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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, often carried out over a limited range of operating conditions and still they need to be validated against other experimental data. 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 $\phi=0.6 \div 1.7$, $p\_u=1 \div 50$ atm and $T\_u=298 \div 800$ K. The purpose is to provide simple and workable expressions for modeling the laminar flame

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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 literature is also provided.

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Laminar Flame Speed Correlations for Methane, Ethane, Propane and their Mixtures, and Natural Gas and Gasoline for Spark-Ignition Engine Simulations

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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, often carried out over a limited range of operating conditions and still they need to be validated against other experimental data. 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 $\phi = 0.6 - 1.7$, $p_u = 1 - 50\ atm$ and $T_u = 298 - 800\ K$. 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 literature is also provided.

1. 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 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 sixty 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 towards 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 provided too. 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.

2. Overview of empirical correlations available in literature

In the last sixty 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 et al. [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, isooctane 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:

$$\alpha(\phi) = 2.18 - 0.8(\phi - 1)$$
$$\beta(\phi) = 0.16 + 0.22(\phi - 1). \tag{2}$$

112 **Table 1.** *Coefficients of Equation (3) proposed by Metghalchi et al. [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

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

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

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

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

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127 An alternative formulation for evaluating the term $S_{L0}(\phi)$ of Equation (1) was derived by Elia et al.
128 [28]. They developed a correlation by fitting their experimental data of the laminar burning velocity
129 of methane/air mixtures, obtained using a spherical constant volume combustion vessel with fuel-
130 to-air equivalence ratio varying from 0.8 to 1.2, as:

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

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

142 Another work in which the power law formulation was employed coming from measurements of
143 laminar spherical expanding flames of methane/air mixtures is the work carried out by Gu et al. [19],

where reference values of $p_0 = 1\ atm$ and $T_0 = 300\ K$ were also 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 in the range of initial temperatures from $298\ K$ to $498\ K$ and initial pressures from $1\ atm$ 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:

$$S_L(p_u) = 0.133(p_u)^{-0.461}, \qquad T_u = 300\ K, \qquad 1\ atm \leq p_u \leq 60\ atm$$

$$S_L(T_u) = 325 e^{\left(-\frac{9439}{T_u + 1089}\right)}, \qquad p_u = 1\ atm, \qquad 300\ K \leq T_u \leq 700\ K. \qquad (7)$$

In addition, they found that the exponent α increased linearly with increase of initial temperature, while the exponent β decreased exponentially whit increase of initial pressure. Consequently, they formulated the following correlations for the two exponents, as:

$$\alpha(T_u) = 1.39 + 0.0006 T_u, \qquad 300\ K \leq T_u \leq 700\ K$$

$$\beta(p_u) = 0.226 e^{-\frac{p_u}{0.841}} - 0.511, \quad 1\ atm \leq p_u \leq 60\ atm. \qquad (8)$$

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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 φ=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

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 et al. [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

168 of the initial mixture temperatures is the largest available in literature ($300 - 850\ K$), the technique
169 used for the numerical simulations was validated by means of comparisons with experimental data
170 for methane only for the room pressure case and for two different unburnt temperatures, namely
171 300 and $400\ K$. The following expressions for calculating methane's laminar flame speed, based on
172 work by Liao et al. [31] (which is discussed later in this section) was suggested:

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

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

$$S_{L0}(\phi) = Z\ W\ \phi^\eta\ e^{-\xi(\phi-\sigma)^2}, \tag{10}$$

177 where W, η and ξ are constants for a given fuel, and $Z = 1$ for single constituent fuels. The constants
178 for methane/air and propane/air mixtures proposed in [10] are listed in Table 3. In such work, the
179 power law dependence of the laminar burning velocity on the unburnt mixture pressure and
180 temperature was used, but a practical analytical expression for the exponents α and β, as functions
181 of the equivalence ratio, was not proposed. Instead, constant values for α and β were proposed,
182 namely, respectively, 2 and -0.5 for methane and 1.77 and -0.2 for propane.

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

187 Recently, Dirrenberger et al. [32] adopted such a formulation for the prediction of the laminar flame
188 velocity of the components of natural gas, methane, ethane, propane and n-butane, as well as for
189 binary and tertiary mixtures of these compounds, which had been proposed as surrogates for
190 natural gas. In that study, the measurements were performed by using the heat flux method at
191 atmospheric conditions. The mixture strength covered the largest range available in literature,
192 namely from 0.6 to 2.1. The values of the parameters that they proposed for pure methane and
193 propane are given in Table 3. For the methane case their parameters were far from those previously

194
Table 3. Coefficients for Gülder's exponential formulation for different fuels.

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

195

proposed by Gülder, but are very close to those found by other researchers, i.e., 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]:

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

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 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.

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 the subscript 1 refers to parameters calculated for one component, i.e. ethane, and subscript 2 to the other one, i.e., 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 Shannxi 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 $p_u = p_0 = 1atm$), 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.

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

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223 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 et al. [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 (the natural gas investigated by Liao et al. [31] was composed of 96.16% of methane).

230 Metghalchi et al.'s expression [20] was derived for propane and therefore, no direct comparisons can be done with the other two. However, the expression proposed by Metghalchi et al. [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)).

236 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 $353\ K$) who used the stagnation jet-wall flame configuration and Particle Image Velocimetry.
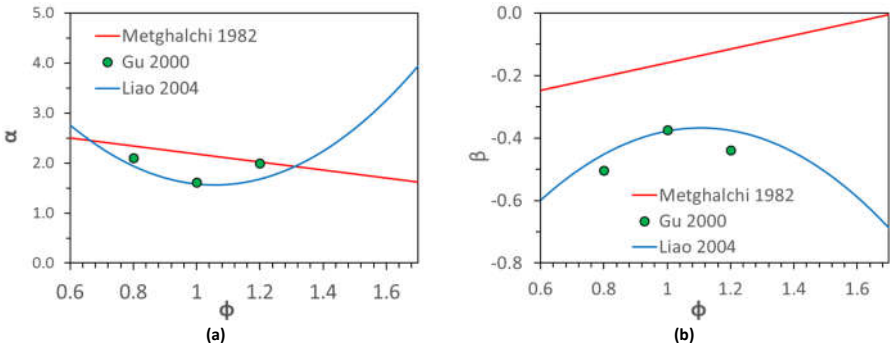
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Figure 1. *Dependence of exponents α and β upon equivalence ratio ϕ.*

249 In the same study, the temperature dependence of the laminar burning velocity was also shown for
250 gasoline, for equivalence ratios varying from 0.7 to 1.3 and for temperatures between $298\ K$ and
251 $358\ K$. The temperature dependence of gasoline was compared to the empirical formulations by
252 Gülder [10] and Metghalchi et al. [20] and a correlation implemented in GT-Power based on the
253 publication by Takashi et al. [44]. The result was that none of the correlations captured the
254 temperature dependence of gasoline. A second-order polynomial, as in Liao et al. [31] (Equation
255 (13)) was then suggested to represent the power exponent α:

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

256 Unfortunately, no correlation for $S_{L0}(\phi)$, or for pressure dependence was provided.

257 Experimental measurements of laminar flame speeds obtained by various workers are compared
258 and analyzed next for each of the fuels considered in this study. A comparison with the above
259 discussed empirical correlations is then reported.

3. Results and comparisons

261 The empirical correlations developed for pure methane and propane, methane/ethane and
262 methane/propane mixtures, as well as for natural gas and gasoline are discussed next.

263 Results for the considered studies are listed in tables, together with the method that was used, the
264 range of the equivalence ratios, pressures and temperatures that were explored, and the fuels that
265 were considered in the specific study.

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

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

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

274

3.1. Methane

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

279

Table 5. Literature considered for methane.

| Ref.	Authors	Year	Fuels	φ	T_u [K]	p_u [atm]	Method
[17]	Ryan	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	1998	Methane	0.6-1.35	298	0.5-4	Outwardly spherical bomb
[19]	Gu	2000	Methane	0.8, 1 and 1.2	300-400	1-10	Constant-pressure spherical comb. chamber / Correlations
[28]	Elia	2001	Methane, Methane/diluent mixtures	0.8-1.2	298-550	0.75-70	Spherical vessel / Correlations
[48]	Rozenchan	2002	Methane, Methane/Oxygen, Methane/Oxygen/Helium	0.6-1.4	298	1-60	Constant-pressure combustion chamber
[49]	Bosschaart	2004	Methane, Ethane, Propane, n-Butane	0.6-1.6	293-360	1	Heat flux
[29]	Han	2006	Methane, Methane/Hydrogen/Carbon monoxide mixtures	1	298-498	1-5	Cylindrical bomb / Correlations
[40]	Coppens	2007	Methane/Hydrogen/Nitrogen mixtures	0.7-1.5	300	1	Heat flux
[30]	Ouimette	2009	Methane, Wood Syngas	0.5-1.5	300-850	1-15.2	Burner for Syngas Numerical simulations for CH$_4$ / Correlations
[50]	Bourque	2010	Methane, Natural gas	0.7-1.3	298	1-4	Cylindrical bomb with Schlieren setup
[51]	Park	2011	Methane, C$_1$-C$_4$/Hydrogen/CO mixtures	0.5-1.3	298 (and 423)	1-4	Counter flow
[52]	Lowry	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	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	2015	Methane	0.7-1.4	298-443	1-5	Constant-volume bomb / Correlations
[53]	Li	2015	Methane/n-Heptane mixtures	0.7-1.5	358-428	1	outwardly cylindrical constant-volume combustion chamber

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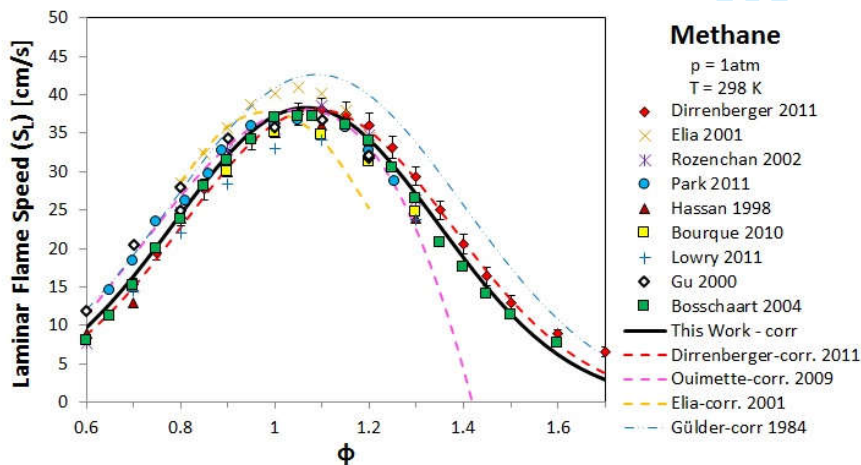
Figure 2 Methane laminar flame speed at room conditions. Marks: experimental data; dashed lines: correlations available in literature; solid line: empirical correlation proposed in this work.

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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$_3$H$_8$	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$_3$H$_8$	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

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] ad 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 et al. [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) depict 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.
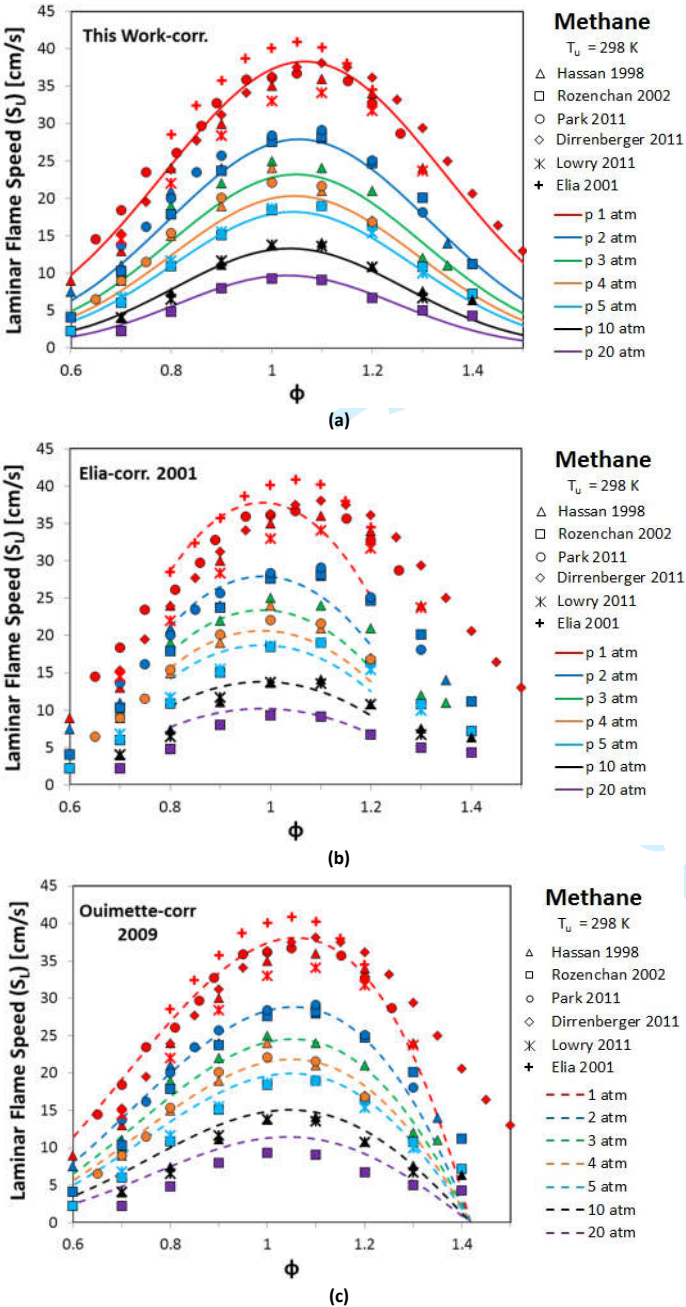
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Figure 3. Methane laminar flame speed at room temperature and different pressures. Correlation in this work (a) and correlations of Elia et al. [28] *(b) and Ouimette et al.* [30] *(c). Marks: experimental data; solid line: empirical correlation proposed in this work; dashed lines: other correlations.*

In Figure 4(b) results for the case of $\phi = 1.2$ are shown. This case is one of the most studied at high pressure, as can be inferred from 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 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.
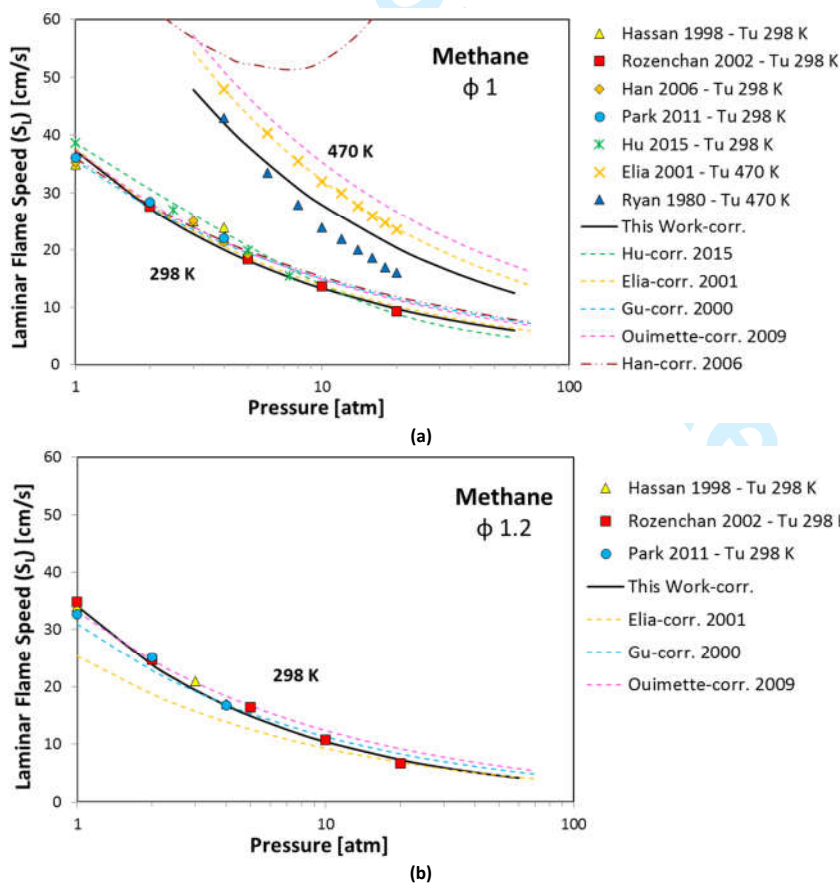

(a)

(b)

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 literature; solid line: empirical correlation proposed in this work.*

333 The initial temperature effects are summarized for the stoichiometric case in Figure 6. Different
334 unburnt mixture pressures are considered as well. At room pressure, the agreement with the
335 experimental data is good, while the other empirical correlations considered tend to overestimate
336 the temperature influence. At higher pressures, the agreement can be considered satisfactory.

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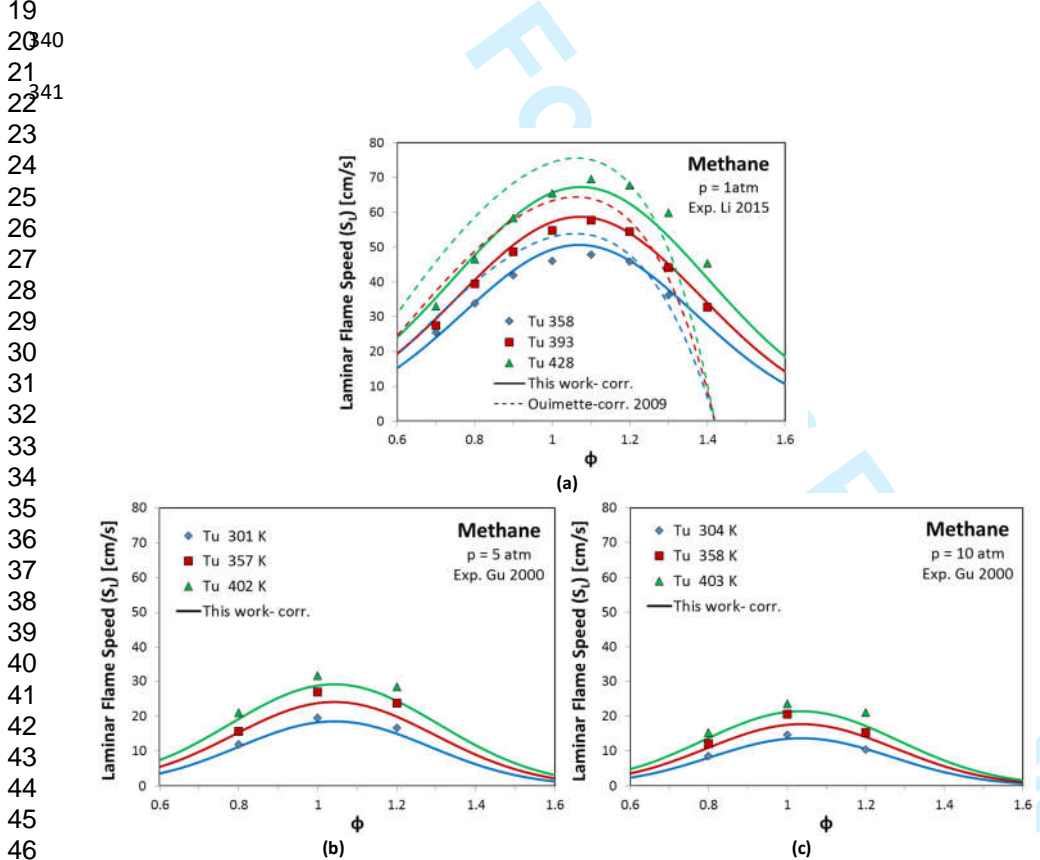339 analysis.

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341

342 **Figure 5.** *Methane laminar flame speed at high temperatures and pressures at different equivalence ratios. Comparisons with*
343 *experimental data available at 1 (a), 5 (b) and 10 (c) atm. Marks: experimental data; dashed lines: correlations available in*
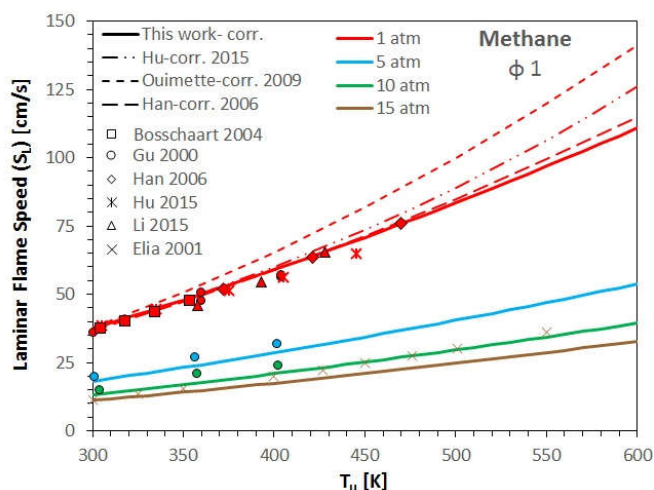344 *literature; solid line: empirical correlation proposed in this work.*

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 literature; solid line: empirical correlation proposed in this work.

3.2. Propane

Propane is usually used in many applications, e.g., laboratory studies of oxidation processes and 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 have 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 literature are also plotted. Similar findings as those for methane are seen for the correlations proposed by Gülder [10] ad 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 measurements gave values considerably higher than all the others.

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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	1961	Methane, Propane, Ethylene, Acetylene, Hydrogen	1	298	0.2-20	Constant-volume bomb
[56]	Chase	1963	Acetylene, Propane	0.8 and 1	280-520	0.5-2.3	Burner
[12]	Metghalchi	1980	Propane	0.8-1.5	298-750	0.4-40	Spherical bomb
[20]	Metghalchi	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	1998	Methane, Ethane, Propane	0.6-1.5	298	1	Counter flow, opposed-jet technique
[58]	Hassan	1998	Ethane, Ethylene, Propane	0.8-1.6	298	0.5-4	Outwardly spherical bomb
[59]	Jomaas	2005	Acetylene, Ethylene, Ethane, Propylene, Propane	0.6-2	298	1-5	Spherical bomb
[60]	Tang	2008	Propane/Hydrogen	0.8 and 1.2	300-440	1-50	Spherical vessel
[33]	Huzayyin	2008	LPG, Propane	0.7-2.2	295-400	0.5-4	Cylindrical bomb / Correlations
[61]	Razus	2010	Propane	vol.% 2.50-6.20	298-423	0.3-1.2	Spherical vessel
[52]	Lowry	2011	Methane, Ethane, Propane Methane/Ethane, Methane/Propane	0.7-1.3	298	1-10	Constant-volume vessel with Schlieren optical setup
[32]	Dirrenberger	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

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

376 Figure 9 summarizes the effect of pressure for stoichiometric propane. Experiments carried out
377 before the 80s give a faster laminar burning velocity, since they did not consider stretch effects. In

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378 addition, Figure 9 shows that the value chosen by Gülder [10] for exponent β, which was in perfect
379 agreement with his experiments, gives the fastest laminar flame speed among those that are
380 proposed in this study. Conversely, the linear form by Metghalchi et al. [20] tends to underestimate
381 the pressure effects in stoichiometric conditions.
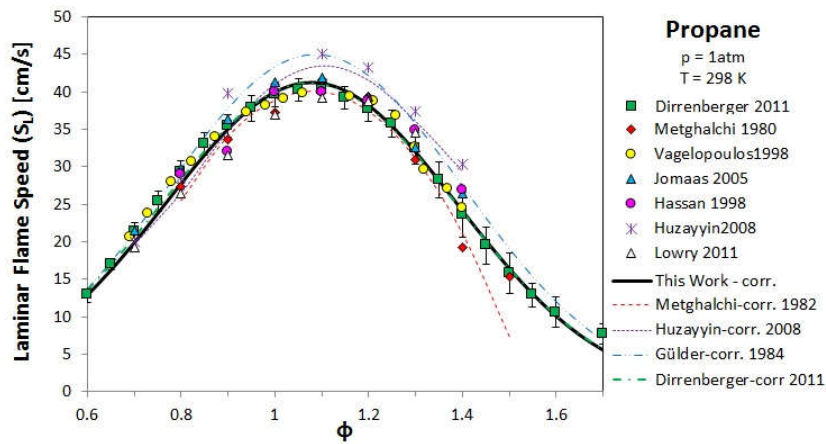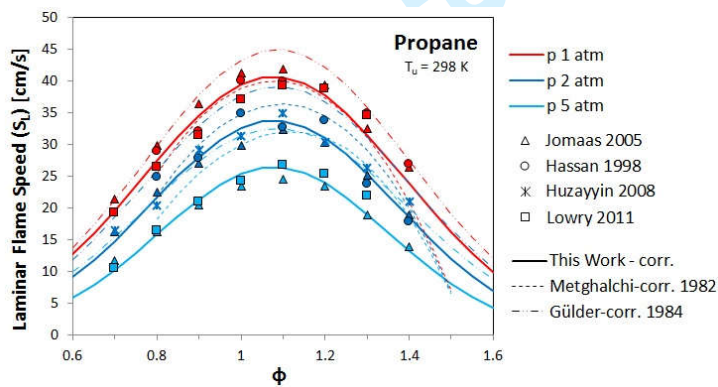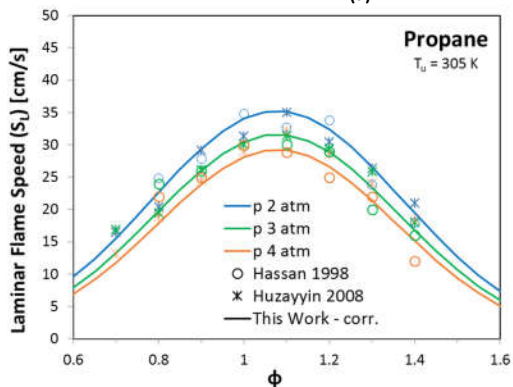
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Figure 7. *Propane laminar flame speed at room conditions. Marks: experimental data; dashed lines: correlations available in literature; solid line: empirical correlation proposed in this work.*

(a)

(b)

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 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 et al. [20] the temperature influence is overestimated for rich mixtures and underestimated for near-stoichiometric and slightly lean conditions.

3.3. 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].

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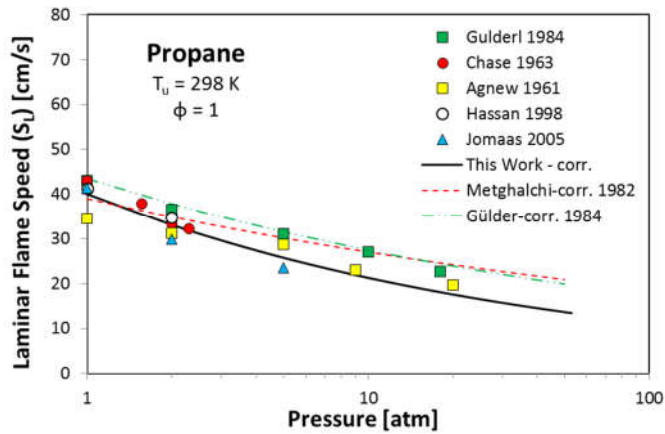

Figure 9. *Initial pressure influence on propane laminar flame speed at stoichiometric conditions. Marks: experimental data; dashed lines: correlations available in literature; solid line: empirical correlation proposed in this work.*
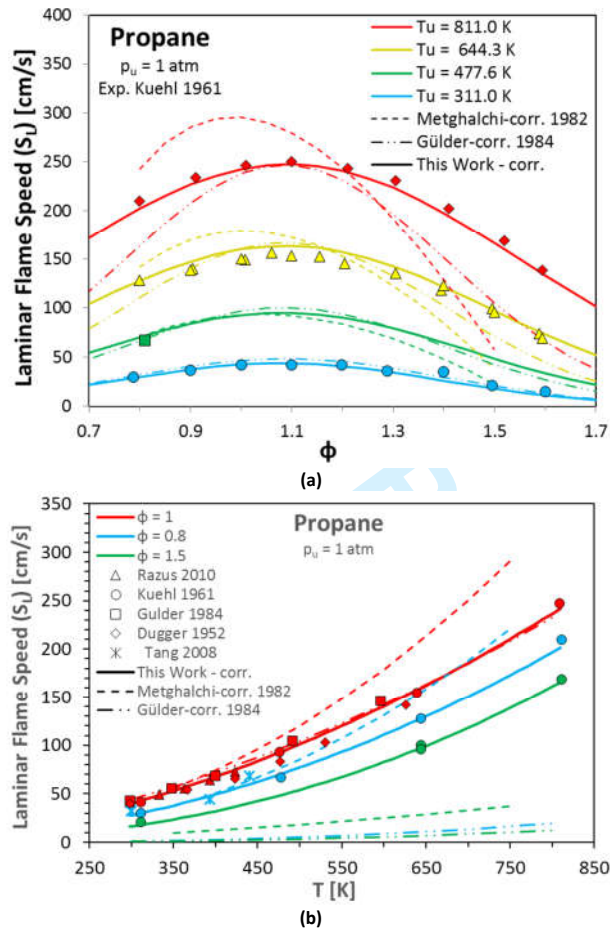
(a)

(b)

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.

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 well the experimental results for lean and rich mixtures, but overestimated flame velocities near stoichiometry. This is because their modifications considered only the 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:

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

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

3.4. Natural Gas

Natural gas is increasingly used as an alternative to petroleum fuels in internal combustion engines and industrial power plants [63–67] because of its smaller environmental effects compared to diesel and gasoline [4,65,68–71], as well as for economic reasons [72]. New combustion techniques [37,73] are related to the use of natural gas, as well as their control strategies [74–77],

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 10. The works considered are listed in Table 11.

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

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

| Ref.	Authors	year	Fuels	Phi	T_u [K]	p_u [atm]	Method
[78]	Kishore	2008	Methane, Ethane, Methane/Ethane mixtures	0.7-1.3	307	1	Heat flux
[52]	Lowry	2011	Methane, Ethane, Propane Methane/Ethane, Methane/Propane	0.7-1.3	298	1-10	Constant-volume vessel with schlieren optical setup
[32]	Dirrenberger	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
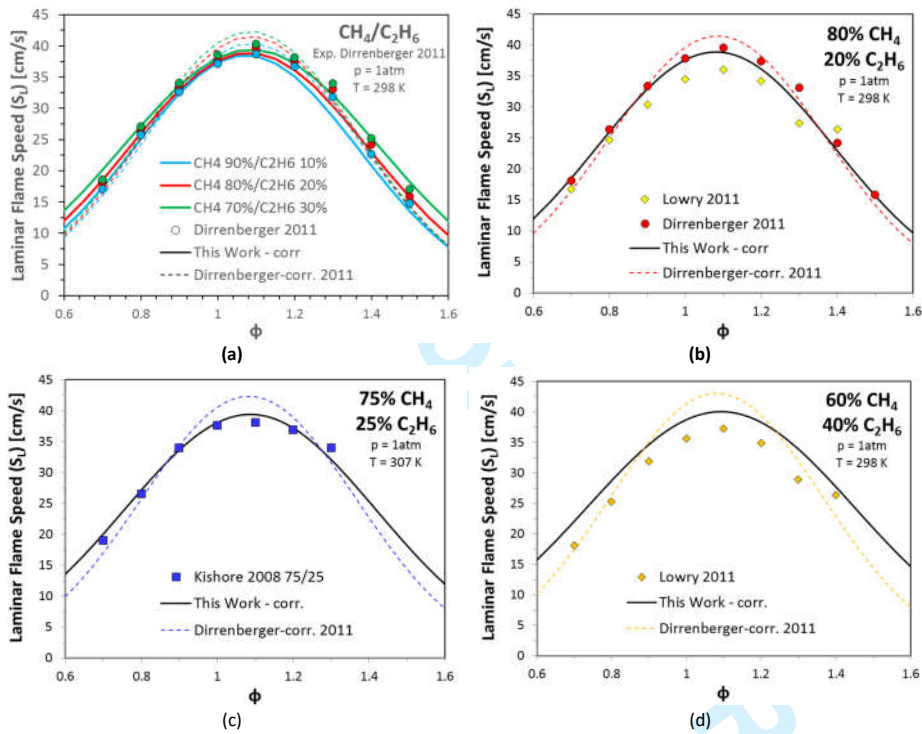
Figure 11. *Laminar flame speed of methane/ethane mixtures at room conditions, considering different ethane content in methane. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al. [32]; solid line: empirical correlation proposed in this work.*
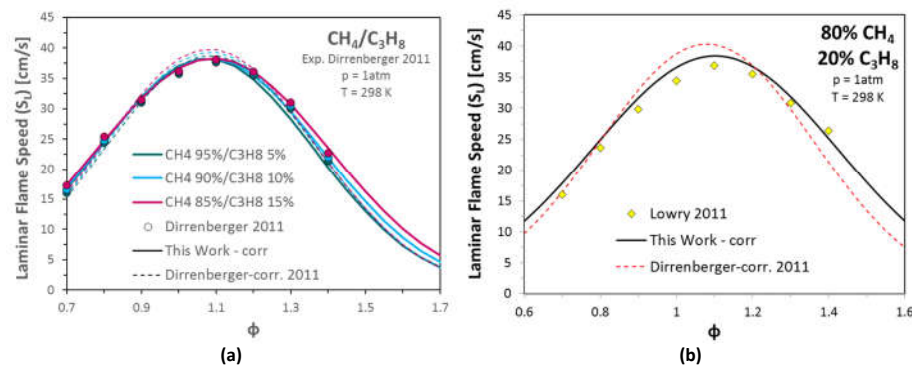
Figure 12. *Laminar flame speed of methane/propane mixtures at room conditions, considering different ethane content in methane. Marks: experimental data; dashed lines: correlations available in literature; solid line: empirical correlation proposed in this work.*

Table 11. Literature considered for natural gas.

Ref.	Authors	year	Fuels	Phi	T_u [K]	p_u [atm]	Method
[31]	Liao	2004	Shannxi Natural Gas	0.6-1.4	300-400	0.5-1.5	Spherical bomb
[50]	Bourque	2010	Methane, Natural gas	0.7-1.3	298	1-4	Cylindrical bomb with Schlieren setup
[32]	Dirrenberger	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-C_4H_{10}	n-C_4H_{10}	i-C_5H_{12}	n-C_5H_{12}	CO_2	N_2	others
[32]	Dirrenberger	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	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	2004	Shannxi	96.16	1.096	-	-	-	-	-			2.74

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 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 Dirrenberger et al. correlation [32] shoed an agreement comparable to that achievable with the present proposed correlation when compared 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).

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(a)

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Figure 13. *Laminar flame speeds of different natural gases at room conditions. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al.* [32]*; solid line: empirical correlation proposed in this work.*
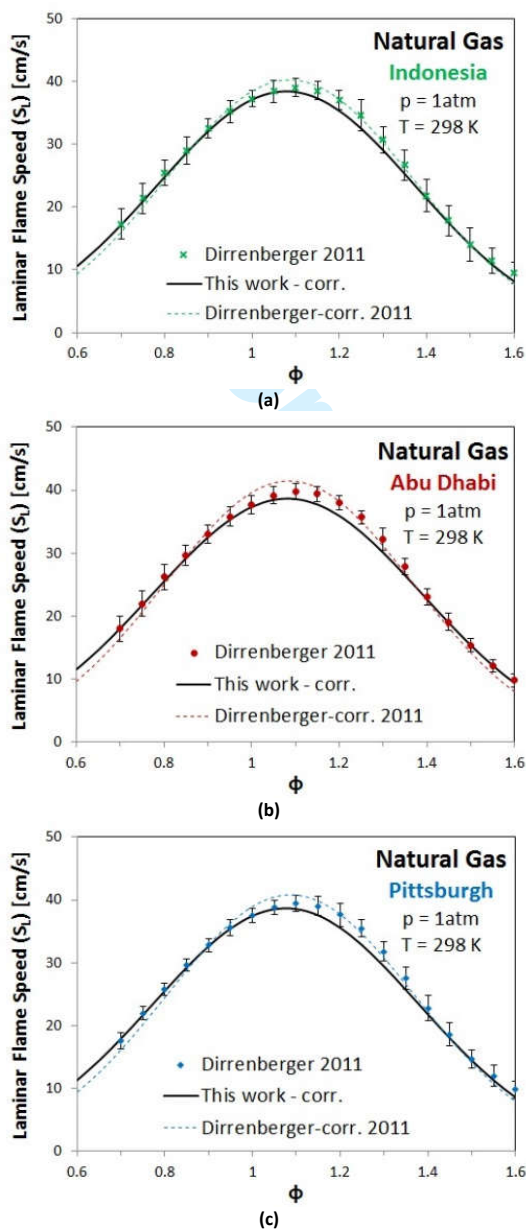
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482 Very few works have investigated the influence of initial pressure and temperature on natural gas
483 laminar flame speed. Figure 15(a) shows results for different initial pressures and Figure 15(b)
484 different initial temperatures. Only the stoichiometric case has been investigated. The derived
485 values for the coefficients of exponents α and β in Equation (16) are listed in Table 7. Liao et al. [31]
486 investigated the temperature influence (the coefficients of exponent α are the same proposed by
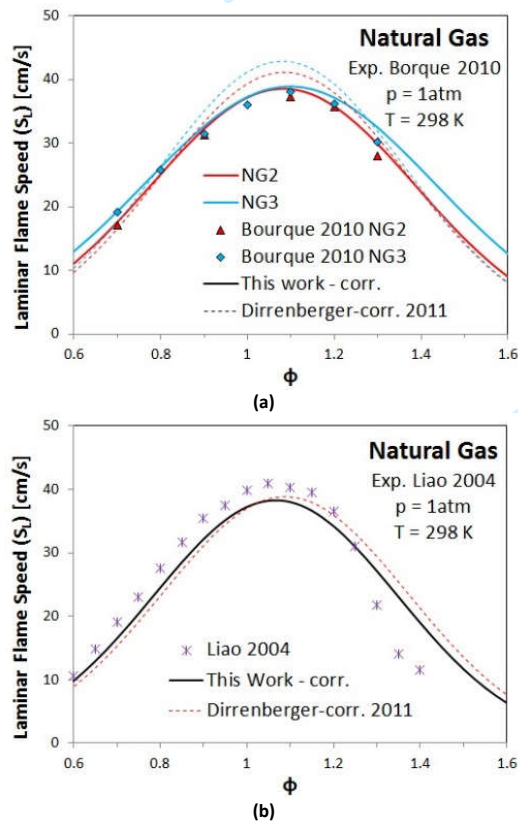487 Liao et al. [31]).

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(a)

(b)

490 *Figure 14. Laminar flame speed of different natural gases measured by Bourque et al. [50] (a) and Liao et al. [31] at room*
491 *conditions. Marks: experimental data; dashed lines: correlations proposed by Dirrenberger et al. [32]; solid line: empirical*
492 *correlation proposed in this work.*
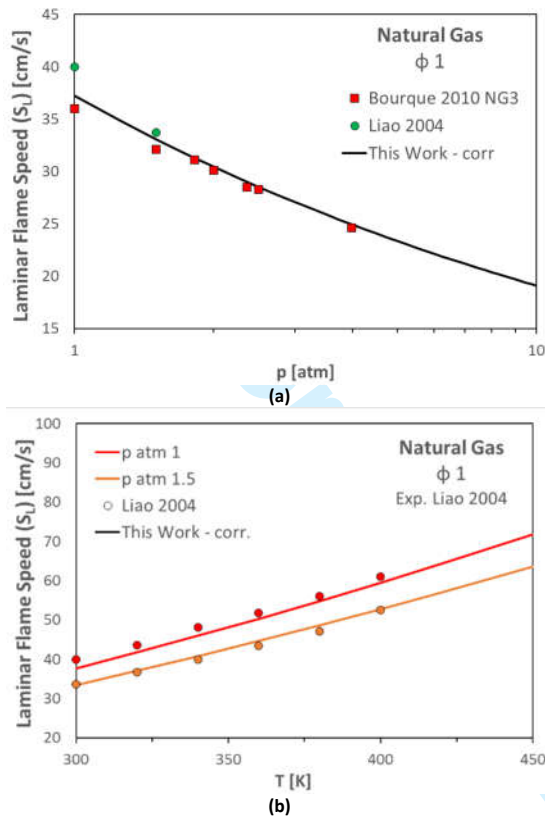
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Figure 15. Initial pressure (a) and temperature (b) influence on natural gas laminar flame speed at stoichiometric conditions. Marks: experimental data; solid lines: empirical correlation proposed in this work.

3.5. 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.

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For gasoline, the term $S_{L0}(\phi, \chi)$ is modeled in the same way as for methane and propane. This 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 results obtained by using the coefficients reported in Table 6 for gasoline, with experimental measurements carried out at 358 and 353 K and room pressure found in literature.

Table 13. Literature considered for gasoline.

Ref.	Authors	year	Fuels	Phi	T [K]	p [atm]	Method
[43]	Zhao	2003	Gasoline (CR-87)	0.6-1.4	353 & 500	1	Single jet-wall stagnation flame
[79]	Jerzembeck	2009	n-Heptane, Isooctane, PRF 87, Gasoline	0.7-1.2	373	10-25	Constant volume bomb
[80]	Tian	2010	2,5-Dimethylfuran, Ethanol, Gasoline (EN228)	0.6-2.0	323-373	1	Constant volume vessel and high speed schlieren visualization
[14]	Sileghem	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	2014	iso-octane/n-heptane/ toluene mixture Gasoline (TAE7000)	0.6-1.5	358	1	Heat flux method

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, different coefficients from those suggested by Sileghem et al. [14] are proposed here, since additional experimental measurements are considerate in this analysis, as reported in Figure 17(b).

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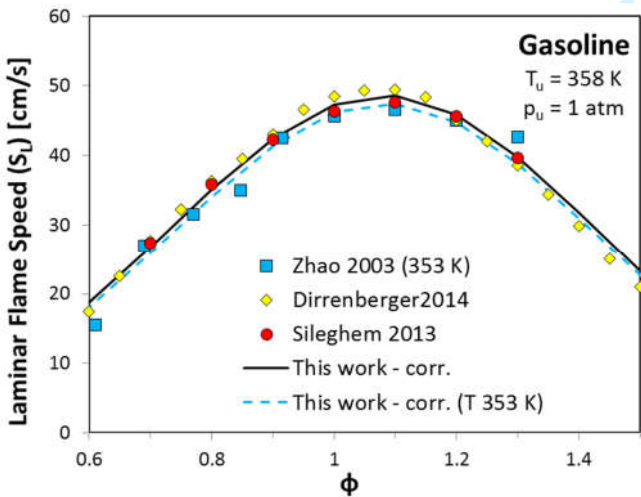

Figure 16. Gasoline laminar flame speed at room pressure and at two different temperatures. Marks: experimental data; lines: empirical correlation proposed in this work.
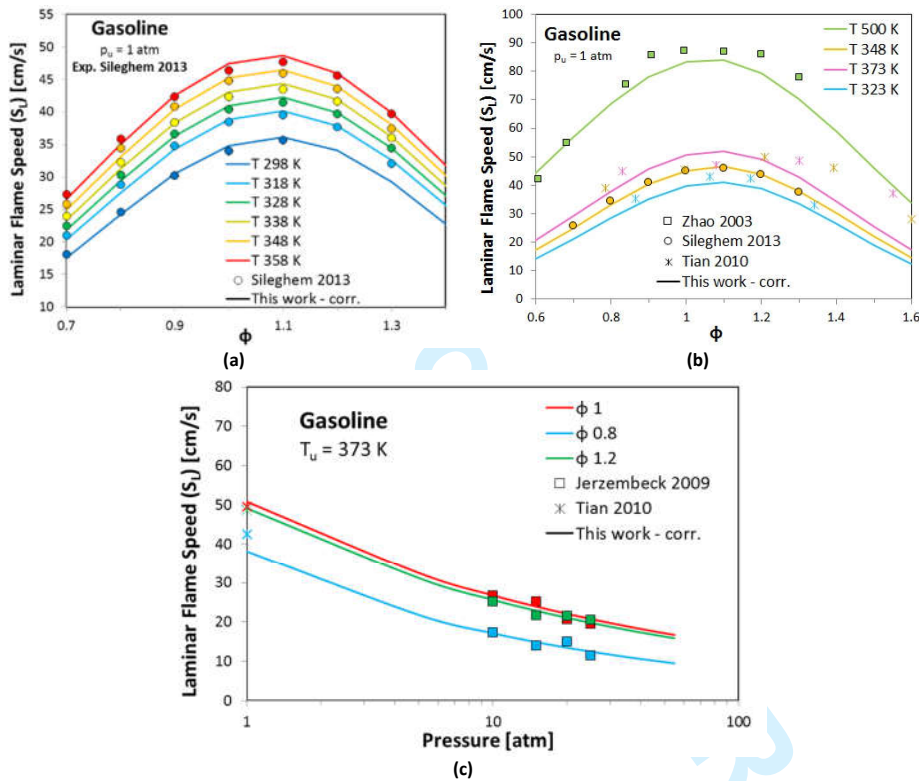
Figure 17. *Initial temperature influence on gasoline laminar flame speed at room pressure (a) and (b). Initial pressure influence at three different equivalence ratios and at an initial temperature of 373 K (c). Marks: experimental data; lines: empirical correlation proposed in this work.*

4. Conclusions

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 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.

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Empirical correlations available in 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:

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

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 W, 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 was 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 the 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 for the unburnt mixture pressure and temperature ranges considered. Coefficients W, η, ξ and σ obtained for gasoline are reported in Table 14.

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

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

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

576 **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$_3$H$_8$	42.11	-0.25	5.24	1.10
Gasoline	36.82	-0.22	4.86	1.11

577

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

Fuel	ν	τ	ε	Ω
CH$_4$/ C$_2$H$_6$	0.20	1.50	0.95	0.09
CH$_4$/ C$_3$H$_8$	0.10	1.50	1.30	0.20

579

580 **Table 16 Co**efficients 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$_3$H$_8$	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

581

582 **Nomenclature**

ϕ	Equivalence ratio
p_0	Room pressure
p_u	Unburned mixture pressure
T_0	Room temperature
T_u	Unburned mixture temperature
S_L	Laminar flame speed
S_{L0}	Laminar flame speed at room conditions
α	Temperature influence exponent
β	Temperature influence exponent
$B_m, B_2, \phi_m \ and \ S_{u0}$	Coefficients in Metghalchi et al.'s correlations
$\alpha_0, \alpha_1 \ and \ \alpha_2$	Coefficients for exponent α
$b_0, b_1 \ and \ b_2$	Coefficients for exponent β
$Z, W, \eta, \xi \ and \ \sigma$	Coefficients in "Gülder's formulation" for pure compounds
$\nu, \tau, \varepsilon \ and \ \Omega$	Coefficients in "Gülder's formulation" for fuel mixtures
χ	Volume fraction of other compounds in methane fuel mixtures

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