Laminar flame speed correlations for methane, ethane, propane and their mixtures, and natural gas and gasoline for spark-ignition engine simulations

Riccardo Amirante[1], Elia Distaso[1,2], Paolo Tamburrano[1] and Rolf D Reitz[2]

Abstract
The laminar flame speed plays an important role in spark-ignition engines, as well as in many other combustion applications, such as in designing burners and predicting explosions. For this reason, it has been object of extensive research. Analytical correlations that allow it to be calculated have been developed and are used in engine simulations. They are usually preferred to detailed chemical kinetic models for saving computational time. Therefore, an accurate as possible formulation for such expressions is needed for successful simulations. However, many previous empirical correlations have been based on a limited set of experimental measurements, which have been often carried out over a limited range of operating conditions. Thus, it can result in low accuracy and usability. In this study, measurements of laminar flame speeds obtained by several workers are collected, compared and critically analyzed with the aim to develop more accurate empirical correlations for laminar flame speeds as a function of equivalence ratio and unburned mixture temperature and pressure over a wide range of operating conditions, namely $\phi = 0.6-1.7$, $p_u = 1-50\,\text{atm}$ and $T_u = 298-800\,\text{K}$. The purpose is to provide simple and workable expressions for modeling the laminar flame speed of practical fuels used in spark-ignition engines. Pure compounds, such as methane and propane and binary mixtures of methane/ethane and methane/propane, as well as more complex fuels including natural gas and gasoline, are considered. A comparison with available empirical correlations in the literature is also provided.

Introduction

A crucial aspect for the reliability of many multidimensional combustion models for spark-ignition engines simulations is represented by the laminar flame speed estimation, which is essential for the accurate prediction of the turbulent burning velocity of the fuel–air mixture[1–5] and, hence, for an adequate representation of the whole combustion process taking place within the engine.

Flame propagation in spark-ignition engines involves time and spatial scales which cannot be typically captured with practical finite volume methods.[1,2,6] Therefore, several dedicated numerical models have been implemented to evaluate the turbulent burning velocity once the spark has been triggered.[7–9] These models rely on empirical or semi-empirical correlations of laminar flame speeds which are derived from experimental measurements.[1,5,10]

Several combustion phenomena depend on the laminar flame speed, such as the turbulent flame structure and speed, various modes of flame front instabilities, flame extinction through heat loss and stretch and flame stabilization.[11] In addition to engine applications, it plays a primary role in many other combustion applications, such as in the design of burners and in the prediction of explosions.[7,9] For these reasons, it has long been the subject of extensive experimental and theoretical investigation, over a wide range of operating conditions.[11]

In an internal combustion engine, the laminar burning velocity plays a crucial role in determining the spark

[1]Department of Mechanics, Mathematics & Management, Polytechnic University of Bari, Bari, Italy
[2]Engine Research Center, University of Wisconsin–Madison, Madison, WI, USA

Corresponding author:
Elia Distaso, Department of Mechanics, Mathematics & Management, Polytechnic University of Bari, Via Re David 200, 70125 Bari, Italy.
Email: elia.distaso@poliba.it

advance and influences the cycle-to-cycle fluctuations, the thickness of wall quench layers (which are the primary source of unburned hydrocarbons), as well as the minimum energy to ignite the charge, which affects the range of equivalence ratio over which an engine can operate.[12]

The laminar flame speed is an intrinsic property that is a function of the unburnt mixture composition, temperature and pressure. Although encouraging progress has been made in developing detailed chemical kinetic models for its prediction, such models are still extremely complex and require significant computational effort for solving the mass, species and energy conservation equations coupled with chemistry.[1,13] In addition, they can fail outside the range in which they have been validated against experimental data, or if the grid resolution chosen for the simulation is not appropriate for the specific case.[14]

Thus, analytical correlations of the laminar flame speeds as a function of equivalence ratio, pressure and temperature are preferred in engine practical simulations. Moreover, they are more easily implemented in CFD codes than tabulated data. Their use in spark-ignition engine simulations still allows the use of detailed chemical kinetics for modeling the post-flame chemistry and the end-gas chemistry, which does not require high resolution, thus saving computational time.[1,2,6]

In order that an analytical formulation (as well as a chemical kinetics mechanism) can be considered reliable for many possible conditions, it must be validated against a large body of data. Thus, in this work, experimental measurements of laminar flame speeds carried out by several workers are compared and critically evaluated. As Ranzi et al.[11] pointed out when summarized the experimental laminar burning velocities of methane/air mixtures for the previous 60 years, it was not until the mid-1980s that Wu and Law[15] noted the importance of the stretch effects in the experimental determination of laminar flame speeds and proposed a rational approach toward their elimination. Therefore, the attention was focused only on recent experimental studies (when available) in which a higher accuracy is guaranteed by the fact that they consider the stretch effects in the determination of the laminar flame speed. In addition, published predictions of empirical correlations derived by other workers have also been considered and compared with those proposed in this work for a more exhaustive analysis.

In the present study, empirical correlations are presented for laminar flame speeds as functions of equivalence ratio and unburned mixture temperature and pressure over a wide range of operating conditions, namely $\phi = 0.6 - 1.7$, $p_u = 1 - 50$ atm and $T_u = 298 - 800$ K. The fuels considered include promising cleaner alternatives to gasoline for the future, including natural gas. However, since pure compounds represent a good starting point, correlations for methane and propane are also provided. Then, together with natural gas, binary mixtures of methane/ethane and methane/propane are also considered. Finally, an empirical correlation for gasoline is also provided. The aim is to offer a simple, but accurate method for determining the laminar burning velocity for a wide range of equivalence ratios, temperatures and pressures that it is suitable for engine simulation applications.

Overview of empirical correlations available in the literature

In the last 60 years, various forms of empirical and semi-empirical functional relationships have been proposed for the laminar burning velocity. These semi-empirical relationships are based either on the thermal flame propagation theory of Zel'dovich/Frank-Kamenetsky/Semenov[12,16–22] or on the active species diffusion theory.[23–26] Wholly empirical correlations are instead exclusively derived from the interpolation of experimental measurements within the operating range over which they were carried out.

The "Arrhenius form," upon which many semi-empirical formulations are based, is very sensitive to the adiabatic flame temperature, which is in turn sensitive to the thermodynamic model used to calculate it.[20] In addition, it seems that the Arrhenius parameters do not depend consistently on the equivalence ratio, but rather they vary erratically with it.[20] As a consequence, sometimes the interpolation and extrapolation process can be very difficult, with a possible inability to produce smooth variations with equivalence ratio or temperature.[10,20]

The simplest alternative and the most widely used form of the wholly empirical correlation is the so-called power law formula, adopted by many investigators[10,12–14,19,27–33]

$$S_L(\phi, T_u, p_u) = S_{L0} \left(\frac{T_u}{T_0}\right)^{\alpha} \left(\frac{p_u}{p_0}\right)^{\beta} \tag{1}$$

where S_{L0} is the velocity measured at $T_u = T_0$ and $p_u = p_0$ for a given equivalence ratio ϕ, and α and β are constants or mixture strength-dependent terms.

One of the most known studies in which such a form was employed is the work by Metghalchi and Keck.[20] It was derived from measurements carried out in a constant-volume vessel for fuel-to-air equivalence ratios $\varphi = 0.8 - 1.5$, over pressure and temperature ranges of $p_u = 0.4 - 50$ atm and $T_u = 298 - 750$ K. After testing their correlation against experimental measurements of the laminar burning velocity of methanol, propane, iso-octane and indolene, they concluded that the temperature and pressure exponents α and β were independent of fuel type within their estimated experimental error and could be represented by the expressions

$$\begin{aligned} \alpha(\phi) &= 2.18 - 0.8(\phi - 1) \\ \beta(\phi) &= 0.16 + 0.22(\phi - 1) \end{aligned} \tag{2}$$

Table 1. Coefficients of equation (3) proposed by Metghalchi and Keck[20] for propane/air mixtures.

Fuel	T (K)	P (atm)	Φ_m	B_m (cm/s)	B_2 (cm/s)
C_3H_8	298	1	1.08	34.22	−138.65
	350−700	1−50	1.08	40.11	−186.48

In addition, they found that the reference velocities S_{L0} were a weak function of fuel type and could be fit by a second-order polynomial of the form

$$S_{L0}(\phi) = B_m + B_2(\phi - \phi_m)^2 \qquad (3)$$

where the parameters B_m, B_2 and ϕ_m are given in Table 1 for propane, which is also of interest in the present study. In equation (1), they considered $p_0 = 1$ atm and $T_0 = 298$ K and recommended its use for applications in the ranges: $p_u = 1-50$ atm and $T_u = 350-700$ K. However, at room temperature that interpolation underestimated burning velocities. Therefore, they proposed different and more appropriate values for coefficients B_m and B_2 for room conditions, which are reported as well in Table 1.

Beside the fact that two sets of coefficients must be provided for each fuel, another major limitation is that equation (3) predicts negative flame speeds for lean ($\phi < 0.6$) and rich ($\phi > 1.6$) mixtures. It is acceptable for simulations of premixed flames near stoichiometric conditions, but is not applicable for stratified charge combustion in direct injection spark-ignition engines with gasoline or gaseous fuels, that is, natural gas.[34–37]

An alternative formulation for evaluating the term $S_{L0}(\phi)$ of equation (1) was derived by Elia et al.[28] They developed a correlation by fitting their experimental data of the laminar burning velocity of methane/air mixtures, obtained using a spherical constant-volume combustion vessel with fuel-to-air equivalence ratio varying from 0.8 to 1.2, as

$$S_{L0}(\phi) = S_{u0}(\alpha_0 + \alpha_1\phi + \alpha_2\phi^2) \qquad (4)$$

where $S_{u0} = 37.5$ cm/s, $\alpha_0 = -5.883$, $\alpha_1 = 14.003$, $\alpha_2 = -7.115$. In the same study, after analyzing measurements in which the unburned gas pressure was varied from 0.75 to 70 atm and the temperature from 298 to 550 K, they suggested the use of a fixed value for the coefficients α and β, namely 1.857 and −0.435, respectively. The results appeared to be in good agreement with their experimental data, but some discrepancies with other works have been recorded. In addition, it is widely recognized that the exponents α and β vary with equivalence ratio and a possible dependence on pressure and the temperature could exist too.[10,29,38,39] Therefore, for a more appropriate formulation, expressions for α and β, at least as functions of only ϕ, are also needed. Furthermore, the correlation proposed by Elia et al.[28] for S_{L0} shows a problem similar to that highlighted for Metghalchi and Keck,[20] regarding the

Table 2. Coefficients for equation (1) by Gu et al.[19] for methane/air mixtures in the ranges of 300–400 K and 1–10 atm for three different equivalence ratios, namely $\phi = 0.8$, 1.0 and 1.2.

ϕ	S_{u0} (cm/s)	α	β
0.8	25.9	2.105	−0.504
1.0	36.0	1.612	−0.374
1.2	31.4	2.000	−0.438

generation of negative values outside the tested equivalence ratio range.

Another work in which the power law formulation was employed was carried out by Gu et al.,[19]. It was based on measurements of laminar spherical expanding flames of methane/air mixtures. In this work reference values of $p_0 = 1$ atm and $T_0 = 300$ K were assumed. What the authors of this study suggested were three expressions for equation (1) in which the parameters S_{u0}, α and β were optimized in the ranges of $300-400$ K and $1-10$ atm for three different equivalence ratios, namely $\phi = 0.8, 1.0$ and 1.2. The values that they proposed are reported in Table 2.

Other researchers developed correlations for stoichiometric mixtures, including Han et al.[29] and more recently Hu et al.[13] The first[29] used a preheated cylindrical combustion chamber to measure the laminar burning velocity of methane/air mixtures, with initial temperatures ranging from 298 to 498 K and initial pressures from 1 to 5 atm. The derived empirical formulation was

$$S_L = 36.11\left(\frac{T_u}{T_0}\right)^\alpha \left(\frac{p_u}{p_0}\right)^{-0.37} \qquad (5)$$

where $p_0 = 1$ atm and $T_0 = 300$ K and the temperature exponent α depends on pressure as

$$\alpha(p_u) = 1.5365 + 0.1165 p_u \qquad (6)$$

Hu et al.[13] quantified the laminar flame speed dependence upon pressure and temperature as

$$\begin{aligned}
&S_L(p_u) = 0.133(p_u)^{-0.461}, \\
&T_u = 300\,\text{K}, \quad 1\,\text{atm} \leqslant p_u \leqslant 60\,\text{atm} \\
&S_L(T_u) = 325e^{\left(-\frac{9439}{T_u + 1089}\right)}, \\
&p_u = 1\,\text{atm}, \quad 300\,\text{K} \leqslant T_u \leqslant 700\,\text{K}
\end{aligned} \qquad (7)$$

In addition, they found that the exponent α increased linearly with increase of initial temperature,

Table 3. Coefficients for Gülder's exponential formulation for different fuels.

Ref.	Authors	Year	Fuel	Z	W (cm/s)	η	ξ	σ
10	Gülder	1984	CH_4	1	42.2	0.15	5.18	1.075
			C_3H_8	1	44.6	0.12	4.95	1.075
32	Dirrenberger et al.	2011	CH_4	1	38.638	−0.15	6.2706	1.1
			C_3H_8	1	42.2012	−0.3104	5.1455	1.1
40	Coppens et al.	2007	CH_4	1	39.0542	−0.4333	6.0157	1.1

while the exponent β decreased exponentially with increase of initial pressure. Consequently, they formulated the following correlations for the two exponents, as

$$\alpha(T_u) = 1.39 + 0.0006T_u, \qquad 300\,\text{K} \leqslant T_u \leqslant 700\,\text{K}$$
$$\beta(p_u) = 0.226e^{-\frac{p_u}{0.841}} - 0.511, \quad 1\,\text{atm} \leqslant p_u \leqslant 60\,\text{atm}$$
$$(8)$$

The last expressions were validated by Hu et al.[13] at high temperatures and pressures by comparison with numerical simulations. A similar strategy was adopted by Ouimette and Seers[30] who calculated numerically the laminar flame speed for both methane and a synthetic gas using PREMIX. For both fuels, they provided a correlation in the form of equation (1) exclusively based on the calculations. Although the calculations were performed at different equivalence ratios and pressures and the explored range of the initial mixture temperatures is the largest available in the literature (300−850 K), the technique used for the numerical simulations was validated by means of comparisons with experimental data for methane only for the room pressure case and for two different unburnt temperatures, namely 300 and 400 K. The following expressions for calculating methane's laminar flame speed based on work by Liao et al.[31] (which is discussed later in this section) were suggested

$$S_{L0}(\phi) = -204.6\phi^3 + 428.9\phi^2 - 220.2\phi + 33.3$$
$$\alpha(\phi) = 4.3\phi^2 - 9.0\phi + 6.6 \qquad (9)$$
$$\beta(\phi) = -0.7\phi^2 + 1.4\phi - 1.1$$

A completely different approach for determining the term $S_{L0}(\phi)$ in equation (1), which represents a practical solution to the intrinsic problem of polynomial forms, was proposed by Gülder,[10] who chose the following empirical expression to represent the room temperature burning velocity of methane, propane and other fuels considered in his work

$$S_{L0}(\phi) = ZW\phi^\eta e^{-\xi(\phi-\sigma)^2} \qquad (10)$$

where W, η and ξ are constants for a given fuel, and $Z = 1$ for single constituent fuels. The constants for methane/air and propane/air mixtures proposed in Gülder[10] are listed in Table 3. In such work, the power law dependence of the laminar burning velocity on the unburnt mixture pressure and temperature was used,

but a practical analytical expression for the exponents α and β, as functions of the equivalence ratio, was not proposed. Instead, constant values for α and β were proposed, namely, respectively, 2 and −0.5 for methane and 1.77 and −0.2 for propane.

Gülder's formulation for the evaluation of S_{L0} has also been adopted in the correlations proposed in the present work, since it appears to be the most promising expression among all the analyzed solutions. The power law formula is then chosen for taking into account the influence of pressure and temperature.

Recently, Dirrenberger et al.[32] adopted such a formulation for the prediction of the laminar flame velocity of the components of natural gas, methane, ethane, propane and n-butane, as well as for binary and tertiary mixtures of these compounds, which had been proposed as surrogates for natural gas. In that study, the measurements were performed using the heat flux method at atmospheric conditions. The mixture strength covered the largest range available in the literature, namely from 0.6 to 2.1. The values of the parameters that they proposed for pure methane and propane are given in Table 3. For the methane case, their parameters were far from those previously proposed by Gülder, but are very close to those found by other researchers, that is, Coppens et al.,[40] which are reported as well in Table 3. They proposed a correlation valid for a natural gas surrogate mixture of methane, ethane and propane. This ternary mixture correlation was a combination of the expressions obtained for binary methane/ethane and methane/propane mixtures, which used the correlation, derived from Coppens et al.[40]

$$S_{L0}(\phi, \chi) = (1 + \nu\chi^\tau)W\phi^\eta e^{-\xi(\phi-\sigma-\Omega\chi)^2} \qquad (11)$$

The term Z, present in Gülder's formulation (equation (10)), assumes the value $(1 + \nu\chi^\tau)$ to take into account the presence of other compounds in methane. χ is the amount of the other gas in the fuel mixture. The additional term $\Omega\chi$ in the exponent allows to reproduce the shift of the maximum of the laminar flame velocity's dependence with the additional gas concentration. When χ is zero, the original Gülder's formulation for pure compounds is obtained. The coefficients ν, τ and Ω derived from the experimental data interpolation are reported in Table 4. Good agreement was found for lean and rich mixtures, but the correlation overestimated flame velocities near stoichiometry.

Table 4. Coefficients proposed by Dirrenberger et al.[32] for binary mixtures.

Fuel	ν	τ	Ω
CH_4/C_2H_6	0.2103	0.545	−0.0191
CH_4/C_3H_8	0.2129	0.8312	−0.0439

By combining these results, they formulated the following correlation for ternary mixtures

$$S_{L0}(\phi, \chi_1, \chi_2) = \left(1 + \nu_1\chi_1^{\tau_1}\right)\left(1 + \nu_2\chi_2^{\tau_2}\right)$$
$$W\phi^\eta e^{-\xi(\phi - \sigma - \Omega_1\chi_1 - \Omega_2\chi_2)^2} \tag{12}$$

where subscript 1 refers to parameters calculated for one component, that is, ethane, and subscript 2 to the other one, that is, propane. Once again, if $\chi_1 = \chi_2 = 0$, the correlation for pure fuels is obtained. If either $\chi_1 = 0$ or $\chi_2 = 0$, then the previous binary mixtures formulation is derived.

Liao et al.[31] studied the dependence of the exponents α and β upon the equivalence ratio of a Chinese Natural Gas (from the north of Shaanxi Province), when the mixture strength was varied from 0.6 to 1.4, in the case of spherically expanding flames of natural gas/air mixtures. Initial pressures of 0.05, 0.1 and 0.15 MPa, and preheat temperatures ranging from 300 to 400 K were considered. They proposed the following second-order polynomial form

$$\alpha(\phi) = 5.75\phi^2 - 12.15\phi + 7.98$$
$$\beta(\phi) = -0.905\phi^2 + 2\phi - 1.473 \tag{13}$$

A simple third-order polynomial expression was used to fit their data at ambient conditions ($T_u = T_0 = 300$ K and $p_u = p_0 = 1$ atm), namely

$$S_{L0}(\phi) = -177.43\phi^3 + 340.77\phi^2 - 123.66\phi - 0.2297 \tag{14}$$

which does not consider the influence of the natural gas composition, since was not varied during the tests.

The expressions provided by Liao et al.[31] for the exponentials α and β appear to be the most convincing among the forms analyzed so far, since they can well reproduce influence of pressure and temperature on the lean and rich sides, in comparison to the stoichiometric case. This is appreciable from Figure 1 in which the various solutions proposed for the calculation of α and β developed by Metghalchi and Keck,[20] Gu et al.[19] and Liao et al.[31] are compared. The results by Gu et al.[19] and Liao et al.[31] look very close to each other, coherently with the fact that the natural gas investigated by Liao et al.[31] was composed of 96.16% of methane. Metghalchi and Keck's[20] expression was derived for propane, and therefore, no direct comparisons can be done with the other two. However, the expression proposed by Metghalchi and Keck[20] has a linear form, since it was derived by considering only three different equivalence ratios near stoichiometric conditions. Thus, the temperature influence is overestimated for rich and lean mixtures and underestimated for near-stoichiometric conditions (Figure 1(a)). For the same reason, the opposite is true for the pressure influence (Figure 1(b)).

The laminar burning velocity of gasoline, similarly to natural gas, has not been investigated as extensively as other pure compounds. Gasoline is a complex fuel mixture, with large variations in compositions between different commercial gasolines. As a result, there is no fixed laminar burning velocity. This also explains why it is currently not possible to represent the complex chemistry of gasoline in a chemical kinetic model and surrogates are usually used.[14,41] However, Sileghem et al.[14] recently measured the laminar burning velocities for a gasoline (Exxon 708629-60) using the heat flux method on a flat flame adiabatic burner and they found good agreement both with the data of Dirrenberger et al.,[42] who used the same method to measure the laminar burning velocity, and with Zhao et al.[43] (at 353 K)
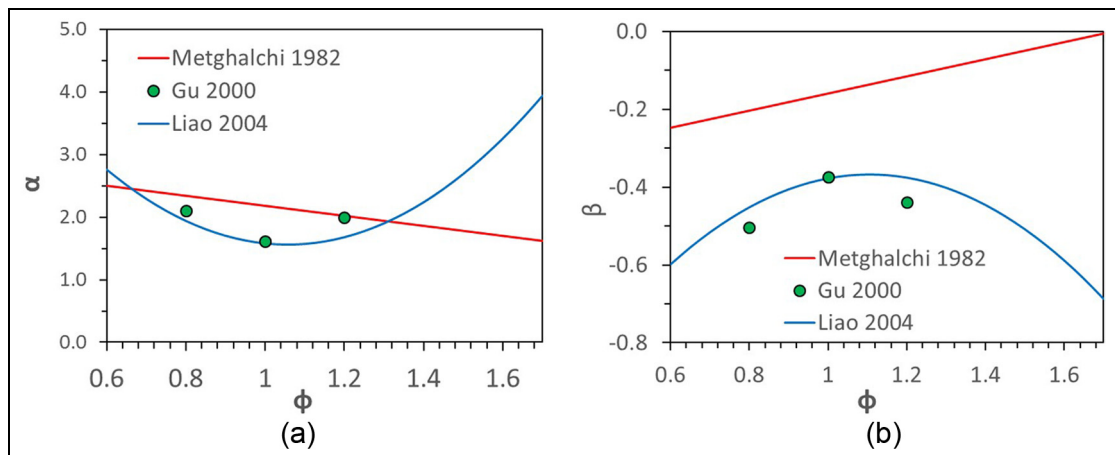

Figure 1. Dependence of exponents α (a) and β (b) upon equivalence ratio φ. Comparison between the solutions proposed by Metghalchi and Keck[20] (red line), Gu et al.[19] (green dots) and Liao et al.[31] (blue line).

who used the stagnation jet-wall flame configuration and particle image velocimetry.

In the same study, the temperature dependence of the laminar burning velocity was also shown for gasoline, for equivalence ratios varying from 0.7 to 1.3 and for temperatures between 298 and 358 K. The temperature dependence of gasoline was compared to the empirical formulations by Gülder[10] and Metghalchi and Keck[20] and a correlation implemented in GT-Power based on the publication by Takashi and Kimitoshi[44] The result was that none of the correlations captured the temperature dependence of gasoline. A second-order polynomial, as in Liao et al.[31] (equation (13)), was then suggested to represent the power exponent α

$$\alpha(\phi) = 3.28\phi^2 - 7.52\phi + 5.93 \tag{15}$$

Unfortunately, no correlation for $S_{L0}(\phi)$ or for pressure dependence was provided. Experimental measurements of laminar flame speeds obtained by various workers are compared and analyzed next for each of the fuels considered in this study. A comparison with the above-discussed empirical correlations is then reported.

Results and comparisons

The empirical correlations developed for pure methane and propane, methane/ethane and methane/propane mixtures, as well as for natural gas and gasoline, are discussed next. Results for the considered studies are listed in tables, together with the method that was used, the range of the equivalence ratios, pressures and temperatures that were explored, and the fuels that were considered in the specific study.

As previously mentioned, all the correlations proposed in this work have the "power law" form of equation (1), with $p_0 = 1$ atm and $T_0 = 298$ K. For all the fuels considered, the $S_{L0}(\phi)$ term is represented using the "Gülder's exponential formulation" of equation (10), while the exponents α and β were considered to be functions of the mixture strength ϕ and the second-order polynomial fitting proposed by Liao et al.[31] (equation (13)) is considered, namely

$$\begin{aligned} \alpha(\phi) &= a_2\phi^2 - a_1\phi + a_0 \\ \beta(\phi) &= -b_2\phi^2 + b_1\phi - b_0 \end{aligned} \tag{16}$$

An iterative reweighted least squares algorithm[45] was performed for the parameter estimation. In particular, a nonlinear regression using ordinary least squares, coupled with the Levenberg–Marquardt algorithm,[46] was adopted.

Methane

Methane is often considered as a reference gas for combustion studies and it has been studied for a large range of conditions. However, the data start to become scarce at high pressures and temperatures, due to experimental difficulties.

The literature experiments and empirical correlations for methane are listed in Table 5. Figure 2 shows the results of curve fitting for the methane laminar flame speed at 1 atm and 298 K, for equivalence ratio ranging from 0.6 to 1.7. The black solid line represents $S_{L0}(\phi)$ when the coefficients proposed in Table 6 are adopted.

Figure 2 also offers a comparison between the present and other empirical correlations. Gülder[10] and Dirrenberger et al.[32] used the same form that is considered in the present study (equation (10)), but with different coefficients (listed in Table 3). The correlation proposed by Elia et al.[28] (equation (4)) does not reproduce the trend in a satisfactory way, while that proposed by Ouimette and Seers[30] (equation (9)) is closer to the best fit, although it overestimates the flame speed on the rich side and does not follow the trend for equivalence ratios larger than 1.3 (above 1.4 it gives negative results, which must be avoided).

Figure 3 summarizes pressure effects over the range of equivalence ratios considered. The obtained coefficients for the exponents α and β in equation (16) are listed in Table 7. Figure 3(a) shows good agreement with the various experiments and with pressures ranging from 1 to 20 atm. Figure 3(b) and (c) depicts the above-mentioned limitations shown by Elia and Ouimette's correlations. In addition, the latter overestimates the laminar burning velocity for all equivalence ratios at higher pressures.

Figure 4(a) summarizes the effect of the pressure for the stoichiometric case. In this case, it was also possible to compare the result with the correlations proposed by Han et al.[29] (equations (5) and (6)), Gu et al.[19] (Table 2) and Hu et al.[13] (equations (7) and (8)). The results appear to be close for ambient temperature conditions. Han, Gu and Ouimette's correlations underestimate the pressure influence, while Hu's correlation overestimates it in the low-pressure range. More problems arise when the correlations are compared with available data at higher initial temperatures. Han's correlation is not able to match the trend anymore, while Ouimette's formulation produces too high values at all considered initial pressures and highlights the limits of a correlation exclusively based on numerical results. Elia's correlation shows good agreement with their own data, but some discrepancies with other experiments start to appear.

In Figure 4(b), results for the case of $\phi = 1.2$ are shown. This is one of the most studied high pressure cases, as can be inferred from the data reported in Table 5. Analogous conclusions to those derived for the stoichiometric case regarding the correlations proposed by Gu and Ouimette can be made and Elia's correlation produces too low values at lower pressures.

Figure 5 reports the effects of an increase of unburnt mixture temperature, considering different initial pressures. For the case of room pressure (Figure 5(a)), a

Table 5. Literature considered for methane.

Ref.	Authors	Year	Fuels	ϕ	T_u (K)	p_u (atm)	Method
17	Ryan and Lestz	1980	Methane, n-heptane, methanol and propane	0.8–1	470–570	1–20	Constant-volume bomb
10	Gülder	1984	Methane, propane, isooctane, methanol, ethanol, alcohol/isooctane and alcohol/water	0.7–1.4	298–800	1–20	Constant-volume bomb, correlations/correlations
47	Hassan et al.	1998	Methane	0.6–1.35	298	0.5–4	Outwardly spherical bomb
19	Gu et al.	2000	Methane	0.8, 1 and 1.2	300–400	1–10	Constant-pressure spherical comb. chamber/correlations
28	Elia et al.	2001	Methane, methane/diluent mixtures	0.8–1.2	298–550	0.75–70	Spherical vessel/correlations
48	Rozenchan et al.	2002	Methane, methane/oxygen, methane/oxygen/helium	0.6–1.4	298	1–60	Constant-pressure combustion chamber
49	Bosschaart and De Goey	2004	Methane, ethane, propane, n-butane	0.6–1.6	293–360	1	Heat flux
29	Han et al.	2006	Methane, methane/hydrogen/carbon monoxide mixtures	1	298–498	1–5	Cylindrical bomb/correlations
40	Coppens et al.	2007	Methane/hydrogen/nitrogen mixtures	0.7–1.5	300	1	Heat flux
30	Ouimette and Seers	2009	Methane, wood syngas	0.5–1.5	300–850	1–15.2	Burner for syngas numerical simulations for CH_4/correlations
50	Bourque et al.	2010	Methane, natural gas	0.7–1.3	298	1–4	Cylindrical bomb with schlieren setup
51	Park et al.	2011	Methane, C_1-C_4/hydrogen/CO mixtures	0.5–1.3	298 (and 423)	1–4	Counter flow
52	Lowry et al.	2011	Methane, ethane, propane, methane/ethane and methane/propane mixtures	0.7–1.3	298	1–10	Constant-volume vessel with schlieren optical setup
32	Dirrenberger et al.	2011	Methane, ethane, propane, n-butane methane/ethane and methane/propane mixtures, natural gas	0.6–2.1	298	1	Heat flux method with flame adiabatic burner/correlations
13	Hu et al.	2015	Methane	0.7–1.4	298–443	1–5	Constant-volume bomb/correlations
53	Li et al.	2015	Methane/n-heptane mixtures	0.7–1.5	358–428	1	Outwardly cylindrical constant-volume combustion chamber

Table 6. Coefficients proposed for the term $S_{L0}(\phi)$ in equation (10) for methane, propane and gasoline.

Fuel	Z	W (cm/s)	η	ξ	σ
CH_4	1	38.85	-0.20	6.45	1.08
C_3H_8	1	42.11	-0.25	5.24	1.10
Gasoline	1	36.82	-0.22	4.86	1.11

Table 7. Coefficients proposed for exponents α and β in equation (16) for methane, propane, natural gas and gasoline.

Fuel	a_2	a_1	a_0	b_2	b_1	b_0
CH_4	4.9199	10.287	6.9258	1.3712	2.6808	1.7492
C_3H_8	2.7620	5.8808	4.9221	0.9250	2.0000	1.3560
Natural gas	5.7500	12.150	7.9800	0.9250	2.0000	1.3650
Gasoline	3.2800	7.5200	5.9300	0.9250	2.0120	1.3650

comparison with Ouimette's correlation is also reported, which is not able to reproduce the pressure influence in any of the equivalence ratios considered. However, the present correlation shows good agreement in all considered cases.

The initial temperature effects are summarized for the stoichiometric case in Figure 6. Different unburnt mixture pressures are considered as well. At room pressure, the agreement with the experimental data is good,
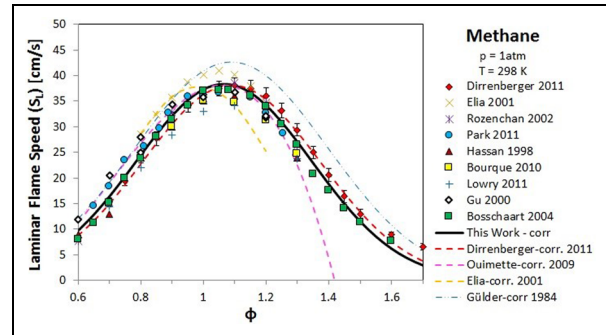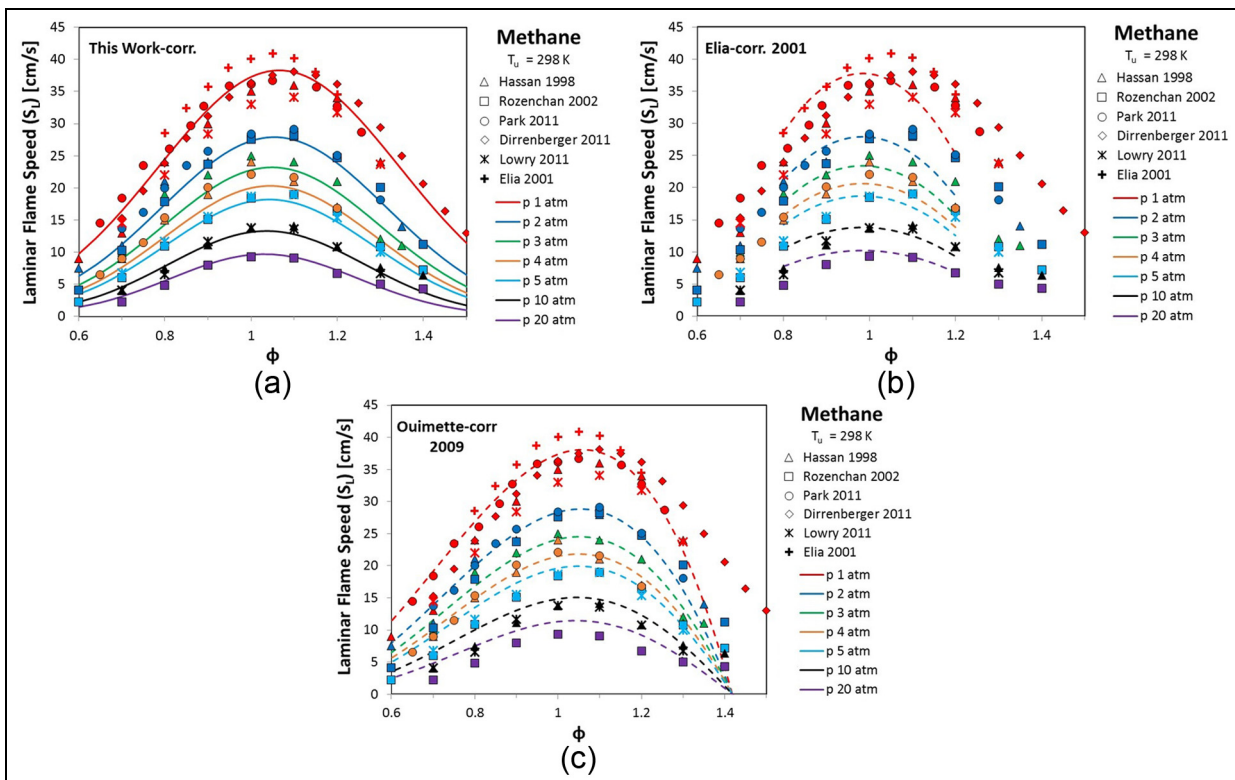

Figure 2. Methane laminar flame speed at room conditions. Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.

while the other empirical correlations considered tend to overestimate the temperature influence. At higher pressures, the agreement can be considered satisfactory.

However, the data become scarce when lean and rich mixtures are considered. Therefore, measurements that consider high initial pressures and temperatures are needed for a deeper analysis.

Propane

Propane is usually used in many applications, for example, laboratory studies of oxidation processes and

Figure 3. Methane laminar flame speed at room temperature and different pressures. Correlation in this work (a) and correlations of Elia et al.[28] (b) and Ouimette and Seers[30] (c). Marks: experimental data; solid line: empirical correlation proposed in this work; dashed lines: other correlations.
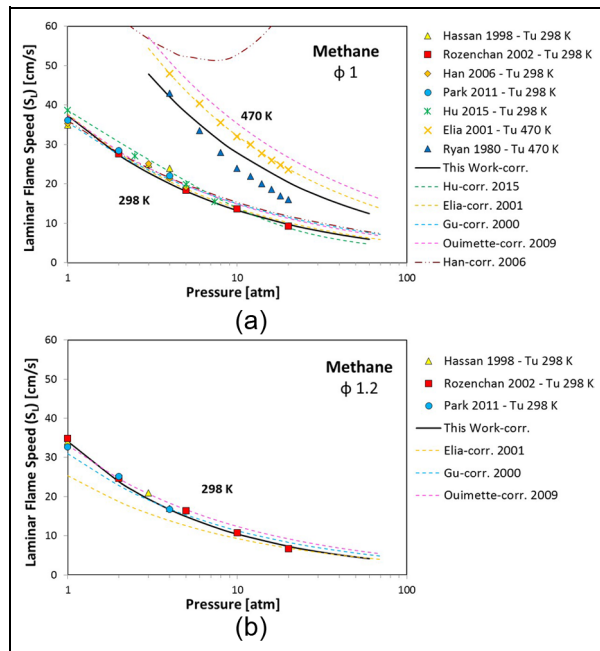
Figure 4. Initial pressure influence on methane laminar flame speed, considering an equivalence ratio equal to 1 (a) and 1.2 (b). Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.

internal combustion engines. Unlike hydrocarbons fuel with simple structures such as methane and ethane, the thermochemical and combustion properties of propane are similar in many ways to those of a more complex practical fuel.[10] For this reason, a considerable body of studies has been focused on the measurement of its laminar burning velocity. As previously done for methane, the literature considered for propane in the present work is summarized and listed in Table 8.

The result of the curve fitting for propane at 1 atm and 298 K is reported in Figure 7 and the coefficients for propane are listed in Table 6. Some of the empirical correlations available in the literature are also plotted. Similar findings as those for methane are seen for the correlations proposed by Gülder[10] and Dirrenberger et al.[32] The well-known Metghalchi's correlation (equation (3)) is not far from the best fit, but cannot be adopted for equivalence ratios lower than 0.8 and larger than 1.5 due to its second-order polynomial form. The correlation proposed by Huzayyin et al.[33] consisted of a fourth-order polynomial expression for $S_{L0}(\phi)$ and although it shows reasonable agreement with the measurements from which it was developed, a large discrepancy appears when it is compared with all the other data considered, since their experimental
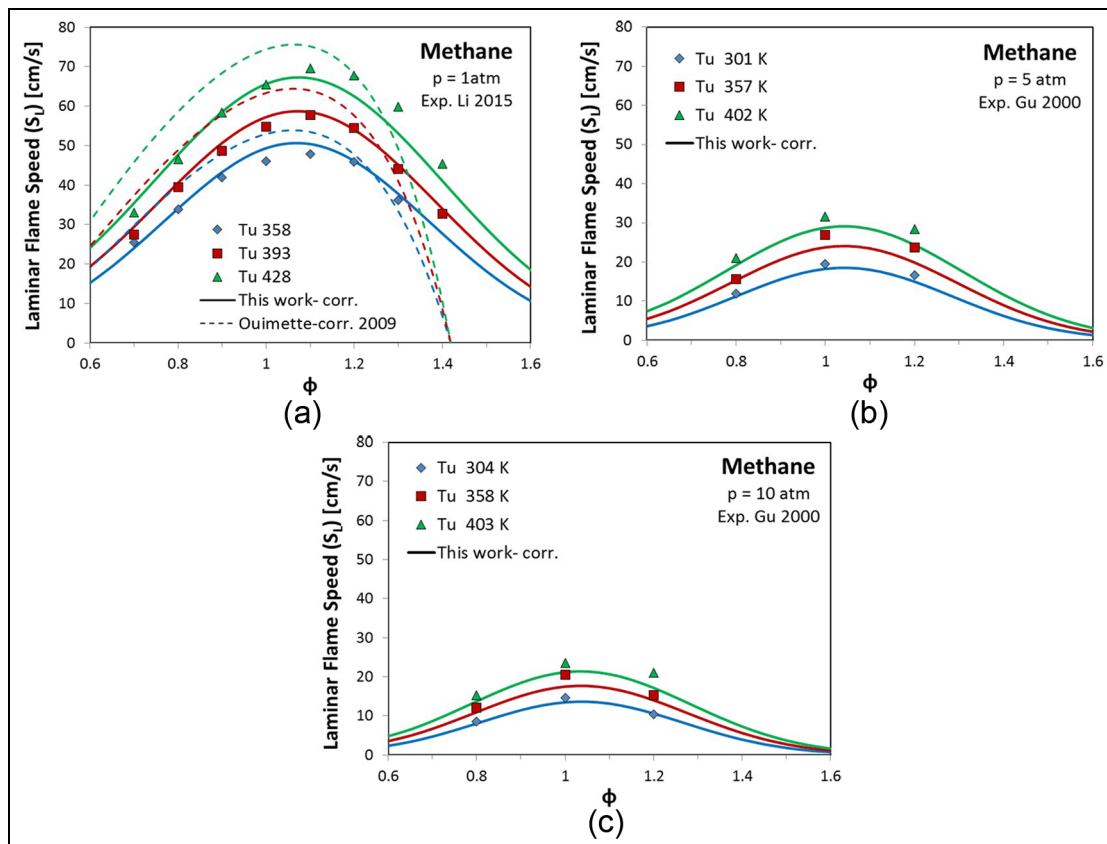

Figure 5. Methane laminar flame speed at high temperatures and pressures at different equivalence ratios. Comparisons with experimental data available at (a) 1 atm, (b) 5 atm and (c) 10 atm. Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.
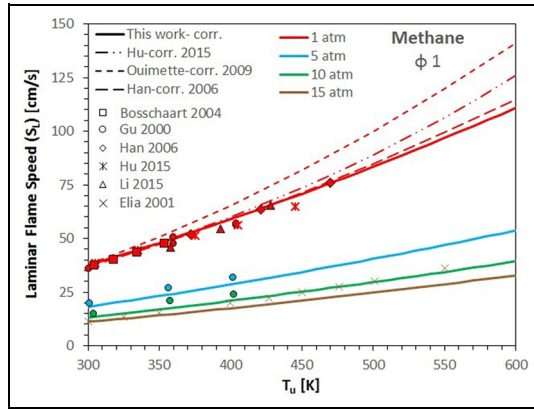
Figure 6. Initial temperature influence on methane laminar flame for the stoichiometric case and for different initial pressures. Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.
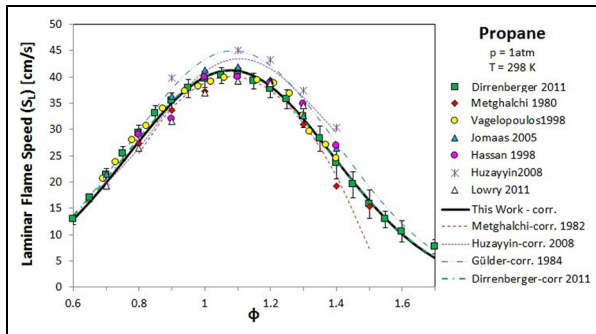

Figure 7. Propane laminar flame speed at room conditions. Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.

measurements gave values considerably higher than all the others.

The effects of initial pressure are provided in Figure 8. The obtained coefficients for the exponents α and β in equation (16) are listed in Table 7. Figure 8 shows good agreement with all the various literature experiments in which the pressure was varied for 1–5 atm. Since Gülder[10] proposed constant values for both α and β, his correlation is not able to reproduce pressure effects at different equivalence ratios, as shown in Figure 8(a). Metghalchi and Keck[20] proposed a linear expression for both α and β, and the comparison with experimental data (Figure 8(a)) makes visible the above-discussed limitations of such an expression.

Figure 9 summarizes the effect of pressure for stoichiometric propane. Experiments carried out before the 80s give a faster laminar burning velocity, since they did not consider stretch effects. In addition, Figure 9 shows that the value chosen by Gülder[10] for exponent β, which was in perfect agreement with his experiments, gives the fastest laminar flame speed among those that

Table 8. Literature considered for propane.

| Ref.	Authors	Year	Fuels	ϕ	T_u (K)	p_u (atm)	Method
18	Dugger	1952	Methane, propane, ethylene	0.7–1.5	200–617	1–11	Burner with total area of outer edge of the cone shadow
54	Kuehl	1961	Propane	0.7–1.7	311–866.5	0.23–1	Modified slot burner with square burner tube
55	Agnew and Graiff	1961	Methane, propane, ethylene, acetylene, hydrogen	1	298	0.2–20	Constant-volume bomb
56	Chase and Weinberg	1963	Acetylene, propane	0.8 and 1	280–520	0.5–2.3	Burner
12	Metghalchi and Keck	1980	Propane	0.8–1.5	298–750	0.4–40	Spherical bomb
20	Metghalchi and Keck	1982	Methanol, isooctane, indolene	0.8–1.5	298–700	0.4–50	Constant-volume bomb/correlations
10	Gülder	1984	Methane, propane, isooctane, methanol, ethanol, alcohol/isooctane, alcohol/water	0.7–1.4	298–800	1–20	Constant-volume bomb/correlations
57	Vagelopoulos and Egolfopoulos	1998	Methane, ethane, propane	0.6–1.5	298	1	Counter flow, opposed-jet technique
58	Hassan et al.	1998	Ethane, ethylene, propane	0.8–1.6	298	0.5–4	Outwardly spherical bomb
59	Jomaas et al.	2005	Acetylene, ethylene, ethane, propylene, propane	0.6–2	298	1–5	Spherical bomb
60	Tang et al.	2008	Propane/hydrogen	0.8 and 1.2	300–440	1–50	Spherical vessel
33	Huzayyin et al.	2008	LPG, propane	0.7–2.2	295–400	0.5–4	Cylindrical bomb/correlations
61	Razus et al.	2010	Propane	vol.%2.50–6.20	298–423	0.3–1.2	Spherical vessel
52	Lowry et al.	2011	Methane, ethane, propane methane/ethane, methane/propane	0.7–1.3	298	1–10	Constant-volume vessel with schlieren optical setup
32	Dirrenberger et al.	2011	Methane, ethane, propane, n-butane, methane/ethane, methane/propane, natural gas	0.6–2.1	298	1	Heat flux method with flame adiabatic burner/correlations
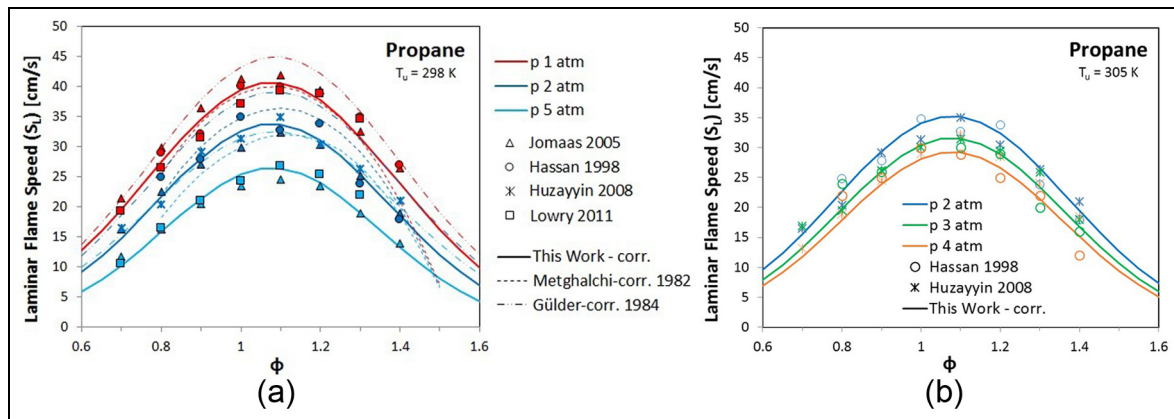
Figure 8. Initial pressure influence on propane laminar flame speed at room temperature (a) and 305 K (b). Marks: experimental data; solid line: empirical correlation proposed in this work; dashed lines: other correlations.
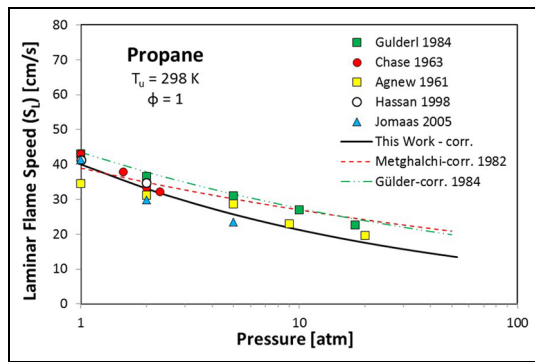

Figure 9. Initial pressure influence on propane laminar flame speed at stoichiometric conditions. Marks: experimental data; dashed lines: correlations available in the literature; solid line: empirical correlation proposed in this work.
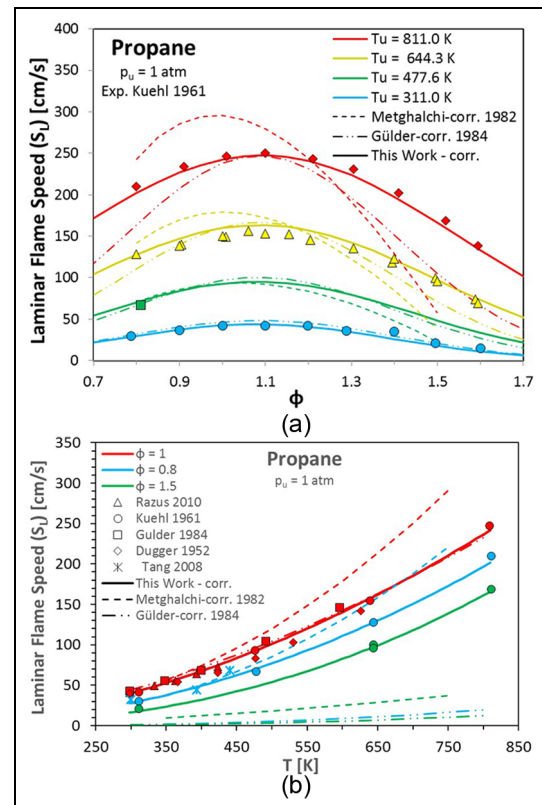

Figure 10. Initial temperature influence on propane laminar flame speed at room pressure, considering an equivalence ratio ranging from 0.7 to 1.7 (a). Comparison with other available data for equivalence ratio equal to 0.8, 1 and 1.5 (b). Marks: experimental data; solid line: empirical correlation proposed in this work; dashed lines: other correlations.

are proposed in this study. Conversely, the linear form by Metghalchi and Keck[20] tends to underestimate the pressure effects in stoichiometric conditions.

Figure 10(a) depicts the effects of an increase of unburnt mixture temperature, for initial pressure equal to 1 atm. In Figure 10(b), the effects are summarized for three different equivalence ratios, namely 0.8, 1 and 1.5, and good agreement is obtained by the present correlation. The Gülder[10] value for β is inappropriate when the initial temperature is increased, and with the linear form of Metghalchi and Keck,[20] the temperature influence is overestimated for rich mixtures and underestimated for near-stoichiometric and slightly lean conditions.

Methane/propane and methane/ethane mixtures

A study of binary mixtures of methane with ethane and propane allows the development of empirical correlations to reproduce the laminar flame speed of different types of natural gas, since the methane volume fraction can vary between 55.8% and 98.1%, ethane can vary between 0.5% and 13.3% by volume, and propane can vary between 0% and 23.7% by volume.[62]

Dirrenberger et al.[32] proposed a modified version of Gülder's expression to take into account the presence of another compound with methane. They found that their correlation reproduced the experimental results for lean and rich mixtures in a satisfactory way, but it overestimated flame velocities near stoichiometry. This is because their modifications considered only the

Table 9. Coefficients of equation (17) for binary mixtures.

Fuel	ν	τ	ε	Ω
CH_4/C_2H_6	0.20	1.50	0.95	0.09
CH_4/C_3H_8	0.10	1.50	1.30	0.20

influence on the peak amplitude and position. From their experimental data, it seems that the lean and the rich side are more sensitive to the addition of another compound. Therefore, in order to take into account such behavior, the coefficient η in equation (11) has

been multiplied by the term $(1 - \chi)^\varepsilon$, resulting in the following expression

$$S_{L0}(\phi, \chi) = (1 + \nu\chi^\tau)W\phi^{\eta(1-\chi)^\varepsilon}e^{-\xi(\phi-\sigma-\Omega\chi)^2} \quad (17)$$

The coefficients ν, τ, ε and Ω derived in this study for methane/ethane and methane/propane mixtures are reported in Table 9. The terms η, ξ and σ refer to pure methane (Table 6).

The works that were considered for the analysis are reported in Table 10. Figure 11 shows the results for different fractions of ethane in methane, while Figure 12 refers to methane/propane mixtures. From Figures

Table 10. Literature considered for methane/ethane and methane/propane mixtures.

| Ref.	Authors	Year	Fuels	Phi	T_u (K)	p_u (atm)	Method
63	Kishore et al.	2008	Methane, ethane, methane/ethane mixtures	0.7–1.3	307	1	Heat flux
52	Lowry et al.	2011	Methane, ethane, propane methane/ethane, methane/propane	0.7–1.3	298	1–10	Constant-volume vessel with schlieren optical setup
32	Dirrenberger et al.	2011	Methane, ethane, propane, n-butane, methane/ethane, methane/propane, natural gas	0.6–2.1	298	1	Heat flux method with flame adiabatic burner/correlations
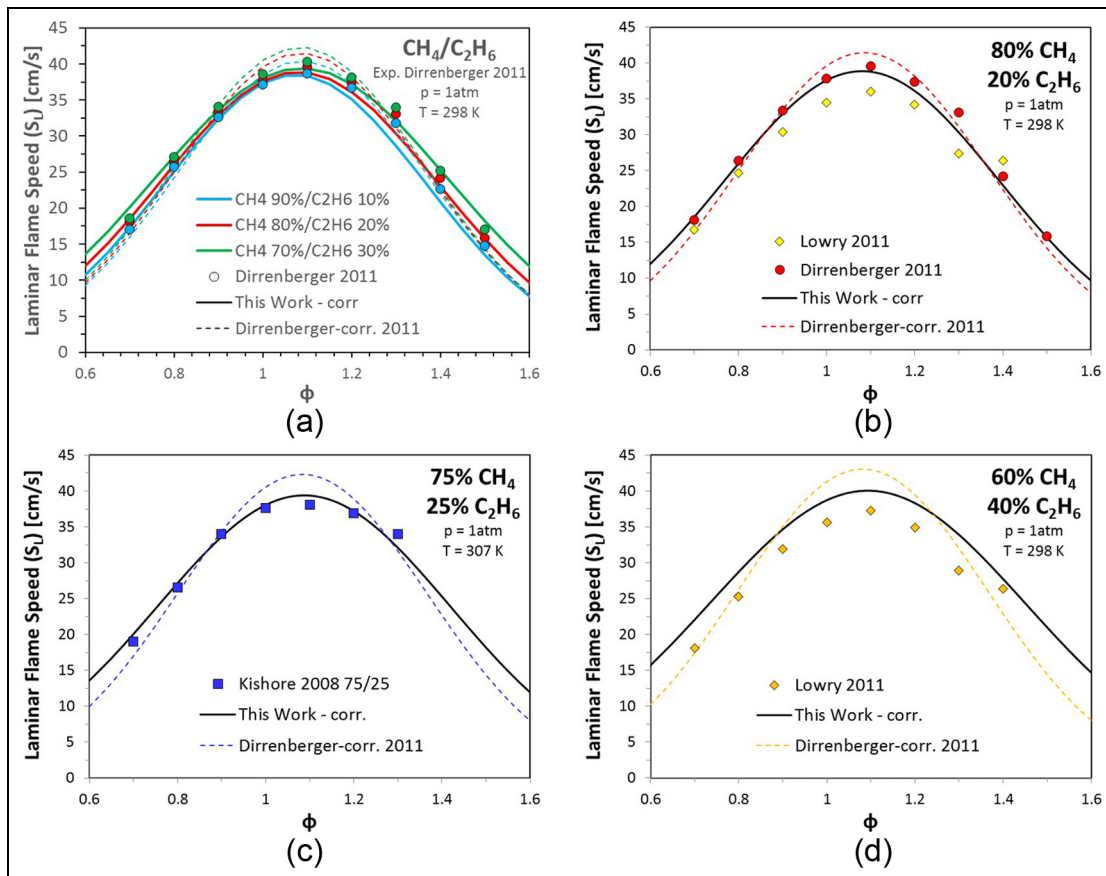
Figure 11. Laminar flame speed of methane/ethane mixtures at room conditions, considering different ethane contents in methane. Marks: experimental measurements carried out by Dirrenberger et al.[32] (a), Lowry et al.[52] and Dirrenberger et al.[32] (b), Kishore et al.[63] (c), Lowroy et al.[52] (d); dashed lines: correlation proposed by Dirrenberger et al.[32]; solid line: empirical correlation proposed in this work.
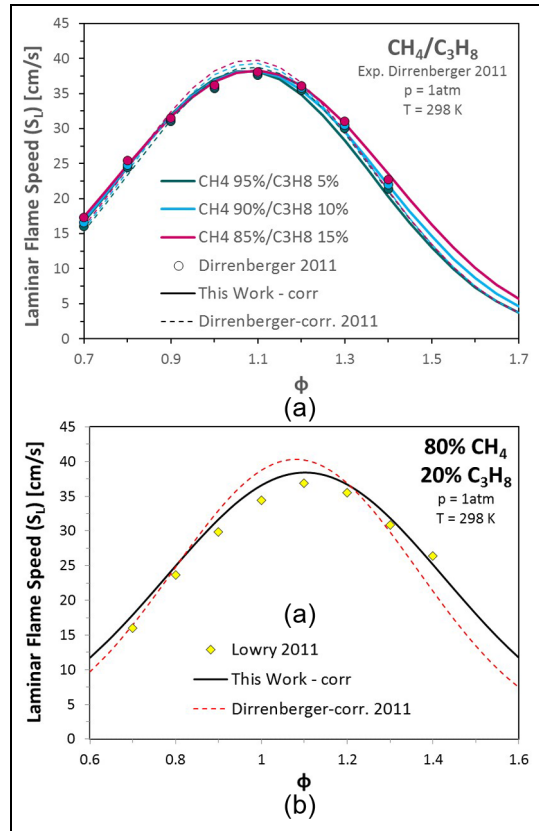
Figure 12. Laminar flame speed of methane/propane mixtures at room conditions, considering different ethane contents in methane. Marks: experimental measurements carried out by Dirrenberger et al.[32] (a), Lowry et al.[52] (b); dashed lines: correlation proposed by Dirrenberger et al.[32]; solid line: empirical correlation proposed in this work.

11(a) and 12(a), it is seen that the proposed correlation reproduces the experimental trends better than the formulation proposed by Dirrenberger et al.,[32] for all equivalence ratios considered. It captures the greater

sensitivity to the addition of other compounds in methane for lean and rich mixtures.

Figure 11(b)–(d) offers a comparison with other experimental measurements for methane/ethane, and Figure 12(b) compares methane/propane mixtures. The overall agreement can be considered satisfactory.

Natural gas

Natural gas is increasingly used as an alternative to petroleum fuels in internal combustion engines and industrial power plants[64–68] because of its smaller environmental effects compared to diesel and gasoline,[4,66,69–72] as well as for economic reasons.[73] New combustion techniques[37,74] are related to the use of natural gas, as well as their control strategies.[75–78] Dirrenberger et al.[32] proposed a correlation valid for a natural gas surrogate mixture of methane, ethane and propane, which was obtained by combining the expressions derived for binary methane/ethane and methane/propane mixtures. The same approach has been adopted in this study, resulting in the following expression

$$S_{L0}(\phi, \chi_1, \chi_2) = (1 + \nu\chi_1^{\tau_1})(1 + \nu\chi_2^{\tau_2})$$
$$W\phi^{\eta(1-\chi_1)^{\varepsilon_1}(1-\chi_2)^{\varepsilon_2}} e^{-\xi(\phi-\sigma-\Omega_1\chi_1-\Omega_2\chi_2)^2} \quad (18)$$

in which the terms W, η, ξ and σ refer to pure methane (Table 6), while the coefficients ν, τ, ε and Ω for ethane and propane are the same as derived in the previous section, and are reported in Table 9. The works considered are listed in Table 11.

Dirrenberger et al.[32] studied three surrogate mixtures with compositions close to those of three representative natural gases: Indonesia, Abu Dhabi and Pittsburgh; Table 12 shows the exact composition of these natural gases. In such study, they were represented by the

Table 11. Literature considered for natural gas.

Ref.	Authors	Year	Fuels	Phi	T_u (K)	p_u (atm)	Method
31	Liao et al.	2004	Shaanxi natural gas	0.6–1.4	300–400	0.5–1.5	Spherical bomb
50	Bourque et al.	2010	Methane, natural gas	0.7–1.3	298	1–4	Cylindrical bomb with schlieren setup
32	Dirrenberger et al.	2011	Methane, ethane, propane, butane, methane/ethane, methane/propane, natural gas	0.6–2.1	298	1	Heat flux method with flame adiabatic burner/correlations

Table 12. Composition of different natural gases (% volume) considered.

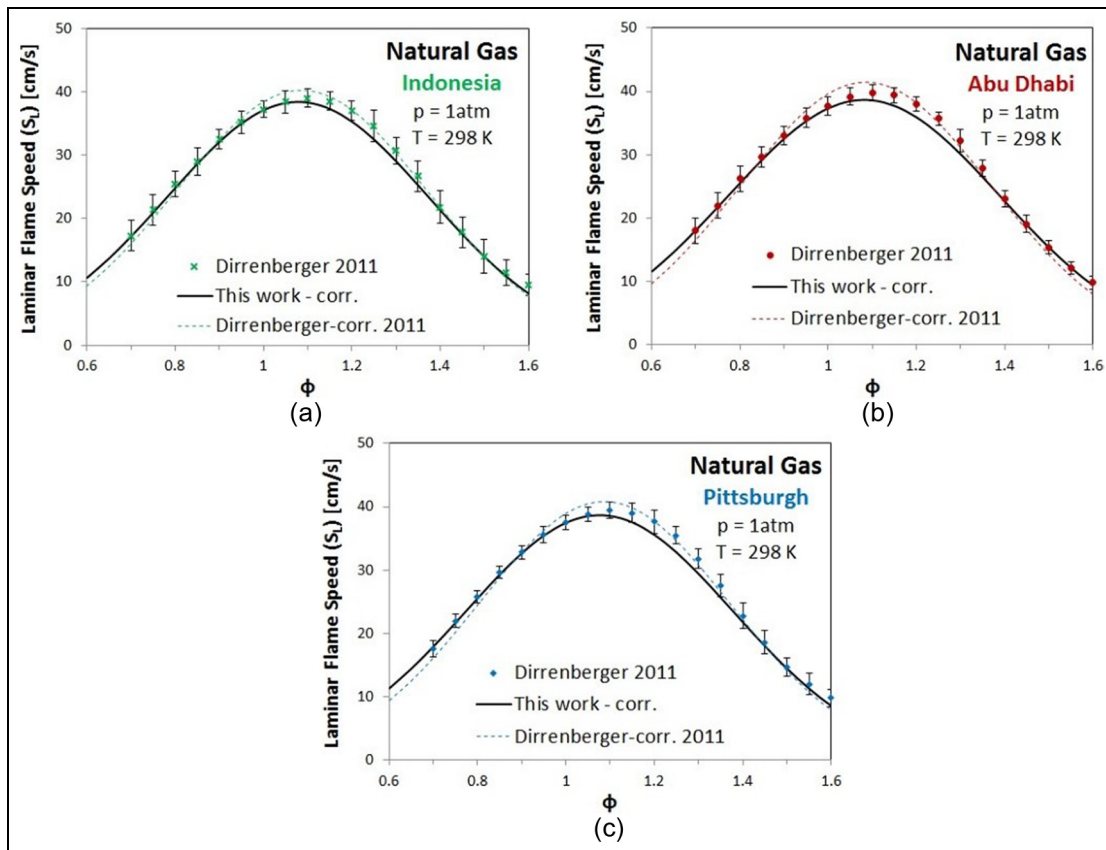| Ref.	Authors	Year	Fuels	CH_4	C_2H_6	C_3H_8	$i\text{-}C_4H_{10}$	$n\text{-}C_4H_{10}$	$i\text{-}C_5H_{12}$	$n\text{-}C_5H_{12}$	CO_2	N_2	Others
32	Dirrenberger et al.	2011	Indonesia	89.91	5.44	3.16	1	0.75	0.03	–	–	0.04	–
			Abu Dhabi	82.07	15.86	1.89	–	0.06	–	–	–	0.05	–
			Pittsburgh	85.00	14.0	–	–	–	–	–	–	1.00	–
50	Bourque et al.	2010	NG2	81.25	10.0	5.0	–	2.50	–	1.25	–	–	–
			NG3	62.50	20.0	10.0	–	5.00	–	2.50	–	–	–
31	Liao et al.	2004	Shaanxi	96.16	1.096	–	–	–	–	–	–	–	2.74

Figure 13. Laminar flame speeds of different natural gases at room conditions. Marks: experimental measurements carried out by Dirrenberger et al.[32]; dashed lines: correlation proposed by Dirrenberger et al.[32]; solid line: empirical correlation proposed in this work.

following mixtures: 90% CH_4, 6% C_2H_6 and 4% C_3H_8 Indonesia; 82% CH_4, 16% C_2H_6 and 2% C_3H_8 Abu Dhabi and 85% CH_4 and 15% C_2H_6 Pittsburgh. The results for each natural gas are reported in Figure 13, together with a comparison with the empirical correlation proposed by Dirrenberger et al.[32] (equation (12)). The dependence upon the equivalence ratio and the fuel composition is well captured by the present proposed correlation and it shows better agreement, especially near stoichiometry.

Table 12 also reports the composition of the two natural gas mixtures that were the focus of the study by Bourque et al.;[50] they were represented by the following mixtures: 85% CH_4, 10% C_2H_6 and 5% C_3H_8 for NG2; 70% CH_4, 20% C_2H_6 and 10% C_3H_8 for NG3. The results for each natural gas are reported in Figure 14(a). The correlation proposed by Dirrenberger et al.[32] showed a good agreement with their own data, but discrepancies start to appear when it is compared with the measurements carried out by Bourque et al.[50] In particular, it overestimates the maximum flame speed. Therefore, the correlation proposed in this study shows an overall better agreement with a larger set of experiments.

Figure 14(b) reports results for the natural gas investigated by Liao et al.[31] Its composition is reported in Table 12 as well. It was represented by considering 98.9% CH_4 and 1.1% C_2H_6. The values of the laminar flame speed reported in this study appear to be higher than those of the cases previously investigated, even though the natural gas was composed almost exclusively of methane. These experimental measurements report values that are higher than those presented previously for pure methane. Therefore, it is hard to judge the results shown in Figure 14(b).

Very few works have investigated the influence of initial pressure and temperature on natural gas laminar flame speed. Figure 15(a) shows results for different initial pressures and Figure 15(b) shows different initial temperatures. Only the stoichiometric case has been investigated. The derived values for the coefficients of exponents α and β in equation (16) are listed in Table 7. Liao et al.[31] investigated the temperature influence (the coefficients of exponent α are the same proposed by Liao et al.[31]).

Gasoline

Similarly to the case of natural gas mixtures, much less data than for methane and propane are available for fuels with low vapor pressure. Laminar flame speed data of commercial gasoline are summarized in Table 13.
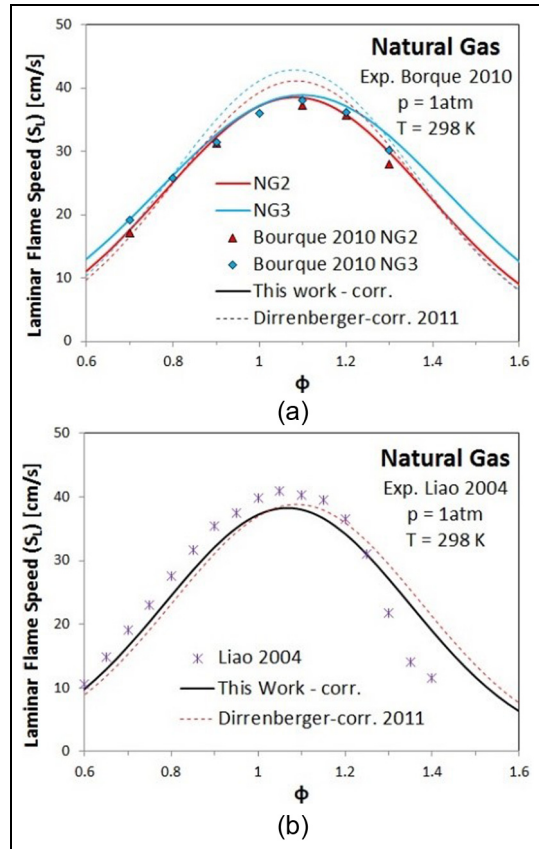
Figure 14. Laminar flame speed of different natural gases measured by Bourque et al.[50] (a) and Liao et al.[31] at room conditions. Marks: experimental data; dashed lines: correlation proposed by Dirrenberger et al.;[32] solid line: empirical correlation proposed in this work.

Figure 15. Initial pressure (a) and temperature (b) influence on natural gas laminar flame speed at stoichiometric conditions. Marks: experimental data; solid lines: empirical correlation proposed in this work.

For gasoline, the term $S_{L0}(\phi, \chi)$ is modeled in the same way as for methane and propane. This is because variations in the composition of gasoline have not been taken into account in any flame speed measurements. An approach similar to that adopted for natural gas would be preferable, but, to make it possible, further experimental investigations are needed in which a variety of gasolines is investigated. Figure 16 compares the present proposed correlation for gasoline (obtained using the coefficients reported in Table 6) with experimental measurements found in the
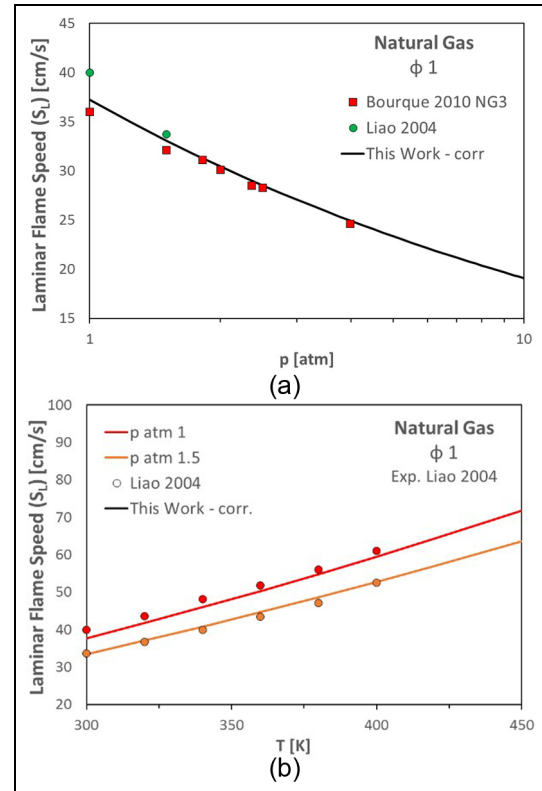
literature and carried out at 358 and 353 K and room pressure.

Sileghem et al.[14] investigated the influence of the initial temperature from 298 to 358 K at room pressure. Their experimental data are reported in Figure 17(a). They used the same second-order polynomial form for the exponents α and β of equation (16) for fitting the data. However, the coefficients derived in the present work differ from those suggested by Sileghem et al.[14] and this is due to the fact that additional experimental measurements were considered in this analysis, as shown in Figure 17(b).

Jerzembeck et al.[79] carried out measurements for unburnt mixture pressures higher than 1 atm, as

Table 13. Literature considered for gasoline.

| Ref.	Authors	Year	Fuels	Phi	T (K)	p (atm)	Method
43	Zhao et al.	2003	Gasoline (CR-87)	0.6–1.4	353 and 500	1	Single jet-wall stagnation flame
79	Jerzembeck et al.	2009	n-heptane, isooctane, PRF 87, gasoline	0.7–1.2	373	10–25	Constant-volume bomb
80	Tian et al.	2010	2,5-Dimethylfuran, ethanol, gasoline (EN228)	0.6–2.0	323–373	1	Constant-volume vessel and high speed schlieren visualization
14	Sileghem et al.	2013	Isooctane, n-heptane, toluene, TRF, gasoline (Exxon 708629-60)	0.7–1.3	298–358	1	Heat flux method on a flat flame adiabatic burner
42	Dirrenberger et al.	2014	Isooctane/n-heptane/toluene mixture gasoline (TAE7000)	0.6–1.5	358	1	Heat flux method
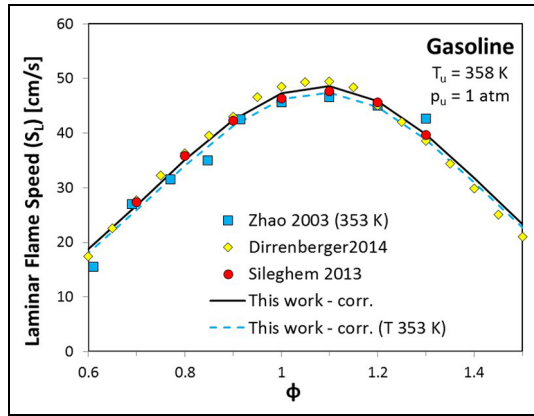
Figure 16. Gasoline laminar flame speed at room pressure and at two different temperatures. Marks: experimental data; lines: empirical correlation proposed in this work.

reported in Figure 17(c). Measurements were available at three equivalence ratios and at an initial temperature of 373 K, and the resulting coefficients for exponents α and β are reported in Table 7.

Conclusion

The present study provides simple and workable expressions, suitable for spark-ignition engine simulations, that allow laminar flame speed calculations of some practical fuels. Pure compounds, such as methane and propane, binary mixtures of methane/ethane and methane/propane, as well as more complex fuels like natural gas and gasoline, were considered. Knowing the behavior of the laminar flame speed, as a function of the unburnt mixture strength, temperature and pressure, is essential for an efficient and reliable simulation of the combustion process that occurs in a spark-ignition engine.

Measurements of laminar flame speeds in the literature were collected and used to develop empirical correlations for the laminar flame speed for equivalence ratios from 0.6 to 1.7, pressures between 1 and 50 atm and temperature from 298 to 800 K.

Empirical correlations available in the literature were also considered and it was highlighted that many of them were generally not able to give good agreement with recent experimental data. This because they were generally based on a single set of measurements and fail outside the considered experimental range. The correlations proposed in this study showed an overall better agreement with a larger set of experiments and over a wide range of operating conditions.

The correlations proposed in this work are here summarized for the reader's convenience. Each of them is based on the "power law" form
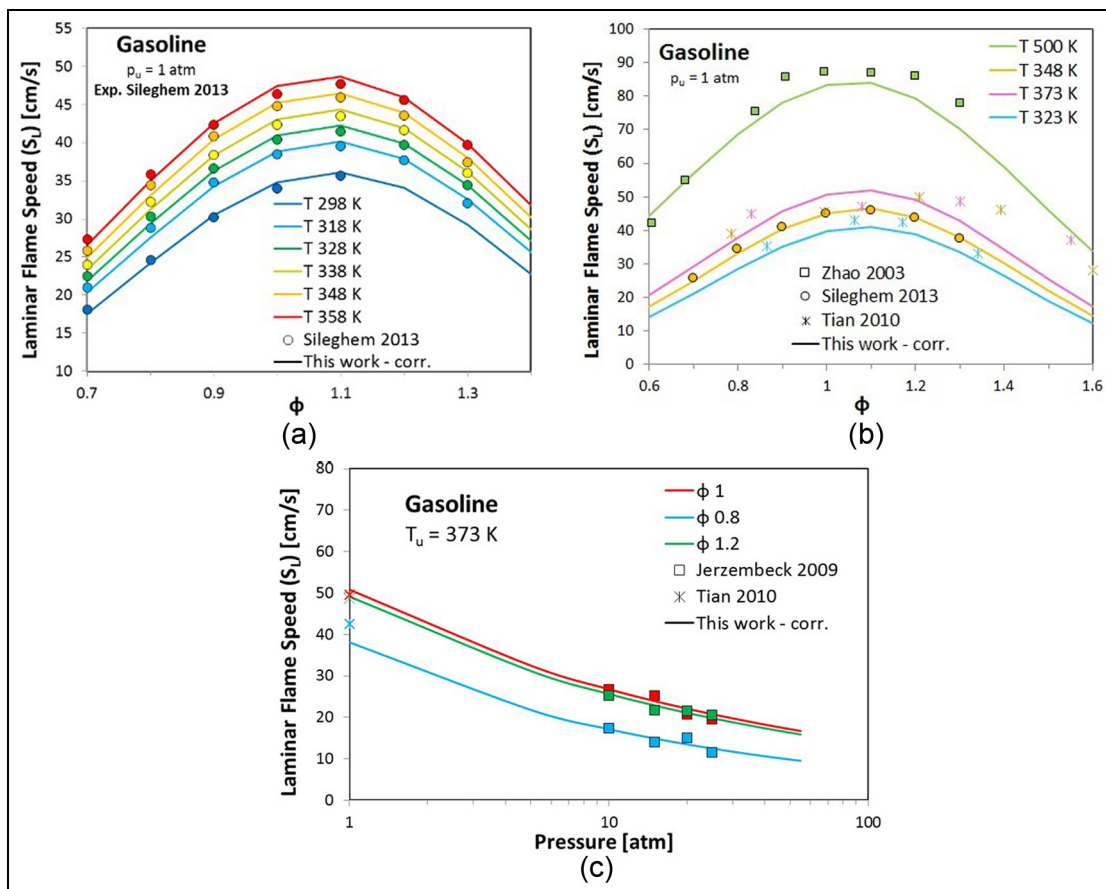

Figure 17. Initial temperature influence on gasoline laminar flame speed at room pressure (a) and (b). Initial pressure influence at three different equivalence ratios and at an initial temperature of 373 K (c). Marks: experimental data; lines: empirical correlation proposed in this work.

Table 14. Coefficients proposed for the term $S_{L0}(\phi)$ for methane, propane and gasoline.

Fuel	W (cm/s)	η	ξ	σ
CH_4	38.85	−0.20	6.45	1.08
C_3H_8	42.11	−0.25	5.24	1.10
Gasoline	36.82	−0.22	4.86	1.11

Table 15. Coefficients for binary mixtures of methane/ethane and methane/propane.

Fuel	ν	τ	ε	Ω
CH_4/C_2H_6	0.20	1.50	0.95	0.09
CH_4/C_3H_8	0.10	1.50	1.30	0.20

Table 16. Coefficients proposed for exponents α and β for methane, propane, natural gas and gasoline.

Fuel	a_2	a_1	a_0	b_2	b_1	b_0
CH_4	4.9199	10.287	6.9258	1.3712	2.6808	1.7492
C_3H_8	2.7620	5.8808	4.9221	0.9250	2.0000	1.3560
Natural gas	5.7500	12.150	7.9800	0.9250	2.0000	1.3650
Gasoline	3.2800	7.5200	5.9300	0.9250	2.0120	1.3650

$$S_L(\phi, T_u, p_u) = S_{L0}\left(\frac{T_u}{T_0}\right)^\alpha \left(\frac{p_u}{p_0}\right)^\beta$$

with $p_0 = 1$ atm and $T_0 = 298$ K.

For pure compounds, such as methane and propane, the laminar flame speed at ambient condition $S_{L0}(\phi)$ is represented using "Gülder's exponential formulation," as follows

$$S_{L0}(\phi) = W\phi^\eta e^{-\xi(\phi-\sigma)^2}$$

The coefficients η, ξ and σ derived from the experimental data interpolation are reported in Table 14.

For binary and ternary mixtures, it was shown that the influence that the amount of the secondary compounds has on the mixture laminar flame speed is different at different equivalence ratios which has not been considered in previous formulations. Therefore, in the case of binary mixtures, the following modified expression for the term S_{L0} was proposed and better overall agreements with all the experimental data were obtained

$$S_{L0}(\phi, \chi) = (1 + \nu\chi^\tau)W\phi^{\eta(1-\chi)^\varepsilon} e^{-\xi(\phi-\sigma-\Omega\chi)^2}$$

The coefficients ν, τ, ε and Ω derived in this study for methane/ethane and methane/propane mixtures are reported in Table 15, while the terms W, η, ξ and σ refer to pure methane (Table 14).

An improved formulation for S_{L0} was developed for natural gas, which was modeled as a ternary mixture of methane, ethane and propane. And it appears as follows

$$S_{L0}(\phi, \chi_1, \chi_2) = (1 + \nu\chi_1^{\tau_1})(1 + \nu\chi_2^{\tau_2})$$
$$W\phi^{\eta(1-\chi_1)^{\varepsilon_1}(1-\chi_2)^{\varepsilon_2}} e^{-\xi(\phi-\sigma-\Omega_1\chi_1-\Omega_2\chi_2)^2}$$

where subscript 1 refers to parameters calculated for ethane, and subscript 2 refers to propane (Table 15). The terms W, η, ξ and σ refer to pure methane (Table 14). Comparisons with experimental data on natural gases having different compositions confirmed the obtained improvements.

Gasoline was treated as a single component fuel. However, the proposed correlation resulted in agreement with all the available data taken from the literature. And, a good agreement was ensured over the entire ranges of unburnt mixture pressure and temperature considered. Coefficients η, ξ and σ obtained for gasoline are reported in Table 14.

The exponents α and β for the "power law" were functions of mixture strength ϕ and a second-order polynomial fitting was considered

$$\alpha(\phi) = a_2\phi^2 - a_1\phi + a_0$$
$$\beta(\phi) = -b_2\phi^2 + b_1\phi - b_0$$

The coefficients that appear in the previous formulation for exponents α and β are reported in Table 16 for each of the fuels considered in the present work.

Declaration of conflicting interests

Funding

References

1. Liang L and Reitz RD. Spark ignition engine combustion modeling using a level set method with detailed chemistry. SAE paper 2006-01-0243, 2006.
2. Tan ZC and Reitz RD. Modeling ignition and combustion in spark-ignition engines using a level set method. SAE paper 2003-01-0722, 2003.
3. Tan Z, Kong S-C and Reitz RD. Modeling premixed and direct injection SI engine combustion using the G-equation model. SAE paper 2003-01-1843, 2003.
4. Amirante R, Distaso E, Tamburrano P and Reitz RD. Measured and predicted soot particle emissions from natural gas engines. SAE paper 2015-24-2518, 2015.
5. Singh S, Liang L, Kong S-C and Reitz RD. Development of a flame propagation model for dual-fuel partially premixed compression ignition engines. *Int J Engine Res* 2006; 7: 65–75.

6. Perini F, Ra Y, Hiraoka K, Nomura K, Yuuki A, Oda Y, et al. An efficient level-set flame propagation model for hybrid unstructured grids using the G-equation. *SAE Int J Engine* 2016; 9: 1409–1424.
7. Buckmaster J, Clavin P, Liñán A, Matalon M, Peters N, Sivashinsky G, et al. Combustion theory and modeling. *P Combust Inst* 2005; 30: 1–19.
8. Brandl A, Pfitzner M, Mooney JD, Durst B and Kern W. Comparison of combustion models and assessment of their applicability to the simulation of premixed turbulent combustion in IC-engines. *Flow Turbul Combust* 2005; 75: 335–350.
9. Veynante D and Vervisch L. Turbulent combustion modeling. *Prog Energ Combust* 2002; 28: 193–266.
10. Gülder ÖL. Correlations of laminar combustion data for alternative S.I. engine fuels. SAE paper 841000, 1984.
11. Ranzi E, Frassoldati A, Grana R, Cuoci A, Faravelli T, Kelley AP, et al. Hierarchical and comparative kinetic modeling of laminar flame speeds of hydrocarbon and oxygenated fuels. *Prog Energ Combust* 2012; 38: 468–501.
12. Metghalchi M and Keck JC. Laminar burning velocity of propane-air mixtures at high temperature and pressure. *Combust Flame* 1980; 38: 143–154.
13. Hu E, Li X, Meng X, Chen Y, Cheng Y, Xie Y, et al. Laminar flame speeds and ignition delay times of methane–air mixtures at elevated temperatures and pressures. *Fuel* 2015; 158: 1–10.
14. Sileghem L, Alekseev VA, Vancoillie J, Van Geem KM, Nilsson EJK, Verhelst S, et al. Laminar burning velocity of gasoline and the gasoline surrogate components isooctane, n-heptane and toluene. *Fuel* 2013; 112: 355–365.
15. Wu CK and Law CK. On the determination of laminar flame speeds from stretched flames. *Symp Int Combust* 1985; 20: 1941–1949.
16. Lavoie GA. Correlations of combustion data for S. I. engine calculations—laminar flame speed, quench distance and global reaction rates. SAE paper 780229, 1978.
17. Ryan TW and Lestz SS. The laminar burning velocity of isooctane, n-heptane, methanol, methane, and propane at elevated temperature and pressures in the presence of a diluent. SAE paper 800103, 1980.
18. Dugger GL. Effect of initial mixture temperature on flame speed of methane-air, propane-air, and ethylene-air mixtures, 1952, https://ntrs.nasa.gov/search.jsp?R = 19930092107
19. Gu XJ, Haq MZ, Lawes M and Woolley R. Laminar burning velocity and Markstein lengths of methane–air mixtures. *Combust Flame* 2000; 121: 41–58.
20. Metghalchi M and Keck JC. Burning velocities of mixtures of air with methanol, isooctane, and indolene at high pressure and temperature. *Combust Flame* 1982; 48: 191–210.
21. Gülder OL. Burning velocities of ethanol-air and ethanol-water-air mixtures. *Progr Astronaut Aero* 1984; 95: 181–197.
22. Koda S, Oda K, Hirano M, Hirano T and Akita K. Burning characteristics of methanol-water-air mixtures in a constant volume combustion vessel. *Combust Flame* 1982; 46: 17–28.
23. Van Tiggelen A and Deckers J. Chain branching and flame propagation. *Symp Int Combust* 1957; 6: 61–66.
24. Ferguson CR and Keck JC. On laminar flame quenching and its application to spark ignition engines. *Combust Flame* 1977; 28: 197–205.
25. Tabaczynski RJ, Ferguson CR and Radhakrishnan K. A turbulent entrainment model for spark-ignition engine combustion. SAE paper 770647, 1977.
26. Tanford C. The role of free atoms and radicals in burner flames. *Symp Combust Flame Explos Phenom* 1948; 3: 140–146.
27. Andrews GE and Bradley D. The burning velocity of methane-air mixtures. *Combust Flame* 1972; 19: 275–288.
28. Elia M, Ulinski M and Metghalchi M. Laminar burning velocity of methane–air–diluent mixtures. *J Eng Gas Turb Power* 2001; 123: 190–196.
29. Han P, Checkel MD, Fleck BA and Nowicki NL. Burning velocity of methane/diluent mixture with reformer gas addition. *Fuel* 2007; 86: 585–596.
30. Ouimette P and Seers P. Numerical comparison of premixed laminar flame velocity of methane and wood syngas. *Fuel* 2009; 88: 528–533.
31. Liao SY, Jiang DM and Cheng Q. Determination of laminar burning velocities for natural gas. *Fuel* 2004; 83: 1247–1250.
32. Dirrenberger P, Gall L, Bounaceur R, Herbinet O, Glaude P, Konnov A, et al. Measurements of laminar flame velocity for components of natural gas. *Energ Fuel* 2011; 25: 3875–3884.
33. Huzayyin AS, Moneib HA, Shehatta MS and Attia AMA. Laminar burning velocity and explosion index of LPG–air and propane–air mixtures. *Fuel* 2008; 87: 39–57.
34. Choi M, Song J and Park S. Modeling of the fuel injection and combustion process in a CNG direct injection engine. *Fuel* 2016; 179: 168–178.
35. Shiga S, Ozone S, Machacon HTC, Karasawa T, Nakamura H, Ueda T, et al. A study of the combustion and emission characteristics of compressed-natural-gas direct-injection stratified combustion using a rapid-compression-machine. *Combust Flame* 2002; 129: 1–10.
36. Zeng K, Huang Z, Liu B, Liu L, Jiang D, Ren Y, et al. Combustion characteristics of a direct-injection natural gas engine under various fuel injection timings. *Appl Therm Eng* 2006; 26: 806–813.
37. Hofmann P, Hofherr T, Hoffmann G and Preuhs J-F. Potential of CNG direct injection for downsizing engines. *MTZ Worldw* 2016; 77: 28–35.
38. Goswami M, Derks SCR, Coumans K, Slikker WJ, de Andrade Oliveira MH, Bastiaans RJM, et al. The effect of elevated pressures on the laminar burning velocity of methane + air mixtures. *Combust Flame* 2013; 160: 1627–1635.
39. Rallis CJ and Garforth AM. The determination of laminar burning velocity. *Prog Energ Combust* 1980; 6: 303–329.
40. Coppens FHV, De Ruyck J and Konnov AA. The effects of composition on burning velocity and nitric oxide formation in laminar premixed flames of $CH_4 + H_2 + O_2 + N_2$. *Combust Flame* 2007; 149: 409–417.
41. Pitz WJ, Cernansky NP, Dryer FL, Egolfopoulos FN, Farrell JT, Friend DG and Pitsch H. Development of an experimental database and chemical kinetic models for surrogate gasoline fuels. SAE paper 2007-01-0175, 2007.
42. Dirrenberger P, Glaude P-A, Bounaceur R, Le Gall H, da Cruz AP, Konnov AA, et al. Laminar burning velocity of gasolines with addition of ethanol. *Fuel* 2014; 115: 162–169.
43. Zhao Z, Conley JP, Kazakov A and Dryer FL. Burning velocities of real gasoline fuel at 353 K and 500 K. *SAE Transactions* 2003; 112: 2624–2629.

44. Takashi H and Kimitoshi T. *Laminar flame speeds of ethanol, n-heptane, iso-octane air mixtures*. International Federation of Automotive Engineering Societies (FISITA), Technical report no. F2006SC40, 2006.

45. Holland PW and Welsch RE. Robust regression using iteratively reweighted least-squares. *Commun Stat: Theor M* 1977; 6: 813–827.

46. Seber GAF and Wild CJ. *Nonlinear regression*. Hoboken, NJ: Wiley-Interscience, 2003.

47. Hassan MI, Aung KT and Faeth GM. Measured and predicted properties of laminar premixed methane/air flames at various pressures. *Combust Flame* 1998; 115: 539–550.

48. Rozenchan G, Zhu DL, Law CK and Tse SD. Outward propagation, burning velocities, and chemical effects of methane flames up to 60 ATM. *P Combust Inst* 2002; 29: 1461–1470.

49. Bosschaart KJ and De Goey LPH. The laminar burning velocity of flames propagating in mixtures of hydrocarbons and air measured with the heat flux method. *Combust Flame* 2004; 136: 261–269.

50. Bourque G, Healy D, Curran H, Zinner C, Kalitan D, de Vries J, et al. Ignition and flame speed kinetics of two natural gas blends with high levels of heavier hydrocarbons. *J Eng Gas Turb Power* 2009; 132: 021504.

51. Park O, Veloo PS, Liu N and Egolfopoulos FN. Combustion characteristics of alternative gaseous fuels. *P Combust Inst* 2011; 33: 887–894.

52. Lowry W, de Vries J, Krejci M, Petersen E, Serinyel Z, Metcalfe W, et al. Laminar flame speed measurements and modeling of pure alkanes and alkane blends at elevated pressures. *J Eng Gas Turb Power* 2011; 133: 091501.

53. Li G, Liang J, Zhang Z, Tian L, Cai Y and Tian L. Experimental investigation on laminar burning velocities and Markstein lengths of premixed methane–*n*-heptane–air mixtures. *Energ Fuel* 2015; 29: 4549–4556.

54. Kuehl DK. Laminar-burning velocities of propane-air mixtures. *Symp Int Combust* 1961; 8: 510–521.

55. Agnew JT and Graiff LB. The pressure dependence of laminar burning velocity by the spherical bomb method. *Combust Flame* 1961; 5: 209–219.

56. Chase JD and Weinberg FJ. The acetylene decomposition flame and the deduction of reaction mechanism from "global" flame kinetics. *P Roy Soc A: Math Phy* 1963; 275: 411–430.

57. Vagelopoulos CM and Egolfopoulos FN. Direct experimental determination of laminar flame speeds. *Symp Int Combust* 1998; 27: 513–519.

58. Hassan MI, Aung KT, Kwon OC and Faeth GM. Properties of laminar premixed hydrocarbon/air flames at various pressures. *J Propul Power* 1998; 14: 479–488.

59. Jomaas G, Zheng XL, Zhu DL and Law CK. Experimental determination of counterflow ignition temperatures and laminar flame speeds of C_2–C_3 hydrocarbons at atmospheric and elevated pressures. *P Combust Inst* 2005; 30: 193–200.

60. Tang C, He J, Huang Z, Jin C, Wang J, Wang X, et al. Measurements of laminar burning velocities and Markstein lengths of propane–hydrogen–air mixtures at elevated pressures and temperatures. *Int J Hydrogen Energ* 2008; 33: 7274–7285.

61. Razus D, Oancea D, Brinzea V, Mitu M and Movileanu C. Experimental and computed burning velocities of propane–air mixtures. *Energ Convers Manage* 2010; 51: 2979–2984.

62. Liss WE, Thrasher WH, Steinmetz GF, Chowdiah P and Attari A. *Variability of natural gas composition in select major metropolitan areas of the United States*. Institute of Gas, Chicago, IL, GRI report no. 92/0153, 1992.

63. Kishore VR, Duhan N, Ravi MR and Ray A. Measurement of adiabatic burning velocity in natural gas-like mixtures. *Exp Therm Fluid Sci* 2008; 33: 10–16.

64. Wang Q, Chen X, Jha AN and Rogers H. Natural gas from shale formation—the evolution, evidences and challenges of shale gas revolution in United States. *Renew Sust Energ Rev* 2014; 30: 1–28.

65. Amirante R, Cassone E, Distaso E and Tamburrano P. Overview on recent developments in energy storage: mechanical, electrochemical and hydrogen technologies. *Energ Convers Manage* 2017; 132: 372–387.

66. Cho HM and He BQ. Spark ignition natural gas engines—a review. *Energ Convers Manage* 2007; 48: 608–618.

67. Amirante R, Distaso E, Di Iorio S, Sementa P, Tamburrano P, Vaglieco BM, et al. Effects of natural gas composition on performance and regulated, greenhouse gas and particulate emissions in spark-ignition engines. *Energ Convers Manage* 2017; 143: 338–347.

68. Amirante R, Coratella C, Distaso E and Tamburrano P. A small size combined system for the production of energy from renewable sources and unconventional fuels. *Energy Proced* 2015; 81: 240–248.

69. Jayaratne ER, Ristovski ZD, Meyer N and Morawska L. Particle and gaseous emissions from compressed natural gas and ultralow sulphur diesel-fuelled buses at four steady engine loads. *Sci Total Environ* 2009; 407: 2845–2852.

70. Ayala A, Kado N, Okamoto R and Gebe M. CNG and diesel transit bus emissions in review. In: *Proceedings of the 9th diesel engine emissions reduction conference work 2003*, Newport, RI, 24–28 August 2003, p.6. California, US: California Environmental Protection Agency, Air Resources Board Sacramento and El Monte; Davis, US: Department of Environmental Toxicology, University of California.

71. Amirante R, Distaso E, Di Iorio S, Napolitano M, Sementa P, Tamburrano P, et al. Effects of lubricant oil on particulate emissions from port-fuel and direct-injection spark-ignition engines. *Int J Engine Res*. Epub ahead of print 3 May 2017. DOI: 10.1177/1468087417706602.

72. Hallquist ÅM, Jerksjö M, Fallgren H, Westerlund J and SjödinÅ. Particle and gaseous emissions from individual diesel and CNG buses. *Atmos Chem Phys* 2013; 13: 5337–5350.

73. Pratson LF, Haerer D and Patiño-Echeverri D. Fuel prices, emission standards, and generation costs for coal vs natural gas power plants. *Environ Sci Technol* 2013; 47: 4926–4933.

74. Ryan Walker N, Wissink ML, DelVescovo DA and Reitz RD. Natural gas for high load dual-fuel reactivity controlled compression ignition in heavy-duty engines. *J Energ Resour: ASME* 2015; 137: 042202.

75. Amirante R, Casavola C, Distaso E and Tamburrano P. Towards the development of the in-cylinder pressure measurement based on the strain gauge technique for

internal combustion engines. SAE paper 2015-24-2419, 2015.

76. Wermuth N, Najt PM and Yun H. *Control strategy for transitions between homogeneous-charge compression-ignition and spark-ignition combustion modes.* Patent 8.955.492, USA, 2015.

77. Benajes J, Molina S, Garcia A, Monsalve-Serrano J and Durrett R. Performance and engine-out emissions evaluation of the double injection strategy applied to the gasoline partially premixed compression ignition spark assisted combustion concept. *Appl Energ* 2014; 134: 90–101.

78. Amirante R, Coratella C, Distaso E, Rossini G and Tamburrano P. Optical device for measuring the injectors opening in common rail systems. *Int J Automot Techn* 2017; 18: 729−742.

79. Jerzembeck S, Peters N, Pepiot-Desjardins P and Pitsch H. Laminar burning velocities at high pressure for primary reference fuels and gasoline: experimental and numerical investigation. *Combust Flame* 2009; 156: 292–301.

80. Tian G, Daniel R, Li H, Xu H, Shuai S and Richards P. Laminar burning velocities of 2,5-dimethylfuran compared with ethanol and gasoline. *Energ Fuel* 2010; 24: 3898–3905.

Appendix 1

Notation

b_0, b_1 and b_2	coefficients for exponent β
B_m, B_2, ϕ_m and S_{u0}	coefficients in Metghalchi et al.'s correlations
p_u	unburned mixture pressure
p_0	room pressure
S_L	laminar flame speed
S_{L0}	laminar flame speed at room conditions
T_u	unburned mixture temperature
T_0	room temperature
Z, W, η, ξ and σ	coefficients in "Gülder's formulation" for pure compounds
α	temperature influence exponent
α_0, α_1 and α_2	coefficients for exponent α
β	temperature influence exponent
ν, τ, ε and Ω	coefficients in "Gülder's formulation" for fuel mixtures
ϕ	equivalence ratio
χ	volume fraction of other compounds in methane fuel mixtures