is valid over a more extended range of stretch rate than recognized.

$S_L°$ of Methane + Air and Propane + Air Mixtures

As references, we present $S_L°(\phi)$ for methane + air and propane + air mixtures, shown in Figs. 4 and 5, respectively. We first note that the present data for propane + air covers a substantially extended range in ϕ on the rich side. As expected, these data level off with increasing ϕ because of the asymmetrical nature of the definition of ϕ relative to the concentrations of fuel and air.

We next compare our data with those of the recently proposed technique of Yamaoka and Tsuji [8]. Specifically, these authors established
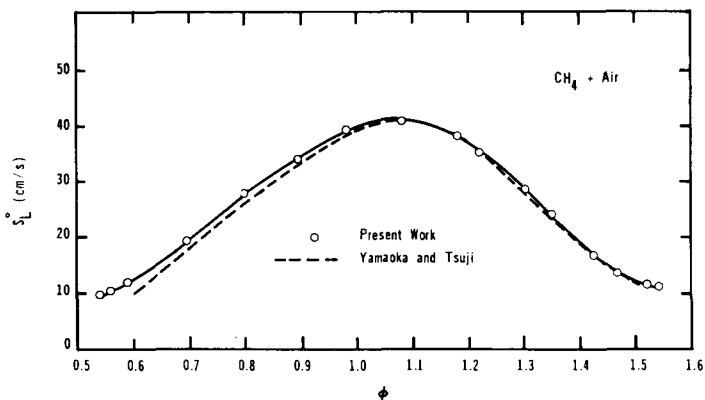
Fig. 4. Laminar flame speed, $S_L°$, of methane + air mixtures, and comparison with the data of Yamaoka and Tsuji.
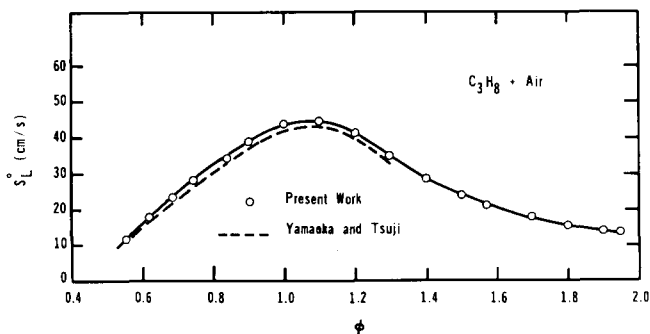

Fig. 5. Laminar flame speed, $S_L°$, of propane + air mixtures, and comparison with the data of Yamaoka and Tsuji.

a binary premixed flame configuration by eject-ing a combustible mixture from a porous cylin-der into the countercurrent flow of another mixture which may or may not have the same stoichiometry. By gradually reducing the ejec-tion velocity from the cylinder, the inner flame can be brought close to the cylinder until there is sufficient heat loss to it that the bulk flame characteristics, such as the flame location, change drastically. The ejection velocity at which such drastic changes occur, which essen-tially signifies the arrival of the upstream boundary of the preheat zone at the cylinder surface, is identified in Ref. [8] as the laminar flame speed.

Figures 4 and 5 show that our data are close to those of Ref. [8]. The data of Ref. [8] exhibit slightly smaller values, possibly because of the unavoidable small amount of heat and radical loss to the cylinder surface. The close agree-ment does support the viability of these two entirely different methods in determining the laminar flame speed without complications due to stretch and heat loss.

If we now accept the validity of the methodol-ogy and data of Ref. [8], then their experimental observation suggests the following important point. That is, in the study of premixed flame propagation, the importance of radical back diffusion relative to heat conduction is fre-quently raised. This concern is well justified because of the role of the radicals as chain

carriers and because the important radical, H, is highly diffusive and should have a much longer range of influence than the preheat zone thickness. If this is the case, then we would expect the radical diffusion zone to impact the cylinder surface before the preheat zone, thereby leading to a lowering of the flame speed but not necessarily the flame temperature. The results of Yamaoka and Tsuji, however, show that drastic changes in the flame speed and the temperature occur simultaneously. This therefore implies that the *effective* radical diffusion zone is at most as thick as the preheat zone.

$S_L°$ of Hydrocarbon + Hydrogen + Air Flames

Because of the presence of three reactants in the mixture, it is necessary to first decide on the stoichiometric parameters for meaningful data reduction. To do so, we first note that since H_2 is present only in small quantities, there should be enough air to facilitate its complete oxidation. Thus if the mole fractions of the hydrocarbon fuel, hydrogen, and air are, respectively, C_F, C_H, and C_A, with $C_F + C_H + C_A = 1$, then in order to oxidize totally C_H amount of H_2, we will need $C_H/(C_H/C_A)_{st}$ amount of air, where $(C_H/C_A)_{st} = 0.418$ is the stoichiometric hydrogen-to-air molar ratio. If the remaining air is used to oxidize the hydrocarbon, we can then define an effective fuel/air equivalence ratio ϕ_F as

$$\phi_F = \frac{C_F/[C_A - C_H/(C_H/C_A)_{st}]}{(C_F/C_A)_{st}} ,$$

where $(C_F/C_A)_{st}$ is the stoichiometric fuel-to-air molar ratio, which is 0.105 and 0.0418 for methane + air and propane + air mixtures, respectively.

To indicate the relative amount of hydrogen addition, we form the ratio

$$R_H = \frac{C_H + C_H/(C_H/C_A)_{st}}{C_F + [C_A - C_H/(C_H/C_A)_{st}]} .$$

The numerator of R_H is the amount of hydrogen plus the amount of air needed to oxidizer it totally, while the denominator is the amount of

hydrocarbon fuel plus the amount of air available for its oxidation.

Thus ϕ_F and R_H are the two parameters to be used to indicate the concentrations of hydrocarbon and hydrogen. It is important to emphasize that these two parameters do not represent the actual stoichiometry during the reaction; for example, the hydrocarbon fuel obviously has access to the total amount of air present. However, as will be demonstrated subsequently, these two intuitively based parameters do facilitate data reduction and correlation.

Figures 6 and 7 show the measured $S_L°$ as a function of ϕ_F, with R_H as a parameter, for methane- and propane-based mixtures, respectively. For $R_H = 0$, we have the reference hydrocarbon + air case. With hydrogen addition and therefore increasing R_H, $S_L°$ increases, as is reasonable to expect. For different R_H, the variation of $S_L°$ with ϕ_F remains qualitatively the same, with the maxima occurring around a narrow range of ϕ_F.

Figures 8–11 show an alternate plot of $S_L°$, with R_H being the independent variable and ϕ_F the parameter. The advantage of such a plot is obvious in that $S_L°$ is seen to vary linearly with R_H over the range of investigation. Furthermore, the slopes of these curves also do not vary too much from each other.

The upper limit in R_H in our investigation is constrained by the propensity of the flame to flashback. However, for the leanest propane case ($\phi_F = 0.5$), with great caution we were able to extend the investigation to $R_H = 1.0$. Figure 12 shows that the linear behavior is sustained.

Based on the behavior of Figs. 8–12, it is then logical to correlate $S_L°$ according to

$$S_L°(\phi_F, R_H) = S_L°(\phi_F, 0) + \alpha_F(\phi_F)R_H, \qquad (1)$$

where $S_L°(\phi_F, 0) = S_L°(\phi = \phi_F)$ is the laminar flame speed without hydrogen addition, as given by Figs. 4 and 5, and $\alpha_F(\phi_F)$ describes the variation of the linear slopes α_F with ϕ_F. For the methane case there is almost no variation in α_F such that

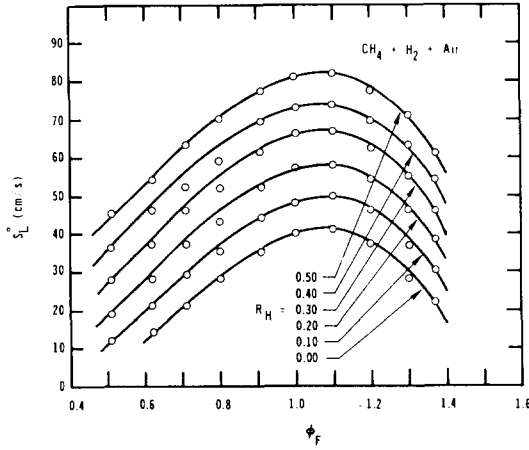$$\alpha_{methane} \simeq 80 \text{ cm/s}.$$

Fig. 6. Laminar flame speed, $S_L°$, of methane + hydrogen + air mixtures as a function of ϕ_F, with R_H as parameter.
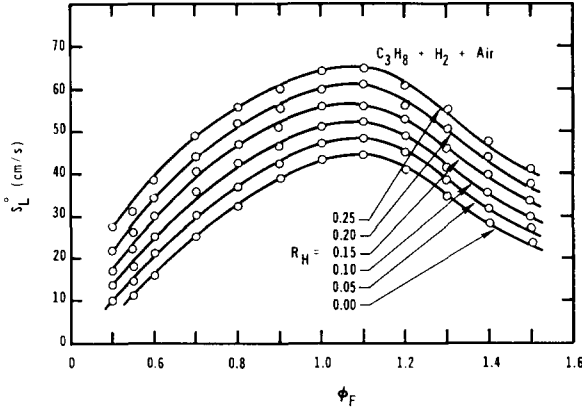

Fig. 7. Laminar flame speed, $S_L°$, of propane + hydrogen + air mixtures as a function of ϕ_F, with R_H as parameter.

For the propane case $\alpha_F \simeq 90$ cm/s for $\phi_F \leq 1$ and $\alpha_F \simeq 82$ cm/s for $\phi_F \geq 1.1$. Since these variations are still quite small, for the sake of simplicity we can set

$$\alpha_{propane} \simeq 86 \text{ cm/s.}$$

In fact, very little error would be incurred even if we write in general

$$\alpha_F = 83 \text{ cm/s}$$

for methane- and propane-based mixtures. Thus Eq. (1) becomes

$$S_L°(\phi, \ R_H) \simeq S_L°(\phi = \phi_F) + 83R_H \quad \text{(cm/s),} \quad (2)$$

such that the effect of hydrogen addition can be represented by the single parameter R_H. Equations (1) or (2) is expected to be of general practical utility. It may also be of interest to explore whether these relations also hold for other hydrocarbon + air mixtures with hydrogen addition.

One possible explanation for the linear behavior is the stoichiometrically small amount of hydrogen present relative to the total amount of air. For example, even for the $R_H = 1.0$, $\phi_F =$
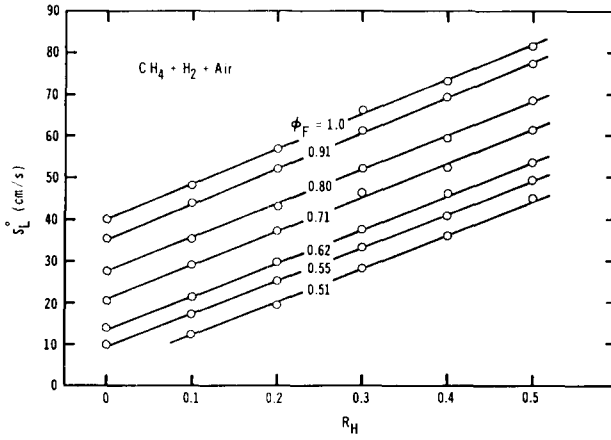
Fig. 8. Laminar flame speed, $S_L{}^\circ$, of methane + hydrogen + air mixtures as a function of R_H, with ϕ_F as parameter, for lean values of ϕ_F.
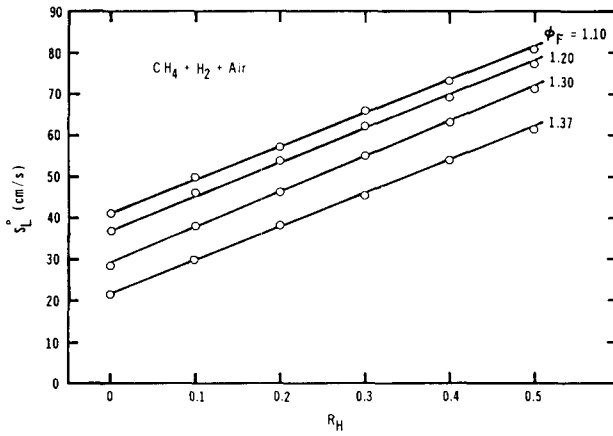

Fig. 9. Laminar flame speed, $S_L{}^\circ$, of methane + hydrogen + air mixtures as a function of R_H, with ϕ_F as parameter, for rich values of ϕ_F.
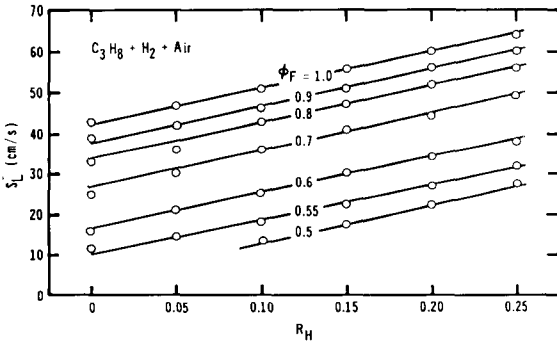

Fig. 10. Laminar flame speed, $S_L{}^\circ$, of propane + hydrogen + air mixtures as a function of R_H, with ϕ_F as parameter, for lean values of ϕ_F.
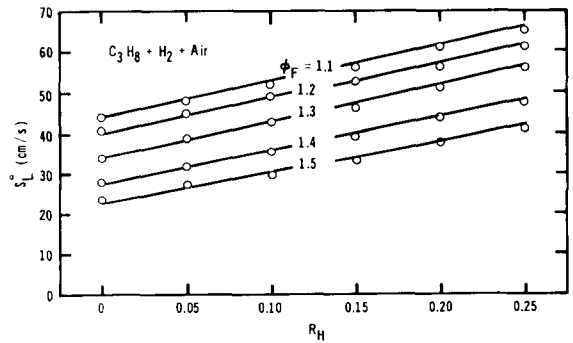

Fig. 11. Laminar flame speed, $S_L{}^\circ$, of propane + hydrogen + air mixtures as a function of R_H, with ϕ_F as parameter, for rich values of ϕ_F.
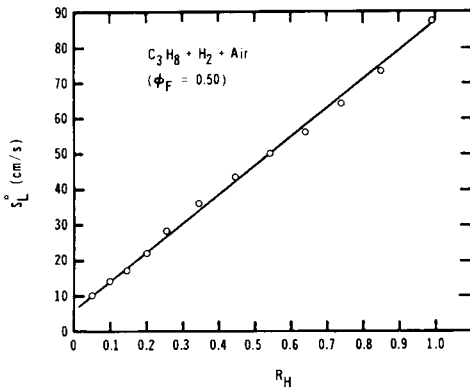
Fig. 12. Laminar flame speed, $S_L°$, of propane + hydrogen + air mixtures as a function of R_H, with $\phi_F = 0.5$, demonstrating the extended range of R_H in which the linear behavior holds.

0.5 propane case, the equivalence ratio of hydrogen relative to total air is only 0.63. Noting that since the maximum flame speed of hydrogen + air mixtures is at $\phi = 1.8$, the presence of hydrogen in the present experiments is basically a perturbation to the hydrocarbon + air stoichiometry, thereby resulting in a linear correction factor. It must, however, also be emphasized that although the amount of hydrogen addition is small on the basis of stoichiometry, it is quite significant in terms of the volumetric fraction.

A direct comparison of our measured flame speeds with those of Milton and Keck [1] is not possible because our data were obtained at overall off-stoichiometric conditions while theirs were determined at stoichiometric concentrations. However, by using the linear relation of Eq. (1), the extrapolated values of $S_L°$ for $\phi_F = 1$ and $R_H = 1$, which correspond to the stoichiometric situation, are 120 cm/s and 129 cm/s for methane- and propane-based mixtures, respectively. These compare favorably with the corresponding atmospheric values of 110 cm/s and 120 cm/s as repored in Table 4 of Milton and Keck [1].

IV. CONCLUSIONS

The specific contributions of the present investigation are the following. First, we have reconfirmed the methodology of Wu and Law [2] for the determination of the laminar flame speed $S_L°$ by performing the experiment over extensive ranges of parameters and without downstream heat loss. Through such an effort we have accurately determined $S_L°$ for methane + air and propane + air mixtures over an extended range of ϕ. We have also shown that these values agree well with those of Yamaoka and Tsuji [8], thereby providing support for the viability of both methodologies in which stretch and heat loss effects are either eliminated or minimized.

Having thus validated our methodology, the laminar flame speeds of methane + air and propane + air mixtures with the addition of stoichiometrically small amounts of hydrogen are determined, and are found to be linearly correlatable with the hydrogen concentration parameter R_H. Such a relation is expected to be useful from practical considerations.

REFERENCES

1. Milton, B. E., and Keck, J. C., *Combust. Flame* 58:13–22 (1984).
2. Wu, C. K., and Law, C. K., to appear in *Proc. of Twentieth Combustion Symposium*, 1985.
3. Matalon, M., and Matkowsky, B. J., *J. Fluid Mech.* 124:239–249 (1982).
4. Chung, S. H., *Trans. Korean Society of Mech. Eng.* 9:250–258 (1985).
5. Law, C. K., Paper presented at the Eastern Section Meeting of the Combustion Institute, Clearwater Beach, Florida, 1984.
6. Ishizuka, S., Miyasaka, K., and Law, C. K., *Combust. Flame* 45:293–308 (1982).
7. Sivashinsky, G. I., Law, C. K., and Joulin, G., *Combust. Sci. Tech.* 28:155–159 (1982).
8. Yamaoka, I., and Tsuji, H., to appear in *Proc. of Twentieth Combustion Symposium*, 1985.