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A model for the laminar flame speed of binary fuel blends and its application to methane/hydrogen mixtures

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ABSTRACT

Recent studies have demonstrated promising performance of adding hydrogen to methane in internal combustion engines and substantial attention has been devoted to binary fuel blends. Due to the strong nonlinearity of chemical reaction process, the laminar flame speed of binary fuel blends cannot be obtained from linear combination of the laminar flame speed of each individual fuel constituent. In this study, theoretical analysis is conducted for a planar premixed flame of binary fuel blends and a model for the laminar flame speed is developed. The model shows that the laminar flame speed of binary fuel blends depends on the square of the laminar flame speed of each individual fuel component. This model can predict the laminar flame speed of binary fuel blends when three laminar flame speeds are available: two for each individual fuel component and the third one for the fuel blends at one selected blending ratio. The performance of this model as well as models reported in the literature is assessed for methane/hydrogen mixtures. It is demonstrated that good agreements with calculations or measurements can be achieved by the present model prediction. Moreover, it is found that the present model also works for other binary fuel blends besides methane/hydrogen.

1. Introduction

Binary fuel blends are widely utilized in developing high-efficiency and low-emission internal combustion engines [1] as well as in the development of surrogate fuel models [2]. For example, recent studies [3–5] have demonstrated the promising performance of adding hydrogen to methane in internal combustion engines and indicated a definite advantage in blending hydrogen. In order to understand the combustion properties of hydrogen enriched methane and to develop high-performance combustion engines utilizing hydrogen blended natural gas, fundamental investigation on the ignition, flame propagation, flame stability, and extinction of methane/hydrogen dual fuel is essential. In this study, the laminar flame speed of binary fuel blends is investigated.

The laminar flame speed (also called the laminar burning velocity) is defined as the speed relative to the unburned gas, with which a planar, one-dimensional flame front travels along the normal to its surface [6]. It is one of the most important parameters of a combustible mixture. The laminar flame speed affects the fuel burning rate in internal combustion engines and the engine's performance and emissions [7].

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Because of the fundamental significance of the laminar flame speed, substantial attention has been given to the determination of laminar flame speed (see Refs. [8,9] and references therein).

In the literature, there are different theoretical models on the laminar flame speed, including the thermal theory, diffusion theory, and asymptotic theory [6,10–12]. One-step global reaction model is usually used in the theoretical analysis. To include the detailed reaction mechanism for the fuel/air system, the conservation equations for mass, species, and energy can be solved by computational techniques. The CHEMKIN-PREMIX code [13] is popularly used in the combustion community to calculate the laminar flame speed. Experimentally, various approaches utilizing different flame configurations, reviewed in Ref. [14], have been developed to measure the laminar flame speed. Currently, the laminar flame speed measured in experiments becomes extremely important for developing and validating chemical kinetic mechanisms for different fuels [15–17].

Most of the studies mentioned above were focused on the laminar flame speed of a single fuel component. Due to the recent interests in the application of fuel blends in internal combustion engines [1], many studies were conducted for the laminar flame speed of binary fuel blends, among which hydrocarbon fuel with hydrogen addition was the most extensively studied [18–31]. However, most of these studies only reported the laminar flame speed of fuel blends from experimental measurements or numerical simulations, and the model to predict the laminar flame speed of fuel blends was not proposed.

Due to the strong nonlinearity of chemical reaction process, the laminar flame speed of binary fuel blends cannot be obtained from the linear combination of the laminar flame speed of each individual fuel constituent. By introducing a parameter, R_H, representing nonlinearly the relative amount of hydrogen addition to the hydrogen/hydrocarbon dual fuel, Law and coworkers [18,30] proposed a linear correlation to predict the laminar flame speed of binary fuel blends:

$$S_L(\phi, R_H) = S_L(\phi, 0) + k(\phi) \cdot R_H \tag{1}$$

where ϕ is the equivalence ratio. $S_L(\phi, 0)$ and $S_L(\phi, R_H)$ represent the laminar flame speed of hydrogen and that of the hydrogen/hydrocarbon dual fuel, respectively. The coefficient, $k(\phi)$, is obtained from the linear fitting [18,30]. The linear relationship holds only for small value of R_H (usually R_H should be less than unity). As will be demonstrated in this study (see Fig. 2b), the linear relationship cannot be used when the content of hydrogen in the fuel blends is close to 100%. Moreover, the linear correlation proposed in Refs. [18,30] works only for hydrogen/hydrocarbon fuel blends and it is not extended for fuel blends containing two types of hydrocarbon fuels.

Recently, Di Sarli and Di Benedetto [21] proposed a Le Chatelier's Rule-like formula to predict the laminar flame speed of hydrogen/methane mixtures:

$$S_L(\phi, a) = \left[a/S_{L\_H2}(\phi) + (1-a)/S_{L\_CH4}(\phi)\right]^{-1} \tag{2}$$

where ϕ is the equivalence ratio and a is the volume fraction (blending ratio) of hydrogen in the hydrogen/hydrocarbon dual fuel. $S_L(\phi, a)$, $S_{L\_H2}(\phi)$, and $S_{L\_CH4}(\phi)$ denote the laminar flame speeds of hydrogen/hydrocarbon dual fuel, pure hydrogen, and pure methane, respectively. Unlike the model given by Eq. (1) which has a coefficient, $k(\phi)$, needing to be determined, the model in Eq. (2) is parameter free. Unfortunately, the above Le Chatelier's Rule-like formula cannot give satisfactory prediction for rich fuel blends [21]. Moreover, as shown in this study (see Fig. 6b), the model given by Eq. (2) cannot work for other binary fuel blends.

The objectives of this study are to develop a model which can predict the laminar flame speed of binary fuel blends and to use this model for methane/hydrogen mixtures. First, the planar premixed flame of binary fuel blends is analyzed and a model for the laminar flame speed is proposed. The performance of this model is then assessed for methane/hydrogen binary fuel blends at different equivalence ratios, pressures, and temperatures. Finally, the conclusions are drawn and the limitation of the present model is discussed.

2. Model derivation

An adiabatic, planar, one-dimensional, premixed flame of binary fuel blends is considered in the theoretical analysis. Following the same way in which the planar flame of a single fuel is analyzed [11], one-step, irreversible, global reaction is assumed for each fuel component of the binary fuel blends:

$$F_i \rightarrow P, \omega_i = \rho Y_i A_i \exp\left(-\frac{E_i}{R^0 T}\right) \quad \text{for } i = 1, 2 \tag{3}$$

where ω_i, Y_i, E_i and A_i are, respectively, the reaction rate, mass fraction, activation energy, and pre-factor of the Arrhenius law for the i^{th} fuel component. ρ is the density, R^0 the universal gas constant, and T the temperature.

For the adiabatic, planar, one-dimensional flame, the governing equations for the temperature and mass fractions of the two fuel components are

$$mC_P \frac{dT}{dx} = \lambda \frac{d^2 T}{dx^2} + \omega_1 q_1 + \omega_2 q_2 \tag{4}$$

$$m \frac{dY_i}{dx} = (\rho D)_i \frac{d^2 Y_i}{dx^2} - \omega_i \quad \text{for } i = 1, 2 \tag{5}$$

where x, C_p, λ, q_i, and D_i are respectively, the spatial coordinate, specific heat capacity at constant pressure, thermal conductivity, the chemical heat release per unit mass of the i^{th} fuel component, and the mass diffusivity of the i^{th} fuel component. According to the continuity equation, the mass flux, $m = \rho u$, is constant. The laminar flame speed of the binary fuel blends is $S_L = m/\rho_u$, where ρ_u is the density of the unburned mixture.

We assume both fuels are totally consumed in the reaction zone. Therefore, the boundary conditions are

$$x \rightarrow -\infty, \quad T = T_u, \quad Y_1 = Y_{1,u}, \quad Y_2 = Y_{2,u} \tag{6}$$

$$x \rightarrow +\infty, \quad T = T_b, \quad Y_1 = 0, \quad Y_2 = 0 \tag{7}$$

where T_u and $Y_{i,u}$ are respectively, the temperature and mass fraction of the i^{th} fuel component in the unburned mixture. T_b is the adiabatic flame temperature. After integrating the linear combination of the governing equations (through which the reaction terms are eliminated), we have the following expression for the adiabatic flame temperature

$$T_b = T_u + (q_1 Y_{1,u} + q_2 Y_{2,u})/C_P \tag{8}$$

Due to the energy conservation across the flame, the heat needed for increasing the temperature of the mixture from T_u to T_b is equal to the chemical heat liberated in the reaction zone. Therefore, we have

$$mC_P(T_b - T_u) = \delta(q_1\overline{\omega}_1 + q_2\overline{\omega}_2) \tag{9}$$

in which the chemical heat release rate is approximated to be proportional to the thickness of the reaction zone, δ, and the mean reaction rate, $\overline{\omega}_i$.

In order to evaluate the mean reaction rate, two characteristic length scales shown in Fig. 1 are introduced: l_T for heat conduction and l_{Di} for mass diffusion of the i^{th} fuel component. In the preheat zone, the chemical reaction can be neglected due to the large activation energy of the reaction process [6,10,11,32]. From the convection–diffusion equations (Eqs. (4) and (5) without the reaction terms) we have the following relationships for the characteristic length scales [6,10,11]:

$$l_T = \lambda/(mC_P), \quad Le_i = l_T/l_{Di} \tag{10}$$

where $Le_i = \lambda/(\rho C_p D_i)$ is the Lewis number of the i^{th} fuel component. It is well known that the thickness of the reaction zone is much less than the characteristic length for heat conduction, i.e. $\delta = l_T/\beta$, with β being the Zel'dovich number [6,10,11]. According to Fig. 1, the fuel mass fraction can be approximated to be linearly distributed in the preheat and reaction zones. Therefore, the mean mass fraction of the i^{th} fuel component in the reaction zone can be approximated as

$$\overline{Y}_i \approx Y_{i,u}\delta/(2l_{Di}) = Y_{i,u}Le_i/(2\beta) \tag{11}$$

The temperature in the reaction zone is close to the adiabatic flame temperature ($T \approx T_b$). Therefore, according to Eqs. (3) and (11), the mean reaction rate can be approximated as

$$\overline{\omega}_i = \rho\frac{Le_i}{\beta}\frac{Y_{i,u}}{2}A_i\exp\left(-\frac{E_i}{R^0T_b}\right) \tag{12}$$

Substituting Eqs. (8), (10) and (12) and $\delta = l_T/\beta$ into Eq. (9) yields

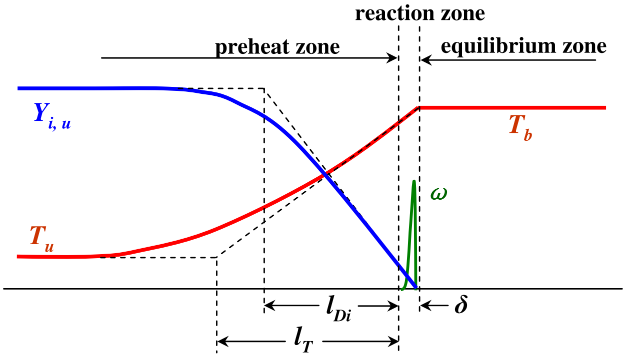

Fig. 1 – The schematic flame structure.

$$m^2(q_1Y_{1,u} + q_2Y_{2,u}) = \sum_{i=1}^{2}\left[\frac{Le_i\lambda}{\beta^2C_P}\frac{q_i\rho Y_{i,u}A_i}{2}\exp\left(-\frac{E_i}{R^0T_b}\right)\right] \tag{13}$$

Following the same procedure, the following relationship can be derived for a planar, one-dimensional, premixed flame of a single fuel component

$$m_i^2 q_i Y_{i,u} = \frac{Le_i\lambda\rho}{\beta^2C_P}\frac{q_iY_{i,u}A_i}{2}\exp\left(-\frac{E_i}{R^0T_b}\right) \quad \text{for } i = 1, 2 \tag{14}$$

where $m_i = \rho_{u,i}S_{L,i}$ with $S_{L,i}$ being the laminar flame speed of the i^{th} fuel component. Substituting Eq. (14) into (13) yields

$$m^2 = (q_1Y_{1,u}m_1^2 + q_2Y_{2,u}m_2^2)/(q_1Y_{1,u} + q_2Y_{2,u}) \tag{15}$$

The laminar flame speed of binary fuel blends can be obtained as $S_L = m/\rho_u$. Therefore, according to Eq. (15), the laminar flame speed of binary fuel blends depends on the square of the laminar flame speed weighted by the chemical heat release of each individual fuel constituent. It is noted that Eq. (15) is derived from the energy conservation given by Eq. (9). In the derivation, approximations are made for the mean fuel mass fractions (Eq. 11) and the mean reaction rate (Eq. 12) in the reaction zone. However, these approximations do not affect the result: the same expression as Eq. (15) can be obtained from the large activation energy asymptotic analysis. Since the effect of preferential diffusion (represented by the Lewis number, Le_i) is already included in the expression for the laminar flame speed of a single fuel component (see Eq. 14), there is no parameter for the diffusivity in the model given by Eq. (15).

The theoretical analysis presented above can be readily extended to fuel blends of multi components. Similar to Eq. (15), we have the following expression for the mass flux

$$m^2 = \sum_i(q_iY_{i,u}m_i^2)/\sum_i(q_iY_{i,u}) \tag{16}$$

3. Model application

The model given by Eq. (15) was used to predict the laminar flame speed of binary fuel blends. Unfortunately, good agreement was not achieved. The main reason is that the adiabatic flame temperature of binary fuel blends is not the same as that of each individual fuel component (i.e. T_b in Eq. (13) is different from T_b in Eq. (14)). Moreover, the chemical interactions between these two fuels are not considered since the one-step global reaction is assumed for each fuel component (see Eq. (3)). We modify the model in Eq. (15) into the following form

$$m^2 = (cY_{1,u}m_1^2 + Y_{2,u}m_2^2)/(cY_{1,u} + Y_{2,u}) \tag{17}$$

in which the coefficient c is a free parameter. Since q_1 and q_2 are constants, the ratio q_1/q_2 in Eq. (15) is in fact included in the coefficient c. The densities of fuel blends and each individual fuel component are readily available. In order to predict the laminar flame speed of binary fuel blends, S_L, according to Eq. (17), we need to know the coefficient c as well as the laminar flame speeds of each individual fuel component, $S_{L,1}$ and $S_{L,2}$ ($m_i = \rho_{u,i}S_{L,i}$ for $i = 1, 2$). The coefficient c can be evaluated using the available date on the laminar flame speed of the binary fuel blends. It is noted that the value found for c is far from q_1/q_2, which might be caused by the facts that the adiabatic flame temperature of binary fuel blends is not the same as that of each individual fuel component and that the laminar flame speed is very sensitive to the adiabatic flame temperature.

Usually, the dual fuel blending is characterized by the volume fraction (blending ratio), a, which changes from zero (only containing one type of individual fuel component) to unity (only containing the other individual fuel component). The ratio, $Y_{1,u}/Y_{2,u}$, in Eq. (17) can be replaced by the following relationship

$$Y_{1,u}/Y_{2,u} = (1-a)W_1/(aW_2) \tag{18}$$

where W_1 and W_2 are the molecular weight of the two fuel components. Therefore, according to Eqs. (17) and (18), the laminar flame speed of binary fuel blends, S_L, as a function of the blending ratio, a, can be obtained.

The model given by Eq. (17) is applied to the binary fuel blends of methane/hydrogen. The laminar flame speed of CH_4/H_2 mixtures at different equivalence ratios, pressures, and temperatures are calculated using the CHEMKIN-PREMIX code [13]. The detailed chemical mechanism, GRI Mech 3.0 [33], and the multi-component diffusion model including the Soret effect are used in the simulation. The computational domain is $0 \le x \le 20$ cm and the flame front is located around the position $x = 10$ cm. Adaptive mesh refinement is used so that most of the grid points are in the reaction zone. The number of grid points is kept to be above 700 so that the flame structure is well resolved and the calculated laminar flame speed is grid-independent.

Fig. 2(a) demonstrates the performance of the model given by Eq. (17) for stoichiometric CH_4/H_2 mixtures at normal pressure and temperature. The coefficient c is determined by ensuring that the laminar flame speed from model prediction and that calculated from PREMIX are the same at one selected blending ratio. For example, the solid line in Fig. 2(a) indicates that the coefficient should be $c = 1.50$ so that at $a = 0.6$, the same laminar flame speed is predicted by the model and by PREMIX. As shown in Fig. 2(a), good agreement is achieved by the model prediction though the flame speed changes nonlinearly with the hydrogen volume fraction. Moreover, it is demonstrated that the model prediction is not very sensitive to the coefficient c while the coefficient itself changes with the selected blending ratio, a. As a result, the selected blending ratio (which is used to determine the coefficient, c, of the model) does not strongly affect the model prediction. This is important for practical application of the present model since there is no stringent requirement on the value of the blending ratio selected as the base data for determining the coefficient, c. When flame speeds at different blending ratios are available, the coefficient c can be determined by minimizing the difference between the laminar flame speeds from model prediction and those calculated from PREMIX or measured in experiments.

The performance of the models in Refs. [18,21] given by Eqs. (1) and (2), respectively, is demonstrated in Fig. 2(b). It is seen that the linear model proposed by Law and coworkers [18] only work for $a < 0.7$ and it cannot be used when the content of hydrogen in the fuel blends is close to 100%. In fact, it was found that the linear relationship holds for small value of R_H [18]. As $a \to 1$, R_H becomes infinity and thus the linear model in Ref. [18] does not work. Fig. 2(b) shows that the Le Chatelier's Rule-like model proposed in Ref. [21] gives nearly the same prediction as the model given by Eq. (17) in this work. However, as will be demonstrated in Fig. 6(b), the model in Ref. [21] works only for methane/hydrogen and cannot work for other binary fuel blends such as methane/dimethyl ether.

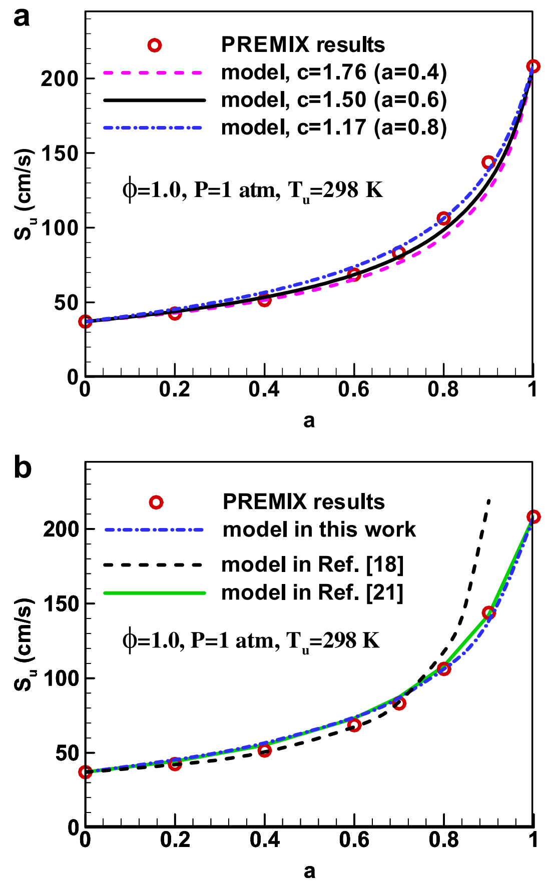

Fig. 2 – Change of the laminar flame speeds of stoichiometric CH_4/H_2 mixtures with the hydrogen volume fraction in the fuel blends: (a), prediction by the model in the present work; (b), prediction by models in Refs. [18,21].

Besides the results for stoichiometric mixtures under normal pressure and temperature, Fig. 3 presents the results at different equivalence ratios, pressures, and initial temperatures, in which the coefficient, c, is determined by the requirement that at $a = 0.8$ the same laminar flame speed is predicted by the model and by PREMIX. It is demonstrated that the laminar flame speeds predicted by the model given by Eq. (17) agree well with those calculated from PREMIX. Moreover, it is found that the coefficient, c, is affected by the equivalence ratio (ϕ) and pressure (P): c increases monotonically with the value of $|\phi\text{-}1|$ and P. The initial temperature is found to have little influence on the value of c.

All the laminar flame speeds of CH_4/H_2 mixtures at 104 different initial conditions (different hydrogen volume fractions $a = 0.2$, 0.4, 0.6, 0.8, or 0.9; equivalence ratios $\phi = 0.6$, 0.8, 1.0, 1.2, or 1.4; pressures $P = 0.5$, 0.8, 1, 2, 3, 4, or 5 atm; and initial temperatures $T_u = 298$ or 365 K) are presented in Fig. 4.

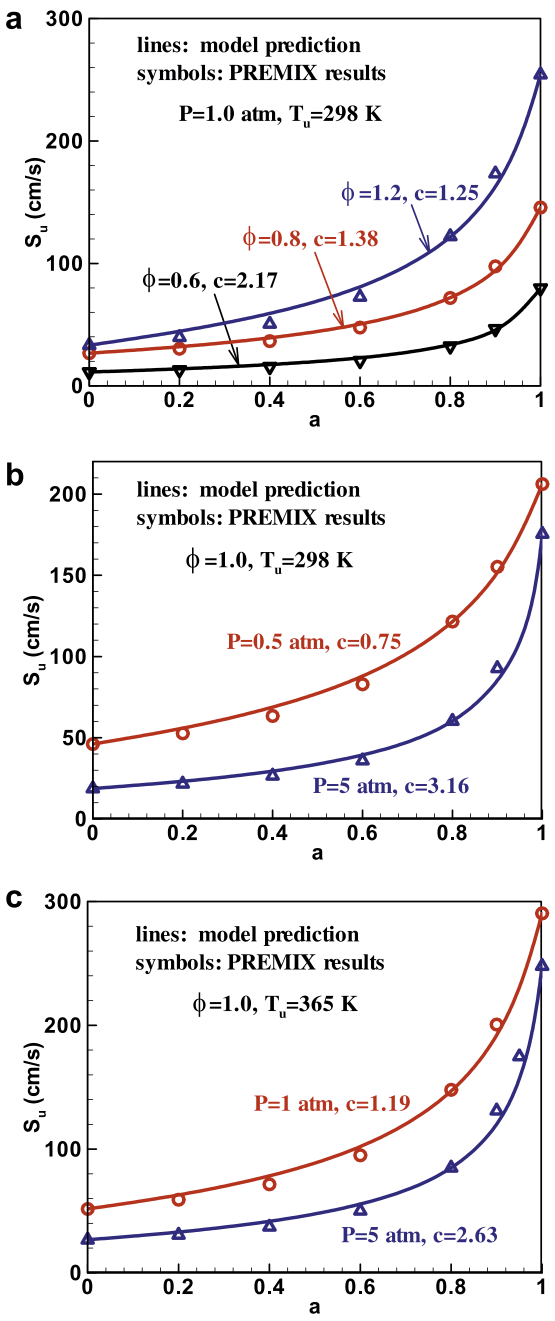

Fig. 3 – Change of the laminar flame speeds of CH_4/H_2 mixtures with the hydrogen volume fraction at (a) different equivalence ratios, (b) different pressures, and (c) elevated temperature.

It is noted that the results for pure methane ($a = 0$) and pure hydrogen ($a = 1$) are not included since the model prediction is the same as the PREMIX results for $a = 0$ and $a = 1$ according to Eqs. (17) and (18). The coefficient of determination, R^2, is shown to be 0.982. From this point, the model given by Eq. (17) can well predict the laminar flame speed of CH_4/H_2 binary fuel blends. Therefore, according to results shown in Figs. 2–4, the present model can be used for CH_4/H_2 binary fuel blends at normal and elevated temperatures and pressures.

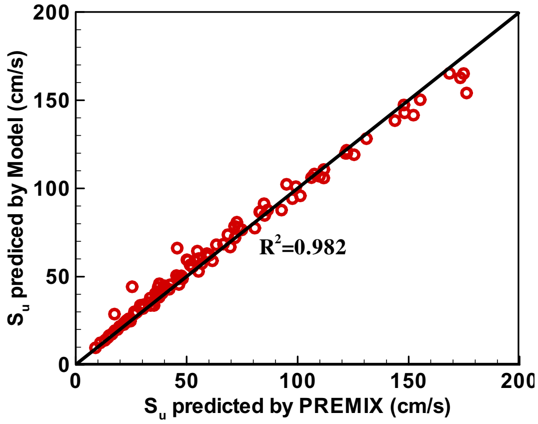

Fig. 4 – Comparison of the laminar flame speeds of CH_4/H_2 mixtures predicted by the model given by Eq. (17) with those calculated from PREMIX.

Besides the simulation results from PREMIX, the laminar flame speeds of CH_4/H_2 mixtures measured by Huang et al. [20] are also used to demonstrate the performance of the model (Eq. (17)) proposed in this study. Besides the laminar flame speeds of pure methane ($a = 0$) and pure hydrogen ($a = 1$), the coefficient c in the model needs to be obtained. Similarly, the coefficient c is determined by ensuring that the laminar flame speed from

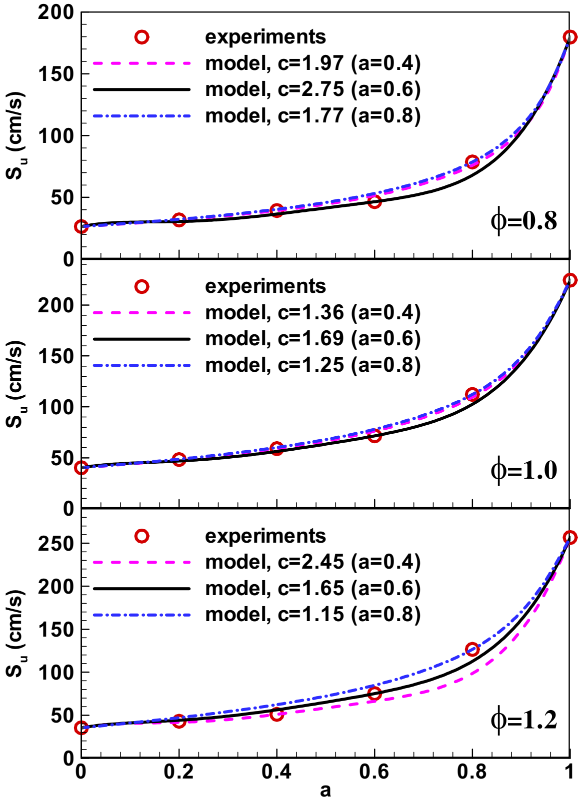

Fig. 5 – Change of the laminar flame speeds of CH_4/H_2 mixtures with the hydrogen volume fraction in the fuel blends (T_u = 298 K, P = 1 atm) Experimental data are from Ref. [20].

model prediction equals to that measured by Huang et al. [20] at one selected blending ratio. The comparison between the model prediction and experimental measurements is shown in Fig. 5. It is seen that good agreement is achieved by the model prediction. Therefore, with the help of the model (Eq. (17)) proposed in this study, the laminar flame speed of binary fuel blends can be predicted and only three laminar flame speeds need to be measured for each condition (equivalence ratio, pressure, and initial temperature): two for pure individual fuel component and the third one for the fuel blends at one selected blending ratio which is used to determine the coefficient *c*. Similar to Fig. 2, Fig. 5 shows that the model prediction is not very sensitive to the coefficient *c* although the coefficient itself changes with the selected blending ratio, *a*.

Besides methane/hydrogen dual-fuel, we also consider the binary fuel blends of methane and dimethyl ether (DME, CH_3OCH_3). The detailed chemical mechanism for DME developed by Zhao et al. [34] was shown to be able to accurately predict the laminar flame speed of both methane/air and DME/air mixtures [22]. Therefore, it is used in the PREMIX calculations. Fig. 6(a) shows that the laminar flame speeds predicted by the model given by Eq. (17) agree very well with those calculated from PREMIX. Compared to CH_4/H_2, the model prediction is significantly improved for CH_4/DME. This is reasonable since the laminar flame speed difference between pure hydrogen and pure methane can reach 200 cm/s while that between pure DME and pure methane is within 15 cm/s. It was found that the laminar flame speeds of large hydrocarbon fuels are similar [35]. Therefore, the present model, Eq. (17), can also be used to predict the laminar flame speed of binary fuel blends of large hydrocarbons. The performance of the present model, Eq. (17), and the Le Chatelier's Rule-like model, Eq. (2), proposed in Ref. [21] is compared in Fig. 6(b). It is seen that the model in Ref. [21] cannot work for CH_4/DME dual-fuel.

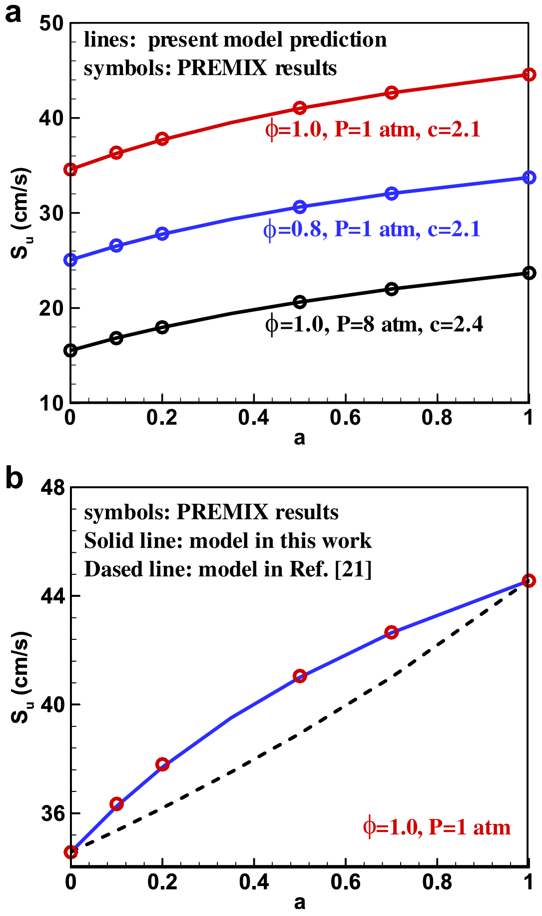

Fig. 6 – Change of the laminar flame speeds of CH_4/DME mixtures with the DME volume fraction in the fuel blends (T_u = 298 K): (a), prediction by the model in the present work; (b), prediction by the model in Ref. [21].

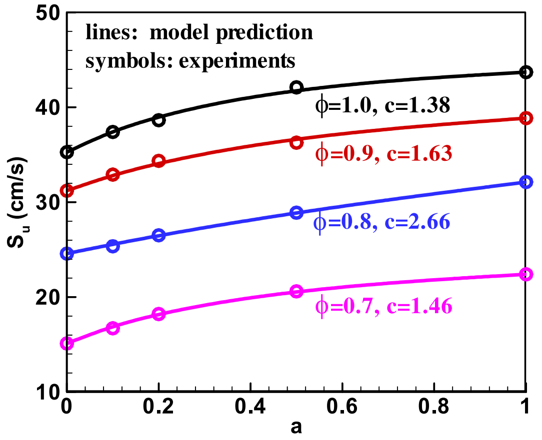

Fig. 7 – Change of the laminar flame speeds of CH_4/DME mixtures with the DME volume fraction in the fuel blends (T_u = 298 K, P = 1 atm). Experimental data are from Ref. [22].

The laminar flame speeds of CH_4/DME mixtures measured in experiments [22] are also used to demonstrate the performance of the model (Eq. 17). As shown in Fig. 7, very good agreement between model prediction and experimental measurements is also achieved.

Besides methane/hydrogen and methane/DME, other binary fuel blends such as propane/hydrogen and n-dodecane/toluene are also studied and it is found that the present model, Eq. (17), can predict the laminar flame speeds of all these binary fuel blends.

4. Concluding remarks

The laminar flame speed of binary fuel blends is investigated here through the one-dimensional, adiabatic, planar, premixed flame. Based on the energy conservation (Eq. 9), the correlation (Eq. 15) for the laminar flame speed of binary fuel blends is derived and its modified form (Eq. 17) is proposed by introducing a free parameter *c*. The model shows that the laminar flame speed of binary fuel blends depends on the square of the laminar flame speed of each individual fuel component. The performance of this model is assessed for methane/hydrogen binary fuel blends at different initial conditions and good agreements with calculations or measurements are found to be achieved by the model prediction. Therefore, the present model can be used for CH_4/H_2 binary fuel blends at normal and elevated temperatures and pressures. Moreover, it is demonstrated that the present model also works for methane/DME and other binary fuel blends.

The shortcoming of the present model is that it is not parameter free: it contains the coefficient *c*. In order to predict the laminar flame speed of binary fuel blends according to the present model, we need know the coefficient *c* as well as the laminar flame speed of each individual fuel component. Therefore, the laminar flame speed at one selected blending ratio is needed to determine the coefficient *c*. Nevertheless, it is demonstrated that the selected blending ratio does not strongly affect the model prediction and thus for practical application of the present model there is no stringent requirement on the value of the blending ratio selected as the base data for determining the coefficient.

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