Inferring laminar burning properties from spherical expanding flames: the pitfalls of an established approach

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Abstract. The procedure of deriving laminar burning parameters from high-speed, high-resolution shadowgraph recordings of spherical expanding flames allows to obtain experimental data in lifelike conditions of high pressure and temperature. Straightforward in principle, relatively simple to implement, the approach has been applied in several labs worldwide, to the extent of achieving a “common practice” status, sometimes resembling a codified protocol. Simple as it may look on paper, yet the technique hides a number of pitfalls, which can impair the end result. After the introduction of solid-state high-speed cameras (CCD, then CMOS), gathering high-speed videos of single combustion events became a breeze: this, along with the possibility of easy stockpiling of data, lend to underrate data analysis: sort of trading the accuracy of measurement for the repeatability. Another critical phase is dealing with the stretch, which affects any real flame: measured data must be processed to obtain the unstretched flame speed and, ultimately, the laminar burning velocity. The relationship between flame speed and stretch will be discussed, being a key factor for the deconvolution of experimental, stretch-affected data.

In the present work a critical discussion is proposed, from experimental data acquisition and processing to stretch analysis: the underlying hypotheses of each step will be used as the guidance to a “good” rather than “common” practice. Reference will be made to a specific test case: the combustion of CH₄ in air at P₀ = 6 bar, spanning the whole flammability range.

1. Introduction
Pursuing CO₂ reduction led to the availability of several gas mixtures (e.g. syngas or biogas), as opposed to the traditional fossil fuels. Their composition is far from fixed or standardized, therefore efficient exploitation in energy conversion systems asks for their characterization as fuels: this implies evaluation of the laminar burning velocity and the Markstein length. The accurate determination of these parameters is still a matter of debate, even if it has been a hot research topic in the last 50 years [1-7], as highlighted in a couple of extensive reviews [7, 8]. Spherical expanding flames offer a number of advantages: they allow high-pressure testing are conceptually simple to analyze. Nevertheless, what can appear as a straightforward approach is actually a delicate evaluation process, punctuated by subtle pitfalls: failing to realize this may lead to less than accurate results [10-12]. Many authors focused on the effects of ignition [3, 9], radiation and buoyancy [13], confinement and chamber geometry [14], not to mention instability [15]. Yet the core of the process leading from the experimental data to the laminar parameters (Su₀ and Luₐ) is the stretch analysis [11]. If the laminar burning velocity is the ideal speed at which a planar flame front propagates normally to its surface, the flame speed in real system is affected by stretch: the last is the temporal flame surface deformation, which conditions the propagation, stability...
and structure of the flame. Evaluating the functional relation between flame speed and stretch remains the big challenge in the quest for accurate laminar burning properties.

In the present work, a specific test case [CH\textsubscript{4} – air, P\textsubscript{0} = 6 bar, T\textsubscript{0} = 298K \phi = \phi\textsubscript{min} ÷ \phi\textsubscript{max}] will be used as the thread for a critical analysis of the estimate of laminar burning parameters, from data acquisition and processing to stretch analysis: the underlying hypotheses of each step will be used to highlight the implications of the specific procedures, and an estimate of the related accuracy will be given.

2. Theoretical background

The laminar burning velocity \( S\textsubscript{u0} \) is the propagation speed of a steady, laminar, one-dimensional, planar, unstretched, adiabatic flame, travelling a premixed mixture, and is relative to the unburned gas. Given the difficulty to respect all of the above-mentioned conditions in practical systems, the measurement strategy relies on pursuing most of them by clever experiment design, and to deal with the rest through a series of theoretical assumptions: this approach is the basis of the experimental determination of laminar burning velocity.

As mentioned earlier [7, 8], constant-pressure spherical expanding flames historically got increasing success, offering simple geometrical configuration and easy definition of stretch, with the virtually unique asset of high-pressure measurements. No matter how plain, the path to laminar burning velocity from the measurement of spherical expanding flames asks for a series of steps, illustrated in the following.

The starting data consist of the time evolution of the flame radius \( R\textsubscript{f} = R\textsubscript{f}(t) \), as inferred from high-speed visualization: the burned flame speed \( S\textsubscript{b} \) (also called propagation speed) can then be evaluated as:

\[
S\textsubscript{b} = \frac{dR\textsubscript{f}}{dt} \tag{1}
\]

This speed includes the stretch effects associated to the propagation of a flame surface, which experiences curvature and flow dynamic strain [16-19]. The flame stretch \( K \) is defined as the relative rate of change of the flame area: for a spherically expanding laminar flame, it can be expressed as:

\[
K = \frac{1}{A} \frac{dA}{dt} = \frac{2}{R\textsubscript{f}} \frac{dR\textsubscript{f}}{dt} = 2 \frac{S\textsubscript{b}}{R\textsubscript{f}} \tag{2}
\]

The relationship between flame speed and stretch has been thoroughly investigated: its expression depends on the number and nature of the related assumptions. Historically, a linear formulation has been suggested [16,19]: it derives from asymptotic analysis and has found wide application. It can be expressed in a dimensional form, suitable for data reduction, as follows:

\[
S\textsubscript{b} = S\textsubscript{b0} - L\textsubscript{b} \cdot K \tag{3}
\]

where \( S\textsubscript{b0} \) is the value assumed by \( S\textsubscript{b} \) at \( K = 0 \), and \( L\textsubscript{b} \) is the burned gas Markstein length. The latter indicates how and to what extent the flame is influenced by the stretch. Positive \( L\textsubscript{b} \) are associated to flames with speed decreasing with stretch (which are stable), while in the case of negative \( L\textsubscript{b} \) the flame speed tends to increase with stretch, becoming unstable; moreover, the magnitude of \( L\textsubscript{b} \) indicates to what extent the flame propagation is influenced by the stretch.

Back in 1983, Frankel and Sivashinsky [20] suggested the following equation for spherical flames with thermal expansion:

\[
S\textsubscript{b} = S\textsubscript{b0} - S\textsubscript{b0}L\textsubscript{b} \cdot \frac{2}{R\textsubscript{f}} \tag{4}
\]

where \( S\textsubscript{b} \) is given by equation (1). According to equation (4), \( S\textsubscript{b} \) varies linearly with flame curvature \( 2/R\textsubscript{f} \).
Equation (3) shows a linear relationship between $S_b$ and the stretch rate $K$. It is obtained assuming a small deviation of the flame speed from a planar, adiabatic value. In a more general case (no small stretch, no density variation, adiabatic and quasi-steady flame), the propagation speed can be expressed as [21,22]:

$$\left(\frac{S_b}{S_{b0}}\right)^2 \ln \left(\frac{S_b}{S_{b0}}\right)^2 = -\frac{2L_0K}{S_{b0}}$$

(5)

The models given by equations (3) (4) and (5) can be used to extract the unstretched burned flame speed and the Markstein length from spherical expanding flames. These models can be classified on the basis of the relationship between $S_b$ and $K$: in this framework, equation (3) will be referred to as “linear model”, while equations (4) and (5) as “nonlinear models”.

Being known the unstretched burned flame speed $S_{b0}$, the laminar burning velocity $S_u$ can be obtained as follows:

$$S_u = S_{b0} \frac{\rho_b}{\rho_u}$$

(6)

where $\rho_b$ is the density of burned gases and $\rho_u$ the density of unburned gases. Equation (6) holds true in the hypotheses of perfect gases, isobaric, adiabatic and equilibrium conditions

3. Experimental layout and procedures

The general arrangement of the experimental layout is shown in figure 1: a detailed description is given in [23]. The heart of the DHARMA (Device for Hydrogen-Air Reaction Mode Analysis) laboratory is a constant-volume test reactor, made of stainless steel (AISI 316): the cylindrical chamber (i.d. = 70 mm, h = 90 mm, aspect ratio = 1.29) is rated for pressure ≤ 20 MPa (static).

A total of 6 optical accesses are available: the larger viewports (Ø 65 mm) are located normal to the chamber axis; smaller ports (Ø 35 mm) are positioned on the chamber side, along two orthogonal axes.

![Figure 1. Layout of the experimental apparatus.](image-url)
Hi-grade quartz windows (Ø 85 mm, 30 mm thick) are installed in the main ports. Four additional service ports are available for the intake and exhaust of the gases.

The mixture is ignited with an automotive inductive ignition system (energy ≤ 60 mJ): the spark energy is set adjusting the time of charge (dwell time). The electrical discharge takes place in the center of the chamber between two pointed-tip tungsten electrodes (Ø 1 mm, gap = 1 mm).

A high-frequency dynamic pressure transducer detects pressure history during the combustion events. A metal-shielded, type K thermocouple is used to monitor the temperature of the gases, save for the combustion phase.

The gas handling system was designed to prepare mixtures of variable composition with high accuracy. High purity gases (CH₄: 99.9995%, H₂: 99.999%, CO: 99.999%, CO₂: 99.998%, N₂: 99.9995%, dry air: 99.999%) are used to prepare the mixtures, relying on the partial pressures method: the gases are metered by solenoid valves, controlled by a high-resolution (100 MHz) counter/timer board. The pressure is monitored by two high-accuracy (<0.08% FS) pressure transmitters, operating in the range 0–1 bar and 0–30 bar. The maximum value of total pressure is 30 bar. After each test, the system is vented, purged with N₂ and pumped down to 10⁻² mbar. Systems operation is fully automated under digital control, to maximize safety and repeatability of the tests: the entire lab conforms to current safety standards.

A parallel-beam direct shadowgraph layout is used for high-speed data collection, based on state-of-the-art devices and highly optimized optic arrangement. A Diode-Pumped Solid-State c.w. laser (Laser Quantum mod. Opus 2W @532nm) is used as the light source. A Photron SA-5 camera (1024x1024 pixel, 12 bit, ≤1000000 fps, ≥ 368 ns) is used for high-speed, time-resolved imaging, together with a low distortion Nikon 60 mm f/2.8D AF Micro.

4. Data acquisition and processing

Figure 2 shows a selection of frames from high speed recordings, obtained at different equivalence ratios for the test case of CH₄ combustion in air at P₀=6 bar and T₀=298 K. For every equivalence ratio ϕ, the flame front is smooth and devoid of any sign of instability. Given the camera resolution and the magnification ratio, the spatial resolution of these images is 65 µm: since the useful range of flame radii (as defined later) is 4–10 mm, this translates in a measurement error 0.6÷1.6%.

![Flame Fronts](image)

**Figure 2.** Flame fronts of CH₄-air mixtures at different ϕ; T₀ = 298 K, P₀ = 6 bar.

ROI: 27x27 mm, 7000 frame/s, exp. time: 1 µs.
The temporal resolution can be considered equal to the exposure time, that is 1 µs. The framing rate of these tests is 7000 fps: this allows to follow the flame evolution with adequate accuracy: the number of useful frames for the fastest burning flames (φ=1.0) is ~50, and increases as φ gets far from stoichiometric. These figures are suitable for meaningful data analysis, as described in the following.

An image processing routine has been implemented to infer the flame radius $R_f$ from the shadowgraph data. Assuming the luminous front in the shadowgraph corresponds to the radius on the unburned gas side [24-25], the flame contour is traced frame by frame, and the area of the projected flame ball is evaluated; the radius is estimated as that of a circle of equal area to the flame. This procedure assumes the flame being axisymmetric and encompasses (i) the inclusion of the electrodes in the flame ball and (ii) the fitting of the projected flame contour with an ellipse. The latter gives an immediate check on how much the flame shape is far from being spherical, while the former does not represent a relevant error, given the pointed tips of the electrodes.

The quantitative assessment of these effects is given in figures 3 and 4, for the case of φ=1.0. As shown in figure 3, in the useful range of flame radii (defined in the following), the electrodes represent at worst 1.25% of the measured volume. Likewise, as shown in figure 4, if one selects the circularity (defined as $4\pi A/P^2$) as the shape descriptor of the flame, it turns out being >98% in the range of interest: this validates the assumption of spherical flame.

The above-described routine allows the determination of the time evolution of the flame radius $R_f = R_f(t)$. Typical outputs are illustrated in figure 5, for two lean and two rich CH$_4$-air mixtures.

These data sets cannot be used in their entirety for the application of the above-described theory, since they are affected by a number of phenomena, which make the underlying hypotheses simply not applicable. Lipatnikov et al. [11] mention no less than seven of what they call “perturbations”, which make actual flame growth deviate from theory: (a) effect of the spark ignition on early stage flame kernel; (b) confinement-induced flow; (c) radiation heat losses; (d) non-uniformity of product density; (e) the compression of unburned gas and products; (f) flame instabilities and (g) flame stretching.

As stated by Bradley et al. [3], the early stages of the flame kernel growth are affected by the spark energy release (a), and must be discarded: this defines a lower limit radius, the actual value of which depends on the ignition type and energy, on φ, etc. [9]. In the present tests, spark energy was set at 20 mJ (electrical): being delivered by an inductive ignition coil, only a small fraction of this energy is released in the plasma breakdown phase, which controls the early kernel growth: the extent of the ignition disturbances can be accordingly limited [23]. As a matter of fact, analysis of the flame radii reveals no discernible evidence of ignition-induced effects: nonetheless a conservative criterion was applied, based on the morphology of the flame: flames with a circularity lower than 0.98 were discarded.

This criterion meets also the need to avoid the issue of non-uniform density of burned gas (d) in small kernels, associated to the high front curvature.
The main reason to set an upper limit to the flame radii arises from the major hypothesis behind the experimental approach: isobaric flame propagation (e). By monitoring the chamber pressure during the combustion event, the constant pressure phase can be defined, and the corresponding radii range be set.

In addition, closed chambers are likely to show up the effect of confinement on the expanding flame, as the front approaches the chamber wall (b). Burke et al. [14] suggested to keep \( R_f < 0.3R_{chamber} \) to avoid the effects of confinement in a cylindrical chamber.

Finally, the range of flame radii may need truncating due to the onset of instabilities on the flame surface (f), which depend on several parameters (species, \( \phi \), \( P_0 \), Lewis number \( Le \), etc.).

According to the above comments, setting the upper and lower limits of the radii range allows to deal with five out of the seven perturbations mentioned earlier. The remaining ones are the radiation heat losses (c) and the stretch effects (g). The former will not be discussed here, but become relevant in the case of small \( S_b^0 \) and/or close to the flammability limits [10-12], while the latter will be the object of thorough analysis in the following. In the current tests, the bounding values for the data set were 4 mm and \(-10 \) mm: the resulting data range is highlighted in figure 5 with red symbols.

5. Stretch analysis

5.1. Linear models

The basic approach for the linear analysis consists of a series of steps: a polynomial fit of \( R_f = R_f(t) \) is first performed; differentiation of the fit allows to obtain the stretched burned flame speed \( S_b \) after equation (1); being known \( S_b \) and \( R_f \), the stretch rate \( K \) can be evaluated after equation (2); according to equation (3), linear-fit extrapolation of the flame speed to \( K = 0 \) gives the unstretched flame speed \( S_b^0 \), while the slope of the fit allows to estimate the Markstein length \( L_b \). The critical issue in this methodology resides in the polynomial differentiation, which is highly sensitive to the choice of the original data interval: this may introduce numerical noise and lead to distorted profiles of \( S_b \), affecting both \( S_b^0 \) and \( L_b \).

5.1.1. Linear model I. In 2007 Burluka et al. [26] suggested an analytical procedure to infer \( S_b^0 \) and \( L_b \) directly from the temporal evolution of the flame radius. Substitution of equation (2) into equation (3) gives, after integration:

\[
R_f(t) - R_f(t_0) + 2L_b \ln \left( \frac{R_f(t)}{R_f(t_0)} \right) = S_b(t - t_0)
\]  

(7)

The problem reduces to the optimization of the least square function:

\[
\Psi(L_b, S_b^0) = \sum_{i=1}^{N} \left( t_i - t_0 - \frac{R_i - R_0}{S_b^0} - 2 \cdot \frac{L_b}{S_b^0} \ln \left( \frac{R_i}{R_0} \right) \right)^2
\]  

(8)
This is easily implemented, no data differentiation is performed, and no singularity is present (the case of $L_b=0$ is allowed). The critical point of this approach lies in the selection of the initial conditions ($t_0$ and $R_0$). In order to circumvent this issue, we modified the procedure, performing $N$ estimates ($N = \text{no. of data points}$), choosing each time a different value of $r_0$ from the available set. The final values of $S_{b0}$ and $L_b$ are then obtained as the average of the $N$ values of $S_{b0}$ and $L_b$ evaluated as described above. Even with a large number of data points ($t, R$), the method demonstrated to be fast and robust.

5.1.2. Linear model II. In 2009 Tahtouh et al. [25] suggested an elegant analytical methodology which offers the exact solution of the linear equation (3). Equations (2) and (3) can be rearranged as follows:

$$\frac{dR}{dt} = S_{b0} - 2L_b \cdot \frac{dR}{Rdt}$$  \hspace{1cm} (9)

The solution of equation (9) is:

$$R(t) = 2L_bW(Z)$$  \hspace{1cm} (10)

where $W$ is the Lambert function, and $Z$ is given by:

$$Z = e^{\frac{S_{b0}+C_1}{2L_b}}$$  \hspace{1cm} (11)

where $C_1$ is a constant to be determined. The values of $S_{b0}$ and $L_b$ (and $C_1$) are obtained minimizing the following equation:

$$\sum_{i=1}^{N} \left( R_{f_i}(t) - R(t) \right)^2 = \sum_{i=1}^{N} \left( R_{f_i}(t) - 2L_bW(Z) \right)^2$$  \hspace{1cm} (12)

where $R_f$ is the experimentally measured radius. The Lambert function $W$ is a multi-valued function [27]: if $Z$ is real, for $-1/e \leq Z < 0$ $W(Z)$ can assume two possible real values. The branch satisfying the condition $-1 \leq W(Z)$ is termed the principal branch and denoted $W_0(Z)$. The branch satisfying the condition $W(Z) \leq -1$ is called the alternate branch and denoted $W_{-1}(Z)$. For $L_b > 0$ (that is $Z > 0$), the principal branch of the Lambert function ($W_0$) must be used, while for $L_b < 0$, $W_{-1}(Z)$ should be used.

This method delivers directly $S_{b0}$ and $L_b$ for each test case, no differentiation is needed and all the data are used simultaneously, with the same weight. The major drawback is that the function is not defined for $L_b=0$: a preliminary guess is needed on the sign of $L_b$, in order to use the principal $W_0(Z)$ or the alternate branch $W_{-1}(Z)$ of the Lambert function. An independent a priori estimate of the sign of $L_b$ is therefore needed, to be performed with a different method. Moreover, when $L_b$ has very small absolute value, the model may become unstable: if the preliminary estimate suggests a value with the wrong sign, the wrong branch of the Lambert function will be selected and the method will fail to converge.

5.2. Nonlinear models

5.2.1. Nonlinear model I. Substitution of equation (1) into equation (4) gives:

$$\frac{dR_f}{dt} = S_{b0}^0 - S_{b0}^0 L_b \cdot \frac{2}{R_f}$$  \hspace{1cm} (13)

This equation can be analytically solved and the values of $S_{b0}^0$ and $L_b$ can be obtained by a nonlinear least-square method, without the need of numerical differentiation. This is a simple and robust formulation, offering the best compromise between accuracy and reliability.
5.2.2. **Nonlinear model II.** In 2007 Kelley & Law [28] suggested an analytical solution of equation (5), allowing to obtain directly $S_b$ and $L_b$ from the raw data of flame radius as a function of time $R_f(t)$.

\[
t = A \left[ E_1(\ln \tau^2) - \left( \frac{\tau^2 \ln \tau}{2} \right)^{-1} \right] + C
\]  

(14)

where:

\[
A = \frac{2L_e}{S_b^0}, \quad R_f = \frac{2L_b}{\tau \ln \tau}, \quad E_1(x) = \int_{-x}^{\infty} \frac{e^{-z}}{z} dz
\]  

(15)

Equation (14) is suitable for nonlinear least-square regression, allowing $A$, $L_b$ and $C$ to be determined. $	au \in [1/e, 1]$ for $L_b > 0$ and $\tau \in [1, \infty]$ for $L_b < 0$.

In the present work, we replaced the expression for $E_1$ with the following:

\[
E_1(x) = \ln \left( \frac{x}{2} \right) + \sum_{k=1}^{\infty} (-1)^k \frac{x^k}{k \cdot k!}
\]  

(16)

which results always defined in a real domain, even for $\tau \in [1/e, 1]$.

The model has the unquestionable advantage of offering an explicit analytical solution, and handles well the cases of nonlinear behaviour of $S_b$ vs. $K$.

Nevertheless, the expression asks for some care. The transformed time variable has different spacing from the original data set: at every step, the outputs from the algorithm have to be interpolated in order to be compared with the input data. This may be a source of interpolation noise (error). Even if the case of $L_b = 0$ offers no problem, a preliminary estimate of the starting values will improve the convergence of the solver. The region $\tau \in [1/e, e]$ can be tricky for the model to handle.

Table 1 summarizes the above-described models.

| author(s)                        | type of model | solution          | short form |
|----------------------------------|---------------|-------------------|------------|
| Burluka et al. (2007)            | linear        | explicit          | LM I       |
| Tahtouh et al. (2009)            | linear        | through least-square | LM II     |
| Frankel & Sivashinsky (1983)     | nonlinear     | through least-square | NM I       |
| Kelley & Law (2007)              | nonlinear     | through least-square | NM II       |

5.3. **Model comparison**

In the following the comparison is proposed and discussed between the linear and nonlinear approaches to data analysis. Two different methods are evaluated to solve the linear equation (3); the solutions of nonlinear equations (4) and (5), as suggested in literature, are tested also in terms of mathematical stiffness.

The starting data consist of the time evolution of the flame radius $R_f = R_f(t)$, as inferred from high-speed shadowgraph recordings of spherical expanding flames.

In figure 6 and 7 the comparison is proposed between the different models, along with the experimental data.

Two examples of lean mixtures ($\phi = 0.7$ and 0.9) are shown in figure 6, highlighting an almost linear variation of the burned flame speed $S_b$ as a function of the stretch rate $K$. LM I and LM II deliver virtually the same values for $S_b$ and $L_b$, which are very close to the results obtained with either NM I or NM II. Actually, at $\phi = 0.7$ and $\phi = 0.9$ the Markstein length is very close to 0, standing for weak sensitivity of flame speed on stretch: therefore, a linear model can be meaningfully applied and offers accurate results.
Figure 6. Comparison of stretch models for lean CH$_4$-air mixtures. $T_0 = 298$ K, $P_0 = 6$ bar.

Figure 7 shows two examples of rich mixtures ($\phi = 1.3$ and 1.5): as the equivalence ratio increases, the relationship between the burned flame speed and the stretch deviates from linearity. The difference between LM I (or LM II) and the nonlinear models indicates that a linear model is no more valid: as a matter of fact, Markstein lengths are much larger (by an order of magnitude), corresponding to regimes of flame evolution much more sensitive to stretch. In these conditions, application of either LM I or LM II leads to overestimation of $L_b$ and consequently of $S_{b0}$. Comparison of NM I and NM II reveals that the experimental data are typically better fitted by the former model: NM II tends to underestimate either the Markstein length or the burned flame speed.

The overall behaviour of $S_b$ vs. $K$, spanning the whole flammability range is shown in figure 8. As anticipated by the examples discussed above, CH$_4$-air mixtures exhibit stronger nonlinearity as the equivalence ratio $\phi$ is increased. These results are in perfect agreement with the findings of Halter et al., obtained with the same mixture at ambient temperature and pressure [22]. The dashed lines represent the results obtained with NM I, which effectively and accurately describe the flame-stretch interaction, whatever the equivalence ratio.

Figure 7. Comparison of stretch models for rich CH$_4$-air mixtures. $T_0 = 298$ K, $P_0 = 6$ bar.
Figure 8. Evolution of burned flame speed with the stretch rate in the complete flammability range of CH₄-air mixtures. T₀ = 298 K, P₀ = 6 bar.

Figure 9 summarizes the burned gas Markstein length and the laminar burning velocity, evaluated after equation (6), as a function of φ. The graphs include the results for the various models, LM I, LM II, NM I and NM II, allowing for an immediate comparison of their validity and accuracy. As remarked above, the Markstein length increases with the equivalence ratio: it’s negative for very lean mixtures and changes sign at φ = 0.8 ÷ 0.9. For equivalence ratios φ ≤ 1.2, |Lₘ| < 0.5 mm and the dependence of the flame speed on stretch can be considered linear with good approximation. For φ > 1.2, this relationship becomes nonlinear: as a consequence, linear models tend to overestimate Lₘ and, in turn, the unstretched burned flame speed Sₘ₀.

This ultimately leads to overestimation of the laminar burning velocity, as evidenced in figure 9 (right): the values of Sₘ₀ obtained with linear and nonlinear models are virtually coincident up to φ = 1.2, but for richer mixtures an increasing difference is noted between LM’s and NM’s.

Figure 9. Markstein length (left) and laminar burning velocity (right) as a function of φ, as obtained by the tested stretch models. CH₄-air, T₀ = 298 K, P₀ = 6 bar.
These findings are represented in figure 10, in terms of relative differences: assuming NM I as the reference, the error induced by the use of the other models can be defined as:

$$\varepsilon_i = \frac{S_{0,i} - S_{0,\text{NM I}}}{S_{0,\text{NM I}}}$$ (17)

where $S_{0,\text{NM I}}$ is the laminar burning velocity evaluated with NM I, and $S_{0,i}$ the velocity obtained with model $i$ ($i =$ LM I, LM II, NM II). Save for the case of $\phi = 0.6$, where higher relative errors can be expected, due to the low absolute value of the velocity involved, $\varepsilon_i$ keeps limited to a few percent up to $\phi = 1.2$, then for LM I and LM II exponentially increases, reaching ~20% at $\phi = 1.5$. Errors as high as 200%, obtained with LM I at the rich flammability limit ($\phi = 1.6$), stand for the definite inaccuracy of linear models for rich CH$_4$-air mixtures. While both linear models tend to overestimate the laminar burning velocity, the method suggested by Kelley & Law (NM II) leans towards underestimation, even if the absolute value of $\varepsilon_i$ does not exceed 5% for $\phi \leq 1.5$.

These results confirm the conclusions of Chen [29], who carried out an in-depth comparison of linear and nonlinear models against both simulated data and experimental results. He concluded that NM I is the most accurate model for analyzing data with positive Markstein length, which is the case for most of the methane-air mixtures within the flammability interval.

6. Final comments
A critical analysis was proposed of the experimental evaluation of laminar burning parameters from spherical expanding flames at constant pressure. The discussion involved dwelling on the design of the experimental setup, on data collection and processing, and on the stretch analysis. CH$_4$-air combustion ($T_0 = 298$ K, $P_0 = 6$ bar, $\phi_{\text{min}} < \phi < \phi_{\text{max}}$) was used as test case.

Crucial in the experimental practice is the precision of the mixture, which, in the case of partial pressure approach, relies on the accuracy of the devices used for pressure monitoring. The shape and size of the test chamber affect the useful range of measurement: the smaller the chamber, the smaller the isobaric phase and, consequently, the number of useful data. Proper choice of the acquisition system (frame rate and spatial resolution) can either compensate a constrained data range or allow adequate topological accuracy. As far as the processing is concerned, most of the perturbations that may impair the theory application can be dealt with through careful identification of the radius range suitable for subsequent analyses. Overall, paying proper attention to the described point, an accuracy of a few percent is not out of reach.
With a sound data set $R_f(t)$ available, the last and most delicate phase is left: the deconvolution of flame speed from stretch. Four methodologies have been compared to the purpose: LM I and LM II were based on the hypothesis of a linear dependence of $S_b$ on $K$; NM I and NM II were based on a more complex, nonlinear relation. All the tested models offer the appeal of an analytical solution, allowing to infer $S_b$ and $L_b$ directly from the experimental data $R_f(t)$.

The main findings can be summarized as follows:

- NM I demonstrated to be the most accurate model in the case of CH₄, which has a positive Markstein length $L_b$ (corresponding to a Lewis number $Le \sim 1$) in most of the flammability range.
- The behaviour of $S_b$ vs. $K$ is linear up to $\phi \sim 1.2$. For $\phi > 1.2$ the application of a linear model gives rise to growing errors: for $\phi = 1.5$ the error on $S_b^0$ can be as high as 20%.

The stretch analysis remains the critical phase in the evaluation of laminar burning parameters, yet careful (preliminary) consideration of mixture properties (e.g. the $Le$) can keep accuracy within 5%.

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