

THE USE OF EXPANDING SPHERICAL FLAMES TO DETERMINE BURNING VELOCITIES AND STRETCH EFFECTS IN HYDROGEN/AIR MIXTURES

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A new technique is presented for determining burning velocities and stretch effects in laminar flames, and applied to hydrogen/air mixtures. The speeds of expanding spherical flames, measured by high-speed schlieren cine-photography, are shown to vary with flame radius. A simple phenomenological model has been developed to analyse the data and obtain the 1-D flame speed by extrapolation to infinite radius.

A computer program has also been developed for detailed simulation of expanding flames. The validity of the simple model has been tested by using it to analyse the results of the detailed simulations. The true 1-D flame speeds in this case are known from planar flame modelling using the same kinetic scheme. The simple model predicted flame speeds within 2% of the true values over most of the stoichiometric range. This demonstrates that our extrapolation procedure is sound and will produce reliable results when applied to experimental data.

Since the flame speeds derived from experiments are 1-D values, multiplying them by the density ratio gives 1-D burning velocities (S_u^0). Burning velocities are reported for mixtures containing 9 to 68% hydrogen. The maximum S_u^0 is 2.85 ms^{-1} , considerably less than most burner-derived values. The discrepancies can be explained by flow divergence and stretch effects perturbing burner measurements.

Planar flame modelling reproduces the experimental burning velocities to within 3% over most of the stoichiometric range.

The rate at which the measured flame speed approaches its limiting value depends on flame thickness and flame stretch. By subtracting the flame thickness term, Markstein lengths can be derived. Again values are given across the whole stoichiometric range. They are negative in lean mixtures (i.e. stretch increases the burning rate) but positive in stoichiometric and rich (stretch reducing burning rate). This is in line with predictions based on Lewis number considerations.

Introduction

It is now well established¹⁻⁵ that stretch effects can exert a significant influence on flame propagation rates, but few attempts have so far been made to quantify them. A related issue is the reliability of much of the current information on burning velocities. With the new insights into stretch and divergence effects comes the realisation that many measurements of burning velocity are flawed.^{1,6} Flow

divergence by itself may not modify the burning rate but does mean that burning velocity measurements must be referred to some plane or surface in the flame to remove ambiguity. The choice of reference then matters. In addition, the presence of flow divergence or flame motion induces stretch which may well perturb the burning rate. New methods for determining burning velocities have appeared in recent years with claims to circumvent such difficulties.^{1,7} We propose another.

Much of the previous work on stretch has employed flames in stagnation flows. This geometry is popular because stretch in stagnation flow is, at least in principle, well characterised and easy to quantify. But we have argued elsewhere⁸ that the presence of the flame strongly perturbs the flow pattern and may lead to incorrect burning velocities. Moreover, Kee et al⁹ have demonstrated that flow patterns produced using experimental arrangements with nozzle burners positioned fairly close to the stagnation surface are significantly different from the theoretical ideal. Thus it seems that the stagnation flow configuration is not ideally suited for such studies.

Expanding spherical flames represent another case where stretch is well characterised and easily determined. Because their geometry is so simple, they escape the type of complications mentioned above. A number of theoretical analyses of spherical flames incorporating stretch have been presented,¹⁰⁻¹² though these have all assumed very simple, one-step, chemistry. Expanding flames have also been modelled using more realistic chemistry.¹³⁻¹⁶ While these studies provide useful insights, few quantitative conclusions may be drawn from them. There are no previous reports of expanding flames being used for the experimental study of stretch effects.

Expanding flames at both constant pressure and constant volume have been used for many years to measure burning velocity. There have been many refinements to the techniques and considerations of complicating factors, particularly for constant volume work.¹⁷ But one notable feature of these analyses is that none has included considerations of stretch. Yet the fact that expanding flames experience stretch has been apparent since Palm-Leis and Strehlow reported¹⁸ that the flame speeds of lean propane/air flames increased with flame radius, while for rich flames they fell.

Despite their relative neglect in recent years, expanding spherical flames at constant pressure, because of their simplicity, offer considerable advantages for the experimenter. In this paper, we examine a novel way of using them to measure burning velocities and stretch effects. We present a new analysis of expanding flames and establish its validity by comparing it with results of detailed computer modelling. The new analysis of the flame radius vs time records is easy to apply and capable of yielding reliable results. We demonstrate its application to hydrogen/air flames, presenting data on burning velocities and Markstein lengths. Markstein lengths are increasingly being used as a measure of the effect of stretch on burning velocity.

Analysis Method

We now derive a simple expression for the radius as a function of time of an expanding spherical

flame at constant pressure. Two effects, flame stretch and flame thickness, are included which prevent the relationship from being a simple linear one. We stress that our analysis is not intended to be a theory of spherical flame propagation; it is rather an attempt to explain phenomenologically the influence of low stretch rates on laminar flame propagation. It is a simplified approach in that we assume that the flame structure is essentially that of a one-dimensional (1-D) planar flame. Our analysis is therefore expected to be valid at large radii, but not at very small radii where the quasi 1-D assumptions will not hold. Later in the paper we demonstrate its range of validity.

Stretch in expanding spherical flames arises from change of curvature with time. The general definition of stretch¹⁹

$$\Gamma = (1/A) dA/dt, \quad (1)$$

when applied to expanding spherical flames, becomes

$$\Gamma = (2/r) dr/dt \quad (2)$$

where A is the area of the flame, r is its radius and t is time. Asymptotic analyses^{11,20} and detailed modelling^{21,22} show a linear relationship between stretch and burning velocity in the low stretch regime, and we assume

$$S_{u,r} = S_u^0 - L\Gamma. \quad (3)$$

Here $S_{u,r}$ is the burning velocity referred to the reference surface, S_u^0 is the 1-D burning velocity we wish to determine and L is the Markstein length. Theory¹¹ suggests that L is independent of whether stretch is manifested as strain or curvature.

The concept of a reference surface is needed to remove ambiguity when applying the conventional (1-D) definition of burning velocity to flames in divergent flows.^{1,6,8,23} We locate the reference surface by comparing a 1-D planar with a stationary spherical flame which, although in a divergent flow, is unstretched.¹¹ Its position is where the varying mass flux in the spherical flame equals the constant value of the 1-D planar flame. $S_{u,r}$ is the mass flux at this surface divided by the cold gas density ρ_u . Any change in the position of the reference surface as the flame is stretched is likely to be too small to affect the experimental results.

There is still confusion over the correct location of the reference surface, with many workers continuing to use the cold boundary. We show later that for hydrogen/air flames our reference surface is within the reaction zone, in agreement with the general conclusions of Fristrom²³ and Dixon-Lewis and Islam.⁶

For an expanding spherical flame, the mass of the gas inside the reference surface at radius r is

$$m = (4/3)\pi r^3 \bar{\rho}_b$$

where $\bar{\rho}_b$ is the mean burnt gas density. The mass flux crossing this surface is

$$\begin{aligned} M &= (dm/dt)/(4\pi r^2) \\ &= [\bar{\rho}_b + (r/3) d\bar{\rho}_b/dr] dr/dt \\ &= \rho_b^0 f(r) dr/dt \end{aligned} \quad (4)$$

where ρ_b^0 is the 1-D burnt gas density and

$$f(r) = [\bar{\rho}_b + (r/3) d\bar{\rho}_b/dr]/\rho_b^0 \quad (5)$$

is a density correction function. Following Fristrom,²³ we assume from geometrical considerations that this has the form

$$f(r) = 1 + k/r \quad (6)$$

where k is a constant which we determine by modelling. In fact, k varies slightly with radius and we use a mean value determined over the range $r = 5$ to 40 mm. The standard deviation of k over this range is about 5%.

The mass flux at the reference surface differs from its 1-D value because of stretch. Combining Eqs. (2) and (3) and incorporating the conservation of mass, we have

$$M = \rho_b^0 S_b^0 - (2 \rho_u L/r) dr/dt \quad (7)$$

where S_b^0 is the 1-D burnt gas value of flame speed. Equating (4) and (7) and rearranging gives

$$dr/dt = S_b^0/(f(r) + \ell/r) \quad (8)$$

where $\ell = 2L\rho_u/\rho_b^0$. Substituting (6) in (8) gives

$$dr/dt = S_b^0/(1 + b/r) \quad (9)$$

where

$$b = k + \ell.$$

We see later that negative values of b are obtained in lean hydrogen/air mixtures. Equation (9) breaks down when $r \leq -b$ so the fitting procedure is limited to larger radii for these mixtures.

Integrating Eq. (9) gives the final equation

$$r + b \ln r = S_b^0 t + \text{constant}. \quad (10)$$

The experimentally visualised flame radius will normally be different from the reference surface radius of this analysis. But the difference is a second order effect and will be negligible when $r \gg |b|$. We show later (Fig. 5) that over most of the stoichiometric range $|b|$ is less than 2 mm.

A common practice in previous studies has been

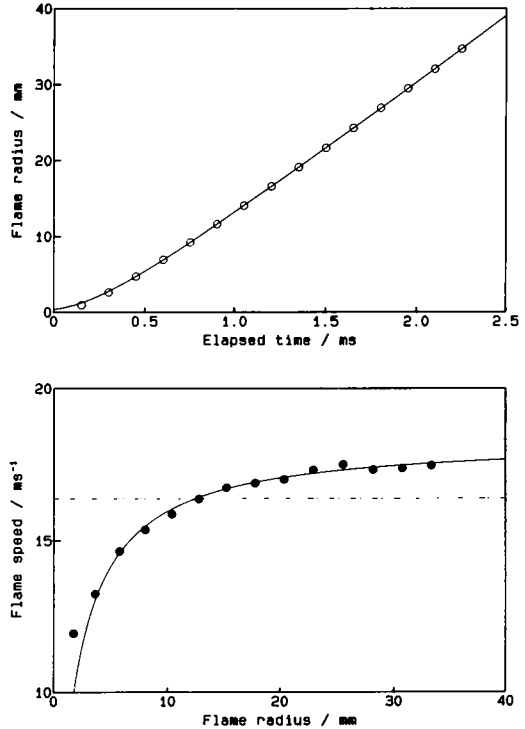


FIG. 1. a) Typical radius vs time data for expanding spherical flame. Points: experimental data. Line: Eq. (10) with $S_b^0 = 18.31 \text{ ms}^{-1}$, $b = 1.49 \text{ mm}$. Stoichiometry = 1.4 (41% hydrogen).

b) Typical flame speed vs radius data for expanding spherical flame. Points: time derivative calculated from radius vs time. Solid line: Eq. (9) with S_b^0 and b as above. Dotted line: limiting flame speed S_b^0 . Dashed line: flame speed obtained by fitting straight line to data in Fig. 1a. Same run as above.

to derive constant flame speeds from radius vs time data. This corresponds to the assumption that $b = 0$. On the face of it (see Fig. 1a), this seems entirely reasonable if the early ignition phase is ignored. However, the closer analysis in Fig. 1b reveals a more complex picture. (In none of the flames studied here did the r vs t curves appear non-linear to the casual glance.) The true S_b^0 is then obtained by fitting a curve of the form of Eq. (10) to the data, where S_b^0 is one of the fitted parameters. For the data in Fig. 1, this yields a value of 18.31 ms^{-1} , compared with the traditional best straight-line value of 16.36 ms^{-1} . Finally, the burning velocity S_u^0 is obtained by multiplying S_b^0 by the calculated density ratio $\sigma = \rho_b^0/\rho_u$. This is a valid procedure because S_b^0 is a 1-D value.

The other parameter obtained by fitting Eq. (10) to the experimental data is b . In order to derive the Markstein length, we need to subtract the den-

sity constant k . Values of k were obtained from computer modelling for each flame as explained later, and the Markstein length is determined from

$$L = (b - k) \sigma / 2. \quad (11)$$

It is worth stressing that although determination of L requires knowledge of the reference surface position, our method for determining burning velocity does not.

Experimental

Flames were burned in a spherical bomb (radius 300 mm) with central ignition, using a spark gap of about 0.7 mm. Data were obtained in the pre-pressure period, with flame radius limited to 35 mm. Flame propagation was recorded by high-speed cinematography (up to 7000 frames per second) of the schlieren image. The camera speed was calibrated by a 1 kHz timing signal.

Gases used were hydrogen (99.95% purity) and cleaned, filtered, compressed air. Mixtures were made by measuring partial pressures and mixed thoroughly by stirring. All runs were performed at a pressure of 1 atmosphere (101.3 kPa) and at 296 ± 1 K. The spark energy was adjusted for each run such that the mixture was just ignited.

The flame radius was measured on each frame of the film, giving a radius vs time record for each run. Equation (10) was fitted to the points by a modified least squares procedure, in which initial points were omitted until the best fit was obtained.

A complicating issue in the case of lean hydrogen/air mixtures is the occurrence of cellular flames. For mixtures containing about 25% hydrogen, onset of cellularity occurred within the measurable range, at a radius of about 30 mm. For the leanest flame (9% hydrogen) onset was at about 11 mm. The analysis was confined to the pre-cellular region, though this did entail some loss of accuracy.

Modelling

Three types of flames were modelled: planar 1-D, stationary spherical and expanding spherical.

For planar and stationary spherical flames, modelling was carried out using the Sandia Laboratories' PREMIX program,²⁴ slightly modified to ensure strict conservation of mass and energy. Because of its particular importance in hydrogen combustion, thermal diffusion (the Soret effect) was included in all computations.

A new program has been developed for modelling time-dependent expanding spherical flames. The program was developed from the Sandia code and retains many of its features. In particular, it pre-

serves the use of a reference frame attached to the flame. In order to allow for changes in the structure of the flame with time, a regridding strategy is included which involves the computation of the optimum grid as part of the time-dependent solution. The main advantage of this approach is that the form of the solution changes little during the course of the calculation, resulting in high computational efficiency.

A reaction scheme was assembled for use in all the simulations. It includes the most recent data²⁵⁻²⁸ for the important reactions $H + O_2 = OH + O$, $O + H_2 = OH + H$, $OH + H_2 = H_2O + H$ and $H + O_2 + M = HO_2 + M$. Full details of the expanding flame program and the reaction scheme will be given elsewhere.

Results and Discussion

Comparison of Simple and Detailed Expanding Flame Models:

The detailed model of expanding spherical flames produced results which agreed well with experimental data, although we have insufficient space to show this. More significant for our present purposes is the comparison of the dr/dt vs r behaviour predicted by our simple model (Eq. (9)) with that produced by detailed computer modelling. Figure 2 shows that for most radii the agreement is very good. In particular, the "limiting" flame speed predicted by Eq. (10) is very close to that obtained by the detailed model in a planar geometry. For all stoichiometries greater than 0.7, it is within 2% of the planar value. This has two important consequences. First, it means that, for the first time, measured burning velocities can be shown to be good estimates of the true 1-D values. Second, the assumptions in the simple model appear to be justified, so valid Markstein lengths will be derived.

The detailed modelling also shows that the simple model fails below some radius which is usually small. This is not surprising, since the assumptions would be expected to break down at high values of stretch and curvature. However, in very lean mixtures this radius becomes large, suggesting that the assumptions in our simple model are approaching the limits of their validity.

Burning Velocities:

Having established that the method of deriving burning velocities is sound, we now proceed to the experimental results. These are shown in Fig. 3, where we have plotted burning velocity against stoichiometry, defined as $(\text{mole \% } H_2) / (\text{mole \% } H_2)_{\text{stoich}}$. The maximum burning velocity obtained was 2.85 ms^{-1} at a stoichiometry of 1.4 (41% H_2).

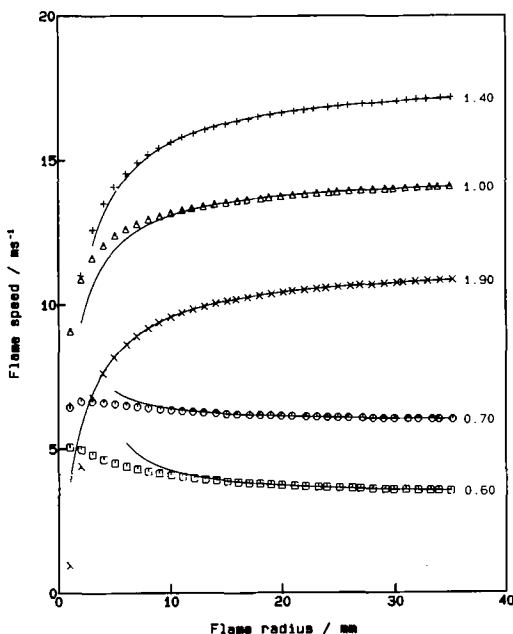


FIG. 2. Computed flame speed vs radius for hydrogen/air flames. Symbols: detailed model. Lines: Eq. (9) with fitted values of S_b^0 and b . Stoichiometries are listed opposite curves.

The stoichiometric result was 2.13 ms^{-1} . Overall accuracy of the results is estimated at $\pm 2.5\%$, though reproducibility was about 1%. Also shown in Fig. 3 are the burning velocities predicted by planar 1-D modelling. The agreement is remarkably good (within 3% over most of the range) considering that no arbitrary changes have been made in the reaction scheme to improve the fit. We attribute the poor agreement in very lean mixtures to error in the experimental data arising from inapplicability of the simple model.

The best-fit polynomial curve through the points is reproduced in Fig. 4 as curve G, where it is compared with a sample of data by other workers which is representative of the spread in values. The large differences between the data can be explained by considering two main effects.

Flow divergence between the cold gas and the reference surface can have a strong effect on burner determinations^{1,6,23,29} and always causes the results to be high. Its effect in conical flames will vary inversely with burner radius, and since small burners are generally used for hydrogen burning velocity determination, the effects can be large. Dixon-Lewis²⁹ estimated that it can cause errors of up to 40%.

Flame stretch will exert a secondary influence. Its effect can be estimated roughly, using our

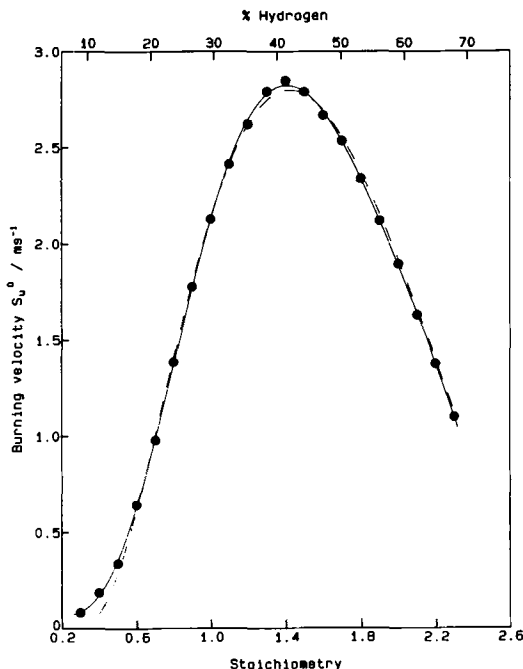


FIG. 3. Burning velocity of hydrogen/air as a function of stoichiometry. Points: experimental results. Solid line: best fit polynomial through experimental data. Broken line: best fit polynomial through planar modelling results. Initial temperature 296 K for both experiment and modelling.

Markstein length data in Fig. 6 and assuming that stretch rates of 1000 s^{-1} are typical of burner flames. At the maximum burning velocity, the effect is small (compared with divergence) but further out is of the order of 10%. Because stretch affects lean and rich flames in opposite directions, the result is a "tilt" in curves A, B and C of Fig. 4.

The button flame data of Günther and Janisch³⁰ (curve A) have the highest maximum burning velocity, exceeding ours by about 25%. The major reason for the difference is probably flow divergence: a back calculation from the methane button flame data in the same paper reveals a stream tube area increase between cold boundary and luminous zone of 20%, so a similar figure for hydrogen does not seem unreasonable. In addition, button flames are positively stretched. This will increase lean burning velocities and decrease rich, tending to tilt the curve relative to ours to produce better agreement on the rich side.

The conical flame data of Wu and Law¹ using nozzle diameters of 5 mm (curve B) and 7 mm (curve C) underline the dependence of divergence effects on burner size. Wu and Law estimated that their maximum burning velocity would be about 2.8 ms^{-1}

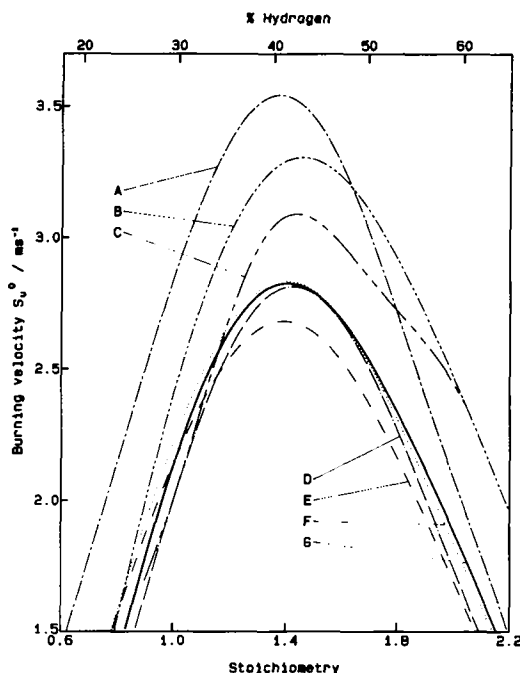


FIG. 4. Comparison of hydrogen/air burning velocities measured by different workers. Line A: Günther & Janisch button flame.³⁰ B: Wu and Law conical flame, 5 mm nozzle.¹ C: Wu and Law conical flame, 7 mm nozzle.¹ D: Scholte and Vaags conical flame, 3 and 5 mm nozzles.³¹ E: Andrews and Bradley constant pressure expanding spherical flame,³² uncorrected. F: Andrews and Bradley constant pressure expanding spherical flame,³² corrected to infinite radius. G: Present work.

if the data were extrapolated to zero curvature, in good agreement with our value of 2.85 ms^{-1} . If conical flames are negatively stretched¹ then the curves will be tilted in the opposite way to curve A, explaining the better agreement with our data on the lean side.

The conical flame data of Scholte and Vaags³¹ (curve D) apparently agree with ours but this may be fortuitous. On the above arguments their results should be high. It may be significant that nozzle diameters of 3 and 5 mm were used but no burner size dependence was uncovered. The reason for this is not clear, but must throw some doubt on their data.

The most recent constant-pressure bomb work is that of Andrews and Bradley.³² They measured their flame speeds at a flame radius of $r_f = 25 \text{ mm}$. If these data are multiplied by the density ratio then the "uncorrected" curve E in Fig. 4 is obtained. The results are considerably lower than ours on the rich side. However, if we use our experimental val-

ues of b (in Fig. 5) to correct these data to infinite radius by multiplying by $(1 + b/r_f)$, then very good agreement with our results is obtained (curve F).

Markstein Lengths:

We first compare the values of b obtained from the experiments with those from the detailed model. In both cases they were obtained by the fitting procedure described earlier. To ensure comparability with the majority of the experiments, only the first 35 mm of flame travel predicted by the detailed expanding flame model were analysed, though the conclusions would be very similar if larger radii had been used. The values of b as a function of stoichiometry are shown in Fig. 5. The modelled b values agree well with the experimental ones except at the extremes of stoichiometry. On the lean side, onset of cellularity restricts the analysis of experimental data to small radii where the simple model does not fit well. Small differences between modelling and experiment will then give rise to disproportionate differences in the fitted values of b .

The b values are converted into Markstein lengths in a three stage process:

1. The reference surface (where the mass fluxes of stationary spherical and planar 1-D flames have the same value) was located by modelling.
2. Values of k were derived by determining the mean gas density inside the reference surface of spherical flames at various radii, and applying Eqs. (5) and (6). For simplicity, density profiles from planar 1-D modelling were employed. As a future refinement, data from the expanding flame model will be used; early indications sug-

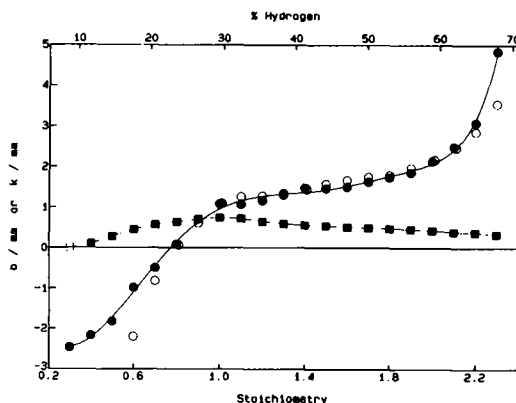


FIG. 5. Parameter b (see Eq. (10)) and density constant k as functions of stoichiometry. ●: b derived from experiments. ○: b derived from detailed model. ■: calculated values of the density constant k . Note: the leanest k value (□) is an estimate.

gest that the differences will not be great. Resulting values of k are plotted in Fig. 5.

3. Markstein lengths were derived from the experimental b and computed k values using Eq. (11).

Resulting Markstein lengths are plotted in Fig. 6. Values are negative (i.e. burning velocity increases with stretch) on the lean side, and positive on the rich. We have observed the same behaviour in methane/air and the opposite in propane/air.

Finally, we have derived approximate Markstein lengths from earlier detailed modelling studies: the expanding spherical flames of Dixon-Lewis¹³ and the stagnation flow flames of Warnatz and Peters.²¹ They are also plotted in Fig. 6. In view of the difficulties involved in determining L , the agreement is highly satisfactory.

Conclusions

1. A new method is presented for determining burning velocities and Markstein lengths. It has been demonstrated that the method gives true 1-D burning velocities.
2. Burning velocities are reported across the whole stoichiometric range of hydrogen/air mixtures (Fig. 3). The maximum value was $2.85 \pm .07 \text{ ms}^{-1}$ at a stoichiometry of 1.4 (41% hydrogen).
3. Markstein lengths are reported for hydrogen/air for the first time across the whole stoichiometric range (Fig. 6). They are in line with predictions based on Lewis number considerations.

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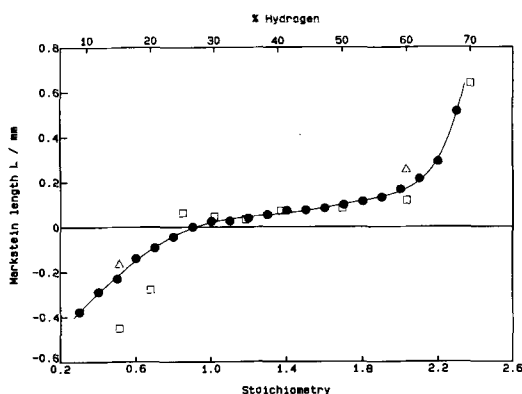


FIG. 6. Markstein lengths for hydrogen/air as a function of stoichiometry. ●: present experimental work. □: derived from modelling by Dixon-Lewis.¹³ △: derived from modelling by Warnatz and Peters.²¹

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