Conference or Workshop Item:

Al-Mughanam, T, Bradley, D, Lawes, M et al. (1 more author) (Accepted: 2015) Burning Velocity Blending Laws for Methane/Air and Hydrogen/Air Blends. In: 25th International Colloquium on the Dynamics of Explosions and Reactive systems, 2015, 02-07 Aug 2015, Leeds, UK.

Burning Velocity Blending Laws for Methane/Air and Hydrogen/Air Blends

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1 Introduction

There is increasing interest in supplementing natural gas supplies with the addition of hydrogen. This has led to a number of experimental [1] and chemical kinetic [2] studies of the burning velocities, u_l, of such blends with air. The interest in this extends beyond these practicalities to the general problem of deriving satisfactory blending laws for fuels with very different chemical kinetics and burning velocities. The present paper explores the application of six different laws for predicting the burning velocities of blends of H_2/air and CH_4/air, with the same equivalence ratio, ϕ.

An early prediction law is that the blend burning velocity is the sum of the products of the mole fraction of each component mixture and its burning velocity [3]. Another law is based on an observed correlation between the heat of reaction of one mole of mixture, Q, and u_l, but is restrictively confined to fuels in the same family. This provides an approximately linear reltionship, for each separate family of fuels [4].

Spalding [5-7] adopted a fractional mass, rather than mole, weighting of burning velocities. He also took into account the differences in heat release rate between mixtures that arise from their different adiabatic burned gas temperatures, T_b. However, the differences in T_b between H_2/air and CH_4/air mixtures in the present study proved too excessive for this approach. Fractional mixture mass weightings for u_l were employed in [8]. The importance of the heat release profile through the flame became clear from Spalding's seminal analytical expressions for u_l, given below for a Lewis number of unity:

$$u_l = \left[\frac{k_u \int_0^1 R(c)\,dc}{\lambda (T_b - T_u) \bar{c}_p^2 \rho_u^2} \right]^{0.5} . \tag{1}$$

Here k_u is the thermal conductivity of unburned gas, T_u, the initial temperature, $\bar{c}_p$, the mean specific heat, and ρ_u the density of unburned gas. With a reaction progress variable, c, given by the fractional temperature rise, the heat release rate source term is $R(c) = (k/k_u) H \dot{m}_A$, with H the heat of reaction of the fuel, A, and $\dot{m}_A$ its mass volumetric rate of burning. λ is the burning velocity

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eigenvalue $= k_u \bar{R}/(T_b - T_u)c_p^2 G$, where $\bar{R}$ is the area under the $R(c)$ curve and G the mass flow rate per unit area. The value of λ is related by an algebraic expression to the value of c at the centroid of the heat release rate profile plotted against c.

Chemical kinetic modelling in [4] showed that with H_2, in comparison with those of CH_4, the peak in the flame heat release moves towards lower values of c, as a consequence of enhanced molecular transport and low temperature reaction, both attributable to H atoms, with a resulting increase in u_l. The kinetic modelling in [2] shows how the blend u_l of H_2/CH_4 flames increases with the concentration of H.

In terms of Eq. (1), as the maximum of the profile moves to lower values of c, λ decreases with the centroid distance, and u_l consequently increases. A correlation was noted in [4] betweeen the centroid distance, and hence the influence of the eigenvalue on u_l, with the product Qu_l. This implies the value of u_l is dependent not only upon Q, but also upon the profile of the heat release rate through the flame, suggesting a blending law in terms of Qu_l rather than of Q. Values of u_l for the component mixtures were plotted against Qu_l. As Q is known for both the component mixtures and the blend, the value of u_l for the blend can be found. However, it was found in the course of the present study that more satisfactory predictions were obtained with values of Q based on one mole of H_2/CH_4 fuel, rather than on one mole of fuel/air mixture.

The high activation energy, asymptotic, assumption [9], that all the heat release occurs at T_b, results in a value of λ of 0.5 and a single value of volumetric heat release rate, q_{max}, at c=1. Equation (1) then becomes:

$$u_l = \left[\frac{2k_u q_{max}}{Z(T_b - T_u)\bar{c}_p^2 \rho_u^2}\right]^{0.5}, \qquad (2)$$

where Z is the Zel'dovich number, $= (T_a/T_b^2)(T_b - T_u)$, with T_a the activation temperature for the heat release. If S(1) represents the product of reactant concentrations, Arrhenius "A" values, heat of reaction, k/k_u, and any effect of non-unity Lewis number, Le, at c=1, then. It can be shown that,

$$u_l \rho_u = \exp(-T_{al}/2T_b), \text{ where} \qquad (3)$$

$$T_{al} = T_a - 2T_b \ln\left(\frac{2k_m S(1)}{Z(T_b - T_u)\bar{c}_p^2}\right)^{1/2}. \qquad (4)$$

Here T_{al} is an activation temperature for the laminar burning mass flux, dependent upon pressure, P, Le, $(T_b\text{-}T_u)$, and the variation of q with c. It follows that:

$$2\ln u_l \rho_u = -T_{al}/T_b, \text{ and } \frac{d\,2\ln u_l \rho_u}{d\,1/T_b} = -T_{al}, \qquad (5)$$

a frequently used expression, extending beyond asymptotic analyses [10,11]. Hirasawa et al. [12] employed a slightly different activation temperature, $\tilde{T}_a$, based on u_l alone rather than $u_l \rho_u$, with:

$$u_l = \exp(-\tilde{T}_a/T_b) \text{ or } \ln u_l = -\tilde{T}_a/T_b. \qquad (6)$$

This expression provides the basis, in [12], of another blending law, based on $\tilde{T}_a$. Values of u_l and T_b are known for each of the comonent fuel/air mixures to be blended, enabling the values of $\tilde{T}_a$ to be found. These are weighted by their respective mole fractions for each component mixture to give $\tilde{T}_a$

for the blend, from which u_l can be found from Eq. (6).

2 Mixture Composition Data and the Blending Laws

Burning velocities of blended component mixtures are presented, for a given ϕ as a function of α, the fraction of H_2 moles in one mole of the fuel, comprised of H_2 and CH_4. It is readily shown that for one mole of fuel the overall composition of the mixture is:

$$\alpha H_2 + (1 - \alpha)CH_4 + (1/0.42\phi)[\alpha + 4(1 - \alpha)] \text{ air} \tag{7}$$

The fraction of (H_2 + air) moles at ϕ in the blend, β, is:

$$\beta = \alpha[1 + 1/(0.42\phi)] \text{ x } \left[\alpha[1 + 1/(0.42\phi)] + (1 - \alpha)(1 + 4/(0.42\phi))\right]^{-1} \tag{8}$$

A further blending law, based on Le Chatelier’s rule and applied specifically to H_2/air and CH_4/air blends in [2], gives a blend u_l of:

$$[\alpha/u_{l\,H2} + (1 - \alpha)/u_{lCH4}]^{-1}. \tag{9}$$

Evaluation of Q, the heat of reaction for one mole of a H_2/CH_4 fuel mixture is found from:

$$Q = \sum_P n(h_f - \Delta h) - \sum_R n(h_f - \Delta h). \tag{10}$$

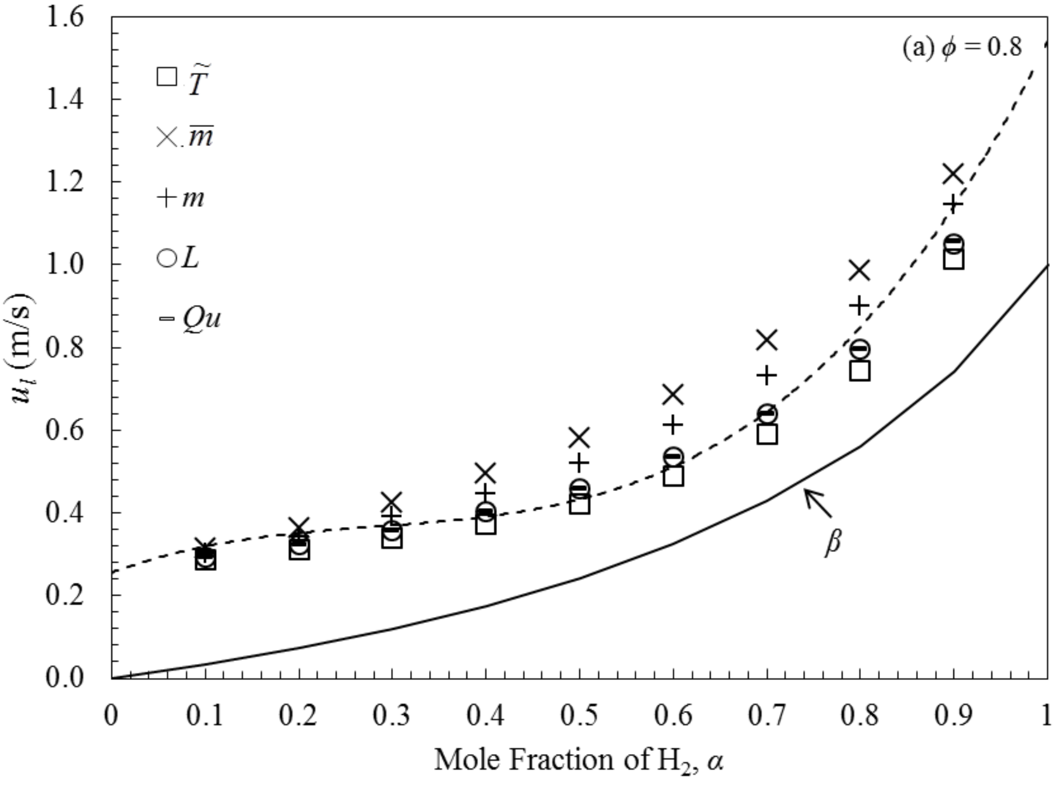
where subscripts R and P refer to reactants and products respectively, n is the number of moles given by Eq. (7), h_f the enthalpy of formation at the standard state conditions of 298K and 0.1 MPa, and Δh the sensible enthalpy. Equilibrium products of combustion at constant pressure and T_b, h_f and Δh are found from the GasEq code [13]. The different blending laws that are scrutinised in the present study are summarised in Table 1.

Table 1: Summary of blending laws investigated.

Blending Law	Reference	Symbol
Fractional mole concentration of each component mixture	Payman and Wheeler [3]	$\overline{m}$
Fractional mass concentration of each component mixture	van Lipzig et al. [8]	m
Q per mole of component mixture	Bradley et al. [4]	Q
u_l plotted against Qu_l per mole of fuel	Present work	Qu
Fractional mole-weighted values of $\tilde{T}_a$ for component mixtures.	Hirasawa et al. [12]	$\tilde{T}$
Eq. (9)	Di Sarli et al. [2]	L

3 Predictions of Blending Laws

Predicted values of u_l for blends of H_2/air and CH_4/air at the same ϕ are compared below with experimentally measured values of u_l of Hu et al. [1], in an explosion bomb, at 303K and 0.1MPa, with ϕ values beween 0.6 and 1.3, and α betweeen 0 and 1.0. Predicted and measured values are shown in Figs. 1 (a) and (b) for ϕ=0.8 and 1.1. Measured values, with maximum errors of 8.6% are shown by the dashed curves, while the solid curves show the way in which β, varies with α. Predicted values are shown by the symbols for the various blending laws. Table 2 summarises the ratios of predicted to measured burning velocities, $u_{lp}/u_{l,}$ for the different blending laws, for ϕ=0.6, 0.8 and 1.1, as a function of α.



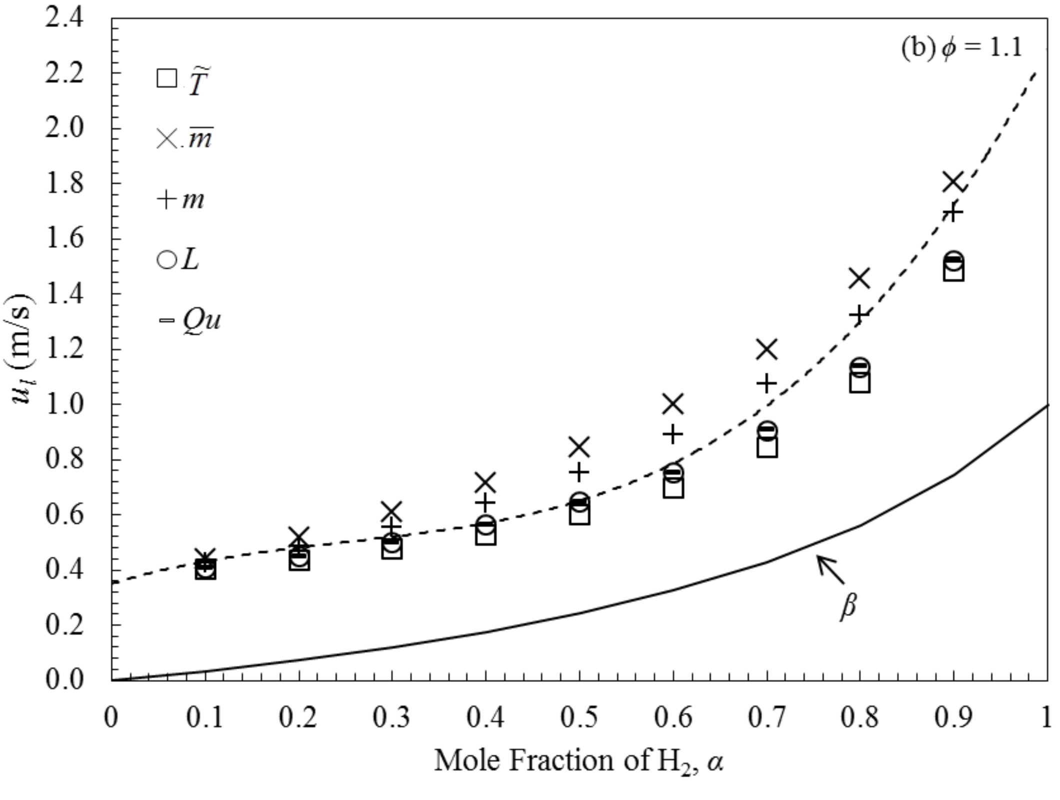


Figure 1. Predicted laminar burning velocities of blended CH_4/air and H_2/air for different mole fractions, α, of H_2 in the CH_4/H_2 fuel mixture. Dashed curve show measured values from Hu et al. [1], solid curve shows variation of β. (a) ϕ=0.8, (b) ϕ=1.1.

Table 2: Ratio of predicted to measured burning velocities, $u_{lp}/u_{l,}$ for different blending laws, for ϕ=0.6, 0.8 and 1.1, as a function of α.

α	$\overline{\mathrm{m}}$			m			Qu			$\tilde{\mathrm{T}}$			L		
	0.6	0.8	1.1	0.6	0.8	1.1	0.6	0.8	1.1	0.6	0.8	1.1	0.6	0.8	1.1
0.1	1.11	1.03	1.08	1.07	1.00	1.04	1.02	0.97	1.00	1.00	0.95	0.98	1.02	0.97	1.00
0.2	1.11	1.08	1.18	1.04	1.01	1.10	0.94	0.96	1.02	0.91	0.92	0.98	0.94	0.96	1.02
0.3	1.15	1.15	1.13	1.04	1.05	1.03	0.91	0.97	0.93	0.87	0.91	0.88	0.91	0.97	0.93
0.4	1.16	1.22	1.21	1.03	1.10	1.09	0.87	1.00	0.96	0.82	0.92	0.90	0.87	0.99	0.96
0.5	1.17	1.31	1.31	1.04	1.17	1.17	0.85	1.04	1.00	0.80	0.96	0.93	0.85	1.04	1.00
0.6	1.28	1.29	1.16	1.13	1.15	1.03	0.91	1.01	0.87	0.86	0.93	0.81	0.91	1.01	0.87
0.7	1.23	1.32	1.29	1.10	1.18	1.15	0.88	1.03	0.97	0.84	0.95	0.91	0.88	1.03	0.97
0.8	1.22	1.17	1.13	1.10	1.07	1.03	0.90	0.95	0.88	0.87	0.88	0.84	0.90	0.94	0.88
0.9	1.15	1.06	1.04	1.08	1.00	0.98	0.93	0.92	0.88	0.93	0.88	0.86	0.93	0.91	0.88
Av.	1.18	1.18	1.17	1.07	1.08	1.07	0.91	0.98	0.95	0.88	0.92	0.90	0.91	0.98	0.95

Table 2 summarises the ratios of predicted to measured burning velocities, $u_{lp}/u_{l,}$ for the different blending laws, for ϕ values of 0.6, 0.8 and 1.1, as a function of α. Values of u_l for the component H_2/air and CH_4/air mixtures at the bracketed values of ϕ are 0.822 m/s (0.6), 1.546 m/s (0.8), 2.3 m/s (1.1) for H_2/air and 0.121 m/s (0.6), 0.27 m/s (0.8), 0.375 m/s (1.1), for CH_4/air.

4 Discussion

Not surprisingly, the predictions of the Q law proved to be the least satisfactory. They increasingly over-predicted the burning velocity as α increased to 0.5, by a factor of 2. This over-prediction arises from the much higher values of u_l for a given Q, for H_2 than for CH_4. Equation (1) shows the importance of the profiles of heat release rate through the flame and the eigenvalue and these correlate with Qu_l [4].

The bottom row of Table 2 shows average values over all values of α for the given ϕ. The $\overline{\mathrm{m}}$ and m laws consistently over-predict the blend velocity, while the $\tilde{\mathrm{T}}$ law consistently under-predicts. The Qu and L laws both perform well, and are surprisingly close in their predictions, but tend to under-predict for ϕ=0.6. The Le Chatelier based law gave good predictions of the computed lean and stoichiometric blend u_l values [2]. Quite large prediction errors became apparent for the richer mixture of ϕ=1.3 and α=0.8, with the Qu law giving a $u_{lp}/u_{l,}$ values as low as 0.59, possibly due to the onset of sooting reactions and departures from GasEq equilibrium.

Figure 1 shows the large range of the u_l blend values. This arises from the large difference in the datum end values of u_l for the pure H_2/air and CH_4/air components. When an intermediate calibrating blend was introduced at α=0.7, the three datum mixtures gave much improved predictions for the intermediate blends.

It was found that the value of Qu_l increased with α, in much the same way as β in Fig.1, while Q declined markedly. This is a consequence of the combination of a higher burning velocity of hydrogen and a lower molar heat of reaction. In practical terms, for molar fractions of H_2 up to 0.5, there is no marked increase in u_l, or in burner power as indicated by Qu_l. For higher values of there is a sharp increase in Qu_l, despite the decline in Q.

Acknowledgements

Erjiang Hu and Zuohua Huang are thanked for providing additional u_l data.

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