INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 37 (2012) 691–697

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HDMR correlations for the laminar burning velocity of premixed $CH_4/H_2/O_2/N_2$ mixtures

Zhenlong Zhao [a,1], *Zheng Chen* [a,b,*]

[a] *State Key Laboratory for Turbulence and Complex Systems, Department of Mechanics and Aerospace Engineering, College of Engineering, Peking University, Beijing 100871, PR China*
[b] *Department of Aeronautics and Astronautics, College of Engineering, Peking University, Beijing 100871, PR China*

ARTICLE INFO

Article history:
Received 21 July 2011
Received in revised form
1 September 2011
Accepted 14 September 2011
Available online 15 October 2011

Keywords:
Laminar burning velocity
$CH_4/H_2/O_2/N_2$ mixture
HDMR correlation

ABSTRACT

Correlations for the laminar burning velocity of premixed $CH_4/H_2/O_2/N_2$ mixtures were developed using the method of High Dimensional Model Representation (HDMR). Based on experiment data over a wide range of conditions reported in the literature, two types of HDMR correlation (i.e. global and piecewise HDMR correlations) were obtained. The performance of these correlations was assessed through comparison with experimental results and the correlation reported in the literature. The laminar burning velocity predicted by the piecewise HDMR correlations was shown to agree very well with those from experiments. Therefore, the piecewise HDMR correlations can be used as an effective replacement for the full chemical mechanism when the prediction of the laminar burning velocity is needed in certain combustion modeling.

1. Introduction

The laminar burning velocity is defined as the speed relative to the unburned gas, at which a planar, one-dimensional flame front travels along the direction normal to its surface. It is one of the most important parameters of a combustible mixture. On a fundamental level, the laminar burning velocity is a critical target for reaction mechanism development and validation [1]. On a practical level, it affects the fuel burning rate in Internal Combustion (IC) engines and hence determines the engine's performance as well as pollutant emissions [2]. Moreover, in spark-ignition engines, the turbulent propagation speed of the flame front is a key parameter in the engine modeling and it can be determined from the laminar burning velocity according to certain models [3]. Therefore, for the past half century, substantial attention has been given to the determination of the laminar burning velocity of different fuel/air mixtures [4–9].

Recently, many studies have been devoted to the development of high-performance combustion engines utilizing hydrogen blended natural gas [10,11] and the laminar burning velocity of hydrogen enriched methane has been studied extensively [12–23,31]. However, significant difficulties lie in the complexity of detailed kinetic mechanism, which render the accurate calculation of laminar burning velocity a great challenge in the numerical modeling. In order to reduce the computational time due to complex chemistry, correlations were developed to predict the laminar burning velocities for mixtures of different compositions (equivalence ratio and dilution) and at different conditions (pressure and

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[*] *Corresponding author*. State Key Laboratory for Turbulence and Complex Systems, Department of Mechanics and Aerospace Engineering, College of Engineering, Peking University, Beijing 100871, PR China. Tel.: +86 (10)6256 8935.

E-mail addresses: zhaozl@pku.edu.cn (Z. Zhao), cz@pku.edu.cn (Z. Chen).

[1] Currently at Massachusetts Institute of Technology, USA.

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doi:10.1016/j.ijhydene.2011.09.086

temperature). For example, Hermanns et al. [24] presented a correlation for the laminar burning velocity of $CH_4/H_2/O_2/N_2$ mixtures based on their experimental data [17]. However, the form of the correlation in Ref. [24] was based on the physical characteristics of laminar flame within different conditions and revised with respect to different factors, such as hydrogen blending, temperature change, dilution, etc. Therefore, this method is not readily applicable to other mixtures and/or working conditions. Besides, the correlation presented in Ref. [24] was obtained mainly through curve fitting and hence the accuracy was highly constrained by the selected forms of nonlinear equations.

Instead of conducting nonlinear regression, the method of High Dimensional Model Representation (HDMR, to be introduced in the next section) [25–28] is utilized here to give more accurate correlations for the laminar burning velocity of $CH_4/H_2/O_2/N_2$ mixtures. Two types of HDMR correlations are constructed based on the experimental data [17]. The performances of these HDMR correlations are assessed through the comparison with the experimental data and the correlation proposed in Ref. [24].

2. The HDMR method and specifications

The High Dimensional Model Representation (HDMR) method was developed by Rabitz et al. [25] to capture the input–output relationship of a complex system. It provides a straightforward approach to explore the input–output mapping relations without requiring a large number of tests. In HDMR, the output variable $f(\mathbf{x})$ is expressed as a finite hierarchical correlated expansion in terms of the normalized input variables $\mathbf{x} = (x_1, x_2, \ldots, x_n)$ in the following form [25]:

$$f(\mathbf{x}) = f_0 + \sum_{i=1}^{n} f_i(x_i) + \sum_{1 \le i < j \le n} f_{ij}(x_i, x_j) + \cdots + f_{12\ldots n}(x_1, x_2, \ldots, x_n) \qquad (1)$$

where the zeroth-order component function f_0 is a constant denoting the mean response; the first-order component function $f_i(x_i)$ represents the individual contribution to the output $f(\mathbf{x})$ acted by the ith input variable; the second-order function $f_{ij}(x_i, x_j)$ describes the cooperative effects of the input variables x_i and x_j upon the output $f(\mathbf{x})$, etc. The last term $f_{12\ldots n}(x_1, x_2, \ldots, x_n)$ reflects any residual nth-order correlated contribution of all input variables to the output $f(\mathbf{x})$.

Systematic optimal procedures [26–29] have been developed to construct the distinct HDMR component functions in Eq. (1). Recently, based on the Random Sampling (RS) of the input variables, the RS-HDMR was introduced by Li et al. [27,28]. It was shown that the RS-HDMR can accurately and efficiently treat high dimensional input–output mapping problems. The RS-HDMR has been successfully utilized in several scientific modeling applications including atmospheric chemistry [26], molecular dynamics simulation [27], and combustion [29,30]. Therefore, the RS-HDMR is used here to construct correlations for the laminar burning velocity of premixed $CH_4/H_2/O_2/N_2$ mixtures.

The intrinsic idea of RS-HDMR is to approximate component functions by optimal weighted orthonormal polynomials $\{\varphi\}$ as [27, 28]

$$\begin{aligned}
f_i(x_i) &\approx \sum_{r=1}^{k} \alpha_r^{(0)i} \varphi_r^i(x_i) \\
f_{ij}(x_i, x_j) &\approx \sum_{r=1}^{k} \left[\alpha_r^{(ij)i} \varphi_r^i(x_i) + \alpha_r^{(ij)j} \varphi_r^j(x_j) \right] + \sum_{p=1}^{l} \sum_{q=1}^{l'} \beta_{pq}^{(0)ij} \varphi_p^i(x_i) \varphi_q^j(x_j) \\
f_{ijk}(x_i, x_j, x_k) &\approx \sum_{r=1}^{k} \left[\alpha_r^{(ijk)i} \varphi_r^i(x_i) + \alpha_r^{(ijk)j} \varphi_r^j(x_j) + \alpha_r^{(ijk)k} \varphi_r^k(x_k) \right] \\
&\quad + \sum_{p=1}^{l} \sum_{q=1}^{l'} \left[\beta_{pq}^{(ijk)ij} \varphi_p^i(x_i) \varphi_q^j(x_j) + \beta_{pq}^{(ijk)ik} \varphi_p^i(x_i) \varphi_q^k(x_k) \right. \\
&\quad \left. + \beta_{pq}^{(ijk)jk} \varphi_p^j(x_j) \varphi_q^k(x_k) \right] + \sum_{p=1}^{m} \sum_{q=1}^{m'} \sum_{r=1}^{m''} \gamma_{pqr}^{(0)ijk} \varphi_p^i(x_i) \varphi_q^j(x_j) \varphi_r^k(x_k)
\end{aligned} \qquad (2)$$

where k, l, l', m, m', m'' are integers. In many cases, the optimal weighted orthonormal polynomials $\{\varphi\}$ are tailored according to the sampling data as:

$$\begin{aligned}
\varphi_1^i(x_i) &= a_1 x_i + a_0 \\
\varphi_2^i(x_i) &= b_2 x_i^2 + b_1 x_i + b_0 \\
\varphi_3^i(x_i) &= c_3 x_i^3 + c_2 x_i^2 + c_1 x_i + c_0
\end{aligned} \qquad (3)$$

The coefficients a_0, a_1, b_0, …, c_3 are determined to ensure the orthonormality of $\{\varphi\}$ for a given set of random samples [27, 28]. In this study, the output variable, $f(\mathbf{x})$, in Eq. (1) is the laminar burning velocity, S_L, of $CH_4/H_2/O_2/N_2$ mixtures at standard temperature and pressure (298 K, 1 atm). The laminar burning velocities of $CH_4/H_2/O_2/N_2$ mixtures at different equivalence ratio ϕ, hydrogen content $\mathscr{R}_{H_2}$ (defined as the volume/molar fraction of hydrogen in the methane/hydrogen mixture) and amount of oxygen $\mathscr{R}_{O_2}$ (defined as the volume/molar fraction of oxygen in the oxygen/nitrogen mixture) were measured by Hermanns [17] using the heat flux method. These data are used as samples to construct HDMR correlations in this study. Therefore, the input variables are $(\phi, \mathscr{R}_{H_2}, \mathscr{R}_{O_2})$ and the following correlation is obtained via the RS-HDMR method

$$S_L = f(\phi, \mathscr{R}_{H_2}, \mathscr{R}_{O_2}) \qquad (4)$$

According to Eqs. (1,2,4), we have the following third-order RS-HDMR expansion for the laminar burning velocity

$$\begin{aligned}
S_L \approx{} & f_0 + \sum_{i=1}^{3} \sum_{r=1}^{3} \alpha_r^i \varphi_r^i(x_i) + \sum_{1 \le i < j \le 3} \sum_{p=1}^{3} \sum_{q=1}^{3} \beta_{pq}^{ij} \varphi_p^i(x_i) \varphi_q^j(x_j) \\
& + \sum_{p=1}^{3} \sum_{q=1}^{3} \sum_{r=1}^{3} \gamma_{pqr}^{123} \varphi_p^1(x_i) \varphi_q^2(x_j) \varphi_r^3(x_k)
\end{aligned} \qquad (5)$$

More details on how to construct the RS-HDMR component functions can be found in Refs. [27,28]. The RS-HDMR component functions are calculated based on 396 sets of data, $(\phi, \mathscr{R}_{H_2}, \mathscr{R}_{O_2}, S_L)$, which cover a wide range of equivalence ratio $(0.6 \le \phi \le 1.5)$, hydrogen content $(0 \le \mathscr{R}_{H_2} \le 40\%)$, and oxygen content $(16\% \le \mathscr{R}_{O_2} \le 20.9\%)$. It is noted that the HDMR correlations cannot predict the laminar burning velocity outside of the input date range. The coefficients for different HDMR correlations developed in this study can be found in the supplemental document.

3. Results and discussions

The original objective of this study was to obtain a global HDMR correlation that would accurately fit the entire set of laminar burning velocity data. However, the accuracy of the global HDMR correlation is constrained by the limited amount of sample data as well as the different behaviors within the fuel-lean ($\phi < 1$) and fuel-rich ($\phi > 1$) regions. In order to improve the accuracy, piecewise HDMR correlations are also obtained.

3.1. *Global HDMR correlation*

The RS-HDMR technique [27,28] is applied here for the prediction of the laminar burning velocity, S_L, of premixed $CH_4/H_2/O_2/N_2$ mixtures according to Eq. (5). Around 75 percent of entire data set ($M = 396$) were randomly selected and used to construct the global HDMR correlation (referred as “used data”). The remaining 25 percent data points (referred to as “test data”) were used to assess the performance of the global HDMR correlation. The results are summarized in Table 1. The accuracy of different order HDMR correlations is reflected by the data portion whose relative errors are less than a given value (5% and 10%). Table 1 shows that the results from a third-order global HDMR correlation lie mostly (over 80%) within five percent relative to the experimental data. Therefore, the third-order HDMR correlation is used to predict the laminar burning velocity of premixed $CH_4/H_2/O_2/N_2$ mixtures. Fig. 1 shows the laminar burning velocity as a function of equivalence ratio at different amounts of hydrogen and oxygen. From Fig. 1 it can be observed that the laminar burning velocities predicted by the third-order global HDMR correlation are in good agreement with those from experiments. Fig. 2 further compares the laminar burning velocities predicted by the global HDMR correlation and those from experiments. The coefficient of determination, R^2, is shown to be 0.984. From this point, the global HDMR correlation can well predict the laminar burning velocity.

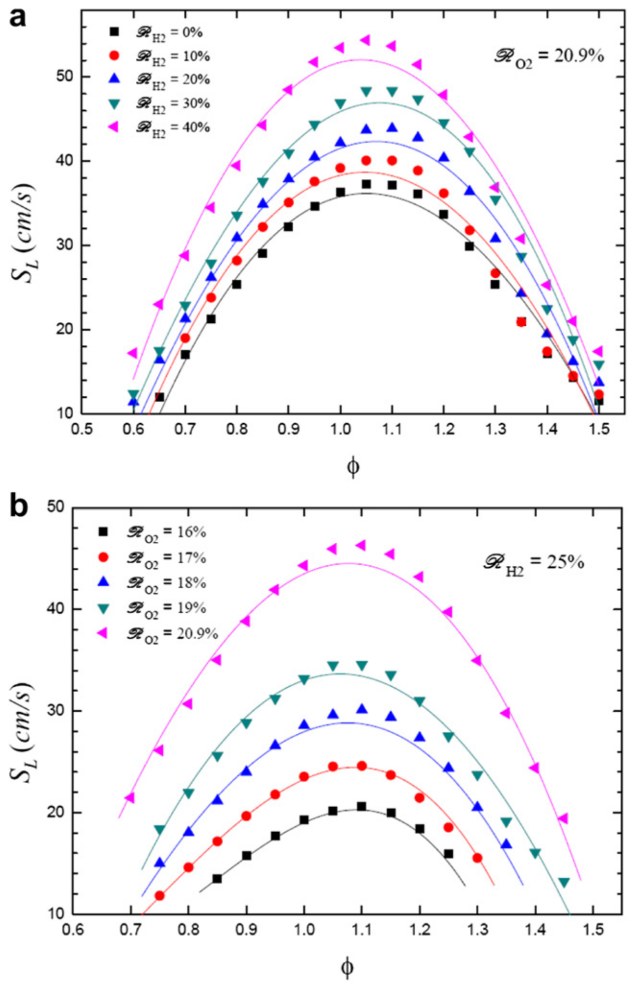

Fig. 1 – Laminar burning velocity as a function of equivalence ratio: (a), $\mathscr{R}_{O_2} = 20.9\%$; (b), $\mathscr{R}_{H_2} = 25\%$. Lines, global HDMR correlation; symbols, experimental results [17].

However, as shown in Fig. 1, the laminar burning velocities for mixtures around stoichiometry are slightly under-predicted by the global HDMR correlation compared to experimental results. This is mainly due to the non-monotonic change of the laminar burning velocity with the equivalence ratio for near-stoichiometric mixtures. In order to improve the accuracy, the whole domain of the equivalence ratio is divided into two sub-domains and different correlations are generated based on the data in each sub-domain.

3.2. *Piecewise HDMR correlation*

Due to the different behaviors of the laminar burning velocity with respect to equivalence ratio at the fuel-lean and fuel-rich regions, two different HDMR correlations were developed separately. The entire data set is divided into two subsets: one for $\phi \leq 1.05$ and the other for $\phi \geq 1.05$. For each subset, around 75 percent of data were used to construct the piecewise correlation via RS-HDMR (“used data”). The remaining 25 percent data points (“test data”) were used to test the performance of the piecewise correlations. Table 2 shows the relative errors of piecewise HDMR correlations. Compared to the global HDMR correlation, the data portion for relative errors less than 5% or 10% is greatly enhanced by using the piecewise HDMR correlations. By comparing Tables 1 and 2, one can

Table 1 – The relative errors of different order global HDMR correlations obtained from the entire data subset.

sample size N (396-N)	Relative error	Data portion					
		Used data			Test data		
		1st order	2nd order	3rd order	1st order	2nd order	3rd order
301 (95)	5%	63.8%	78.7%	81.1%	63.2%	79.0%	80.0%
	10%	87.7%	91.0%	92.0%	83.2%	91.6%	91.6%

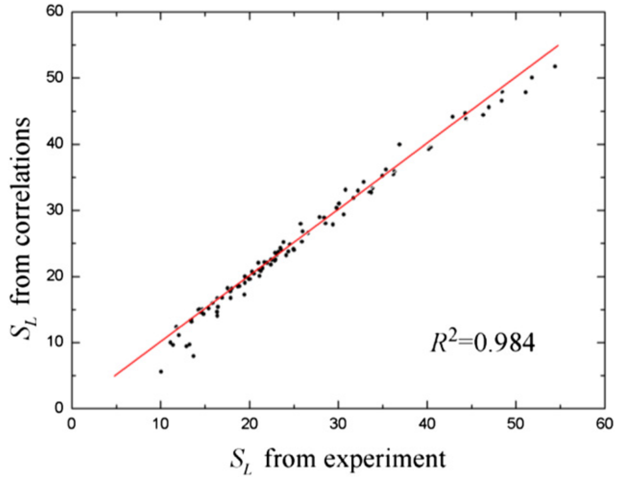

Fig. 2 – Comparison of the laminar burning velocities predicted by the global HDMR correlation with those measured by Hermanns [17].

easily notice that the piecewise HDMR correlation gives a much more accurate prediction of laminar burning velocity. In addition, the second-order piecewise HDMR correlation is even more accurate than the third-order global HMDR correlation. Therefore, the accuracy can be improved using the piecewise correlations for sub-domains as compared to the global HDMR correlation for the whole domain of the equivalence ratio.

Fig. 3 compares the laminar burning velocities predicted by the piecewise HDMR correlations and those from experiments. Very good agreement is observed, as can be expected based on the results from Table 2. The prediction by the global HDMR correlation is also presented for comparison. After using the piecewise HDMR correlations, significant improvement is achieved, especially for near-stoichiometric mixtures. As shown in Fig. 4, the coefficient of determination, R^2, is closer to unity than the global HDMR correlation (see Fig. 2). Therefore, results from the piecewise correlations are more accurate than those from the global HDMR correlation.

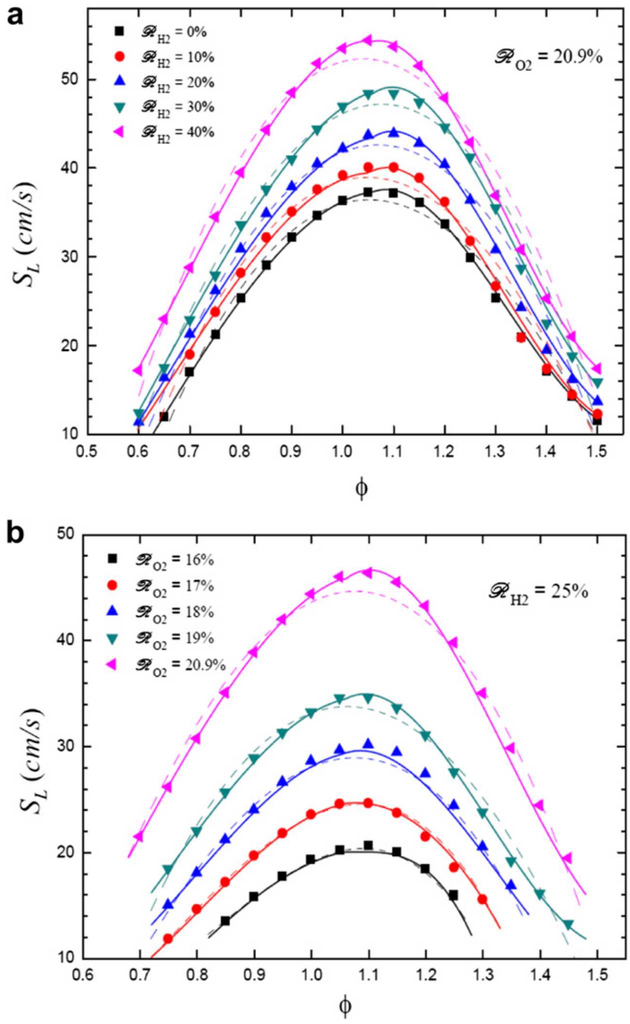

Fig. 3 – Laminar burning velocity as a function of equivalence ratio: (a), $\mathscr{R}_{O_2} = 20.9\%$; (b), $\mathscr{R}_{H_2} = 25\%$. Solid lines, piecewise HDMR correlations; dashed lines, global HDMR correlation; symbols, experimental results [17].

3.3. *Discussions*

In this study, a wide range of conditions including the equivalence ratio from 0.6 to 1.5, hydrogen content in the fuel from 0% to 40%, and oxygen content in the oxidizer from 20.9% down to 16% is covered by the HDMR correlations. As shown in the above sections, two types of HDMR correlation (i.e. global and piecewise HDMR correlations) are constructed to represent the laminar burning velocities as a function of the

Table 2 – The relative errors of different order piecewise HDMR correlations obtained from two data subsets corresponding to fuel-lean and fuel-rich cases.

	Sample size *N*	Relative error	Data portion					
			Used data			Test data		
			1st order	2nd order	3rd order	1st order	2nd order	3rd order
$\phi \leq 1.05$	157 (46)	5%	68.2%	97.5%	100%	60.9%	95.7%	89.1%
		10%	92.4%	100%	100%	93.5%	100%	100%
$\phi > 1.05$	170 (51)	5%	59.4%	90.6%	92.4%	60.8%	90.2%	86.3%
		10%	83.5%	97.7%	98.2%	84.3%	100%	100%

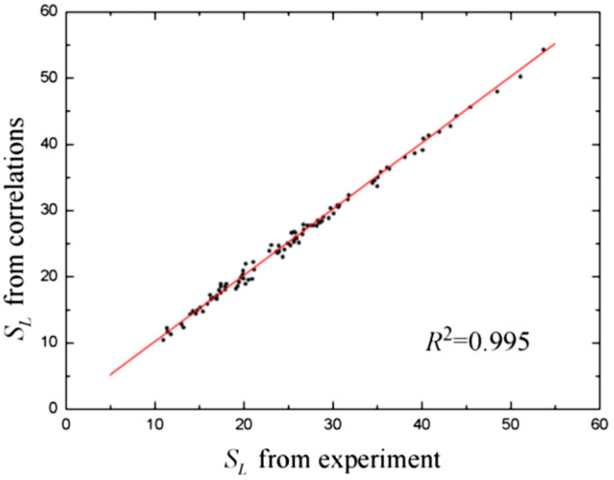

Fig. 4 – Comparison of the laminar burning velocities predicted by the piecewise HDMR correlations with those measured by Hermanns [17].

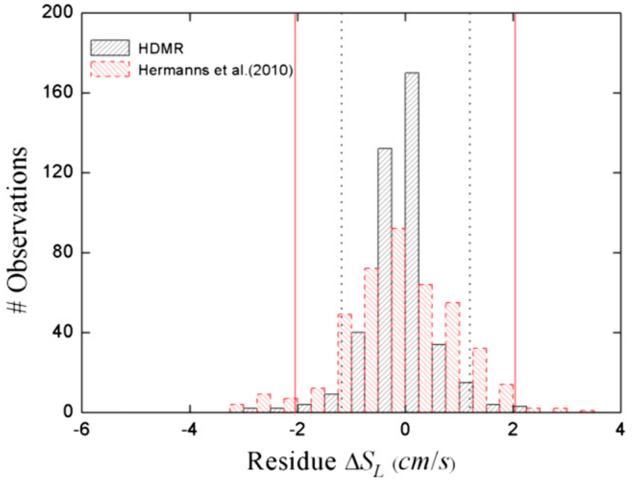

Fig. 6 – Histogram of the residual of the present correlation (experimental data - calculated values). Lines indicate 2σ uncertainty interval (solid lines for results reported by Hermanns et al. [24]; dashed lines for the present study).

equivalence ratio, hydrogen content in the fuel, and the amount of oxygen in the oxidizer. The global HDMR correlation uses a single equation to predict the laminar burning velocities. The advantage of the global HDMR correlation is that only one group of coefficients needs to be evaluated for a wide range of conditions. It can accurately predict the laminar burning velocity of lean and rich mixtures. However slight under-prediction is observed for the near-stoichiometric mixtures. For the piecewise HMDR correlations, two equations are utilized to predict the laminar burning velocities for equivalence ratio within two regions. The accuracy is shown to be greatly improved by replacing the global HDMR correlation with the piecewise HDMR correlations. The data portion for error less than 5% is much larger for the piecewise HDMR correlations than the global HDMR correlation. Nevertheless, the R^2 values of both types of HDMR correlations are close to 0.99 and thus the laminar burning velocity is well predicted by these HDMR correlations.

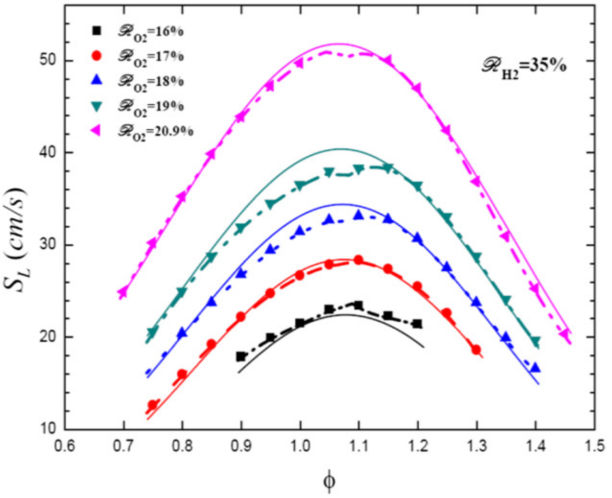

Fig. 5 – Laminar burning velocity as a function of equivalence ratio for $\mathscr{R}_{H_2}$ = 35%. Solid lines, the correlation proposed by Hermanns et al. [24], dash-dotted lines, piecewise HDMR correlations; symbols, experimental results [17].

To further demonstrate the performance of the piecewise HDMR correlations, Fig. 5 compares the laminar burning velocities for $CH_4/H_2/O_2/N_2$ mixtures predicted by the correlation proposed by Hermanns et al. [24] (solid lines), those by the present piecewise HDMR correlations (dashed lines), and the experimental data (symbols). The results from piecewise HDMR correlations are shown to be much closer to the experimental results than those from Hermanns et al. [24]. Therefore, the piecewise HDMR correlations are more accurate than the correlation proposed by Hermanns et al. [24]. The overall uncertainty of the correlation in comparison with the experimental data is shown Fig. 6. In this figure a histogram of the available laminar burning velocity residues (S_L measured in experiments minus S_L predicted by correlations) [24] is plotted. The uncertainty of the piecewise correlations based on a sigma 2σ uncertainty interval is 1.19 cm/s. It is much less than the 2σ uncertainty of 2.02 cm/s of the correlation from Ref. [24] and is close to the reported uncertainty of the experiment (1 cm/s). Since the correlation in Ref. [24] is for the entire equivalence ratio range while each piecewise HDMR correlation works only for half of the entire equilibrium ratio range, we construct two correlations in the same form as that in Ref. [24] for $\phi \leq 1.05$ and $\phi \geq 1.05$, respectively. The overall 2σ uncertainty is 1.78 cm/s, which is still much larger than that from the piecewise HDMR correlations. Therefore, the piecewise HDMR correlations could predict the laminar burning velocities more accurately than the correlation proposed by Hermanns et al. [24]. It should be mentioned that when the correlation in Ref. et al. [24] is compared to the global HDMR correlation, the accuracy is quite comparable. However, the HDMR method still merits over the correlation proposed by Hermanns et al. [24] since no expertise is needed for the determination of the form of the nonlinear equations.

4. Conclusions

HDMR correlations are developed to predict the laminar burning velocity of $CH_4/H_2/O_2/N_2$ mixtures over a wide range of conditions: the equivalence ratio from 0.6 to 1.5, hydrogen content in the fuel from 0% to 40%, and oxygen content in the oxidizer from 20.9% down to 16%. Two types of HDMR correlations are constructed, one being an overall general one covering the full range of the equivalence ratio while the other one a piecewise correlation model. The laminar burning velocities predicted by both the global and piecewise HDMR correlations are shown to agree well with the experimental results. The R^2 values of both types of HDMR correlations are close to 0.99. The results from the piecewise HDMR correlations are shown to be more accurate than either the global HDMR correlation or the one proposed by Hermanns et al. [24]. Therefore, the piecewise HDMR correlations can be used as an effective replacement for the full mechanism when the prediction of the laminar burning velocity is needed in certain combustion modeling. It is noted that in order to cover a complete range of conditions in IC Engines, the HDMR correlations should be extended to conditions with high pressures and temperatures. This will be explored in the future.

Acknowledgments

This work was supported by National Natural Science Foundation of China (NO. 51136005 and NO. 50976003). It is also a pleasure to thank Dr. Hermanns for providing us the experimental data in Ref. [17] and Dr. Gengyuan Li for helpful discussions on the RS-HDMR.

Appendix A. Supplementary material

Supplementary document associated with this article can be found, in the online version, at doi:10.1016/j.ijhydene.2011.09.086.

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