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# Uncertainty in stretch extrapolation of laminar flame speed from expanding spherical flames

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## Abstract

The present work investigated the uncertainties associated with the extrapolation of stretched flames to zero stretch in flame speed measurements using expanding spherical flames. Direct numerical simulations of time evolution of expanding spherical flames from a small ignition kernel to a propagating front with sufficiently large radius provide the relations between stretched flame speed and stretch rate that can be used to assess the uncertainty of extrapolation models. It is found that the uncertainties of flame extrapolation largely depend on the mixture Lewis numbers. While the uncertainty is minimized for stoichiometric H<sub>2</sub>/air and *n*-heptane/air flames, the uncertainty can be as high as 60% for lean H<sub>2</sub>/air mixtures, and 10% for lean and rich *n*-heptane/air mixtures. The present findings show that the weakly stretched flame assumption fails for lean hydrogen mixtures, and give a good explanation to the discrepancies between experiments and model predictions for H<sub>2</sub>/air as well as the discrepancies between measurements of *n*-heptane/air using spherical and counterflow flames. A relation between extrapolation uncertainties and the product of Markstein number and Karlovitz number is provided, which can be useful for uncertainty quantification of future and existing measurements.

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*Keywords:* Flame speed; Uncertainty; Extrapolation; Spherical flame; Stretch

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## 1. Introduction

One of the most important global parameters of a combustion mixture is the propagation speed of the steady, one-dimensional, planar, adiabatic flame, namely the laminar flame speed,  $S_u^0$ . It is

frequently used to validate combustion chemistry and advance our understanding of flame dynamics. Several flame configurations have been used to measure  $S_u^0$ , such as Bunsen flame, flat-burner flame, counterflow flame, and expanding spherical flame. Among these techniques, the expanding spherical flame is proven to be an effective method, especially at elevated pressures, that are typically not accessible using other configurations.

Despite of its simple geometry, expanding spherical flames are subjected to positive flame

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stretch, causing the flame speed to be modified by the nonequidiffusion of heat and mass characterized by non-unity values of the mixture Lewis number ( $Le$ ) and preferential diffusion between fuel and oxidizer. Therefore, the stretch effect needs to be eliminated by extrapolating the measured flame speed to zero stretch, using relations between the local flame speed and stretch. Different stretch extrapolation relations and procedures have been used [1–4], and it is believed that the method for stretch extrapolation and the selection of experimental data for extrapolation is one of the major contributions of uncertainties. Recognizing that the extrapolation equations currently in use were all derived from asymptotic analysis based on various assumptions, such as one-step chemistry scheme, and weakly stretched flames, the validity and systematic uncertainties of these models under different conditions need to be adequately examined. Previous studies have compared the difference among the different models on experimental data [2,3,5]. However, little work is done on model validation against the “true” values.

It is also noted that the uncertainty in the stretch extrapolation of spherical expanding flames can be coupled with or magnified by the uncertainties caused by radiation [6–8], flow compression and confinement effects [4,6,9], especially at large flame radii. Unfortunately, due to the flame instability and limitation of imaging capability, few experiments data are available to understand the uncertainty in stretch extrapolation at large flame radius.

The present work aims to investigate computationally and experimentally the uncertainties of different extrapolation relations for flame speed measurement using the constant-pressure spherical flame method. The focus is on the extrapolation of stretched flame speed to zero stretch at large flame radius. In the present study, the uncertainty quantification is based on the computation results of both the 1-D planar flame and the expanding spherical flame using detailed chemistry without including radiation loss and absorption. The confinement effect in the computation is eliminated by using a large computation

domain. New experiments were also conducted in order to compare with the computation results, and results with low extrapolation uncertainties are reported. Moreover, a criterion in terms of the product of Markstein number and Karlovitz number is provided to reduce the extrapolation uncertainty for practical spherical flame experiments.

## 2. Extrapolation equations

Table 1 summarizes all the five extrapolation equations evaluated in this study. The first one is a linear model based on stretch, denoted as LS. It was proposed by Wu and Law [1], and since then the large scatter in laminar flame speed measurements has been significantly reduced. Such a relation has been commonly used for flame speed measurement using expanding spherical flames. It is a first-order correction of the stretch effect, based on the assumption that  $Le$  is near unity and the flame is weakly stretched. Therefore, some degree of uncertainty using this model in flame speed extrapolation is expected.

The second model is a nonlinear model suggested by Kelley and Law [2]. This relation was also based on weakly flame stretch and derived by Ronney and Sivashinsky [10]. It allows arbitrary  $Le$ , and hence is expected to be more general than the linear model and can be applied for arbitrary mixtures. For the past few years, this model has been used extensively for extrapolating laminar flame speeds from expanding spherical flames. Improved accuracy and performance have been demonstrated in [2,3,5]. Since the nonlinear model is based on quasi-steady flame propagation, we denote it as NQ in this study.

In addition to the NQ model, Chen and Ju derived a nonlinear flame speed and stretch relation of expanding spherical flames by including the effect of both strong stretch and general Lewis number [11] and suggested the third model, another nonlinear extrapolation equation in the limit of large flame radius [3]. Since the suggested model is also a linear relation between flame speed

Table 1

Extrapolation equations considered in this study.  $S_b^0$  and  $L_b$  are the unstretched laminar flame speed and the Markstein length with respect to the burned mixture,  $S_b = dR_f/dt$ ,  $K = (2/R_f)dR_f/dt$  and  $\kappa = 2/R_f$  are the stretched flame speed with respect to the burned gas, the stretch rate and curvature of an expanding spherical flame, and  $R_f$  is the flame radius.

Model	Refs.	Notes	
LS	[1]	Linear model based on stretch	$S_b = S_b^0 - L_b K$
NQ	[2]	Quasi-steady nonlinear model	$\left(\frac{S_b}{S_b^0}\right)^2 \ln\left(\frac{S_b}{S_b^0}\right)^2 = -\frac{2L_b K}{S_b^0}$
LC	[3]	Linear model based on curvature	$S_b = S_b^0 - L_b \kappa = S_b^0 - \frac{2L_b}{R_f}$
NE	[4]	Nonlinear model in expansion form	$\frac{S_b}{S_b^0} \left[ 1 + \frac{2L_b}{R_f} + \frac{4L_b^2}{R_f^2} + \frac{16L_b^3}{3R_f^3} + o^4\left(\frac{L_b}{R_f}\right) \right] = 1$
N3P	–	Nonlinear model with 3 fitting parameters	$\frac{S_b}{S_b^0} = 1 - \frac{2L_b}{R_f} + \frac{C}{R_f^2}$

and curvature, we denote it as LC. This relation has been proposed empirically by Markstein [12]. It was demonstrated in [3] that the difference between the extrapolated laminar flame speeds using LC and NQ is large for mixtures with  $Le > 1$  while they give almost the same results for mixtures with  $Le < 1$ . Chen [3] further showed that the extrapolation curve using LC agrees closer to result of numerically simulated methane flames than NQ, and therefore LC should be used instead of NQ, especially for mixtures with  $Le > 1$ .

To explain the apparent improved accuracy of LC over NQ, Kelley et al. [4] noted that in the analysis of Ronney and Sivashinsky [10], there is another unsteady term. However, including this unsteady term causes the equation to be numerically unstable, thereby providing difficulty in its use for extrapolation. Kelley et al. [4] therefore suggested the use of the expansion form of the relation in terms of inverse power of flame radius. This is the fourth model in Table 1 and it is denoted as NE. It was further explained in [4] that the reason for the improved accuracy of LC over NQ is because in the expansion form LC model has closer agreement with the NE model.

Finally, recognizing that difference among LS, LC, NQ and NE starts from the second-order inverse power of  $R_f$ , in this study we also consider an extrapolation equation with a free parameter on the second-order term. This equation does not assume the curvature of the relation between flame speed and stretch; rather it relies on the experimental data to obtain the curvature. Since this nonlinear equation has three fitting parameters, it is denoted as N3P in this study.

### 3. Numerical and experimental methods

The one-dimensional expanding spherical flame is simulated using the one-dimensional, adaptive simulation of reactive flow (A-SURF) code [13], which has been successfully used in a series of studies on spherical flame initiation and propagation, such as [9,14,15]. A-SURF solves the conservation equations of one-dimensional, multi-component, reactive flow in a spherical coordinate using the finite volume method. The details on the governing equations, numerical schemes, and code validation can be found in [6,13]. To maintain adequate numerical resolution of the moving flame, a multi-level, dynamically adaptive mesh refinement algorithm has been used in A-SURF. In all simulations, we have ensured the confinement effect is negligible by using a large chamber radius (100 cm for the simulations of H<sub>2</sub>/air flame and 25 cm for those of *n*-heptane/air in this study) and only considering flame radius that is less than a fraction of the wall radius [9,14].

The one-dimensional planar steady adiabatic flame is simulated using the PREMIX Code,

which is part of the CHEMKIN package. PREMIX solves the conservation equations of one-dimensional, multi-component, reactive flow in planar coordinate. Therefore, the solution in theory should be equal to that of A-SURF at very large flame radius. To be consistent, simulations with PREMIX and A-SURF used the same chemical kinetic model and transport formulations. For H<sub>2</sub>/air flame simulations both used multi-component model with Soret diffusion, and for *n*-heptane/air flame simulations both used mixture-averaged model without Soret diffusion. Since the present study focuses only the uncertainty associated with stretch extrapolation, both simulations do not include radiation model to suppress the effects of radiation coupling.

Experiments reported in this study were conducted in a dual-chamber, constant-pressure vessel. Detailed specification of the experimental apparatus, procedure and data analysis were reported previously [16]. The apparatus consists of a cylindrical chamber of near-unity aspect ratio radially situated within another cylindrical chamber of substantially larger volume. The inner chamber is filled with the test mixture, and outer chamber is filled with inert gases to match the pressure of the inner chamber. Such a design allows an expanding spherical flame to propagate throughout the inner chamber in essentially an isobaric environment. The vessel allows the flame radius to grow independent of ignition and compression effects from 1.0 cm to 2.0 cm for typical flames.

### 4. Results

From A-SURF simulations, the stretched flame speed relative to the burned gas,  $S_b = dR_f/dt$ , can be calculated by numerically differentiating the flame front history,  $R_f = R_f(t)$ , with  $R_f$  defined as the position of maximum heat release rate. We have calculated the flame radius history by using both the maximum temperature gradient and maximum heat release definition. The relative difference is within 0.2%, indicating that the two definitions give almost identical flame radius history. The stretch rate can also be calculated from the flamefront history,  $K = (2/R_f)dR_f/dt$ . To consistently compare the results at different conditions, normalization is necessary. From PREMIX, the following quantities are determined: the flame speed relative to the burned gas  $S_{b, \text{Premix}}^0$ , the flame thickness  $\delta_L$  of the one-dimensional planar flame, defined based on the maximum gradient of the temperature profile,  $\delta_L = (T_{ad} - T_u)/(dT/dx)_{\text{max}}$ , where  $T_{ad}$  is the adiabatic flame temperature, and  $T_u$  the unburned mixture temperature. This definition of flame thickness was originally proposed by Spalding [17]. It has been shown [18] that this definition gives almost the same results as that based

on full width at half maximum (FWHM) of the heat release profile [19]. It also has been proven useful in studying the aerodynamics of flames, such as turbulent flames [20] and flamefront instability [21]. With  $S_{b, \text{Premix}}^0$  and  $\delta_L$ , the normalized flame speed  $S_b^0/S_{b, \text{Premix}}^0$  and Karlovitz number  $Ka = K\delta_L/S_{b, \text{Premix}}^0$  can then be computed.

#### 4.1. Results on $H_2/\text{air}$

$H_2/\text{air}$  mixtures are most suitable for uncertainty quantification for the following reasons. First, the chemistry is most well understood; therefore deviation from experiments is minimized. Second, since  $H_2$  has distinct transport properties from  $N_2$  and  $O_2$ , the variation of  $Le$  from lean to rich covers a wide range of values (0.3–2.4) and therefore can provide the conservative estimation of extrapolation uncertainty. Third, the simulation is efficient so that large-size flames can be computed within reasonable time.

Fig. 1 plots  $S_b^0/S_{b, \text{Premix}}^0$  versus  $Ka$  for various equivalence ratios for  $H_2/\text{air}$  at 1 atm. The updated high pressure  $H_2\text{--}O_2$  model by Burke et al. [22] were used for both PREMIX and A-SURF simulations. As expected, for lean/rich mixtures the stretched flame speeds, calculated by A-SURF, start from high/low values at small radii (large  $Ka$ ), respectively. As the flame grows and  $Ka$  decreases, the flame speed calculated by A-SURF for lean/rich mixtures decreases/increases, respectively, approaching the solution of PREMIX for all equivalence ratios. This means that simulations using the two codes are consistent and can be used for uncertainty quantifications of different extrapolation models.

To mimic the situations in real experiments, we will only use the data in the range from  $R_f = 1.0$

cm to  $R_f = 2.0$  cm for extrapolation. Fig. 1 plots the corresponding extrapolation curves using the five equations in Table 1. It is seen while the extrapolated flame speeds for rich mixtures ( $Le > 1$ ) typically differ by less than 10% from the PREMIX solutions using all equations, those for lean mixtures ( $Le < 1$ ) for  $\phi < 0.7$  are substantially higher than the PREMIX solutions. N3P model produces results closest to PREMIX solutions; however, the differences are still 10–35%. This large systematic uncertainty means none of the models are valid for mixtures with negative Markstein lengths and will cause significant over-prediction in practice. In addition, for very rich mixtures, for example  $\phi = 5.0$ , LS and N3P turn out to be more accurate than the other three nonlinear relations, which under-predicts by 5–10%.

To investigate the cause of the failure of all equations for lean  $H_2/\text{mixtures}$ , we first compute the 95% confidence intervals [23] of all the fittings shown in Fig. 1. For all five models, the 95% confidence intervals for  $S_b^0$  all fall within 99–101% of the estimates. This means the over-prediction of all models is not due to the statistic insignificance caused by inadequate data. Next, we conduct fittings on a wide range of data from  $R_f = 1.0$  cm to  $R_f = 10.0$  cm for  $\phi = 0.4$ , and the results are shown in Fig. 2. It is seen that the A-SURF data show a strong curvature, which none of the fittings can capture exactly. LS, LC and NE models have significant deviations from the A-SURF data. NQ model has a closer agreement. The N3P model has the closest agreement with the A-SURF data because it has another free parameter on the second-order term on RHS of the N3P model for fitting. Even this, the N3P still has noticeable deviation from A-SURF data and its estimate is 20% higher than the PREMIX result.

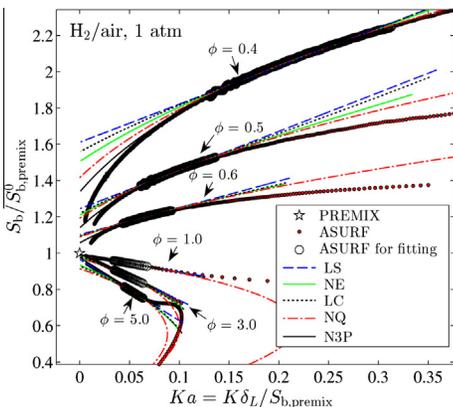


Fig. 1. Comparison between results of various extrapolation models with numerical simulation for various equivalence ratios of  $H_2/\text{air}$  at 1 atm. Data used for fitting is for flame radius from 1 cm to 2 cm.

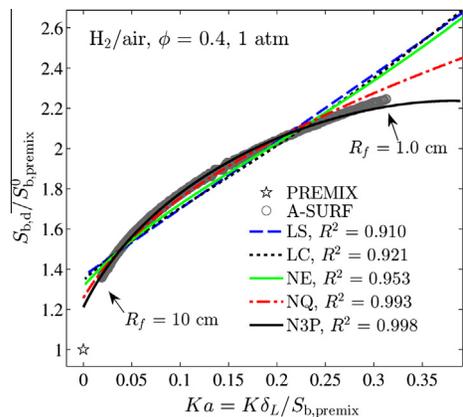


Fig. 2. Comparison of various extrapolation models for  $\phi = 0.4$  of  $H_2/\text{air}$  at 1 atm using a large range of simulation data.

#### 4.2. Burned gas density effect

Another influence of stretch that is frequently neglected is the change of burnt gas density due to the change in the flame temperature. The issue was recently raised in [24]. For  $Le < 1$ , the burned gas density of a spherical flame is reduced due to increased temperature compared to the planar case, which further increases the flame speed due to expansion. Similarly, for  $Le > 1$  flames it further decreases the flame speed. This effect causes the measured flame speed to be different than the real consumption speed,  $S_{b,c}$ , and becomes more sensitive on stretch. Using A-SURF simulations, we have investigated whether this is the cause of the failure of nonlinear extrapolations and quantified its effect on extrapolation results. It turns out this effect does not change the extrapolation uncertainty significantly because the effect is largely diminished at large flame radii. Due to space limit, detailed results of this part of investigation are moved into the [Supplementary material](#).

#### 4.3. Comparison with experiments

We next investigate the performance of extrapolation on experimental data. Experiments were conducted on expanding spherical flames, and the flamefront position was tracked from Schlieren luminosity which is proportional to the density gradient. A Matlab program is written to track the location of maximum luminosity gradient of an image and we define it as the flamefront position. This is consistent with the definition of flame radius in A-SURF simulations. Therefore the experimental data corresponds to the flame speed relative to the burned gas.

Fig. 3 plots the experimental data in comparison to the A-SURF simulations and the extrapolation results based on the experimental data. First, it is seen that the experimental data agree closely with the simulated flame speed at  $\phi = 0.5, 0.6, 1.0$  and  $3.0$ . Such agreement is much smaller than the difference between the extrapolated flame speeds and the PREMIX solutions. This supports the validity of both the A-SURF simulations, experiments as well as the kinetic model. At  $\phi = 0.4$ , the experimental data is slightly lower than the simulated flame speed, indicating influencing factors, such as radiation or chemistry. Second, the extrapolation results based on the experimental data are similar to those based on the flame speeds from A-SURF simulations, *i.e.*, there are still large over prediction and moderate under prediction for lean and rich  $H_2$ /air flames. On the experimental data, the N3P model shows a different behavior compared to its performance on A-SURF data. While its estimates on  $S_b^0$  with A-SURF data have the closest agreement with PREMIX compared to

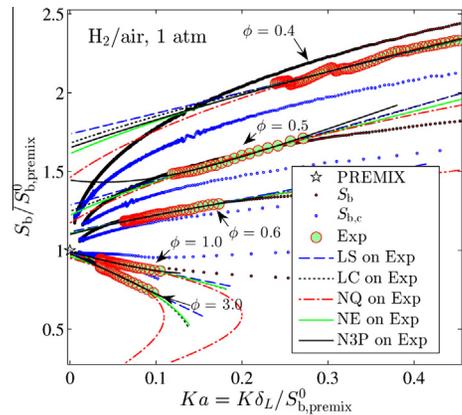


Fig. 3. Extrapolation based on experimental data using various models at various equivalence ratios of  $H_2$ /air at 1 atm. The flame speed  $S_b$ , and consumption flame speed  $S_{b,c}$ , are plotted for comparison. Definition of  $S_{b,c}$  is given in the [Supplementary material](#).

other models, its performance on the experimental data is not predictable. This is because the three-parameter fitting largely relies on the curvature of the data, while experimental data always has inevitable noise. For example, the N3P curve for  $\phi = 0.5$  is non-monotonic at small  $Ka$  which is clearly nonphysical.

#### 4.4. Results on *n*-heptane/air

Opposite to  $H_2$ , the diffusivity of heavy hydrocarbon fuels is much smaller than  $N_2$  and  $O_2$ . The resulting  $Le$  and Markstein lengths is also large but with opposite dependence on  $\phi$ . We have also conducted simulations of *n*-heptane/air flames using the 88-species skeletal mechanism by [25]. Fig. 4 plots the extrapolation results on *n*-heptane/air at 1 atm. It is seen that similar to  $H_2$ /air, the extrapolation results of all models are under predicted for  $\phi < 1.2$  with  $Le > 1$  and over predicted for  $\phi > 1.3$  with  $Le < 1$ . On the other hand, different from  $H_2$ /air, for mixtures with  $\phi > 1.5$  further increasing  $\phi$  does not result in increase of the slope of the flame speed dependence on stretch rate, *i.e.*, decrease in Markstein lengths. The A-SURF curves for  $\phi = 1.7$  and  $\phi = 2.0$  almost overlap. This is because in realistic hydrocarbon flames, fuel quickly decomposes into small fuel fragments before these fragments can be oxidized. Therefore the sensitivity due to  $Le$  and preferential diffusion are controlled by the diffusion of the fuel fragment, rather than the fuel. Despite this, it is still seen that the uncertainty caused by extrapolation can be  $\pm 10\%$  for very lean and rich mixtures, respectively. This uncertainty is expected to be higher for heavier fuels, such as *n*-dodecane, or fuels which do not decompose quickly in flames, such as trimethylbenzene.



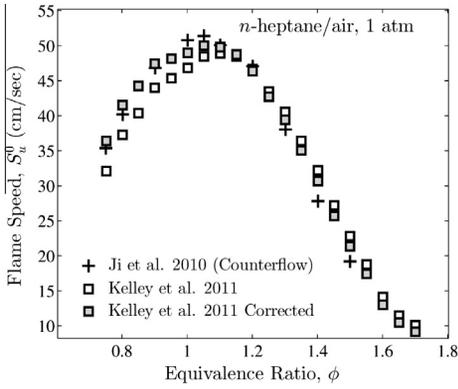


Fig. 6. Comparison of original and corrected *n*-heptane/air flame speeds measured in spherical flames [16] to those measured in counterflow flames [26].

However,  $S_u^0$  for an arbitrary mixture is still a useful quantity in other context, such as study of turbulent combustion, thermal acoustics, that is worth our effort to measure it accurately. Therefore, solutions are needed to reconcile the extrapolation uncertainties. Fundamentally, the variation of flame speeds on stretch comes from two sources [33]: the first one is the  $Le$  and preferential diffusion effects, which manifests them in terms of the Markstein number,  $Ma = L_b/\delta_L$ , while the second source is the normalized stretch rate, *i.e.*, the Karlovitz number  $Ka = K\delta_L/S_b$ . The combined effect is the product of the two,  $MaKa$ . It is important to recognize that the non-linearity of the relation between flame speed and stretch not only depends on the  $Le$ , but also on  $Ka$ . This means that for data at different  $Ka$ , the nonlinearity is different. For example, although we can acquire the flame radius history experimentally from 1 to 2 cm for all equivalence ratios, they correspond to different  $Ka$  at different equivalence ratios. It can be easily shown that,

$$Ma_{Linear}Ka_{mid} = 2L_b/R_{f,mid} \quad (2)$$

where  $Ma_{Linear}$  is the slope of the flame speed on stretch for a set of experimental data, *i.e.*, the Markstein length,  $L_b$ , here corresponding to the LS model, and  $Ka_{mid}$  is the normalized stretch rate at the middle point of the data. Eq. (2) can be easily calculated in any experiments. In Fig. 7, we plot the extrapolation uncertainties of different models versus  $Ma_{Linear}Ka_{mid}$  for both  $H_2$ /air and *n*-heptane/air. It is seen that for  $Ma_{Linear}Ka_{mid} < -0.05$  the extrapolations have significant over-prediction, while for  $Ma_{Linear}Ka_{mid} > 0.15$  the extrapolations also have under predictions up to 10%. Future measurements should be conducted in the range of  $-0.05 < Ma_{Linear}Ka_{mid} < 0.15$  to minimize the extrapolation uncertainties (within  $\pm 5\%$ ). The dependence of uncertainties on  $Ma_{Linear}Ka_{mid}$  can also be used to quantify the

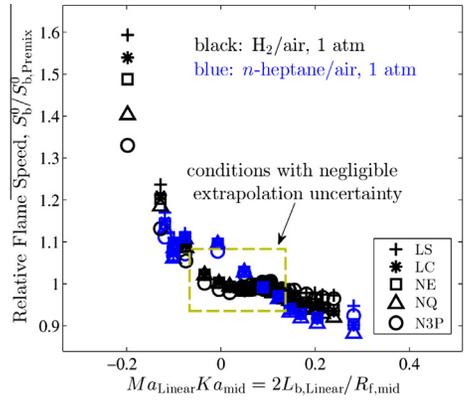


Fig. 7. Extrapolation uncertainties versus  $Ma_{Linear}Ka_{mid}$  for  $H_2$ /air and *n*-heptane/air at 1 atm.

uncertainties of existing measurements and correct them. In practical spherical flame experiments, the flame radius range used in data processing is constrained by ignition and compression effects, and thereby it is difficult to change the value of  $R_{f,mid}$  in Eq. (2). The best way to minimize the extrapolation uncertainties is to set the mixture  $Le$  close to unity, for which  $|L_b|$  approaches zero. Furthermore, Eq. (2) indicates that the extrapolation uncertainty decreases with increasing pressure since  $|L_b|$  becomes smaller at higher pressure.

### 6. Conclusions

The present study quantifies the systematic uncertainties associated with the extrapolation of flame speed measurements using expanding spherical flames. Results show that the uncertainties of extrapolation largely depend on the Lewis number (or Markstein number) and the Karlovitz number. For  $H_2$ /air, it is found that all the existing models have large over predictions (up to 60%) for  $\phi < 0.8$  and finite under predictions for  $\phi > 3.0$  (up to 10%). For *n*-heptane/air, there are under and over predictions (up to 10%) for lean and rich mixtures, respectively. The reason is that none of the existing relations were able to accurately capture the strong nonlinear trends between stretched flame speed and stretch when the Lewis number is appreciably different from unity.

The uncertainties quantified in this study suggest that discrepancies between flame speeds of  $H_2$ /air at 1 atm measured in spherical flames and the corresponding predictions of model by Burke et al. [22] are mostly due to extrapolation error. The extrapolation uncertainties for *n*-heptane/air flame speeds at 1 atm can also explain most of the discrepancies between measurements in spherical flames and those measured in counterflow flames.

The dependence of uncertainties on a controlling parameter  $Ma_{\text{Linear}}Ka_{\text{mid}}$ , which is easily accessible in any experiment, is given. It is suggested future measurements should be in the range  $-0.05 < Ma_{\text{Linear}}Ka_{\text{mid}} < 0.15$  to minimize the extrapolation error; otherwise, corrections are needed to appropriately interpret the data.

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### Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.proci.2014.05.065>.

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