MOLECULAR TRANSPORT EFFECTS OF HYDROCARBON ADDITION ON TURBULENT HYDROGEN FLAME PROPAGATION

S. Muppala[$,1], J.X. Wen[1], N.K. Aluri[2], and F. Dinkelacker[3]

[1]Faculty of Engineering, Kingston University, Roehampton Vale, Friars Avenue, London, Surrey SW15 3DW, United Kingdom.
E-mail: s.muppala@kingston.ac.uk (S Muppala), J.Wen@Kingston.ac.uk (JX Wen)

[2]ALSTOM Ltd, Brown Boveri Strasse, 5401 Baden, Switzerland.
Email: naresh.aluri@power.alstom.com

[3]Institut Fluid- und Thermodynamik, Universität Siegen, Paul-Bonatz-Str. 9-11, 57076 Siegen, Germany.
E-mail: dinkelacker@thv.mb.uni-siegen.de

ABSTRACT

We analytically investigated the influence of light hydrocarbons on turbulent premixed H_2/air atmospheric flames under lean conditions, in view of safe handling of H_2 systems, applications in H_2 powered IC engines and gas turbines, and also with an orientation towards modelling of H_2 combustion. For this purpose, an algebraic flame surface wrinkling model included with pressure and fuel type effects is used. The model predictions of turbulent premixed flames are compared with the set of corresponding experimental data of Kido et al. (Kido, Nakahara et al. 2002). These expanding spherical flame data include H_2–air mixtures doped with CH_4 and C_3H_8, while the overall equivalence ratio of all the fuel/air mixtures is fixed at 0.8 for constant unstretched laminar flame speed of 25 cm/s, by varying N_2 composition. The model predictions show that there is little variation in turbulent flame speed S_T for C_3H_8 additions up to 20-vol%. However, for 50 vol% doping, flame speed decreases by as much as 30 % from 250 cm/s that of pure H_2–air mixtures for turbulence intensity of 200 cm/s. With respect to CH_4, for 50 vol% doping, S_T reduces by only 6 %, cf. pure H_2/air mixture. In the first instance, the substantial decrease of S_T with C_3H_8 addition may be attributed to the increase in the Lewis number of the dual-fuel mixture and proportional restriction of molecular mobility of H_2. That is, this decrease in flame speed can be explained using the concept of leading edges of the turbulent flame brush (Lipatnikov and Chomiak 2005). As these leading edges have mostly positive curvature (convex to the unburned side), preferential-diffusive-thermal instabilities cause recognizable impact on flame speed at higher levels of turbulence, with the effect being very strong for lean H_2 mixtures. The lighter hydrocarbon substitutions tend to suppress the leading flame edges and possibly transition to detonation in confined structures and promote flame front stability of lean turbulent premixed flames. Thus, there is a necessity to develop a predictive reaction model to quantitatively show the strong influence of molecular transport coefficients on S_T.

Keywords: Lean turbulent premixed flame, Reaction subclosure, Local burning velocity, Preferential diffusion, Lewis number

1. Introduction

The advantages of premixed combustion operating in lean conditions have been noteworthy, both in terms of higher thermal efficiency and for lowered NOx emissions due to reduced flame temperatures. However, the major limitation of operating in lean premixed mode is with respect to the flame stability, which has implication for successful design of combustors. While several experimental and numerical studies of premixed turbulent combustion on gaseous hydrocarbons have been reported in the literature, very few contributions investigate on the H_2 safety and combustion, despite its high practical importance. The physics of lean H_2 combustion differs significantly with those of typical light hydrocarbon fuels. In H_2 combustion under lean or very lean conditions, interactions between thermo-diffusive instabilities attributed to the combined Lewis number and preferential diffusion and stretch of laminar premixed flames affect strongly the properties of premixed turbulent flames, notably the local burning velocity near the convex zone of the flamelet.

In a recent study, Chen and Bilger (Chen and Bilger 2004) reported the superadiabaticy and flame front bulges convex toward the unburned premixed H_2 mixture on typical Bunsen configuration. These features are unique to the less-than-unity Lewis number mixtures. Halter et al. (Halter, Chauveau et al. 2007) showed an increase in the turbulent flame speed with addition of H_2 to lean CH_4 turbulent premixed Bunsen flames. Measurements by Law and Kwon (Law and Kwon 2004) emphasize the importance of hydrocarbon doping on H_2 combustion. Nakahara et al. (Nakahara and Kido 1998) carried out extensive experimental studies on lean H_2 spherical flames reporting a relation between the local burning velocity included with preferential diffusion and turbulent flame speed. Measurements by Nakahara et al. indicate that the lean turbulent propagating flames of H_2–air mixtures or hydrogenated mixtures produce higher local displacement turbulent velocity due to the effect of positive stretch. These fuel mixtures have nearly the same laminar burning velocity S_{L0} of 0.25 m/s, and the highest turbulence intensity u' used was 2.0 m/s. The key parameters available from experiment are the local burning velocity S_L and the turbulent flame speed S_T.

The aim of the present study is to analytically investigate the relationship between S_L with preferential diffusion and S_T, using the algebraic flame surface wrinkling (AFSW) model (Muppala, Aluri et al. 2005). Here, we try to understand the influence of addition of higher density fuels CH_4 and C_3H_8 to H_2–air mixtures on turbulent flame speed. For modelling predictions, both unstretched laminar flame speed and local burning velocity are given as inputs separately for two different scenarios. In our earlier studies (Muppala, Aluri et al. 2005), in the AFSW model, the Lewis number of the deficient reactant was defined for single fuels. For multi-component mixtures, an effective Lewis number Le_{eff} is used to describe the mixed properties. The single fuel Lewis number is simply defined as the ratio of thermal diffusivity to molecular diffusivity of the fuel with $Le = \alpha/D$. The Le_{eff} as function of heat release rates and pure fuel Lewis numbers was proposed by Law et al. (Law, Jomaas et al. 2005) as

$$Le_{eff} = 1 + \frac{q_1(Le_1 - 1) + q_2(Le_2 - 1)}{q} \tag{1}$$

Here, Le_{eff} is the weighted average of the Lewis number values Le_1 and Le_2 of the two fuels (CH_4–H_2, and C_3H_8–H_2). q_1 and q_2 are the corresponding heat release rates of the two fuels such as H_2 and C_3H_8, and $q = q_1 + q_2$ (see Table 1). However, a simplified approach for estimation of Le_{eff} may be given as

$$Le_{eff} = \alpha \cdot Le_1 + (1 - \alpha) \cdot Le_2 \tag{2}$$

where, α is a function of molar fractions of two fuels and amounts of oxygen consumed by CH_4 and H_2. That is, α = 2*mole fraction of CH_4/([2*mole fraction of CH_4 + 0.5*mole fraction of H_2]). For lean C_3H_8–H_2 mixtures with fixed equivalence ratio of 0.8, the Le_{eff} value varies between 1.57 and 0.40, which are corresponding Le values for pure C_3H_8 and pure H_2, respec-

tively. As is known, larger Lewis number reflects insensitivity to hydrodynamic (Darrieus-Landau) instability. In the subsequent discussions for sake of brevity, Le_{eff} will be simply termed as Le, unless otherwise stated.

In our previous studies (Muppala, Aluri et al. 2005; Muppala and Papalexandris 2006), for different fuel-air mixtures, the unstretched laminar flame speed S_{L0} was used in the reaction modelling. However, it is important to understand that the local burning velocity of turbulent flames deviate strongly from S_{L0} especially for lean mixtures and it is an important parameter in determining turbulent flame speed. This finding was experimentally confirmed for instance by Kido et al. (Kido, Nakahara et al. 2002). This deviation is attributed to preferential-thermo-diffusive effects. The Lewis number Le of the deficient reactant mixture reflects the different response of the local flamefront propagation to the variation of strain rates (Chen and Bilger 2002). In other words, if the composition of higher density fuel is diminishing as compared to H_2, i.e., for smaller Lewis number, the flame still remains to act robust even at higher strain rates. Thus, higher local burning velocity near the convex part implies greater flamelet resistance to local flame quench. On the other hand, addition of hydrocarbons to H_2-air mixtures leads to reverse effects, as discussed below.

A study of addition of C_3H_8 to freely propagating H_2 spherical flames (Law, Jomaas et al. 2005) found enhanced flame stability characteristics. Law and Kwon (Law and Kwon 2004) have experimentally verified that doping of H_2 mixtures with C_3H_8 delays the onset of flame-front instability in lean outwardly propagating spherical flames. These instabilities are well described by the shift in the Lewis number from less-than-unity to greater-than-unity. The lean C_3H_8 –H_2 (where, $Le < 1$ for lean H_2, $Le > 1$ for lean C_3H_8) mixtures well describes these stabilization and destabilization processes. Law et al. (Law, Jomaas et al. 2005) have also determined the state of the onset of instability using the Peclet number Pe, which is a function of the effective Lewis number of the dual fuel mixture. This non-dimensional critical Peclet number Pe_{cr}, defined as the ratio of the radius of curvature to the laminar flame thickness, is a measure of the critical conditions for the onset of flame wrinkling. For larger Le_{eff} (i.e., attained for relatively abundant presence of C_3H_8 in H_2 mixtures for a shift from $Le < 1$ to $Le > 1$), the critical Peclet number Pe_{cr} increases, which is an indication of delay in onset of hydrodynamic instability. These concepts are wholly relevant to turbulent H_2 combustion, which helps to explain the causes of explosion hazards, considering the high reactivity and diffusivity of H_2. It may also be inferred that the increase in local burning rates of leading structures reflects a strong evidence of possibility of self-turbulization and transition to detonation; for example, in cases such as flame propagation interacting with turbulence generated due to obstacles in confined structures.

The present study investigates for the properties of the local burning velocity of outwardly propagating spherical lean premixed turbulent flames for composite fuel mixtures for $\phi = 0.8$, for turbulent velocity up to 2 m/s (Kido, Nakahara et al. 2002). The composition of nitrogen was varied such that the unstretched laminar flame speed S_{L0} is nearly identical for all the dual-fuel mixtures. A qualitative study is made to understand on the relationship between the maximum local burning velocities on the convex part of the turbulent flame, being the positively stretched flamelet towards the unburned mixture. We also make some quantitative comparisons between the turbulent flame speed correlation results obtained using the maximum local burning velocity and one based on the unstretched laminar flame speed to elucidate the Lewis number effects.

2. Experimental details

Experimental set up and procedures implemented in flame front tracking are briefed below. For further details on experimental set up, procedure and methodology, see (Kido, Nakahara et al. 2002). The combustion chamber used was a constant volume near-spherical chamber with an inner diameter of 100 mm. Premixed mixtures were ignited near the central region of the vessel under the desired turbulence intensity. The measured longitudinal integral length scale l_x was 3 mm for all investigated flames. The turbulent flame speed S_T reported was calculated from pressure-time data using the standard approach $S_T/S_{L0} = (\mathrm{d}p/\mathrm{d}t)_T/(\mathrm{d}p/\mathrm{d}t)_{L0}$. All CH_4–H_2 and

C_3H_8–H_2 mixtures were prepared such that S_{L0} was similar for all investigated mixtures with about 25 cm/s, by varying N_2 content (Table 1).

For CH_4–H_2 mixtures, the Lewis number *Le* is between 0.38 and 0.89, while for C_3H_8 – H_2, maximum *Le* value is 1.59. *Le* is the ratio of thermal diffusivity to mass diffusivity, and the preferential diffusion is the ratio of mass diffusivities of deficient fuel and oxidant in the mixture. Comparison of such combination of two dual-fuel mixtures yields the impact of the preferential-thermo-diffusion. Two examples of turbulent propagating flames are shown in Figure 1. Importantly, Kido et al. (Kido, Nakahara et al. 2002) estimated the mean local burning velocity S_L (included with preferential diffusion) for CH_4–H_2 and C_3H_8 –H_2 mixtures, see Figure 2. The major experimental finding by Kido et al. *(Kido, Nakahara et al. 2002)* was that the S_T values of C_3H_8 –H_2 mixtures are substantially less than those of CH_4–H_2 mixtures, for identical turbulent velocity and unstretched laminar flame speed S_{L0} (see Table 2 and experimental data in Figs. 3-4).

Mixture	Equi. ratio ϕ	Effective *Le*	Molar fraction					S_{L0} cm/s	α_0 mm^2/s	ν_0 mm^2/s
			CH_4	C_3H_8	H_2	O_2	N_2			
CH_4 - H_2										
H - a 0 0	0.8	0.38	1.00	--	0.00	2.50	9.63	25.30	21.05	15.55
H - a 0 2	0.8	0.45	0.80		0.20	2.13	9.56	25.01	22.12	15.75
H - a 0 5	0.8	0.53	0.50		0.50	1.56	7.66	24.95	24.47	16.23
H - a 0 8	0.8	0.70	0.20		0.80	1.00	6.50	25.40	27.81	16.90
H - a 1 0	0.8	0.89	0.00		1.00	0.63	6.25	25.25	30.35	17.40
C_3H_8 - H_2										
H - a 0 0	0.8	0.42	--	1.00	0.00	6.25	25.00	25.40	19.47	14.66
H - a 0 2	0.8	0.55		0.80	0.20	5.13	21.78	25.25	20.04	14.80
H - a 0 5	0.8	0.70		0.50	0.50	3.44	15.13	25.47	21.41	15.13
H - a 0 8	0.8	1.30		0.20	0.80	1.75	9.28	24.74	24.73	15.94
H - a 1 0	0.8	1.57		0.00	1.00	0.63	6.25	25.25	30.35	17.40

Table 1: Properties of premixed mixtures. α_0 and ν_0 are thermal diffusivity and kinematic viscosity (Kido, Nakahara et al. 2002). Numbers indicate doping concentration x 10 in vol%. Example: H-a08: major component H_2 with 20 vol% CH_4.

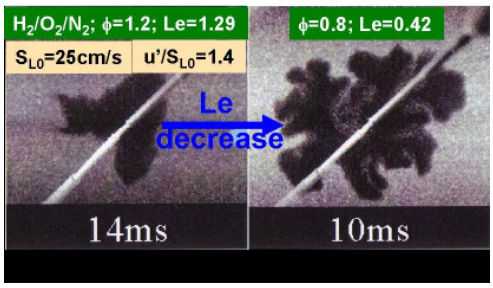

Fig. 1. Experimentally obtained sequential tomograms of propagating flames by Nakahara (Nakahara 2006). The cusps increase with decrease in the Lewis number, under identical conditions.

Results and Discussion

The aim of this work is the validation of theoretical models for large experimental flame data. For that the algebraic flame surface wrinkling model (AFSW model) developed by Muppala et al. (Muppala, Aluri et al. 2005) is used as the basic approach.

The model predictions of S_T have been derived in three modelling scenarios, using:

(1) unstretched laminar flame speed S_{L0} in the basic reaction closure without the preferential-thermo-diffusive instabilities included,

(2) laminar flame speed replaced by mean local burning velocity S_L to predict the preferential diffusion (for more details, see (Nakahara and Kido 1998))

(3) S_{L0} along with exponential *Le* term that is derived from the flame-ball concept of Zel'dovich, to investigate the contribution of the Lewis number effect (for detailed description, see (Muppala, Aluri et al. 2005)),

Comparisons of the predictions are made with the corresponding Kido database on expanding spherical turbulent premixed flames.

In the first scenario, the basic form of the algebraic flame surface wrinkling reaction sub-model is used, which has been found by Muppala et al. for lean CH_4/air flames (Muppala, Aluri et al. 2005).

$$\frac{A_T}{\bar{A}} \approx \frac{S_T}{S_{L0}} = 1 + 0.46\,\mathrm{Re}_t^{0.25}\left(\frac{u'}{S_{L0}}\right)^{0.3} \tag{3}$$

where A_T is the turbulent flame surface area, $\bar{A}$ is the averaged surface area, S_{L0} is the unstretched laminar flame speed, Re_t is the turbulent Reynolds number (= $u'l_x/\nu$). Here, u' is the rms turbulent velocity, l_x the longitudinal integral length scale, and ν the molecular kinematic viscosity, S_T is the turbulent flame speed. The turbulence and flame quantities used in Eq. 3 are derived from the experimentally determined values.

H - a 0 0	*H_2 doped with CH_4,*			*H - a 0 0*	*H_2 doped with C_3H_8*		
u'(m/s)	S_T	l_x	Re_t	u'(m/s)	S_T	l_x	Re_t
0.00	25.3	0.00	0	0.00	25.4	0.00	0
0.49	99.1	2.98	840	0.49	92.0	2.98	84
0.98	148.7	3.37	190	0.98	148.2	3.37	190
1.47	189.0	3.51	297	1.47	190.2	3.51	297
1.97	224.8	3.54	400	1.97	226.9	3.54	400
H-a02	80% H_2 + 20% CH_4			*H-a02*	80% H_2 + 20% C_3H_8		
u'(m/s)	S_T	l_x	Re_t	u'(m/s)	S_T	l_x	Re_t
0.00	25.4	0.00	0	0.00	24.7	0.00	0
0.49	98.0	2.98	87	0.49	83.8	2.98	92
0.98	145.2	3.37	196	0.98	142.4	3.37	208
1.47	185.2	3.51	306	1.47	186.5	3.51	324
1.96	220.9	3.54	411	1.96	222.5	3.54	436
H-a05	50% H_2 + 50% CH_4			*H-a05*	50% H_2 + 50% C_3H_8		
u'(m/s)	S_T	l_x	Re_t	u'(m/s)	S_T	l_x	Re_t
0.00	25.0	0.00	0	0.00	25.5	0.00	0
0.49	84.1	2.98	90	0.49	69.8	2.98	97
0.98	134.2	3.37	204	0.98	109.3	3.37	218
1.47	175.5	3.51	318	1.47	138.0	3.51	341
1.96	207.5	3.54	428	1.96	161.8	3.54	459
H-a08	20% H_2 + 80% CH_4			*H-a08*			
u'(m/s)	S_T	l_x	Re_t	u'(m/s)	S_T	l_x	Re_t
0.00	25.0	0.00	0	0.00	25.3	0.00	0
0.49	70.5	2.98	93	0.49	56.2	2.98	99
0.98	109.1	3.37	210	0.98	84.0	3.37	223
1.47	138.7	3.51	328	1.47	103.1	3.51	349
1.96	170.4	3.54	441	1.96	122.7	3.54	469
H - a 1 0	Pure CH_4			*H - a 1 0*	Pure C_3H_8,		
u'(m/s)	S_T	l_x	Re_t	u'(m/s)	S_T	l_x	Re_t
0	25.3	0.00	0	0	25.4	0.00	0
0.49	70.1	2.98	94	0.49	52.5	2.98	100
0.98	105.8	3.37	212	0.98	75.7	3.37	225
1.47	135.7	3.51	332	1.47	95.3	3.51	352
1.96	158.6	3.54	446	1.96	102.0	3.54	473

Table 2: Experimental expanding spherical turbulent flame data of Kido et al (Kido, Nakahara et al. 2002). Left columns: H_2 doped with CH_4. Right columns: H_2 doped with C_3H_8. Overall equivalence ratio of both CH_4 and C_3H_8 mixtures is 0.8. Numbers indicate doping concentration x 10 in vol%. Example: H-a08: major component H_2 with 20 vol% CH_4. u' is the rms turbulent velocity in m/s, S_T is the turbulent flame speed cm/s, l_x is longitudinal integral length scale in mm, and Re_t is Reynolds number = $l_x\, u'/\nu$, where ν is molecular kinematic viscosity.

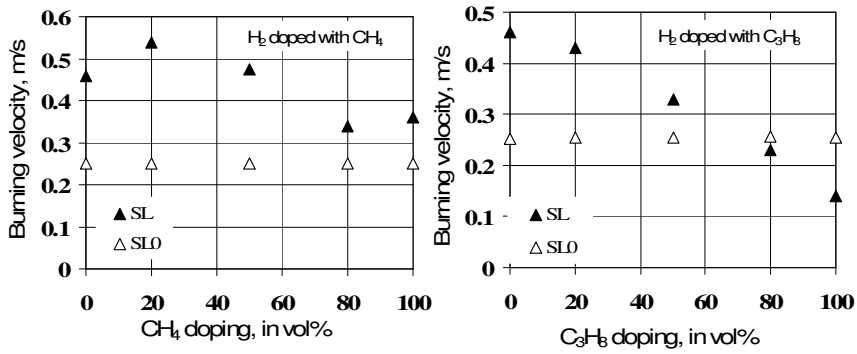

Fig. 2. Mean local burning velocity embedded with preferential diffusion, S_L, in comparison with the unstretched laminar flame speed, S_{L0}: a) H_2, doped with CH_4, b) H_2 doped with C_3H_8. Both speeds were obtained experimentally by Kido et al. (Kido, Nakahara et al. 2002).

Comparative study based on the global quantity S_T shows that the model predictions are close to the experiments for pure CH_4 (Fig. 3a) or C_3H_8 (Fig. 3b) flames, but the differences rise with increase of H_2 content. These large differences are because the model does not account for the preferential and thermo-diffusive effects. Here, the deviation is stronger for higher levels of turbulence, showing that these two effects are evidently visible at higher levels of turbulence.

In the second model scenario, S_{L0} in the basic model is replaced with S_L (Eq. 4).

$$\frac{S_T}{S_L} = 1 + 0.46\,\mathrm{Re}_t^{0.25}\left(\frac{u'}{S_L}\right)^{0.3} \tag{4}$$

Here, S_L is the mean local burning velocity, being an experimentally determined quantity, evaluated near the convex part of the flamelet pointing towards the unburned mixture (Nakahara and Kido 1998). The trick of this approach is that in this quantity the preferential diffusion effects are already included due to the experimental situation.

Note, that S_L is commonly defined as strained laminar flame speed, being modelled with a Markstein number Ma, according to

$$S_{L0}/S_L = 1 + Ma\, Ka_t \tag{5}$$

where, Ka_t is the turbulent Karlovitz number. However, in the following the directly determined laminar flame speed S_L is used.

In the third model scenario, an amended version of the reaction closure with the inclusion of the exponential Lewis number term to exclusively account for the Lewis number effects is applied (Eq. 6).

$$\frac{S_T}{S_{L0}} = 1 + \frac{0.46}{e^{(Le-1)}}\,\mathrm{Re}_t^{0.25}\left(\frac{u'}{S_{L0}}\right)^{0.3} \tag{6}$$

where, the effective Lewis number is Le of the dual fuel mixtures.

In the Figures 4a and 4b the results from the modelling scenarios (2) and (3) are shown together with the experimentally determined S_T. As can be seen here, both modelling approaches are fitting better to the different experimental situations than that based on Eq. 3. This finding indicates that molecular diffusive effects are of importance for the description of turbulent flames even for increasing turbulence intensities. These molecular transport effects, in the first order, may be described either by an effective Lewis number and the corresponding model (Eq. 6) where the Lewis number is included, or by an local burning velocity (Eq. 4). With the addition of CH_4 to H_2, a decreasing trend in S_T was observed, while this fall is relatively small as

against addition of C_3H_8, under identical conditions. As is expected, the difference between S_T values of pure fuels in case of $C_3H_8 - H_2$ was huge and it is as much as 140 cm/s. For lighter fuel mixtures of CH_4–H_2 , this difference is relatively low, 75 cm/s. Additional observations made from Figs. 3 and 4 include, influence of addition of 20 vol% of C_3H_8 (or CH_4) to H_2 is very little on S_T. While doping with 50 vol% of C_3H_8 showed substantial decrease in S_T, in the other case only a nominal decrease was observed. This phenomenon of falling trends is a result of the different molecular transport effects. Moreover, the impact of C_3H_8 for composition of 20 vol% in H_2 mixture has little impact on S_T, as is visible in Figs. 4a & 4b. For the case of C_3H_8 –H_2 mixtures, the mean S_L decreases linearly with addition of C_3H_8. A higher value of S_L was noticed in lighter CH_4–H_2 combination and the maximum S_L differs by 10 cm/s with the corresponding C_3H_8–H_2 mixture. Interestingly, doping effect indicates that for 80% H_2 and 20 vol% CH_4 projects highest S_L.

For lean lighter mixtures with Le<1, the influences of Le and preferential diffusion are equally important. For example, for CH_4–H_2 mixtures, the diffusivity of H_2 (or CH_4) is greater than that of oxidant such that lighter component diffuses and accumulates more towards the convex part of the flamelet facing the unburned mixture. The S_L is a strong function of characteristic time scale of molecular transport due to preferential diffusion. This process causes local virtual increase in equivalence ratio at the leading edge of the flamelet, thus increasing the mean S_L of the lean mixture (Le<1).

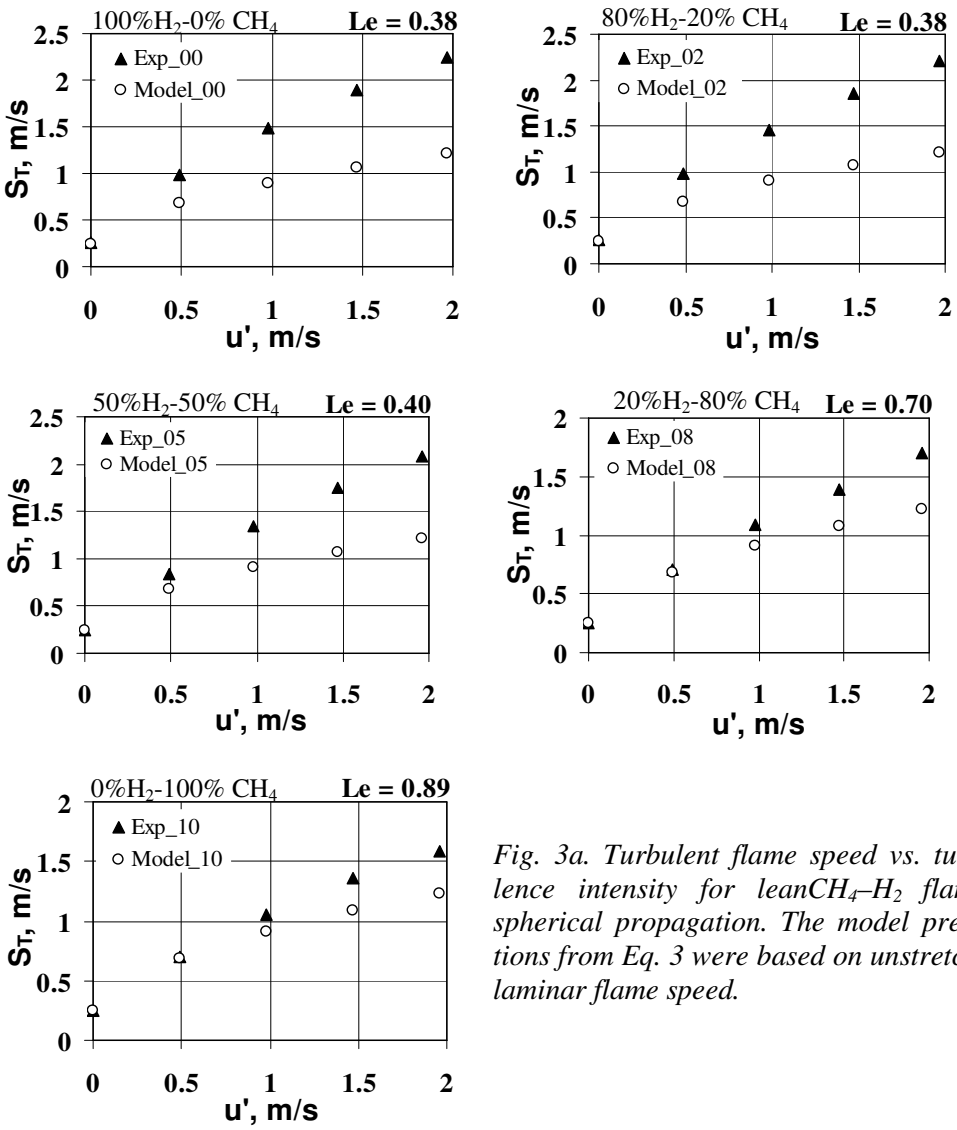

Fig. 3a. Turbulent flame speed vs. turbulence intensity for leanCH_4–H_2 flames, spherical propagation. The model predictions from Eq. 3 were based on unstretched laminar flame speed.

The model predictions using Eqs. 4 and 6 are combinedly shown in Figure. 4a. The correlation data from Eqs. 4 and 6 are in very good quantitative agreement with the corresponding measured data. The correctness of the exponential *Le* term in the S_T model is proved satisfactory for all the single and dual fuel mixtures studied. Inclusion of the *Le* term is especially beneficial if the data for an effective S_L value is not available. The model predictions from Eqs. 4 and 6 are in very good quantitative agreement with the corresponding measured data. Comparison between figures 4a and 4b shows that decrease of S_T is more predominant with addition of C_3H_8 to H_2 than using lighter CH_4. This near-linear decreasing trend in S_T is a result of thermo-diffusive effects that restrain the flame instabilities.

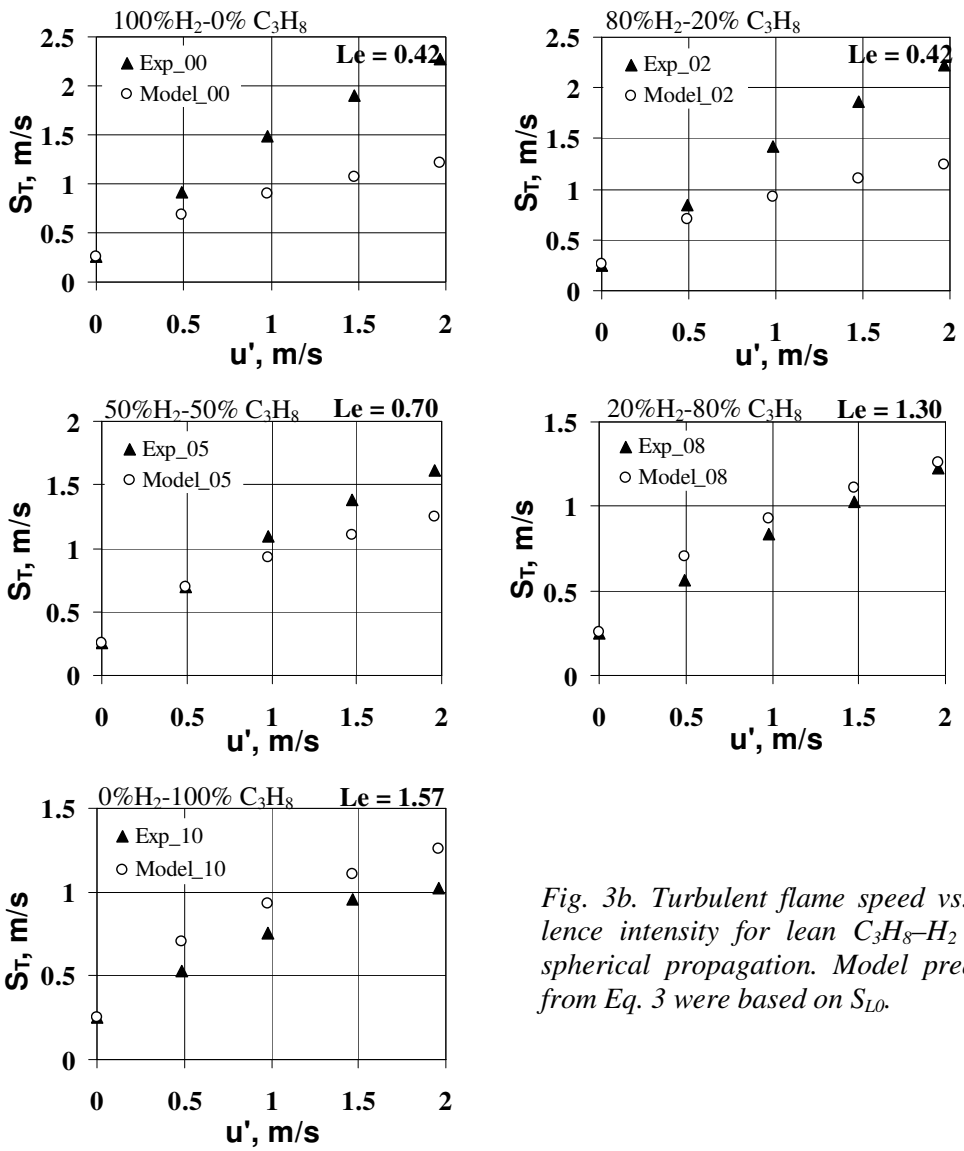

Fig. 3b. Turbulent flame speed vs. turbulence intensity for lean C_3H_8–H_2 flames, spherical propagation. Model predictions from Eq. 3 were based on S_{L0}.

Correlation plots

The correlation plots of turbulent flame speed for both CH_4 –H_2, and C_3H_8 –H_2 mixtures shown in figure 5 shows that experiment and model predictions are in very good agreement with each other. The analytical results are obtained based on the mean local burning velocity instead of the unstretched laminar flame speed, using Eq. 3. Thus, it is shown that the reaction model with suitable modifications is able to reproduce the experimental S_T trends for all the studied mixtures.

Implication for hydrogen flame safety

The high local burning velocities S_L, in spherical hydrogen flames with *Le* much less than unity are due to transient processes, such as the instability of the adiabatic flame ball. Addition of C_3H_8 causes the smaller maximum growth rate and smaller wave number at this rate of the propagating flame resulting in decrease of S_L, and hence in S_T. This decrease in flame speed results in narrowing of the detonability range of pure H_2 mixtures. It has long been understood that the deflagration to detonation transition is a likely phenomenon especially with highly reactive H_2 mixtures. Theoretically, this decrease in flame speed may be quantitatively explained considering the structure of leading kernels of free propagating spherical flames which is known to increase the scale of the convex part of the flamelet propagating rapidly. These leading kernels depend substantially on preferential-thermo-diffusive effects. For less-than-unity Lewis number mixtures with diffusivity of deficient reactant greater than that of the excess reactant, the strongest known perturbations of laminar flames are associated with the critically curved spherical kernel. On other the other hand, Law and Kwon (Law and Kwon 2004) have experimentally demonstrated that the flame instabilities associated with these effects is a strong function of thermal flame thickness. That is, with the addition of C_3H_8 to H_2 mixtures, the increase in flame thickness diminishes the flame instabilities, with the suppression of the formation of hydrodynamic (diffusional-thermal) cells.

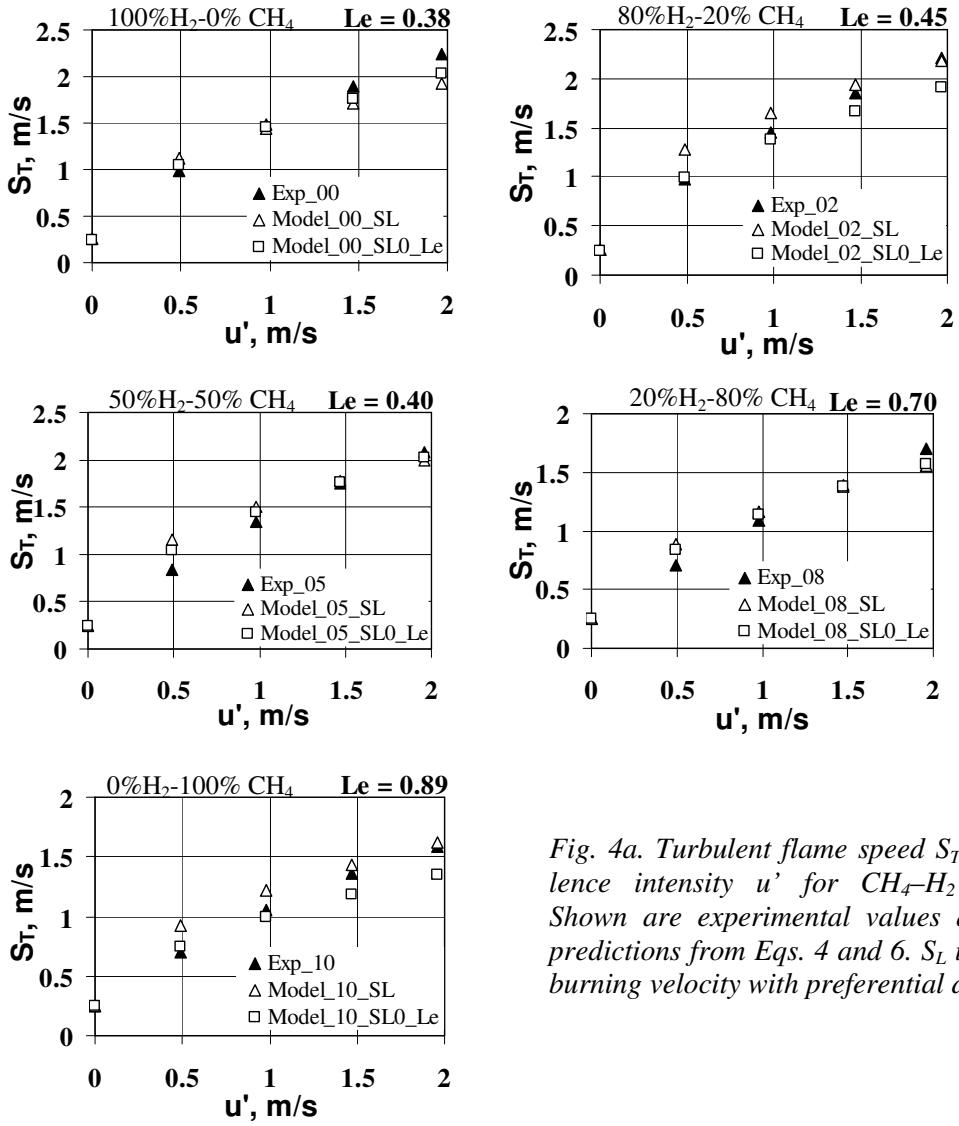

Fig. 4a. Turbulent flame speed S_T vs. turbulence intensity u' for CH_4–H_2 mixtures. Shown are experimental values and model predictions from Eqs. 4 and 6. S_L is the local burning velocity with preferential diffusion.

Our analytical studies based on the AFSW model show a general decreasing trend of flame speeds for hydrocarbon doped H_2 mixtures. These trends indicate that doping is a promising prevention method of deflagration to detonation transition, which is mostly caused by enhancement in flame front propagation. While the Lewis numbers of lean pure CH_4 and H_2 mixtures does not differ significantly; it is believed that CH_4 as a minor constituent will have lesser impact on preferential-thermo-diffusive instabilities. To address the issues of H_2 safety, it will be interesting to numerically quantify the propagation of H_2 flames in real geometries, ducts and narrow spaces. For this, a submodel for chemical time scale that characterizes the leading points based on the flame ball concept may be applicable. (Lipatnikov and Chomiak 2005). This time scale is usually obtained from simulation of critically curved laminar flames invoking a detailed chemistry and is given as an input parameter to model turbulent combustion.

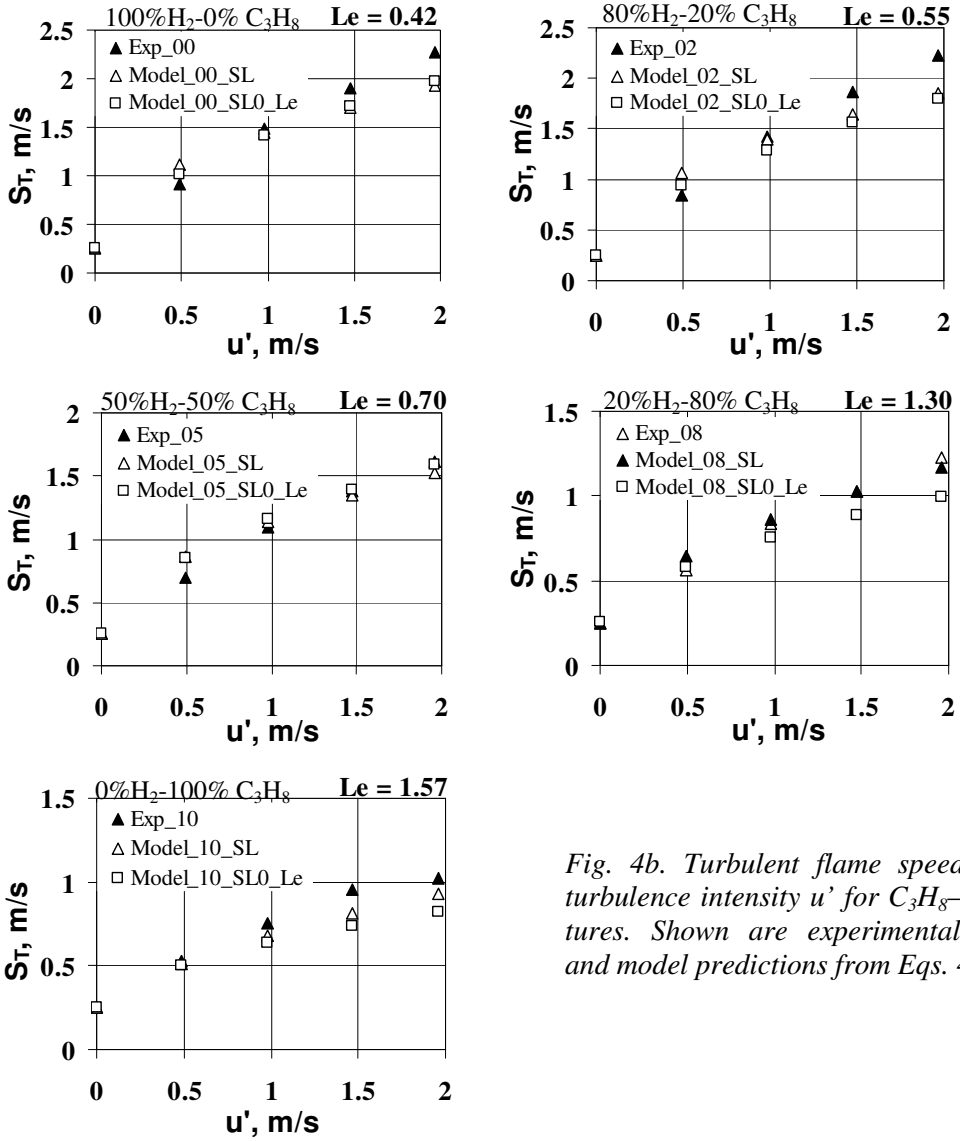

Fig. 4b. Turbulent flame speed S_T vs. turbulence intensity u' for C_3H_8–H_2 mixtures. Shown are experimental values and model predictions from Eqs. 4 and 6.

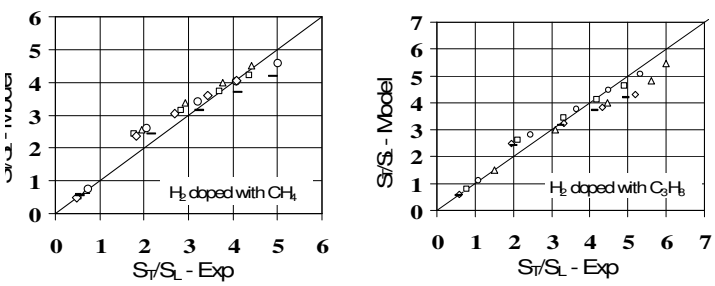

Fig. 5. Correlation plot of experimentally measured vs. model predicted S_T, estimated based on S_L,(Model Eq. 4) for CH_4–H_2 and C_3H_8–H_2 mixtures.

Conclusions

An existing algebraic flame surface wrinkling reaction model was used to investigate the quantitative dependence of turbulent flame speed on molecular transport coefficients for two-component lean fuel (CH_4–H_2 and C_3H_8–H_2) mixtures.

The model predictions were in good quantitative agreement with the corresponding experiments, if either an effective laminar flame speed S_L is applied in the reaction model (Eq. 5), or an exponential Lewis number term of the fuel mixture is used (Eq. 7). The latter approach is a generalisation of earlier findings for single fuels and shows the applicability of the exponential *Le* term in the reaction subclosure model for the mixtures studied.

The hydrocarbon substitutions to H_2 mixtures are expected to suppress the leading flame edges, which are manifested by a decrease in mean local burning velocity, eventually preventing transition to detonation. Addition of hydrocarbons may also promote flame front stability of lean turbulent premixed H_2 flames.

Muppala acknowledges Masaya Nakahara (Kyushu University, Japan) for providing experimental data. Suggestions and insightful inputs by Andrei Lipatnikov (Chalmers University of Technology, Sweden) are greatly acknowledged.

References

Aluri, N. K., S. P. R. Muppala, et al. (2006). "Substantiating a Fractal-based Algebraic Reaction Closure of Premixed Turbulent Combustion for High-Pressure and the Lewis Number Effects." Combust. and Flame **145**: 663-674.

Chen, Y.-C. and R. W. Bilger (2002). "Experimental investigation of three-dimensional flame-front structure in premixed turbulent combustion--I: hydrocarbon/air bunsen flames." Combust. and Flame **131**(4): 400-435.

Chen, Y.-C. and R. W. Bilger (2004). "Experimental investigation of three-dimensional flame-front structure in premixed turbulent combustion: II. Lean hydrogen/air Bunsen flames." Combust. and Flame **138**(1-2): 155-174.

Halter, F., C. Chauveau, et al. (2007). "Characterization of the effects of hydrogen addition in premixed methane/air flames." International Journal of Hydrogen Energy (**in press**).

Kido, H., M. Nakahara, et al. (2002). "Turbulent Burning Velocities of Two-component Fuel Mixtures of Methane, Propane and Hydrogen." JSME International Journal, Series B **45**(2).

Kido, H., M. Nakahara, et al. (2005). "A Study on the Local Flame Displacement Velocity of Premixed Turbulent Flames." JSME International Journal Series B **48**(1): 164-171.

Law, C. K., G. Jomaas, et al. (2005). "Cellular instabilities of expanding hydrogen/propane spherical flames at elevated pressure: theort and experiment." Proceedings of the Combustion Institute **30**: 159-167.

Law, C. K. and O. C. Kwon (2004). "Effects of hydrocarbon substitution on atmospheric hydrogen-air flame propagation." International Journal of Hydrogen Energy **29**: 876-879.

Lipatnikov, A. N. and J. Chomiak (2005). "Molecular transport effects on turbulent flame propagation and structure." Progress in Energy and Combustion Science **31**: 1-71.

Muppala, S. P. R., N. K. Aluri, et al. (2005). "Development of an algebraic reaction rate closure for the numerical calculation of turbulent premixed methane, ethylene and propane/air flames for pressures up to 1.0 MPa." Combust. and Flame **140**: 257-266.

Muppala, S. P. R. and M. V. Papalexandris (2006). A Modelling Approach for Hydrogen-doped Lean Premixed Turbulent Combustion. ASME Congress, Chicago.

Nakahara, M. (2006). "Personal Communication".

Nakahara, M. and H. Kido (1998). "A Study of the Premixed Turbulent Combustion Mechanism Taking the Preferential Diffusion Effect into Consideration." Memoirs of the Faculty of Engineering, Kyushu University **58**(2).